

**UNEDITED PAPERS**

**Proceedings of the Third International Symposium on the  
Protection of the Environment from Ionising Radiation (SPEIR 3)  
held in Darwin, Australia, 22–26 July 2002,  
and organized by  
the Supervising Scientist Division of Environment Australia  
and the Australian Radiation Protection and Nuclear Safety Agency  
in co-operation with the International Atomic Energy Agency**



# **Protection of the Environment from Ionising Radiation**

**The Development and Application of a System  
of Radiation Protection for the Environment**



**INTERNATIONAL ATOMIC ENERGY AGENCY**

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## FOREWORD

In recent years, awareness of the vulnerability of the environment has increased, as evidenced by new and developing international policies for environmental protection, starting with the Rio Declaration of 1992. In the context of ionizing radiation, the existing international approach is largely based on providing for the protection of humans, but this is being critically reviewed in several international fora. It is in this context that the *Third International Symposium on Protection of the Environment from Ionising Radiation* (SPEIR 3) was held between 22 and 26 July 2002, in Darwin, Australia.

The symposium focused on issues related to the development and application of a system of radiation protection for the environment. The symposium programme included sessions dedicated to: ongoing research on the effects, responses and mechanisms of the interactions of ionizing radiation with biota; policy and ethical dimensions of the development of a framework for environmental radiation protection; and the development and use of methods and models for evaluating radiation as a stressor to the environment. Three workshops were held to allow for detailed discussion of each of these subjects.

This symposium was the third in a series. The first International Symposium on Ionising Radiation: Protection of the Natural Environment, was held in Stockholm, Sweden, 20–24 May 1996. This symposium was organized jointly by the Swedish Radiation Protection Institute (SSI) and the Atomic Energy Control Board (AECB) of Canada, and the proceedings were published by the Akademityck AB, Edsbruk, Sweden in 1996. The second International Symposium on Ionizing Radiation: Environmental Protection Approaches for Nuclear Facilities, was held in Ottawa, Canada, 10–14 May 1999, and was organized by the Canadian Nuclear Safety Commission (CNSC), the Supervising Scientists Group of Environment Australia, and the Swedish Radiation Protection Institute (SSI). The proceedings were published in April 2001 by CNSC. This third symposium was organized by the Supervising Scientist Division of Environment Australia and the Australian Radiation Protection and Nuclear Safety Agency, in co-operation with the International Atomic Energy Agency, and supported by the following organizations:

Energy Resources Australia Limited, Australia.  
Canadian Nuclear Safety Commission, Canada.  
Radiation and Environmental Science Centre (RESC), Ireland.  
Swedish Radiation Protection Authority (SSI), Sweden.  
British Nuclear Fuel Limited (BNFL), United Kingdom.  
The Environment Agency, United Kingdom.  
The United States Department of Energy, United States of America.  
European Commission.

The theme of this symposium is closely related to the IAEA's work programme on the development of safety standards on the protection of the environment from the effects of ionizing radiation. The IAEA's programme also has the objective of fostering information exchange and establishing an international consensus on this issue, and its involvement in the organization of this symposium, and the publication of these proceedings, are examples of its activity in this regard. This work is continuing with preparations for the International Conference on the Protection of the Environment from the Effects of Ionizing Radiation, which will be held in Stockholm, Sweden, 6–10 October 2003. The responsible IAEA officer is C. Robinson of the Division of Radiation and Waste Safety.

## *EDITORIAL NOTE*

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# Symposium Opening Speech

## A. Johnston

Supervising Scientist Division, Environment Australia, Darwin, NT, Australia

Ladies and gentlemen, my name is Dr. Arthur Johnston, Supervising Scientist. Please join me in thanking Mr. Ash Dargan of the Larrakia Aboriginal people, once again, for his welcome to his country. I would also like to welcome you to Darwin and to the Third International Symposium on the Protection of the Environment from Ionising Radiation, or SPEIR 3 as we have come to know it.

This Symposium would not have been possible if not for the hard work of the International Organising Committee and the Domestic Organising Committee. The membership of these committees is on the back page of the Symposium Program and I encourage you to take a moment during the next few days to take note of those individuals. In particular, it is appropriate that we recognise the extraordinary efforts of Ms. Sandie Devine who has done such a magnificent job over the past 18 months to make this Symposium happen.

SPEIR 3 has received excellent support from various organisations which must be acknowledged. The Domestic Organising Committee was drawn from the Supervising Scientist Division of Environment Australia and the Australian Radiation Protection and Nuclear Safety Agency. These Australian Federal Government Agencies organised the Symposium in co-operation with the International Atomic Energy Agency. I also wish to recognise the contributions of our sponsors who have provided considerable financial support. They are:

- The International Atomic Energy Agency;
- United States of America Department of Energy;
- Swedish Radiation Protection Authority;
- United Kingdom Environment Agency;
- British Nuclear Fuels Limited;
- Energy Resources of Australia Limited; and
- The Radiation and Environmental Science Centre of the Dublin Institute of Technology.

The Symposium was also supported by the the European Commission and the Canadian Nuclear Safety Commission.

But now to what lays before us. Over the next few days, 48 oral presentations will be made, 15 posters will be presented, and 3 workshops will be completed covering the broad topics:

- *Ionising Radiation and Biota: Effects, Responses and Mechanism;*
- *Frameworks for Environmental Radiation Protection; and*
- *Methods and Models for Evaluating Radiation as a Stressor to the Environment.*

Many conferences and symposia are proud to boast one internationally recognised keynote speaker. We have three of the highest order; Professor Ward Wicker of Colorado State University, Dr. Lars-Erik Holm, Director of the Swedish Radiation Protection Authority, and Professor Jan Pentreath from the University of Reading. In addition to delivering a keynote address, they have each agreed to act as session chairs and lead the Workshops on Day 4 – so we are certainly getting value for money! We also have about 100 delegates, from every corner of the globe representing, I dare say, a very significant proportion of the world's expertise in the emerging field of environmental radiation protection. So we have a recipe for a very successful Symposium. However we still need to combine the ingredients in the right way. The subtitle of the Symposium is "*The Development of a System of Radiation Protection for the Environment*". I ask that each of us focus on that throughout the next few days, and particularly during the workshops, as it is the ultimate goal behind this Symposium, and behind others that have preceded it and that will follow. The challenge is to make progress in identifying the important issues, defining what we know, don't know, and need to know, agreeing on where there is consensus and where there is not, and then close, even if only slightly, the gaps and uncertainties that emerge. I'm confident that we will succeed in meeting that challenge, and also have a lot of fun along the way. On that note, and with the big picture firmly in mind, I take great pleasure in opening the Third International Symposium on the Protection of the Environment from Ionising Radiation and invite Dr. Abel González, Director of the Division of Radiation and Waste Safety of the International Atomic Energy Agency, to discuss the Development of IAEA Policy on the Radiological Protection of the Environment.

# The development of IAEA policy on the radiological protection of the environment

A.J. González

Division of Radiation and Waste Safety, International Atomic Energy Agency, Vienna

**Abstract.** This paper was presented as an opening address of this symposium, on behalf of the International Atomic Energy Agency (the Agency). It comprised an overview of the Agency's responsibilities, related to environmental radiation protection; its historical involvement in this issue; the context of its current work programme; and a number of issues for further consideration.

## 1. INTRODUCTION

The Agency is the organization within the UN family with statutory functions in radiation safety. Its Statute requires the Agency '*...to establish ...standards of safety for protection of health and minimization of danger to life and property...*' [1]. In this context, the Agency is continuously working towards the construction of an international radiation safety regime, which includes legally binding conventions, a corpus of international standards, and provisions for their application. A hierarchy of safety standards exists in which: *Safety Fundamentals* present basic objectives, concepts and principles of safety and protection; *Safety Requirements* establish requirements that must be met to ensure safety, and *Safety Guides* recommend actions, conditions or procedures for meeting the safety requirements. The Agency also undertakes to provide for the application of these standards.

The Agency's current safety standards include Safety Fundamentals on *The Principles of Radioactive Waste Management* [2], which include the following principle: "*Radioactive waste shall be managed in such a way as to provide an acceptable level of protection of the environment*". This principle has also been effectively incorporated in *The Joint Convention On Safety Of Spent Fuel And Radioactive Waste Management* [3], which entered into force in the year 2000. The implications of these commitments on present and future Agency work are explored.

The development of international radiation safety standards is achieved through the interaction of a number of international organisations. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) traditionally provides estimates on the biological effects, attributable to radiation exposure, while the International Commission on Radiological Protection (ICRP) makes basic recommendations on radiation protection, which are incorporated into international radiation safety standards by the Agency, in cooperation with other specialized UN organizations, as appropriate.

## 2. HISTORICAL BACKGROUND

The Agency has a long history of involvement in the field of radiation protection from radioactive materials released into the environment. The wording of its Statute, which requires the '*minimization of danger to life and property...*' [1], may be interpreted as a reference to the '*environment*', as it would now be phrased. In 1958, the UN Conference on the Law of the Sea [4] recommended assigning responsibilities to the Agency for promulgating standards to prevent pollution due to radioactive materials. In 1963, the Agency issued the first international standards for radiation protection [5], and in 1967 revised them with the effect of implicitly affording protection of the environment [6].

In 1972, the Agency established the *definition* and *recommendations* for the London Convention, one of the first international undertakings for the protection of the sea [7]. In 1976, the Agency issued the first report on effects of ionizing radiation on aquatic organisms and ecosystems [8]. This was followed, in 1978, by the establishment of the first international standards for limiting discharges to the environment [9] and, in 1979, by the first international methodology for assessing impacts of radioactivity on aquatic systems [10].

In 1982 the new IAEA international radiation safety standards [11] were issued introducing the concept of the *dose commitment*, the use of which effectively allows for the build up of material in the environment, and thus acts to protect it. In 1982, the Agency issued the first international standards on generic models and parameters for assessing environmental transfer [12], in 1985 the first international standards for evaluating transboundary exposure [13] and, in 1985, the first international consensus document on  $K_{ds}$  in sediments and concentration factors in the marine environment [14].

A major milestone occurred in 1986 when the Agency issued new comprehensive standards for limiting discharges describing *in extenso* the concept of limiting discharges on the basis of dose commitment [15]. Also in 1986, the Agency issued a consensus report on chromosomal aberration analysis for dose assessment, which may have implications for the interpretation of dosimetric work on fauna and flora [16]. In 1988, the Agency issued a report for assessing the impact of deep sea disposal of low level radioactive waste on living marine resources [17]. In 1992 a report on effects of ionizing radiation on plants and animals at levels implied by current radiation protection standards [18] reviewed knowledge, available at that time, on effects of ionizing radiation on species in terrestrial and freshwater aquatic environments.

In 1992, following the United Nations Conference on Environment and Development in Rio de Janeiro, the Agency's role in this field was strengthened. In 1996 the Agency established the first international fundamental principles for radiation safety [19], the first international fundamental principles of radioactive waste management [2], which form the basis for the Joint Convention [3], and the current international radiation safety standards (co-sponsored by FAO, ILO, NEA, PAHO and WHO) [20].

### 3. RECENT AND CURRENT DIRECTIONS

In 1997, Member States adopted, under the auspices of the IAEA, the Joint Convention that includes the following general safety requirement: *'provide for effective protection of individuals, society and the environment, by applying at the national level suitable protective methods as approved by the regulatory body, in the framework of its national legislation which has due regard to internationally endorsed criteria and standards'* [3]. Furthermore, Article 4 of the Convention establishes that *"Each Contracting Party shall take the appropriate steps to ensure that...individuals, society and the environment are adequately protected against radiological hazards"*. This Convention entered into force on 18 June 2001, and the First Review Meeting of the Contracting Parties is expected to take place in November 2003, initiating the first international undertaking to protect the environment against radiation exposure.

In a response to this convention, and the corresponding principle incorporated in the IAEA Fundamentals on Radioactive Waste Management, the Agency's recent work in this area been focused on the development of safety guidance on the application of this principle. In 1999, the Agency issued its first dedicated report on issues related to the protection of the environment from the effects of ionizing radiation [21]. In 2001, the Agency updated its standards for limiting radioactive discharges to the environment [22], and issued the first comprehensive generic models for applying the international guidance for limiting discharges [23]. This rich history of commitment to the control of releases of radioactive materials to the environment, has continued with consideration of issues related specifically to the protection of the environment itself. In the first half of 2002, the Agency issued the first international report on ethical considerations for protecting the environment from the effects of ionizing radiation [24], which is described in more detail in another paper in these Proceedings [25].

Other elements of the Agency's work are also of relevance to an understanding of the levels of radionuclides present in the environment, and of the practical application of international standards in an environmental context. For example, the Agency has compiled inventories of radioactive waste disposals at sea [26], and the first global inventory of 'accidents and losses' at sea involving radioactive materials [27]. The Agency's function to provide for the application of the international standards has resulted in a number of extensive studies aimed at assessing the radiological situation in areas affected by environmental contamination, including: Chernobyl [28], the nuclear testing sites of Bikini Atoll [29], the Atolls of Mururoa and Fangataufa [30], and Semipalatinsk in Kazakhstan [31], as well as the former Soviet Union's dumping area in the Kara Sea [32]. Another mechanism employed is appraisal, and the organisation of international peer-reviews. For example, the

international peer review of the biosphere modelling programme of the US Department of Energy's Yucca Mountain Site Characterization Project [33].

#### 4. DEVELOPMENT OF AN INTERNATIONAL SAFETY REGIME

It is recognized that other international organizations have interests and responsibilities related to environmental radiation protection; notably the United Nations Scientific Committee on Effects of Atomic Radiation and the International Commission on Radiological Protection. The Agency continues to work closely with these organizations with the objective of consolidating a strong international regime for the radiation protection of the environment, comprising legally binding international obligations for controlling discharges into the environment, international standards for limiting discharges, and provisions to ensure their application.

The consolidation of the international safety regime will be facilitated by forthcoming conferences being held by the Agency. The International Conference on Issues and Trends in Radioactive Waste Management will take place in Vienna, 9–13 December 2002. Then, in 2003, the Agency will hold a conference dedicated to the issue of protection of the environment from the effects of ionising radiation. This Conference, which will take place in Stockholm, 6–10 October 2003, will provide a timely opportunity to discuss a number of developments in this area, which will take place during 2003, and to consider their implications for guiding future work at national and international levels.

#### 5. ISSUES TO BE RESOLVED

This policy is being challenged on the basis that, under current circumstances, it might not be sufficient to provide adequate protection to certain 'environments'; e.g. to environments where humans are absent. A notable example of this situation was assessed by the Agency in consideration of the former Soviet Union's dumping site in the Kara Sea, where humans appear to be afforded a greater level of protection than the environment [32]. A number of underlying questions may be formulated as follows:

- Is the aim to protect the human habitat or the wider environment? (The current international standards implicitly refer to species in the 'human habitat', rather than to species in the 'environment'.)
- Is the objective to protect individuals of a given species or the species as a whole? Namely, is it sufficient to protect non-human species as a whole, i.e., collectively? Or should protection be afforded to individual members of the species?
- And finally, what is the applicable ethic?

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# **1. IONISING RADIATION AND BIOTA: EFFECTS, RESPONSES AND MECHANISMS**



# **Effects of ionising radiation on plants and animals: *What we now know and still need to learn*\***

**F.W. Whicker<sup>a</sup>, T.G. Hinton<sup>b</sup>**

<sup>a</sup> Department of Environmental and Radiological Health Sciences, Colorado State University, Fort Collins, CO, United States of America

<sup>b</sup> Savannah River Ecology Laboratory, Aiken, SC, United States of America

**Abstract.** The intent of this presentation was to provide a broad review of the status of existing knowledge on the effects of ionizing radiation on plants and animals, in the context of field rather than laboratory settings, and to offer thoughts on what more we need to learn in order to set protection criteria and test for compliance with greater confidence. General findings from historical studies on designed and accidental irradiation of plant communities and animals, including comparative radiosensitivity and modifying factors, were reviewed. Experimental limitations of most studies and difficulties in response interpretation were discussed in reference to ecological relevance and protection criteria. Recovery of radiation damaged plant communities and animal populations, both during and after radiation exposure, were discussed. The commonly measured responses, mortality and reproduction, were reviewed and questioned as the most radiosensitive, ecologically-relevant endpoints.

Our perceptions of major knowledge gaps on effects of ionizing radiation on the environment were reviewed, and the types of specific studies that appear needed were presented. These needs were discussed under the general headings of dosimetry, damage endpoints, and dose/response relationships. In the area of dosimetry for example, more work is needed on critical species identification, dose model refinements to account for temporal and spatial dose distribution, and RBEs for various damage endpoints. It is known that biological effects such as genomic damage occur in organisms at dose rates well-below those required to obviously impair reproduction. The question is, are such effects harmful to populations in the irradiated generation or in succeeding generations, and if so, under what conditions? Many species and ecosystems have not been experimentally irradiated, either with relatively uniform photon exposures from sealed sources, or from radioactive contamination, which produces large dose variations in space and time. Finally, very little, if anything, is known about whether and to what extent chemical and other stresses interact with the stress of chronic irradiation.

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\* Only an abstract is given here as the full paper was not available.

## **A regulatory framework for environmental protection**

**G.J. Dicus**

US Nuclear Regulatory Commission, Rockville, MD, United States of America

**Abstract.** Industry, regulatory agencies, and the public have been assessing the environmental impacts of regulated, as well as unregulated activities, for many years now. The basic underlying assumption has generally been that the environment is protected through the protection of humankind. In the United States, the environmental regulatory framework has been improved by having a sound executive policy and national regulatory infrastructure, increased consultation with other agencies, changes in the process and timetable for rulemaking changes, and improved communications by the Federal regulators. This paper discusses the various mechanisms in the United States for achieving and maintaining protection of the environment; why regulatory openness and stakeholder involvement is an integral piece of a successful program for protection of the environment; and how international organizations can make a valuable contribution in providing international consensus in the global arena of environmental protection.

Good morning. It is a pleasure for me to be here today to help start off the Symposium's timely discussion of Protection of the Environment from Ionizing Radiation. I am sure some of you attended the Forum in Sicily earlier this year that addressed Radiological Protection of the Environment. When I spoke at that Forum, I focused my comments on several areas, including the development of radiological protection regulations in the United States, the many agencies and branches of government involved in environmental issues, the challenges of maintaining good communication between agencies and the public, the difficulties in finding a path through the morass created by dual regulation, and the emerging challenges to create internationally accepted uniform standards for addressing radiological issues. Today, I would like to expand on a new concept, which I mentioned only briefly in February, that has introduced significant uncertainty in the US legal framework for environmental evaluations and has the potential to make evaluations of environmental impacts much more complex. This relatively new concept is called "Environmental Justice."

However, before discussing Environmental Justice as it is defined and being implemented in the U.S., I will very briefly review with you how our Federal Government reviews major actions that could affect the environment. For over three decades, the Federal government in the United States has reviewed major actions that could affect the environment under the process set forth in the National Environmental Policy Act of 1969 (NEPA). Most of the individual states within the United States have comparable legislation governing state level actions. While some individual environmental evaluations may have remained controversial, the last few decades has seen most government agencies develop an understanding of the basic process for preparing environmental evaluations. Under NEPA, "major federal actions significantly affecting the quality of the human environment" must be accompanied by a detailed environmental impact statement that serves to inform the decision-maker of the potential negative impacts, benefits, and need for the proposed action. NEPA itself does not dictate that any particular balance of benefits versus costs is necessary for ultimate approval of a particular project, but rather constitutes a full disclosure process so that the responsible authority is fully informed prior to finalizing its decision. In the NRC process, members of the public may comment on draft Environmental Impact Statements published for comment and, by meeting certain standards for participation, may participate in a formal proceeding challenging the completeness and accuracy of the proposed Environmental Impact Statement. There are many specific pitfalls and procedural requirements that make hearings on NEPA issues in the United States complex, but what I've just described is a good overall summary of the process.

This relatively predictable process was complicated in 1994 when President Clinton issued an Executive Order introducing the concept of "Environmental Justice" with respect to environmental analyses. Ostensibly not creating any new requirements, this Executive Order directed Executive Agencies to include in environmental analyses a specific consideration of any disparate impact of proposed actions on minority and low-income populations in the United States. Although, as an independent agency, NRC was not required to follow the Executive Order, it followed its traditional approach of voluntarily attempting to meet the intent of the Executive Order to the extent possible.

The concept of Environmental Justice is new to the NEPA process. The underlying concept is inherently laudable. Its goal was to assure that minorities and the financially disadvantaged were not bearing a disproportionate share of environmental impacts from government approved activities. Given the expense of challenging proposed government actions, there is a logic to assuring that those least able to afford challenging actions are not penalized because of those financial limitations.

The IAEA recently published a discussion report that raises, among other ethical considerations of radiological protection of the environment, the issue of Environmental Justice.

As I understand the issue of Environmental Justice as described by IAEA, it is somewhat different than the concept in the U.S. The IAEA concept, like the 1992 Rio Declaration, relates to issues such as liability, and compensation. It considers the balance between benefits and detriment by redistributing the "benefits of actions or policies" or demand compensation for detriment. It further encompasses direct and indirect harm to humans and harm to the environment including inhabitants and habitants. Environmental justice in the U.S. is directly related to socio-cultural protection of disadvantaged and minority populations.

The difficulty is in trying to implement this new concept into the established process for environmental reviews. In general, United States federal agencies have not yet reached a comfort level as to how best to apply the concept of Environmental Justice to evaluations of proposed actions. This is not the traditional environmental review that looks at potential releases and provides an evaluation of the impacts of the proposed project on hypothetical individuals. We all, at least, had some comfort level in looking at potential radiation doses and determining potential impacts on humans and the environment. We have not, however, developed concepts of radiological impact that focus on ethnic or monetary subgroups of affected populations. Initial attempts by NRC to apply this concept quickly demonstrated the difficulty and pitfalls of this new element of environmental reviews.

For example, in one NRC case involving the licensing of a proposed centrifuge enrichment facility, there was an environmental justice concern introduced in the environmental hearing, addressing the expected blocking of a route between some local residences and a local church. The residences affected were in a low income area and many of these individuals did not own cars. The location of the proposed facility rendered the route for walking to a particular church unavailable and alternatives for walking to the church were significantly longer. Ultimately this project was abandoned for a variety of reasons before this particular issue was resolved. It was the first time the issue of Environmental Justice was raised and might have proven to be difficult to resolve.

Although still in litigation and not appropriate for detailed comment given the Commission's role as the ultimate reviewer, an ongoing NRC proceeding is considering the question of whether there can be a subgroup of a minority group. Specifically, we have a group of Native Americans claiming they are entitled to Environmental Justice consideration because they believe the Tribal Government will not fairly distribute profits from a proposed NRC licensed facility within the tribe. The concept of subgroups within recognized minorities and/or low income groups could further complicate environmental evaluations.

What does this mean to those of us who must conduct these evaluations? It means we must ask a different set of questions and apply our health physics and environmental expertise in an expanded and more complex manner. The NRC has developed some guidance for its staff following our initial experiences with applying the concept of Environmental Justice. From this guidance I'd like to note a few of the elements considered in evaluating the question of whether there are disparate impacts on minorities and the poor, when evaluating a potential radiation-related activity.

The first need is to gather information on the populace around a proposed facility. After identifying the minorities and low income groups that are affected by the proposed facility, one must compare their representation within the affected group to that of the larger population. In the United States that can be done by looking at the state population demographics, or several states where the facility is located near state borders, and determining whether there is a higher percentage of a minority and/or low income group in the affected population than in the general population.

The next part of the evaluation must be to determine the impacts on these minority and/or low income populations, as compared to the rest of the affected population. For example, if the poor are more likely to eat fish and game from the affected area, eat locally grown food, or grow their own, it must be determined if this results in a higher radiological impact than for the rest of the affected population.

In the United States such evaluations are not limited to health and safety impacts. Cultural impacts are also considered under NRC guidelines. The example of the affect on access to a church that I mentioned earlier is one example, as would similar access issues related to the ability of the poor and/or minorities to easily reach businesses or work locations. With respect to Native American Tribes, considerations of ancient burial grounds and areas that are considered sacred to the tribes culture must also be considered.

In the United States we also will include potential benefits to these same groups. Our evaluations will consider the financial benefits to minorities and/or low income groups from increased job opportunities and potential increases in property values from the proposed facility. Finally, the evaluation will consider what actions can be taken to mitigate any negative impacts on these specific groups and whether alternative sites may be available for the facility that would have less impacts.

Clearly, as professionals involved in considering the impacts of activities involving radiation that affect the environment, we have a significant role in looking at these types of issues. We are quite capable of providing an evaluation of potential health impacts, based on current knowledge, for an individual who is exposed to a level of exposure from a facility. We are even capable of looking at a worse case scenario and assuming maximum ingestion of locally-grown food or maximum time living and working in the affected area. For example, NRC has included suppositions in some of its evaluations that included individuals having a substantial intake of locally grown food or assuming the affected population is represented by the individual living closest to the facility. Comparing impacts on different populations within the same area, however, is a far more challenging endeavor and will require that we become more knowledgeable about cultural specifics within various affected population groups. In the future, when we ask a question about radiological impacts, we may have to concern ourselves with non-health non-environmental impacts not previously considered. These will present new challenges for us, but will perhaps allow a more complete and meaningful understanding of the impacts of the projects we are considering. While the goal of assuring no one group must shoulder the burden of government projects is laudable, the implementation of Environmental Justice as a method for reaching that goal presents new and complex challenges for the future.

Today's presentations and others during this symposium concern the science of radiation impacts on the environment. Our radiation protection standards and are our regulatory requirements are based generally on the best available science. They are therefore dependent on the work of scientists – the studies, the findings and the interpretations of those findings. Sooner or later, in some fashion, proven out comes will become part of a radiation protection scheme.

But science is only part of the equation. Political and socio-economic factors are also parts of the equation and in the decision making process could take precedence over the science. Environmental justice is an example.

I suggest that it is incumbent on those of you primarily involved in the science to give those of us primarily involved in policy and political arenas the best foundation possible to balance the equation to give science a very strong voice. I wish you good luck and to the organizers of this symposium, thank you and I wish you a successful venture in the next four days.

Thank you.

# **Chronic radionuclide low dose exposure for non-human biota: challenges in establishing links between speciation in the exposure sources, bioaccumulation and biological effects**

## ***Uranium in aquatic ecosystems: a case-study***

**J. Garnier-Laplace, C. Fortin, C. Adam, O. Simon, F.H. Denison**

Institute of Radioprotection and Nuclear Safety (IRSN), Division of Environmental Protection, Laboratory of Experimental Radioecology, Saint-Paul-lez-Durance, France

**Abstract.** In the field of environmental radioprotection, the knowledge gaps concern situations leading to chronic exposure at the lower doses typical of the living conditions of organisms influenced by radioactive releases. For any radionuclide and ecosystem, the specificities of these situations are as followed: (i) various chemical forms occur in the environment as a function of the physico-chemical conditions of the medium; (ii) each transfer from one component to another can lead to a modification of these forms with a “chemical form-specific” mobility and bioavailability; (iii) different categories of non-radioactive toxicants are simultaneously present. In this multipollution context, the biological effects of ionising radiation may be exacerbated or reduced with the potential for action or interaction of all the pollutants present simultaneously. These situations of chronic exposure at low levels are likely to cause toxic responses distinct from those observed after acute exposure at high doses since long-term accumulation mechanisms in cells and tissues may lead to microlocalised accumulation in some target cells or subcellular components. The assessment of these mechanisms is primordial with regard to internal exposure to radionuclides since they increase locally both the radionuclide concentration and the delivered dose, coupling radiological and chemical toxicity. This is the main purpose of the ENVIRHOM research programme, recently launched at IRSN. After a global overview of the experimental strategy and of the first results obtained for phytoplankton and uranium, this paper scans the state of art for uranium within freshwaters and underlines inconsistency encountered when one wants to carry out an Ecological Risk Assessment (ERA) on the chemical or on the radiological standpoint. This example argues for future research needs in order to establish well-defined relationship between chemo-toxicity and radiotoxicity for internal contamination. The operational aim is to bring adequacy between ecological and human health risk assessment for radioactive or “conventionnal” substances.

### 1. ECOLOGICAL RISK ASSESSMENT AND RADIONUCLIDES: BACKGROUND AND GAPS OF KNOWLEDGE

At the present time, the international community is showing a priority need for the development of design, knowledge and methods for ecological risk evaluation in connection with ionising radiation [1, 2, 3]. Within the framework of radioprotection, one of the main challenges is to acquire data necessary for ecological risk assessment for situations where living organisms are chronically exposed to radionuclides present at low level within the different components of the biosphere. In these situations and throughout their lifespan, the non-human biota and each person from the general public are subject to external radiation according to their mode of life within contaminated environment and to internal radiation alongside chemo-toxicity, both as a consequence of the integration processes of radionuclides in living organisms (direct transfer from abiotic compartments and trophic transfers by ingestion). For both of the following cases, the risk is, in particular, a function of the type of radiation:

- in the case of internal contamination, the risk linked to the biological incorporation of  $\alpha$  or  $\beta$  emitting radionuclides will be more important than that linked to the incorporation of radionuclides that mainly emit radiation which is not directly ionising i.e. neutrons and photons (low LET, high penetration);
- in the case of external radiation, this hierarchy works in the opposite direction. The risk that is therefore associated with  $\gamma$  emitters is more important in front of that associated with  $\alpha$  or  $\beta$  emitters. Furthermore, the potentially associated chemo-toxic risk thus becomes insignificant.

Within the framework of point (1), the ENVIRHOM programme, launched last year at the IRSN suggests data acquisition to understand and quantify biological effects involved by the accumulation of radionuclides by living organisms (biological components of ecosystems and the general public for

human populations) in chronic exposure situations [4]. The accumulation mechanisms in cells and tissues may lead to microlocations on some target cells or subcells according to the biochemical behaviour of the studied radionuclide, bringing chemo-toxic phenomena into play and energy deposits of a very different size and flow rate, characterized by heterogeneity at the cell scale. Biological effects may result from these phenomena. These effects on living organisms, man included, are not precisely known to date as the vast majority of available data corresponds to studies performed on high doses of radionuclides, short term exposure and not within a multipollution context (i.e. without taking into account the simultaneous presence of other categories of pollutants: metals, organic micropollutants, ...). Moreover, the risk evaluation due to ionising radiation has never been compared to the traditional ecotoxicity assessment carried out for chemical substances, even though internal contamination by any radionuclide obviously brings together radiotoxicity and chemotoxicity.

ENVIRHOM suggests the assessment of the integration potentialities of radionuclides into the trophic networks from the soil and sediment considered as reservoir compartments of the biosphere likely to be gradually and significantly enriched in radionuclides released into the environment, and acting as secondary source-terms for the other components of the ecosystems. These transfers within ecosystems are characterised by a wide variety, concerning both the biogeochemical behaviour of radionuclides and the feeding strategies of plants and animals. The studies of long term induced biological disturbance as a consequence of bioaccumulation processes will be systematically focused on behaviour, growth and ability to reproduce, without excluding other more subtle effects (such as cytogenetic effects for example). The aim is to go on establishing relationships between the concentration of bioavailable chemical species in the various exposure sources for organisms, and the ecological repercussions, as a result of individual disturbances, in particular in terms of population dynamics and community structure.

In order to limit the framework of this very broad research programme, a list of a limited number of radionuclides has been decided on the basis of a multi-criteria approach. The choice has been made within the list of radionuclides with a significant occurrence in the different source-terms from nuclear installations under normal operating conditions (nuclear power plant, fuel reprocessing plant), storage sites for radioactive wastes, uranium-bearing ore mining sites in operation or after closure, and more generally industries or particular geochemical situations generating a significant increase in naturally occurring radionuclide concentrations in the environment, post-accident situations such as Chernobyl. The first criteria is the type of radiation with selection of  $\alpha$  and  $\beta$  emitters that develop the highest risk associated with internal contamination. The second criteria is the physical period, which should be significant in terms of chronic contamination at human lifespan scale, or 70 years. Thereafter, the remaining nuclides were ranked according to their propensity to react with biomolecules directly dependent on the affinity of radionuclides for hydroxyl groups, thiols and/or phosphates and therefore to bioaccumulate (In general, the tendency to form organic complexes is proportional to the tendency to hydrolyse and the electric charge, and inversely proportional to the ionic radius). The main differences within these biochemical properties help to distinguish two categories of elements: radioactive isotopes of an element (stable isotope or chemical analogous) involved in the constitution of living matter as macro-nutrients or oligo-elements or radioactive isotopes without any known biological function. Applying this selection method, the priority for radionuclides is as followed:  $\alpha$ emitters with three actinids: natural uranium, americium-241 and neptunium-237, long-lived  $\beta$  emitters with technetium-99, iodine-129, selenium-79 and Cs-135. The first experimental development was launched with uranium.

In the first part of the paper, the brief prospective description of the experimental strategy of the ongoing ENVIRHOM programme is given and partly illustrated with some first results concerning phytoplankton. In a second part, the knowledge needed to carry out any ecological risk assessment is listed and illustrated for uranium and freshwater ecosystems. Before conclusion, the need to define for radionuclides a consistent approach with that developed for chemical pollutants for which the targets protected by the regulations are mankind, the fauna and the flora is illustrated by comparing no-effects values for uranium on the chemical and radiological aspects. Illustrations are given for phytoplankton in order to insist on the discrepancy that appears when the approach existing for ecological risk assessment based on the methodology developed at EC [5] and the approach emerging within radioecological risk assessment, are bringing together.

## 2. THE EXPERIMENTAL STRATEGY OF THE ENVIRHOM PROGRAMME: TOWARDS THE IMPROVEMENT OF KNOWLEDGE LINKED TO INTERNAL CONTAMINATION EXEMPLIFIED WITH URANIUM AND FRESHWATER ECOSYSTEMS

### 2.1. Global overview

A few number of biological models have been selected in order to cover a wide range of diversity for feeding strategies and thus biological barriers to be crossed and for bioaccumulation mechanisms likely to be involved in animals and plants. For example, concerning freshwater ecosystems, a unicellular algae exposed to radionuclide within the water column, and several invertebrates were selected such as crayfish and bivalves. The feeding strategies of these latter based on the sediment interstitial water, the water at the water-sediment interface and various particles (phytoplankton, organic detritus, various sedimentary particles) make them particularly well-suited as biological models for the study of the influence of geochemical and biological parameters on bioavailability and on the bioaccumulation processes. Different types of prey-predator trophic relations have been chosen to complete in a simplified way the range of dietary patterns that may occur for consumers while selecting invertebrate (crayfish) as second order consumer, and several species of fish. All these biological models are widely used in toxicological or ecotoxicological studies and more or less extensive data exists on their physiology. They may be considered as generic key stone phyla within ecosystem functioning.

### 2.2. Link between chemical speciation of the radionuclide in the source of exposure and bioaccumulation processes: short term exposure experiments

The bioaccumulation of a pollutant results from the interaction between the physical and chemical variables of the exposure sources (“physical” compartments and food) and those concerning the characteristics relative to living organisms, from molecular scale to the highest level of integration (biocenosis). In any case, the biogeochemical behaviour of pollutants within physical compartments (atmosphere, soil, sediment, water column) controls the capacity for transfer towards organisms. The speciation of the pollutant in the medium is the first factor that regulates its bioavailability and therefore, its bioaccumulation. For metallic pollutants, it is generally admitted that the total aqueous concentration is not a good predictor of bioavailability and its complexation with most dissolved inorganic and organic ligands normally leads to a decrease in bioavailability. Under this assumption and model known as the Free-Ion-Model, pollutant uptake and induced biological response (toxicity) vary as a function of the concentration of the free-metal ion in solution; however, a great number of exceptions exists [6].

Concerning uranium, geochemical model may fairly reliably support the experimental approach as long as enough thermodynamic data is available and its consistency has been verified, at least for reactions with mineral ligands. The exposure media are in the first stage of simplified physico-chemical composition i.e. artificial water of mineral composition in such a manner as to predict in the most reliable way possible, the chemical aqueous forms of U that are likely to be present and to cross over biological barriers. The geochemical speciation code JChess [7] using a database compiled from the OECD/NEA thermochemical data base project [8] was used to perform the solution speciation calculations (Figure 1a). Three main variables are then with a complexification of the water column chemical composition: (1)  $p\text{CO}_2$  and pH (from atmospheric pressure to 10 atm and from acid to basic conditions respectively); (2) competitive cations such as Ca, Mg; (3) presence of ligands such as phosphates, or dissolved organic matter. For these complementary short-duration well-defined laboratory experiments in simplified conditions; biokinetics for short exposure times (in hours for algae, in days for animal models) and characterisation of the input mechanism(s) are investigated.

The concentration range used in total uranium in the exposure sources goes up to a maximum of  $1 \text{ mg}\cdot\text{l}^{-1}$ , a value that may be encountered in an aquatic ecosystem that is influenced by mining discharge.

Where bivalve is concerned, the uranium transfer associated with model mineral particles will be assessed, in the same way as the trophic transfer due to ingestion of phytoplankton [9]. In order to quantify the bioaccumulation processes that may be employed during transfer through ingestion when in a predator-prey relationship, experimental studies on the bivalve (asiatic clam as prey) or the fish

(rainbow trout as predator) will begin with a pharmacokinetic approach in order to model the fate of an alimentary bolus where the main chemical “pools” of uranium in the prey (subcellular fractionation) have been explored.

### **2.3. Bioaccumulation/biological effect link: chronic exposure (long term) experiments**

Based on knowledge gained from previously described experiments, the exposure scenario that defines the most important bioavailability will be chosen to be transposed into experiments that enable simulation of chronic exposure under controlled conditions (significant duration in front of the organisms’ lifespan). During these experiments, the bioaccumulation processes will be investigated in parallel with the involved biological effects. The primary aim of the studies of microlocalisation will be to determine, for a few target organs, whether uranium is evenly distributed in the tissues and cells or whether, on the contrary, it is localised in particular structures. In the latter case, the position of the radionuclides in relation to or within target cells will be established. The chosen observation technique will be electron microscopy in transmission associated with a spectral analysis of X energy dispersion.

Certain biochemical responses will be measured to assess the early effects of stress at cellular or subcellular level, involving dosage and validation techniques borrowed from ecotoxicology. Several (sub)cellular endpoints will be investigated: (1) The responses to oxidative stress (catalase, superoxide dismutase, glutathion transferase, forms of glutathion); (2) The exploration of energy expenditure (adenylate load, glycogen reserves, protein, lipid and glucid content); (3) Other biomarkers of more general effects (induction of metallothioneins (or phytochelatines for plants), of stress proteins (hsp). At the individual scale, the investigation of biological disturbances following bioaccumulation will be undertaken mainly on three essential functions of great importance for the functioning and structure of any ecosystem: the growth, the behavior and the reproduction, this latter including cytogenetic effects on germinal cells. For organisms with a sufficient organisation level (fish), this will be mainly viewed as investigations on the immune system, the central nervous system and the reproductive system.

### **2.4. First results obtained for the unicellular algae model and uranium**

Phytoplankton represents the basic part of the productivity chain, at the lowest trophic level in the freshwater trophic networks. It is therefore a key player in the elements cycle in the ecosystems, especially with regards to their integration into the food chain from the water column.

Two distinct phenomena may be identified for algae: adsorption (metal fixation at the algae surface without penetration of the cellular membrane) and absorption (metal internalisation). Particular attention is given to the difference between these two phenomena. The first is of a chemical nature whereas the second is of a biological nature. By using short exposure time ( $< 1$  h) and low cellular density for the experimental population, it is possible to keep under control the uranium solution chemistry and to achieve the identification of one or more chemical species that govern the uranium/algae interactions.

The first results [10] suggest that uranium adsorption at the surface happens very quickly and reaches a stationary state (equilibrium) in just a few minutes. The absorption, however, increases with exposure time. In addition to this, saturation phenomena are noted when the uranium concentration reached a certain level (some  $\mu\text{M}$  i.e. around  $0.1 \text{ mg/L}$ ), that is to say that the metal internalisation capacity of the algae reaches a maximum level. One other stage was overcome when observing that phosphates (which are found in the environment due to human activities and which are responsible for the eutrophication of waterways), by forming chemical complexes with uranium, unaffected absorption of this metal by the algae. The uranium accumulation is significantly lower in acidic (pH 5) than it is in neutral media (pH 7). This remark leads to important questions as uranium chemistry is greatly altered within this range. Michaelis-Menten kinetic parameters  $K_m$  and  $V_{\max}$  were determined using the Marquardt-Levenberg algorithm to obtain the best fit to the observed uptake levels (see equation and curves in Figure 1b). The half-saturation constant is nearly doubled at pH 7 when the data is analysed based on the total uranium concentrations. Growth toxicity tests, representative of long term exposure (the lifespan for an algae within our experimental conditions is in the order of 10 h), are in progress, underlying the importance of the water quality variables (such as pH). Microlocation data are also expected.



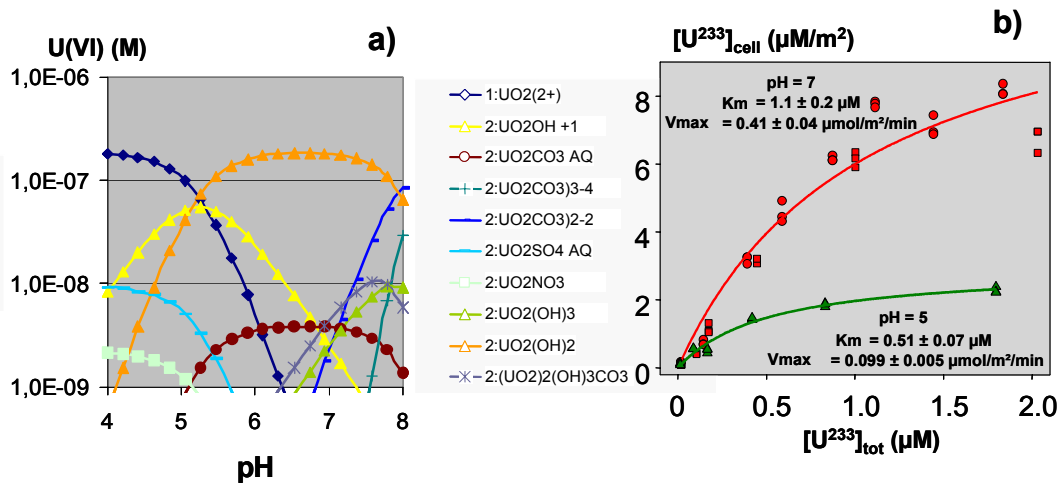


FIG. 1. a) Uranium speciation diagramme in a very simple artificial water in equilibrium with atmospheric CO<sub>2</sub>; b) U intracellular uptake for phytoplankton after 30 minutes of exposure at various total uranium concentrations (● = pH 5; ○ = pH 7). Error bars represent the standard deviation from the average of three measurements.

### 3. CURRENT AVAILABLE DATA FOR ECOLOGICAL RISK ASSESSMENT: STATE OF THE ART FOR URANIUM IN FRESHWATERS

In complementarity with health risk examination, any risk assessment to biota from exposure to radionuclides is to be associated with (1) different source-terms and environmental released scenario, (2) exposure pathways and potential biological effects at different organisation level, (3) estimation of no-effects values and finally, (4) risk calculations as the ratio between predicted concentrations in the source of exposure and estimated no-effects concentration. Concerning the case of internal contamination by any radionuclide, the radiological and the ecotoxicological risk assessments have to be consistent with each other. The whole methodology is exemplified here after with uranium in freshwater ecosystems, underlying discrepancies to solve between the potential risks from the radiological and the chemical toxicity standpoints, giving perspectives for future research needs, as previously overviewed within the ENVIRHOM programme description.

#### 3.1. Source-terms and environmental exposure pathway analysis

Uranium is a naturally occurring element, member of the actinide series. By mass, natural uranium is composed of 99.3% <sup>238</sup>U, in equilibrium with <sup>234</sup>U (therefore, 0.005%) and 0.7% <sup>235</sup>U. Including these three radionuclides, the specific activity for natural uranium is equivalent to 2.6 10<sup>4</sup> Bq/kg. The environmental behaviour of U has been extensively studied and a number of reviews exists in the literature. Its concentrations in terrestrial and aquatic ecosystems may be increased in connection with various anthropogenic contributions, originating from uses throughout the different stages of the nuclear fuel cycle (mines and waste storage sites in particular), and up to agricultural use (phosphate based fertilizers), the medical surroundings, research laboratories and military use of depleted uranium [11]. Several phenomena linked to the biogeochemical behaviour of uranium, in connection with the implementing of physical processes of solid transport (erosion, sedimentation ...) and water transport (colloid and dissolved forms), may lead to the existence of accumulation zones in soils and sediments: horizons that are rich in organic matter and/or iron oxyhydroxides in an oxidising condition, flooded soil or sediments in a reducing condition (uranium is therefore at the (+IV) valency and tends to enter into zones that are rich in organic matter, in sulphur and/or minerals rich in Fe(II)). Uranium's environmental geochemistry quite schematically enables to predict U transport into high Eh zones [U(+VI)] and a deposit by reduction and precipitation in low Eh zones [U(+IV)] [12]. The existence of these accumulation zones may enhance reactions that are likely to occur at the biological interface level and consequently, the mechanisms leading to an implementation of the bioaccumulation processes on various intracellular biological targets in plants and animals. The bioavailability of the radionuclide and its uptake by biota ultimately govern their effects on biota.

At the present time, even if knowledge exists, the operational codes currently used with this purpose, to simulate the likely routes of acute and/or chronic exposure for biota, consider the ecosystems in a very simplified way, at equilibrium, with a homogenous concentration, performing exchanges on the basis of transfer coefficients that characterise the element without distinguishing chemical species or even mobile and bioavailable fractions. The accumulation processes in some areas of the biosphere are not taken into account, in the same way as the bioaccumulation processes and the potentially induced effects on the biocenosis.

### **3.2 Biological Effects: chemical toxicity and radiological toxicity**

The toxic effects linked to pollutants are, as a general rule, closely associated with the processes of bioaccumulation, although since certain of them lead to the sequestration of pollutants in non-toxic form, this may limit the biological consequences within a certain range of concentration. Organisms develop a wide range of biochemical, immunological and physiological responses, according to the concentration level of the pollutant and the duration of exposure. The earliest manifestations may be observed at cellular level or at the level of the individual. They are of three kinds: (1) direct interaction between the toxic element and the biological target(s), (2) effects on the energetic or hormonal metabolism that may have repercussions on growth, fecundity and life-span, or (3) behavioural effects.

For uranium as for the majority of radioactive pollutants, current understanding of radiation effects coupled with chemical effects stems to a large extent on observations made after acute exposures, i.e. at high doses and for short term duration. Data never distinguish the two combined effects and uranium has mainly be studied on the ecotoxicological point of view (Table 1).

At cellular level, various forms of damage may be caused by metals and metalloids according to the conditions of exposure, by means of three mechanisms: (1) binding with intracellular or membranous biomolecules (enzymes, DNA, phospholipids); (2) reaction with the bonds of thiol groups of biomolecules (glutathion, peptides, various proteins); (3) damage to the membranous transport, the stability of the lysosomas and DNA replication [13]. Uranium, like many xenobiotics, induces oxidative stress at cellular level, defined as the full range of deleterious effects linked to active forms of oxygen or oxiradicals [14]. In freshwater animals (bivalves and fishes), it triggers *in vitro* mechanisms of membranous lipidic peroxidation and inhibits the catalytic activity of various enzymes involved in antioxidation defence (superoxide dismutase, catalase, glutathion peroxydase and reductase, etc...). *In vivo*, these mechanisms would also appear to occur with differences between the molluscs that rather react by adaptation of the levels of activity of the antioxidation enzymes and the fishes where this mechanism is linked to adaptation of the glutathion concentration [15, 16]. U accumulates in lysosomes of marine and and freshwater molluscs and crustaceans [17] and mammals [18]. When precipitated as U phosphate microneedles, U damages subcellular structures, such as lysosomal membranes. These membrane peroxydations have also been observed on mammals [15, 19]. Tasat and de Rey [20] suggested that these severe damage to organelles and cell death were mainly explained by lysosomal membrane damage and the subsequent release of hydrolytic enzymes into the cytosol. For fish, recent laboratory studies on *Coregonus clupeaformis* exposed by the trophic pathway with artificial food contaminated by U during a 100-days period, showed the most significant effects were elevations of lipid peroxidation and histopathological lesions in liver and posterior kidney such as tissue necrosis, inflammation [21, 22]. These sub-lethal effects had no effects upon parameters at the whole body level with the experiment duration (growth, morphometrics). Globally, these organs are also considered as target tissue for mammals and for massive concentrations, lesions of the renal tubule cells lead to necrosis and cell death with severe disturbances for renal reabsorption [19]. For lower concentrations, a modification of the cellular energetic metabolism has been reported [23].

TABLE 1. ACUTE TOXICITY DATA FOR URANIUM AND FRESHWATER PLANTS AND ANIMALS

Species	Age/size	Chemical form	Parameters	U-value (mg U/l)	Temp. (°C)	pH	Water quality	Reference
<b>Phytoplankton</b>								
<i>Chlorella sp.</i>	Exponential phase	Uranyl	EC <sub>50</sub> – 72h	0.044 0.078	27	6.5 5.7	2–4 mg/l CaCO <sub>3</sub>	Franklin et al., 2001 [24]
<b>Cladocera</b>								
<i>Daphnia magna</i>	1 j	Uranyl	CL <sub>50</sub> – 48h	5.3–7.6 30–44 30–74	20	8	66–73 mg/l CaCO <sub>3</sub> 126–140 mg/l CaCO <sub>3</sub> 188–205 mg/l CaCO <sub>3</sub>	Poston et al., 1984 [25]
Species from South Hemisphere <sup>2</sup>	<6h	Uranyl sulfate	CL <sub>50</sub> – 24h	0.4–6.4	27	6.6	3.3 mg/l HCO <sub>3</sub>	Bywater et al., 1991 [26]
<b>Bivalve</b>								
<i>Corbicula fluminea</i>	2–2.5cm	Uranyl acetate	CL <sub>50</sub> – 96h	1872	20	7.9	178 mg/l CaCO <sub>3</sub>	Labrot et al., 1996 [27]
<i>Veselinus angasi</i>	0.8–2.5cm	Uranyl sulfate	EC <sub>50</sub> – 48h	0.117	28	5	2.5 mg/l HCO <sub>3</sub>	Brown et al., 1994 [28]
				0.247 0.634 1.228		6	+ 7.5 mg/l COD + 7.5 mg/l COD	Markish et al., 2000 [29]
<b>Cnidaria</b>								
<i>Hydra sp.</i>	Mature	Uranyl sulfate	LOEC – 96h	0.15–0.40	30	6.5	12–20 µS/cm	Hyne et al., 1992 [30]
		Uranyl	EC <sub>50</sub> – 96h	0.114	27	6	6.6 mg/l CaCO <sub>3</sub> –Hardness 4 mg/l CaCO <sub>3</sub> –alkalinity	Riethmuller et al., 2001 [31]
				0.177			165 mg/l CaCO <sub>3</sub> –Hardness 4 mg/l CaCO <sub>3</sub> –alkalinity	
				0.171			165 mg/l CaCO <sub>3</sub> –Hardness 102 mg/l CaCO <sub>3</sub> –alkalinity	
				0.219			330 mg/l CaCO <sub>3</sub> –Hardness 4 mg/l CaCO <sub>3</sub> –alkalinity	
<b>Fish</b>								
<i>Brachydanio rerio</i>		Uranyl acetate	CL <sub>50</sub> – 96h	3.05	20	7.9	178 mg/l CaCO <sub>3</sub>	Labrot et al., 1996 [27]
<i>Pimephales promelas</i>	n.p. <sup>3</sup>	Uranyl	CL <sub>50</sub> – 96h	2.8–3.1 135	n.p.	n.p.	20 mg/l CaCO <sub>3</sub> 400 mg/l CaCO <sub>3</sub>	Tarzwel and Henderson, 1960 [32]
<i>Pimephales promelas</i>	juvenile	Uranyl	CL <sub>50</sub> – 96h	16.7	20	7.9	66–73 mg/l CaCO <sub>3</sub>	Poston et al., 1982 [33]
<i>Oncorhynchus mykiss</i>	n.p.	Uranyl	CL <sub>50</sub> – 96 h	6.2	n.p.	n.p.	Faiblement minéralisée	Davies, 1980 in: Bywater et al., 1991 [26]
<i>Salvelinus fontinalis</i>				8.0				
Species from South Hemisphere <sup>4</sup>	juvenile	Uranyl sulfate	CL <sub>50</sub> – 96 h	0.7–3.5	27	6.6	3.3 mg/l HCO <sub>3</sub>	Bywater et al., 1991 [26]

<sup>1</sup> The test parameter used to quantify toxicity may vary from one experiment to another: the lethal (or effect) concentration for 50% of the individuals after 24 or 96h-exposure period (LC ou EC<sub>50</sub>); the lowest observed effect concentration inhibiting growth (LOEC).

<sup>2</sup> Experiments carried out for 4 species from northern Australia: *Diaphanosoma excisum*, *Latonopsis fasciculata*, *Dadaya macrops*, *Moinodaphnia macleayi*.

<sup>3</sup> Not précised.

<sup>4</sup> Experiments carried out for 6 species from northern Australia: *Melanotaenia nigrans*, *Melanotaenia splendida inornata*, *Craterocephalus marianae*, *Pseudomugil tenelus*, *Ambassis macleayi*.

Concerning radiological effects, uranium is an alpha-emitter and therefore presents an internal hazard for living organisms. However, its low specific activity led researchers to focus on its chemical toxicity. For radioactive substances, a number of literature reviews, mainly based on data from  $\gamma$  external irradiation, have suggested doses of approximately 2.5 and 0.5 mGy·d<sup>-1</sup> respectively for aquatic plants and fish [34] or 10 mGy·d<sup>-1</sup> for aquatic species in general [35] would not endanger populations. Data mainly concerns high doses and acute exposure. For lower doses, reproduction and genotoxicity have mainly been studied, but if effects on reproduction are considered to be the most likely limiting endpoint in terms of survival for the population, genetic damages present some difficulty in interpreting the significance of the effects at the population level. In any case, radionuclides are only seen as different types of particle-emitters and the radiological risk assessment is therefore carried out with the additivity assumption by summing all external and internal sources of radiation. However, concerning internal contamination, and particularly for  $\alpha$  and  $\beta$  particles, the absence of any structured relation between radiotoxicity and chemical toxicity, may bring inadequacy for this assumption and inconsistency for conclusions of ecological risk assessments on the chemical or radiological standpoints. Within this scope, the importance of the bioaccumulation phenomena is primordial with regard to internal exposure by radionuclides since they increase locally both the radionuclide concentration and the biological effect of the delivered dose.

TABLE 2. DISCREPANCY OF NO-EFFECTS VALUES FOR URANIUM AND FRESHWATER PHYTOPLANKTON ON THE ECOTOXICOLOGICAL OR RADIOLOGICAL ASPECTS. CALCULATION ARE CARRIED OUT AS A FIRST APPROACH ACCORDING TO OBTAINED DATA FOR CHLAMYDOMONAS REINHARDTII (SEE EQ. (1) AND FIGURE 1B)

No-effects values	U-value in water (µg/l)	Internal Dose rate for alpha radiation mGy/d	Reference for the PNEC value
PNEC	<b>0.044</b>	$2.56 \cdot 10^{-3}$ (pH 7)	Data analysis according to EC, 1996 [5]
chemical toxicity		$1.33 \cdot 10^{-3}$ (pH 5)	
No-effects values	259 (pH 5) 51 (pH 7)	<b>2.5</b>	Data from this study analysed according to Environment Canada guidelines (2000) [34]

### 3.3. No-effects values: comparison of the “chemical” and “radiological” approaches

The overall approach follows the guidelines in EC (1996) [5]. The central concept is to characterize chronic ecological risk as a quotient of the estimated exposure value divided by the estimated no-effects value. If the quotient is less than unity, the pollutant is not toxic. On the basis of literature effects data (Table 1), adopting the extrapolating method that consists in applying a conservative and protective factor of 1000 if “at least, one short-term L(E)C<sub>50</sub> from each of the three trophic levels of the base-set (fish, daphnia and algae) is available” [5], the no-effects values for freshwaters should be equivalent to 0.044 µg U/l. On the basis of the accumulation results obtained for phytoplankton, a simple calculation of radiation dose rate due to internal contamination by alpha emitting can be carried out for the whole life of a given phytoplanktonic cell. Using Michaelis-menten equations (see Figure 1b) to convert water concentration into cell concentration after a lifespan exposure (around 10 hours for *Chlamydomonas sp.*), one obtains the internal dose rate (DR<sub>intα</sub> in mGy/d) according to the following equation: DR<sub>intα</sub> = DCF<sub>intα</sub> [cell<sub>max</sub>] Eq(1) with:

- DCF<sub>intα</sub>, the internal Dose Conversion Factor equal to  $(1.6 \times 10^{-10} \times \sum_i P_i E_i)$ , where  $1.6 \times 10^{-10}$  corresponds to the conversion Factor between absorbed energy in MeV and mJ, P<sub>i</sub>E<sub>i</sub> are the average yield time energy values for alpha radiation (i.e. 4.55 MeV for natural U);
- [cell<sub>max</sub>], the maximal concentration expressed as Bq/kg fresh weight in an algal cell after a time exposure period equivalent to its life span.

The calculated dose rates corresponding to the chemical predicted no-effects value are reported in Table 2 for two pH conditions ( $2.56 \times 10^{-3}$  and  $1.33 \times 10^{-3}$  mGy/d respectively for pH 7 and 5). The retrocalculation with the same equations on the basis of the “radiological predicted no-effects value” recommended by Environment Canada [34] gives much higher water concentrations than the “chemical PNEC” (51 and 259 µg U/l respectively for pH 7 and 5; see Table 2). The two modes of toxic effects – radiotoxicity and chemical toxicity – are always handled separately, mainly because there are undistinguishable when internal contamination is considered. However, this method consisting in performing separately the risk assessment for non-radioactive and radioactive substances, leads to inconsistent no-effects values. Moreover, the potential combined effects of chemotoxicity and ionizing radiation for a given radionuclide, have never been assessed.

## 5. CONCLUSIONS – PERSPECTIVES

Globally, concerning internal contamination and the resulting potential hazard for both ecosystems and human populations, research are needed to acquire knowledge concerning:

- (1) the consequences of the existence of a radionuclide concentration heterogeneity at the scale of the cell/organ/full organism – this heterogeneity couples chemo-toxicity and radiotoxicity, and basic knowledge does not exist to have well-defined relationship between the different (or not!) toxic effects; moreover, this heterogeneity leads to locally important delivered dose and to calculate it precisely at the scale of (sub)cellular target becomes necessary in order to acquire fiable data to establish dose-effects relationships.

- (2) the consequence of a chronic exposure at low-level coming from the specificities of these situations as followed: (i) various chemical forms occur in the environment as a function of the physico-chemical conditions of the medium; (ii) each transfer from one component to another can lead to a modification of these forms with a “chemical form-specific” mobility and bioavailability; (iii) different categories of non-radioactive toxicants are simultaneously present.
- (3) the need to define for the radionuclides a consistent and integrated approach with that developed for chemical pollutants for which the targets protected by the regulations are mankind, the fauna and the flora.

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# A comparative study of the effect of low doses of ionising radiation on primary cultures from rainbow trout, *Oncorhynchus mykiss*, and Dublin Bay prawn, *Nephrops norvegicus*

F.M. Lyng<sup>a</sup>, P. Olwell<sup>a</sup>, S. Ni Shuilleabhain<sup>a</sup>, A. Mulford<sup>b</sup>, B. Austin<sup>b</sup>, C. Seymour<sup>a</sup>, M. Lyons<sup>c</sup>, D.C. Cottell<sup>d</sup>, C. Mothersill<sup>a</sup>

<sup>a</sup> Radiation and Environmental Science Centre, Dublin Institute of Technology, Dublin, Ireland

<sup>b</sup> Department of Biological Sciences, Heriot Watt University, Edinburgh, Scotland

<sup>c</sup> The Marine Institute, Dublin, Ireland

<sup>d</sup> Electron Microscopy Laboratory, University College Dublin, Dublin, Ireland

**Abstract.** There have been very few studies on comparative radiobiology. Any work that has been done has involved very high doses which are not relevant for environmental exposures. The aim of the present study was to compare the effects of radiation on primary cultures from both rainbow trout and Dublin Bay prawn, *Nephrops norvegicus*.

Primary cultures from the pronephros of rainbow trout and from the haematopoietic tissue of *Nephrops norvegicus* were irradiated *in situ* 2–4 days after explantation using a <sup>60</sup>Co teletherapy unit. The doses used were 0.5 and 5 Gy. The cultures were fixed 4–7 days post irradiation in 2.5% glutaraldehyde, post fixed in 1% OsO<sub>4</sub>, dehydrated in ascending grades of ethanol and embedded in epoxy resin. Thin sections were cut *en face* from the embedded cultures, stained with uranyl acetate and lead citrate and examined using a JEOL 2000 transmission electron microscope.

The irradiated cultures from both rainbow trout and *Nephrops norvegicus* displayed pronounced morphological damage, in particular to the nucleus and mitochondria. Damage to the cytoskeleton was also evident. However, cells from the haematopoietic tissue of *Nephrops norvegicus* appeared to be considerably more radiosensitive than cells from the pronephros of rainbow trout.

These results may have important implications for environmental radiation protection policies.

## 1. INTRODUCTION

There have been few studies on the comparative effects of radiation on different species. Any research that has been done has mainly involved very high radiation doses which are not relevant to environmental exposures.

Very few radiobiological studies have been done on species other than mammals. Among the vertebrates, some research has been completed on reptiles. Altland et al [1] reported the lethal dose of radiation for turtles to be 10–15 Gy and also noted effects on hematopoietic tissue similar to those observed in mammals. Skinks have been shown to survive exposure to 15Gy while iguanas survive 12 Gy [2]. Turner et al [3] found sex, age and dose rate effects in lizards, which may explain variations found in other studies. More recently, Ulsh et al [4] reported that human fibroblasts are 1.7 times more sensitive than turtle (*T. scripta*) fibroblasts to radiation induced chromosomal aberrations.

There are many publications on fish radiobiology due mainly to the use of medaka, *Oryzias latipes*, as a model for studying environmental germ cell mutagenesis [5]. In addition, our group has shown similar radiosensitivities for human and fish cell lines to UV-A and UV-B radiation [6].

Among the invertebrates, there is a suggestion of extremely radioresistant responses. Tunicates (*Botryllus schlosseri*) were exposed to doses of 25 Gy before an immune recognition response was observed [7]. Insect cell lines are known to have radiation dose response curves with D<sub>0</sub> values in the region of 30Gy as compared to 1–2Gy for most mammals. Sponges have been shown to be similarly radioresistant [8].

The overall impression from the available literature is that invertebrates are extremely radioresistant relative to most vertebrates and especially relative to mammals. This has implications for radiological protection policies but may also point to interesting mechanisms which might help in the understanding of the evolution of protective mechanisms. Similarly, fish, as primitive vertebrates, are an important species for radiobiological studies.

Haematopoietic tissue is of considerable interest as it is one of the most sensitive and widely studied tissues in mammalian radiobiology.

Since teleost fish have no lymph nodes and their bones usually have no medullary cavity, haematopoietic tissue is located in the stroma of the spleen and the interstitium of the kidney. The head kidney or pronephros is commonly supposed to be the organ of haemopoiesis analogous to red bone marrow in mammals [9]. The blood forming cells are found in various stages of development, undifferentiated stem cells, blast cells, immature and mature stages of red and white blood cells.

In crustacea, haematopoietic tissue covers the dorsal part of the cardiac stomach or more posteriorly occurs as part of the membrane supporting the heart. The haematopoietic tissue is generally organised into lobules composed of stem cells and maturing hemocytes bounded by an intimal layer. Crustacean hemocytes are thought to be functionally analogous to vertebrate leukocytes. Two major groups are recognised; hyaline hemocytes and granulocytes. The latter group is further divided into small and large granule hemocytes [10].

The aim of the present study was to compare the effects of radiation on primary cultures of haematopoietic tissue from Dublin Bay prawn, *Nephrops norvegicus*, and from rainbow trout, *Oncorhynchus mykiss*.

## 2. MATERIALS AND METHODS

### 2.1. Animals

Healthy males and females of *N. norvegicus* (average length 10–12 cm) were captured by trawling in the Firth of Forth, near Edinburgh, Scotland. After transfer to the laboratory, animals were maintained in aerated seawater (salinity [S]= 33‰) at ~14°C. All the animals were in intermoult stages.

Healthy rainbow trout (200–300g) were obtained from a commercial fish farm. The fish were killed by an overdose of ethyl-4-aminobenzoate.

### 2.2. Primary culture method

The prawns were anaesthetized, and immersed in seawater at 4°C for 50–60 min then dipped briefly in 10% (w/v) sodium hypochlorite followed by several rinses with 70% ethanol to control contamination. The haematopoietic tissue was carefully removed and placed in an antibiotic solution with 10% (w/v) 2X Leibovitz's medium (Gibco BRL), 1% penicillin-streptomycin (Sigma 10 × 4 IU/ml – 10 × 2 µg/ml), 10 µg/ml amphotericin B (Sigma) and 5 µg/ml gentamicin (Sigma). The tissue was washed three times in the antibiotic solution, before chopping into fragments of ~1 mm<sup>3</sup> using razor blades. Small fragments (explants) were placed into sterile disposable 25 cm<sup>2</sup> tissue culture flasks (Nalgene, Nunc), containing 1.5 ml of freshly made medium. To initiate the cultures, the medium was prepared using 70% (w/v) artificial seawater (S = 28‰), containing 10% (w/v) 2x Leibovitz's L-15 medium, 10% foetal bovine serum (FBS; Gibco BRL), glucose (1 g/l) (26), L-proline (0.06 g/l) (22), and 1% penicillin-streptomycin (10 × 4 IU/ml – 10 × 2 µg/ml). Based on previous experience in the laboratory, 5% (v/v) *N. norvegicus* serum was also added to the medium, in order to obtain rapid attachment and growth from the explants. The osmolality of the medium was adjusted to 800 mOsm/kg by means of a Roebbling Camlab osmometer using 6–7% of Chen Salts (NaCl 102.4 g/l, KCl 1.8 g/l, MgSO<sub>4</sub> 10.8 g/l, CaCl<sub>2</sub> 5.1 g/l and MgCl<sub>2</sub> 11.8 g/l). The pH was adjusted to 7.4. The medium was filtered through a vacuum filter using 0.22 µm pore size cellulose membranes (Corning – Sigma) prior to use. The cultures were transported to the Dublin Institute of Technology, incubated at 16°C, and observed daily using an inverted Olympus CK microscope at magnifications of 100x and 200x. Once the cultures were initiated, fresh medium (1 ml) without *N. norvegicus* serum was added on the next day of seeding. At day four, the final volume of medium was made up to 4.5 ml. Cultures were used within one week.



The pronephros, readily identified by its dark red colour, was dissected from the rainbow trout in a sterile cabinet. Mesothelium and fatty tissue were removed and the tissues were subsequently placed in 3–4 baths of medium in order to dilute / remove any microbes that may be present. The explants were placed into sterile disposable 24cm<sup>2</sup> tissue culture flasks (Nalgene, Nunc), containing 2ml of RPMI medium supplemented with 10% fetal calf, 10% horse serum, 20 mM L-glutamine and 1% penicillin-streptomycin. The explants were incubated at 18°C without media change for 16 days.

### **2.3. Irradiation**

Explant cultures were irradiated in situ on the culture flasks. The dose was delivered at room temperature using a Cobalt 60 teletherapy unit at a flask to source distance of 80cm. Under these conditions, the dose rate was approximately 1.9Gy/min during these experiments. After irradiation, the cultures were replaced in a refrigerated incubator set at 16°C for *Nephrops* cultures and 18°C for rainbow trout cultures and maintained there until they were processed 4–7 days later. The doses used for these experiments were 0.5Gy and 5Gy.

### **2.4. Transmission electron microscopy**

The cultures were fixed 4–7 days post irradiation in 2.5% (v/v) glutaraldehyde in 0.1 M phosphate buffer for 1 hour, postfixed in 1% osmium tetroxide in 0.1M phosphate buffer for a further hour, dehydrated in ascending grades of ethanol, and subsequently embedded in epoxy resin. Thick sections for light microscopy (1µm) were cut *en face* parallel to the plane of growth with a glass knife and stained with toluidine blue and examined using a Leica DMLB light microscope. Thin sections (60nm) were then cut *en face* with a diamond knife, stained with uranyl acetate and lead citrate, and examined using a JOEL 2000 transmission electron microscope. Three areas of the cellular outgrowth from three replicate cultures were examined at the ultrastructural level.

## **3. RESULTS**

### **3.1. Nephrops norvegicus haematopoietic tissue**

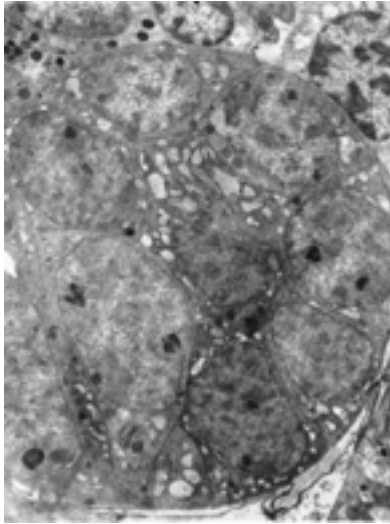
Haematopoietic tissue was composed generally of lobules of densely packed cells including stem cells and maturing hemocytes surrounded by an outer intimal layer (Figure 1). Primary cultures of haematopoietic tissue were found to contain similar cell types as the intact tissue although granulocyte type cells were more common than hyaline hemocyte type cells (Figure 2). Nuclei were regularly shaped and the cells were vacuolated and contained mitochondria with tubulo – vesicular cristae. Ribosomes and α glycogen were freely distributed in the cytosol.

Apoptotic bodies were common in the primary cultures irradiated at 0.5 Gy (Figure 3). Abnormal mitochondrial – RER complexes were also observed frequently. In the cultures irradiated at 5 Gy, a thorough disintegration of the cellular cytoplasm was observed with only occasional disrupted mitochondria and truncated RER evident (Figure 4).

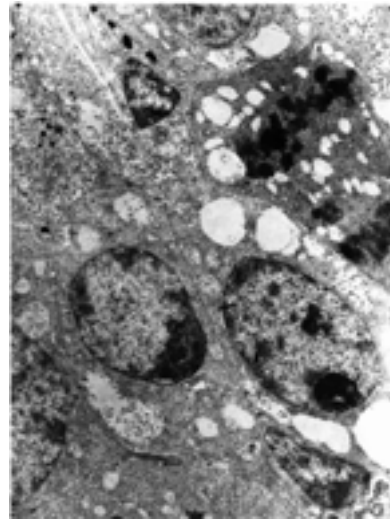
### **3.2. Rainbow trout pronephros**

Tissue from the rainbow trout pronephros was mainly composed of lymphocytes, monocytes, granulocytes and erythrocytes among the cell processes of reticular cells (Figure 5). Primary cultures of pronephros were found to contain similar cell types as the intact tissue (Figure 6). Granulocytes, containing both small and large granules, were found to be of a regular round shape and of a uniform size, approx. 10 µm (Figure 7).

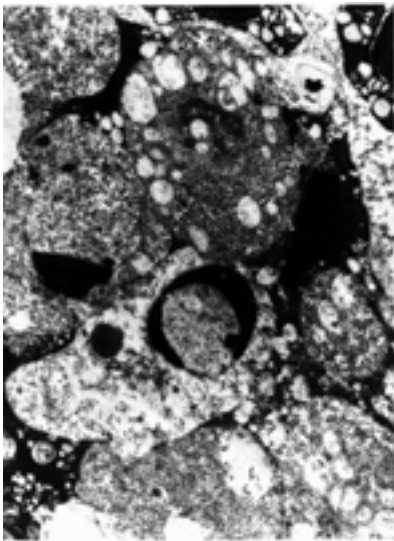
Granulocytes in the cultures irradiated at 0.5 Gy and 5 Gy were consistently found to show an elongation towards a spindle shape and there was considerable variation in size, ranging from 5–20 µm (Figure 8). This effect was more pronounced in the cultures irradiated at the higher dose of 5 Gy.



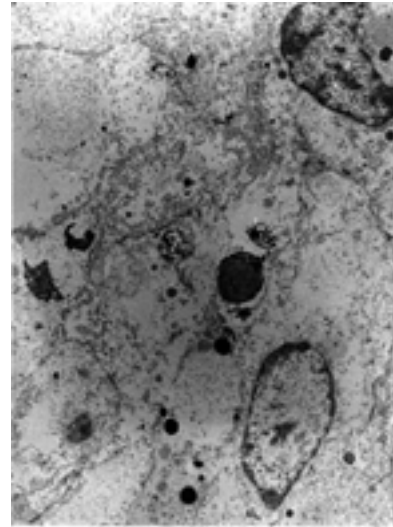
*FIG. 1. Haematopoietic tissue from Nephrops norvegicus showing a variety of cell types surrounded by an outer intimal layer.*



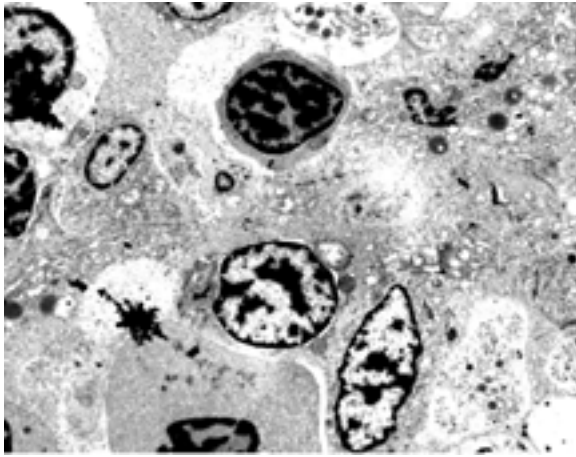
*FIG. 2. Primary culture of Nephrops norvegicus haematopoietic tissue showing similar cell types as the intact tissue. Note mitotic cell on upper right.*



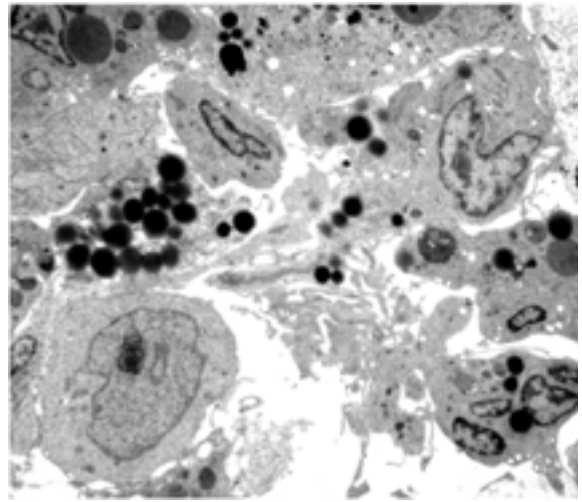
*FIG. 3. Primary culture of Nephrops norvegicus haematopoietic tissue irradiated at 0.5 Gy showing apoptotic bodies.*



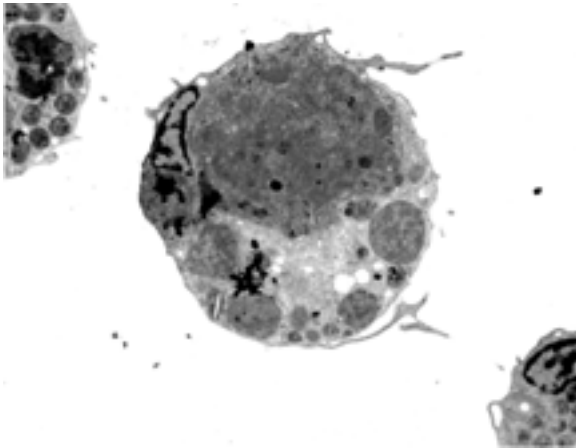
*FIG. 4. Primary culture of Nephrops norvegicus haematopoietic tissue irradiated at 5 Gy showing a thorough disintegration of the cellular cytoplasm.*



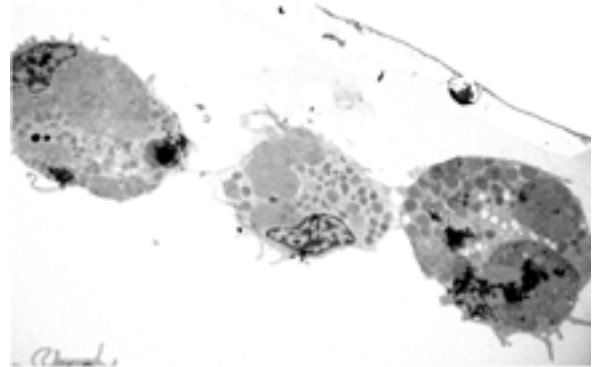
*FIG. 5. Tissue from the rainbow trout pronephros showing lymphocytes, monocytes, granulocytes and erythrocytes among the cell processes of reticular cells.*



*FIG. 6. Primary culture of rainbow trout pronephros showing similar cell types as the intact tissue.*



*FIG. 7. Primary culture of rainbow trout pronephros showing granulocytes, containing both small and large granules.*



*FIG. 8. Primary culture of rainbow trout pronephros irradiated at 5 Gy showing elongated granulocytes with considerable variation in size.*

#### 4. DISCUSSION

Ultrastructural changes are often detectable at lower doses of toxicants where conventional histological examination fails to reveal abnormalities. Electron microscopy allows observation of very early morphological changes whereas histology usually reveals only more severe changes.

In the *Nephrops* cultures exposed to 0.5 Gy  $\gamma$  radiation, changes in cytoplasmic organelles and frequent apoptotic bodies were observed. At 5 Gy, a thorough disintegration of the cellular cytoplasm was seen. The most common feature of the irradiated rainbow trout pronephros cultures was the shape change in the granulocytes. This was apparent at 0.5 Gy but much more pronounced at 5 Gy. There were also significant variations in the size of the granulocytes in the irradiated pronephros, again more pronounced at the higher dose.

Widespread damage or cytoplasmic disintegration of the scale seen in the *Nephrops* cultures was not seen in previous ultrastructural studies carried out in this laboratory with irradiated human primary urothelial cultures [11, 12]. Irregular nuclei, accumulations of lysosomes and lipid droplets were common in human urothelial cultures irradiated at 5Gy.

The shape and size changes seen in the rainbow trout cultures are most likely related to cytoskeletal damage.

The high levels of apoptosis seen in the cultures irradiated at the lower dose suggest a protective response in the *Nephrops* cultures. Terminally damaged cells may be removed by programmed cell death before they pose a threat to the organism as a whole. Selection and proliferation of healthy cells could account for the apparent radioresistance previously reported for invertebrates.

In conclusion, the primary cultures from both rainbow trout and *Nephrops norvegicus* displayed pronounced morphological damage following cobalt 60 gamma irradiation. However, cells from the haematopoietic tissue of *Nephrops norvegicus* appeared to be considerably more radiosensitive than cells from the pronephros of rainbow trout. These results may have important implications for protection of the environment and species other than man.

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# A model for exploring the impact of radiation on fish populations

**D.S. Woodhead**

The Centre for Environment, Fisheries and Aquaculture Science, Lowestoft Laboratory,  
Lowestoft, United Kingdom

**Abstract.** In the general context of protecting the biotic environment, it is frequently the population of organisms at which it is intended that protective measures should be directed. It is well-established, however, that exposure to low-level, chronic irradiation can affect the survival and reproductive capacity of individual plants and animals. Although it is clear that such effects can have implications for the well-being of the populations, it is less apparent (and almost certainly not the case) that the radiation can affect population attributes directly, i.e., without the mediation of effects in individuals. The problem is, therefore, to establish criteria for the limitation of effects in individuals such that the population will also be sufficiently protected. Before this can be achieved, it is necessary to have a means of relating the known effects of radiation in individuals with their possible consequences for the population. A simple Leslie matrix population model approach has been developed to investigate how the effects of radiation in individuals may propagate to produce (or not) a response at the population level. Different species can have different reproductive strategies and life cycles, and may, therefore, respond differently to the same degree of radiation effect on survival and reproductive capacity. It is of interest to investigate their possible responses at the population level, and the matrix population model approach is here applied to two somewhat contrasting fish species – the plaice (*Pleuronectes platessa*) and the thornback ray (*Raja clavata*). The results from the models appear to confirm the relative sensitivities of the two populations, as might be predicted on the basis of their life cycles and reproductive strategies, to the possible effects of radiation on individual fertility, fecundity and mortality.

## 1. INTRODUCTION

It is frequently asserted that measures to protect the environment – the flora and fauna – from the contaminants arising from human activities should be focussed on the population level in the biological hierarchy. It is also the case, however, that the effects of the contaminants mainly, if not entirely, develop from processes that take place in individual organisms. To the extent that the effects of the contaminants in individuals influence the population attributes of age-dependent survival (through increased morbidity and mortality) and age-dependent reproductive capacity (through reductions in fertility (gamete production) and fecundity (the production of viable offspring)), there is a link, probably complex and non-linear, between the individual and the population responses. A third category of effect – an increase in mutation rate – will, for the present, be assumed to be captured by the changes in survival and reproductive capacity; it is recognized, however, that significant contaminant-induced changes in the gene pool would represent a change in the character of the population that should be considered in its own right.

Given – and it is a reasonable assumption – that the population attributes are not directly impacted by radiation, and that the population is to be protected, the question is: what degree of limitation on the direct effects in individuals, i.e., measures to protect the individual, implies no significant consequent impact at the population level? The response is often that there can be no significant effects on the population if there are no significant effects in individuals, although this response, however apparently reasonable, hardly answers the question in a transparent and scientific manner. In practice, the question can be addressed through the development and investigation of the behaviour of models of relevant populations. A simple (not to say, simplistic) Leslie matrix model approach has been developed as an experimental tool to investigate the possible responses of a population to radiation exposure as mediated through the effects at the individual level [1]. This has already been applied to a plaice (*Pleuronectes platessa*) population. This fish can mature at age two years, lives for about 7 years (in the Irish Sea), and each individual mature female can produce many thousands of eggs. This is a life strategy that is often said to be relatively resilient to environmental change (including contaminant effects). A contrasting strategy of later maturity, longer reproductive life and the production of fewer (but more protected) eggs and more highly developed neonates, as exemplified by the thornback ray (*Raja clavata*), is often said to be more sensitive to environmental change. It is the

purpose of this paper to apply the Leslie matrix model approach to these contrasting fish populations, and to compare the possible responses of the populations to radiation-induced effects on individual survival and reproductive capacity.

## 2. THE POPULATION MODEL

The development of the matrix population model for the plaice has already been presented in some detail elsewhere [1]. In brief, the general matrix equation:

$$\begin{pmatrix} n_1 \\ n_2 \\ n_3 \\ \vdots \\ n_s \end{pmatrix} (t+1) = \begin{pmatrix} F_1 & F_2 & F_3 & \dots & F_s \\ P_1 & 0 & 0 & \dots & 0 \\ 0 & P_2 & 0 & \dots & 0 \\ \vdots & \vdots & \dots & \dots & \vdots \\ 0 & 0 & \dots & P_{s-1} & 0 \end{pmatrix} \begin{pmatrix} n_1 \\ n_2 \\ n_3 \\ \vdots \\ n_s \end{pmatrix} (t) \quad (1)$$

allows the initial, age-dependent population at time (t) to be projected forward to time (t+1), provided that the age-dependent fecundities ( $F_i$ ) and survival probabilities ( $P_i$ ) can be estimated (see [2] for a full discussion of matrix models). (In this paper, fecundity is taken to be the production of viable neonates potentially capable of surviving to reproductive maturity; fertility is taken to be a quantitative measure of the production of viable gametes, i.e., sperm and ova that can combine to produce zygotes that have the capacity to pass through embryonic development to produce larvae or post-natal individuals.) For the two fish species considered here, it may be assumed that the time step (t) to (t+1) is one year, as the fish, once mature, spawn annually. For the plaice, this occurs in the period February – April and the fertilised eggs develop in the water column and hatch out after a period of about 20 days; this is followed by a planktonic stage of about 60 days at which time the larvae metamorphose and adopt the adult benthic habit as Group-0 juveniles. There are, thus, three stages, of differing durations and with differing survival probabilities, in the first calendar year of the life of the plaice; of these, the first two must be taken into account in the estimation of the  $F_i$ . For the thornback ray, spawning occurs over the period February to September and an individual female may, depending on size, produce up to 140 eggs, usually in pairs on alternate days. The period of embryonic development is temperature-dependent and can last for 112–144 days. The newly-hatched rays are fully-developed, but miniature, versions of the adults; for the ray, therefore, there is just one stage to be included in the estimation of the  $F_i$ . Although, for both species, the detailed timings are variable, for the purpose of the population model they have been reduced to the standard parameters given in Tables 1 and 2.

### 2.1. The estimation of $F_i$

Fish grow at a variable rates through their lives, and at a given age there is likely to be a range of sizes for each species. The number of eggs produced by a female is, however, more likely to be correlated with size rather than age. For the purpose of the model, therefore, it has been assumed that size does correlate with age so that the  $F_i$  can be estimated for each annual time step.

Table1. Population parameters to implement the Leslie matrix projection model for plaice

Stage	Duration years	$e\mu_1$	$e\mu_2$	$i\mu_1$	$i\mu_2$	Coefficient of natural mortality	Increase of coefficient of natural mortality with age	Coefficient of fishing mortality	$s\mu_2$	Mean length cm	Initial number of fish <sup>a</sup>	Proportion mature
Developing egg	0.055	30 ± 3	(5 ± 0.5) x10 <sup>-4</sup>	-	-	-	-	-	-	-	-	-
Planktonic larvae	0.165	-	-	30 ± 3	(1 ± 0.1) x10 <sup>-3</sup>	-	-	-	-	-	-	-
Post meta- morphic Gp-0	0.780	-	-	-	-	3	0.01	0.00	1.0E-10	-	(2.50E+07)	0.0
Group-I	1	-	-	-	-	0.3 ± 0.03	0.02	0.00	1.0E-07	-	(5.62E+06)	0.0
Group-II	1	-	-	-	-	0.3 ± 0.03	0.03	0.70	7.0E-08	21.9	(1.35E+06)	0.5
Group-III	1	-	-	-	-	0.3 ± 0.03	0.04	0.71	1.0E-06	26.9	2.72E+05	0.9
Group-IV	1	-	-	-	-	0.3 ± 0.03	0.05	0.72	5.0E-06	31.9	1.15E+05	1.0
Group-V	1	-	-	-	-	0.3 ± 0.03	0.06	0.73	1.0E-05	37.0	1.74E+04	1.0
Group-VI	1	-	-	-	-	0.3 ± 0.03	0.07	0.74	5.0E-05	42.0	3.20E+03	1.0
Group-VII	1	-	-	-	-	0.3 ± 0.03	0.08	0.75	1.0E-04	45.0	1.30E+03	1.0

<sup>a</sup> The numbers in parenthesis have been extrapolated from the available information.



Table 2. Population parameters to implement the Leslie matrix projection model for the thornback ray

Stage	Duration years	$\mu_1$	$\mu_2$	$\mu_1$	$\mu_2$	Coefficient of natural mortality	Increase of coefficient of natural mortality with age	Coefficient of fishing mortality	$\mu_2$	Mean length cm	Initial number of fish <sup>a</sup>	Proportion mature
Developing egg	0.334	4.53 $\pm 0.453$	(6.0 $\pm$ 0.60) $\times 10^{-6}$			-	-	-	-	-	-	-
Group-0	0.252	-	-	3.77 $\pm$ 0.377	(6.6 $\pm$ 0.66) $\times 10^{-5}$	-	-	-	-	12.0	-	-
Group-I	1	-	-	-	-	1.57 $\pm$ 0.157	0.001	0.000	2.1E-07	14.9	(482381)	0.00
Group-II	1	-	-	-	-	0.717 $\pm$ 0.0717	0.002	0.000	8.8E-07	25.6	(172210)	0.00
Group-III	1	-	-	-	-	0.168 $\pm$ 0.0168	0.003	0.000	1.7E-06	35.3	61481	0.00
Group-IV	1	-	-	-	-	0.0733 $\pm$ 0.0073	0.004	0.000	6.5E-08	44.3	26240	0.00
Group-V	1	-	-	-	-	0.0733 $\pm$ 0.0073	0.005	0.012	6.5E-08	52.4	15596	0.00
Group-VI	1	-	-	-	-	0.0733 $\pm$ 0.0073	0.006	0.012	6.5E-08	59.9	12785	0.25
Group-VII	1	-	-	-	-	0.0733 $\pm$ 0.0073	0.007	0.012	6.5E-08	66.7	10324	0.50
Group-VIII	1	-	-	-	-	0.0733 $\pm$ 0.0073	0.008	0.012	6.5E-08	73.0	8373	0.75
Group-IX	1	-	-	-	-	0.0733 $\pm$ 0.0073	0.009	0.012	6.5E-08	78.7	5664	1.00
Group-X	1	-	-	-	-	0.243 $\pm$ 0.0243	0.010	0.036	1.1E-06	83.9	3058	1.00
Group-XI	1	-	-	-	-	0.243 $\pm$ 0.0243	0.011	0.036	1.1E-06	88.6	1165	1.00
Group-XII	1	-	-	-	-	0.243 $\pm$ 0.0243	0.012	0.036	1.1E-06	93.0	422	1.00
Group-XIII	1	-	-	-	-	0.243 $\pm$ 0.0243	0.013	0.036	1.1E-06	97.0	349	1.00
Group-XIV	1	-	-	-	-	0.243 $\pm$ 0.0243	0.014	0.036	1.1E-06	100.6	160	1.00
Group-XV	1	-	-	-	-	0.243 $\pm$ 0.0243	0.015	0.036	1.1E-06	103.9	77	1.00

Table 2. continued: Population parameters to implement the Leslie matrix projection model for the thornback ray

Stage	Duration years	$e\mu_1$	$e\mu_2$	$o\mu_1$	$o\mu_2$	Coefficient of natural mortality	Increase of coefficient of natural mortality with age	Coefficient of fishing mortality	$s\mu_2$	Mean length cm	Initial number of fish <sup>a</sup>	Proportion mature
Group-XVI	1	-	-	-	-	0.243 ± 0.0243	0.016	0.036	1.1E-06	106.9	37	1.00
Group-XVII	1	-	-	-	-	0.243 ± 0.0243	0.017	0.036	1.1E-06	109.7	18	1.00
Group-XVIII	1	-	-	-	-	0.243 ± 0.0243	0.018	0.036	1.1E-06	112.3	8	1.00
Group-XIX	1	-	-	-	-	0.243 ± 0.0243	0.019	0.036	1.1E-06	114.6	4	1.00
Group-XX	1	-	-	-	-	0.243 ± 0.0243	0.020	0.036	1.1E-06	116.7	2	1.00
Group-XXI	1	-	-	-	-	0.243 ± 0.0243	0.021	0.036	1.1E-06	118.6	1	1.00
Group-XXII	1	-	-	-	-	0.243 ± 0.0243	0.022	0.036	1.1E-06	120.4	0	1.00
Group-XXIII	1	-	-	-	-	0.243 ± 0.0243	0.023	0.036	1.1E-06	122.0	0	1.00
Group-XXIV	1	-	-	-	-	0.243 ± 0.0243	0.024	0.036	1.1E-06	123.5	0	1.00
Group-XXV	1	-	-	-	-	0.243 ± 0.0243	0.025	0.036	1.1E-06	124.8	0	1.00
Group-XXVI	1	-	-	-	-	0.243 ± 0.0243	0.026	0.036	1.1E-06	126.1	0	1.00
Group-XXVII	1	-	-	-	-	0.243 ± 0.0243	0.027	0.036	1.1E-06	127.2	0	1.00
Group-XXVIII	1	-	-	-	-	0.243 ± 0.0243	0.028	0.036	1.1E-06	128.2	0	1.00
Group-XXIX	1	-	-	-	-	0.243 ± 0.0243	0.029	0.036	1.1E-06	129.2	0	1.00
Group-XXX	1	-	-	-	-	0.243 ± 0.0243	0.030	0.036	1.1E-06	130.0	0	1.00

<sup>a</sup>The numbers in parenthesis have been extrapolated from the available information.

### 2.1.1. The plaice

For the plaice, the available data for the northeast Irish Sea have been used to develop the presumed number-at-age/length (see Table 1) and weight-at-length [1]. Based on data for plaice populations in the North Sea [3], it has been assumed that the regression equation:

$$\ln(F) = -C + 3.11 \ln(L) - 0.218 \ln(t) \quad (2)$$

would be a reasonable basis for estimating the fertility  $F$  (in thousands of eggs per female) from the length  $L$  cm and age  $t$  years; the regression coefficient  $C$  showed substantial variability between years (range 5.996–6.375). To introduce the influence of stochastic environmental conditions into the structure of the model, the values of the coefficients in eq.(2), for each age (length) class in each year, have been randomly taken from the normal distributions:

$$C = 6.0 \pm 0.1; 3.11 \pm 0.11; \text{ and, } 0.218 \pm 0.11,$$

to provide an estimate of the total variability in fertility (note that these distributions have no empirical basis). It has been assumed that the population of female plaice matures progressively from Group-II to Group-IV (see Table 1). During the embryonic and larval development periods, the plaice will experience variable survival probabilities that arise from both inherent and environmental factors that can be independent of, or may be dependent upon, density. In general, the survival at any stage may be modelled as [4]:

$$\frac{dN(t)}{dt} = -\{s\mu_1 + s\mu_2 N(t)\}N(t) \quad (3)$$

where  $s\mu_1$  and  $s\mu_2$  are the density-independent, and density-dependent, survival coefficients for stage  $S$ , respectively. The general solution to eq.(3) provides the number of survivors from stage  $S_{(i)}(t_{(i)})$  to stage  $S_{(i+1)}(t_{(i+1)})$ :

$$N_{S_{(i+1)}}(t_{(i+1)}) = \frac{1}{\frac{S_{(i)}\mu_2}{S_{(i)}\mu_1} [\exp\{S_{(i)}\mu_1 \times (t_{(i+1)} - t_{(i)})\} - 1] + \frac{\exp\{S_{(i)}\mu_1 \times (t_{(i+1)} - t_{(i)})\}}{N_{S_{(i)}}(t_{(i)})}} \quad (4)$$

This equation is applied sequentially to the egg and larval stages to estimate the values of  $F_i$  for each adult age-group. To introduce the element of environmental variability, the relevant parameter values were drawn randomly from the normal distributions adopted to model the plaice egg and larval stages as given in Table 1 (see [1] for full details of the implementation of the population model).

### 2.1.2. The thornback ray

The literature on the growth rate and reproductive biology of the thornback ray is limited and not entirely consistent; the population model will, therefore, be rather more speculative than that for the plaice. The available data have been used to parameterise the von Bertalanffy growth relationship for the thornback ray and, assuming an annual time step, provide estimates of length (cm.)-at-age:

$$L(t+1) = L(t) + \{139 - L(t)\} \{1 - \exp(-0.09)\} \quad (5)$$

where 139 cm. is the asymptotic ultimate length, and 0.09 is the annual growth coefficient [5]. These lengths have been converted to weight (kg) using the relationship given by Holden [6]:

$$W(t) = 3.26 \times 10^{-6} \times L(t)^{3.19} \quad (6)$$

The annual egg production per female has been estimated using the expression developed by Holden [7], adjusted to give an overall lower average value of 100 as suggested by Ryland & Ajayi [5]:

$$N_e = 1.19 L(t) - 10.9 \quad (7)$$

The population of female thornback rays has been assumed to mature progressively from Group-VI to Group-IX (see Table 2). For the thornback ray, there are two stages to be considered in the first year of life for the estimation of  $F_i$  – the developing egg and the Gp-0 hatchlings – and the relevant parameter values, and their distributions (to introduce stochasticity into the model), for application in eq.4, are given in Table 2.

## 2.2. The estimation of $P_i$

The survival of the post-metamorphic plaice, and the thornback rays, depends on the influence of natural (density-independent and -dependent) factors, and, at ages greater than 2 years (plaice) and 5 years (thornback ray), the impact of fishing. The survival for any year  $i$  (or stage  $S_{(i)}$ ) is obtained from eq.4:

$$P_{S_{(i)}} = \frac{N_{S_{(i+1)}}(t_{(i+1)})}{N_{S_{(i)}}t_{(i)}} \quad (8)$$

$$= \frac{1}{\frac{S_{(i)}\mu_2}{S_{(i)}\mu_1} N_{S_{(i)}}(t_{(i)})[\exp\{S_{(i)}\mu_1 \times (t_{(i+1)} - t_{(i)})\} - 1] + \frac{\exp\{S_{(i)}\mu_1 \times (t_{(i+1)} - t_{(i)})\}}{t_{(i)}}}$$

As discussed previously [1], the density-independent mortality has been taken to have three components: a stochastic coefficient of natural mortality; a small coefficient of natural mortality that increases with age; and, a coefficient of fishing mortality that increases slightly with age for the plaice and has two different, and age-dependent, values for the thornback rays. In the operation of the population model, these three components are aggregated to give the single parameter  $S_{(i)}\mu_1$ . The density-dependent coefficient,  $S_{(i)}\mu_2$ , is a singular component. The values adopted for the various parameters, developed from the information available in the literature, are given in Tables 1 and 2 (for the plaice see [1], and for the ray [5]).

## 3. THE IMPLEMENTATION OF THE MODEL

In the development of the matrix population model for the plaice [1], it was noted that, although some of the relationships and parameter values were derived from data concerning real populations, others were not. The model could not, therefore, be taken to provide an accurate representation, in all details, of the real population in the contaminated region of the northeast Irish Sea. The same is undoubtedly true for the thornback ray population model. For both species, it has been assumed that the starting population amounts to approximately 250 tonnes, with corresponding number-at-age and length/weight distributions developed from the available data relating to commercial and/or research vessel catches [5, 8]. The initial numbers of fish in each age group are given in Tables 1 and 2.

## 4. THE BEHAVIOUR OF THE MODEL POPULATIONS

The time-dependent development of the two populations, in terms of the spawning biomass and the number of recruits to the spawning biomass from the spawning occurring 2 (plaice) and 6 (ray) years previously, has been projected. Figure 1 shows the evolution of the plaice and ray populations, both with, and without, the impact of fishing. In the absence of fishing, the number of Gp-II plaice recruits increases by a factor of ~3.5, and the spawning biomass increases from 260 te to about 1100 te; similarly, the number of Gp-VI ray recruits increases by a factor of ~1.5, and the spawning biomass increases from 130 te to about 500 te. With the applied fishing pressure, the number of Gp-II plaice recruits approximately doubles, and the plaice spawning biomass increases marginally to ~350 te; the number of Gp-VI ray recruits increases marginally, and the spawning biomass increases to ~350 te. The initial sharp rise and decline in the ray spawning biomass is probably the consequence of a mismatch between the age/size distribution of the starting population and the aggregate effect of the parameters selected to implement the population model, together with the 6 year generation time; it is clear, however, that the population stabilizes after about 30 years of evolution.

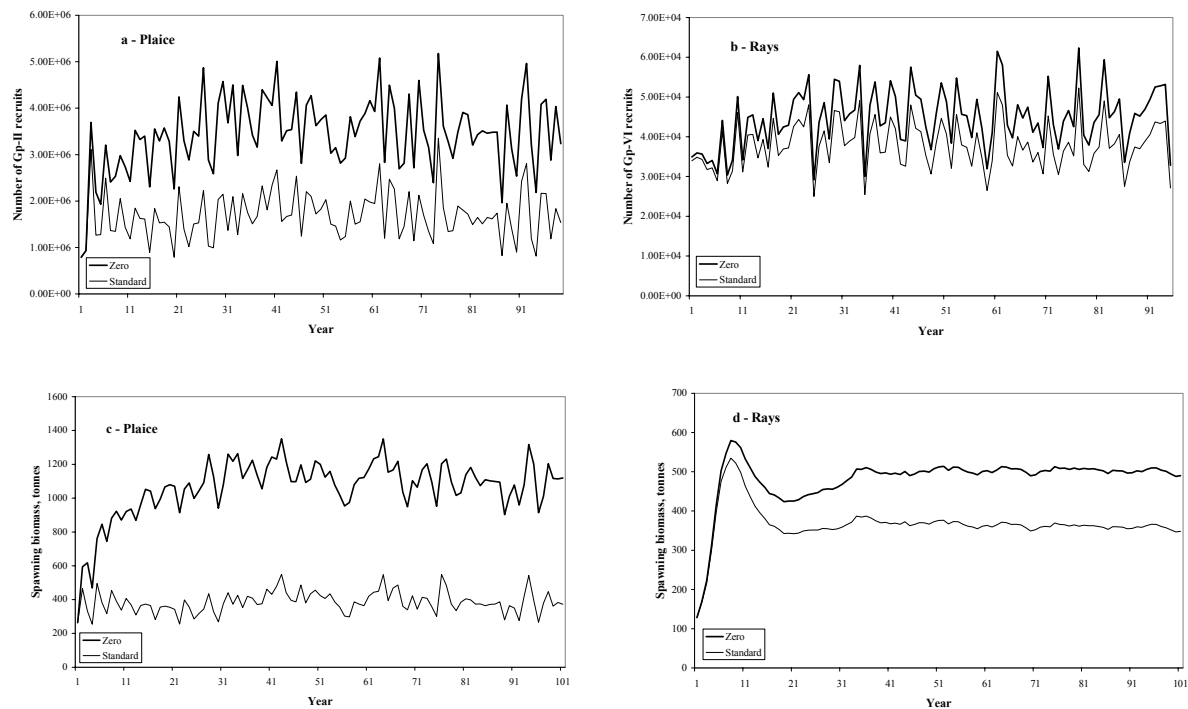


FIG 1. The evolution of the plaice and thornback ray populations with zero and standard fishing pressure. a, c: the recruitment of Gp-II plaice and Gp-VII thornback rays; b,d: the spawning biomass of plaice and thornback rays.

#### 4.1. The evolution of the populations

In the absence of detailed data on the effects of chronic irradiation on the important population attributes – fertility, fecundity and mortality – the approach has been to examine the effects of 0.5, 1, 2, 5 and 10% reductions in fertility (egg output), embryonic survival, and final age-dependent survival.

##### 4.1.1. Fertility

The effects of the reductions in egg production are shown in Figure 2a. In terms of the consequent reductions in both the number of recruits to the breeding population and the spawning biomass, the plaice appears to be rather more sensitive than the ray to a given reduction in egg production. This response is somewhat puzzling, but it seems to imply that, within the parameter spaces applied to each model population, the factors acting after the production of the eggs are more significant in influencing the outcome. The simple comparative magnitudes in the reductions in the spawning biomass and numbers of recruits are not, however, clear indicators of the fates of the respective populations. A closer examination of the trends in the spawning biomass, in terms of the mean annual stochastic rate of growth (see Table 3) indicates that, for a 10% reduction in egg production, the ray population is in terminal decline (a mean annual growth rate < 1); in contrast, the plaice population retains growth potential, although progressively reduced, at all levels of reduced fertility. (It should be noted that, although the mean stochastic growth rate may be > 1, the population does not actually grow indefinitely due to the non-linear interactions between the various density-dependent and density-independent factors.)

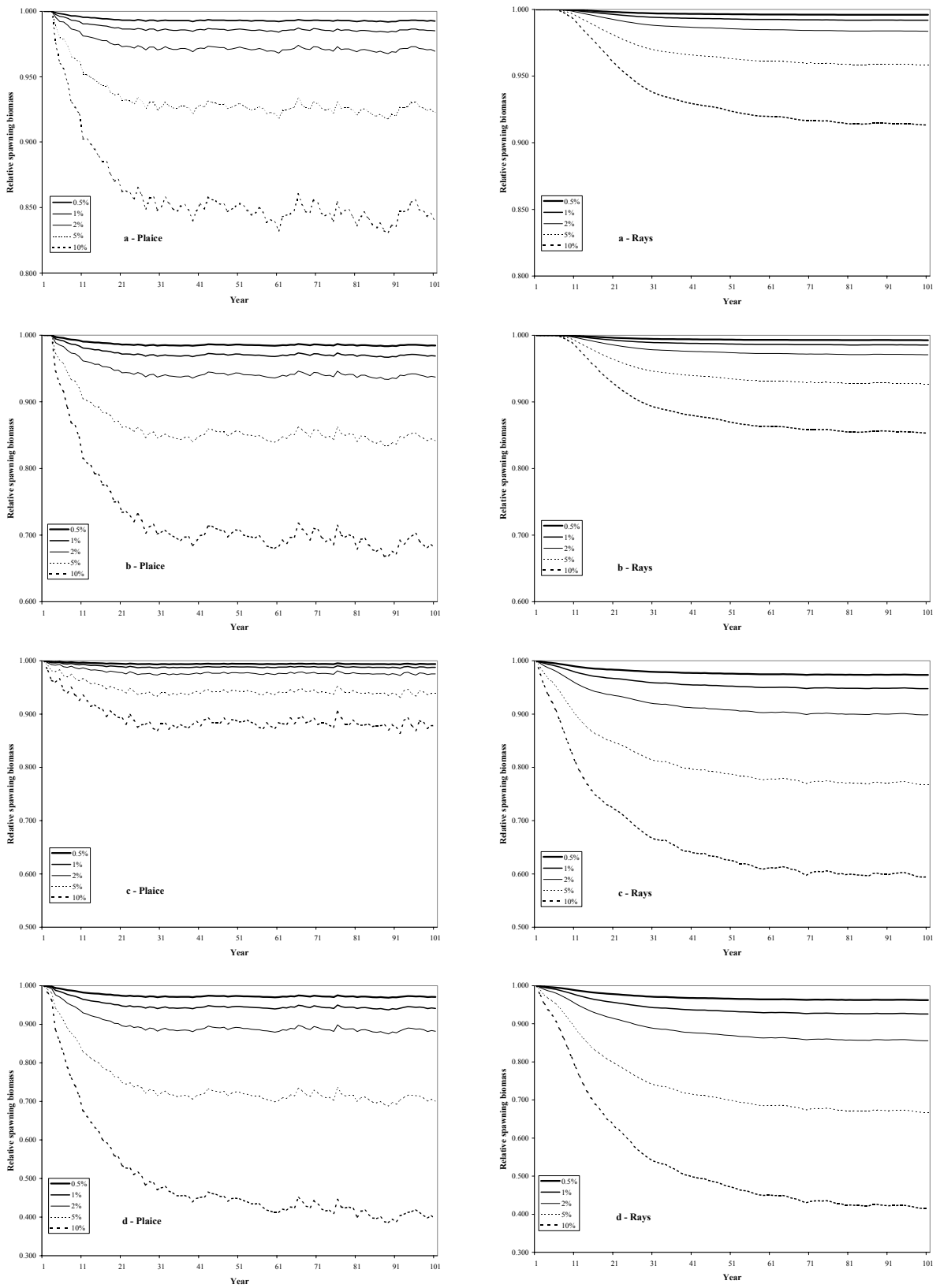


FIG. 2. The evolution of the plaice and thornback ray populations with varying reductions in a: egg production per female; b: embryo survival to hatching; c: final age-dependent survival; and, d: the combined stresses.

TABLE 3. MEAN ANNUAL STOCHASTIC RATE OF SPAWNING BIOMASS GROWTH OVER THE PERIOD 30–100 YEARS, WITH THE INDICATED % CHANGES IN THE POPULATION ATTRIBUTES

<b>Plaice</b>						
Baseline	1.00165	0.5%	1%	2%	5%	10%
Reduced egg production		1.00163	1.00161	1.00157	1.00144	1.00114
Increased coefficient of egg mortality		1.00162	1.00158	1.00150	1.00119	1.00036
Increased final age-dependent mortality		1.00164	1.00163	1.00161	1.00155	1.00143
Combined effects		1.00159	1.00152	1.00134	1.00053	0.99729
<b>Thornback ray</b>						
Baseline	1.00020					
Reduced egg production		1.00019	1.00017	1.00014	1.00004	0.99983
Increased coefficient of egg mortality		1.00018	1.00016	1.00011	0.99994	0.99961
Increased final age-dependent mortality		1.00013	1.00005	0.99990	0.99944	0.99868
Combined effects		1.00009	0.99996	0.99971	0.99881	0.99660

#### 4.1.2. Fecundity

The effects of the reduced embryonic survival to egg hatch are shown in Figure 2b. Again, in terms of the degree of impact on the two measures of population evolution, the plaice appears to be more sensitive than the ray to a given reduction in neonate production. The effects of given reductions in fecundity on the growth potential of the populations is greater than is the case for comparable reductions in fertility (Table 3). The plaice, however, still retains a progressively reduced potential for growth at all levels of reduced fecundity, whereas the thornback ray population is in decline at both 5% and 10% increases in the coefficient of egg mortality (Table 3). It may be concluded, therefore, that the possible effects of radiation may be more significant at this stage of development.

#### 4.1.3. Mortality

On the assumption that the accumulation of radiation exposure over the lifetime of the fish would have a progressive effect on their possible mortality, the age-dependent coefficients of natural mortality have been proportionately increased over the lifetime of the two fish to give the final increases of 0.5, 1, 2, 5 and 10% for Gp-VII plaice and Gp-XXX rays. The effects of these increases in age-dependent mortality are shown in Figure 2c. In contrast to the outcome of effects on the previous two attributes, the ray is substantially more sensitive to increased age-dependent mortality than the plaice. Indeed, in terms of a given degree of impact, an increase in age-dependent mortality has the least effect on the plaice population as compared with reductions in fertility and fecundity. The opposite is true for the thornback ray and its population is in decline at increases in final age-dependent mortality  $\geq 2\%$  (Table 3). Given that the loss of individuals of reproductive age also has the implicit effect of reducing the aggregate fertilities and fecundities of the populations, and that this applies to a greater extent to the longer-lived thornback ray, such an outcome might not be unexpected.

#### 4.1.4. Combined effects

Of course, the effects of radiation will impact all three of the population attributes considered above, although not to the same degree either for each attribute or for each species. There is not, however, sufficient information to quantify the actual relative degrees of effect, and the simple approach has been adopted of considering the aggregated consequences of the same degree of effect on each of the

three attributes. The effects on the evolution of the spawning biomass of the two populations are shown in Figure 2d. Overall, the thornback ray population is more sensitive to the cumulative applied stresses than is the plaice; the two populations are in decline at changes in the attributes  $\geq 1\%$  and  $\geq 10\%$ , respectively.

## 5. CONCLUSIONS

The long-term evolution of the two fish populations, as described by the matrix population models described here, appears to confirm their presumed relative sensitivities to the possible effects of radiation-induced changes in individual fertility, fecundity and mortality. This conclusion is, however, contingent on the degree to which the present parameterisation and implementation of the models accurately describes the features of real populations. Nevertheless, the models do provide a helpful and practical means of exploring the implications, for populations, of radiation damage in the constituent individuals, and effort could be usefully applied to improving the basic biological information required for their implementation.

## ACKNOWLEDGEMENTS

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# Long-term combined impact of $^{90}\text{Sr}$ and $\text{Pb}^{2+}$ on freshwater cladoceran

D.I. Gudkov, M.G. Mardarevich, L.S. Kipnis, A.V. Ponomaryov

Institute of Hydrobiology of the National Academy of Sciences of Ukraine, Kiev, Ukraine

**Abstract.** The effect of  $\text{Pb}^{2+}$  (0.01 and 0.1 mg l<sup>-1</sup>) and  $^{90}\text{Sr}$  (2E+01; 2E+03 and 2E+05 Bq l<sup>-1</sup>) in conditions of long-term two-factor experiment had a negative impact practically upon all biological parameters of freshwater cladoceran *Daphnia magna* shown in decrease of the productional characteristics, delay of growth rate and reduction of lifetime. The level of radiotoxic effect positively correlated with the contents in water of the researched agents and their combinations. At combined impact of the radiating and chemical factors the positive modifying effect of radionuclide on toxic influence of lead ions in range of  $^{90}\text{Sr}$  activity 2E+01–2E+03 Bq l<sup>-1</sup> is registered. It is supposed, that at exposure of ionising radiation in doses 7–70 cGy at daphnia occurs stimulation of protective reactions of organism, interfering realisation of damaging effect of the radiating factor. It is supposed that one of the radioprotective mechanisms is the activation of glutathione system and increase of level of sulfhydryl groups of endogenous glutathione, having the important significance for oxidation-reduction reactions and in fermentative repair of damages. However besides of radioprotective function glutathione takes part in synthesis of metallothioneins, which by chelation are capable to remove heavy metals from a biochemical exchange. Thus, metabolic changes, initially directed on realisation of radioprotective effect, at combined impact of the radiating and chemical factors can carry multifunctional character, being shown in positive modification of toxic influence caused by ions of heavy metals.

## 1. INTRODUCTION

In conditions of global pollution of biosphere the living organisms are exposed to long-term impact of the different man-caused factors. The important place among them occupies the sources of ionising radiation and chemical agents. At the same time the radionuclide contamination modified by a wide spectrum of substances of an organic and inorganic nature is capable to result in biological systems in the most unexpected effects.

In the present studies the biological aspects of separate and combined long-term impact of  $^{90}\text{Sr}$  and  $\text{Pb}^{2+}$  on freshwater cladoceran in condition of two-factor experiment are considered. The changes of lifetime as well as some of morphometrical and productional parameters of *Daphnia magna* Straus were used as criteria of a radiotoxic impact.

*Daphnia* are widely distributed zooplanktonic organisms also are one of a food object for majority species of freshwater fish. In comparison with other species of daphnia *Daphnia magna* Straus is easy enough for cultivation has the large sizes and high sensitivity to toxic substances of a various nature. These characteristics predetermined the use of this species as the most popular object in toxicological researches. Thanking to filtration way of feeding in daphnia organism occurs a concentrating of polluting substances, both from water and from filtered suspensions, that causes the increased reaction of shellfish on the presence of toxic pollutant at water environment [6, 10, 11]. In this connection the biotests with use of *D. magna* are standardised in a number of the countries of the world.

## 2. MATERIALS AND METHODS

The preparation of experiment was carried out taking into account of the recommendations stated in methodical manuals [2, 5, 12]. For experiment selected two-day time juvenile of the synchronised laboratory culture. As environment used missed through a layer of sorbent and settled water with the following hydrochemical parameters: hardness – 4–6 mg-eq l<sup>-1</sup>; dissolved oxygen – 7,8–8,2 mg l<sup>-1</sup>; pH – 7,7–8,2; temperature – 19–21°C. *Daphnia* was kept in 50 ml glasses with two individual in everyone. Experiment realised in 5 reiterations on one generation. Each two day carried out replacement of water. The size control of daphnia was carried out every 5 day by determination of distance from leading edge of a head up to the basis of a thorn. The daily diet of daphnia made up 200–300 thousand cells of *Chlorella* per 1 ml of water.

Estimation of the radiotoxic impact studied at the separate and combined influence of  $^{90}\text{Sr}$  with specific activity  $2\text{E}+01$ ,  $2\text{E}+03$  and  $2\text{E}+05$   $\text{Bq l}^{-1}$  and  $\text{Pb}^{2+}$  in concentration  $0,01$  and  $0,1$   $\text{mg l}^{-1}$ .

The criteria of biological impact were the following parameters: delays of sexual maturity; quantity of litters during life; quantity of juvenile in litters; total quantity of juvenile; dynamics of growth rate and lifetime of daphnia. Experiment carried out during 60 days – up to the moment of death of all researched individuals. The analysis of results of researches realised by comparison of similar parameters of experimental and controls individuals. The contribution of the radiation and chemical factors at change of biological parameters estimated with use of a method of the dispersion analysis [4] and program for statistical data processing "Statistica 5.0" (StatSoft Inc., USA). The absorbed dose rate from an external and internal irradiation of  $^{90}\text{Sr}$  determined according to a method [8].

### 3. RESULTS AND DISCUSSION

During experiment the authentic decrease of daphnia lifetime in all experimental concentration was observed (Figure 1). The value of this parameter was naturally reduced with increase of the contents in water of the researched agents both at separate and at combined impact. The minimal value of average life expectancy is registered at  $\text{Pb}^{2+}$  concentration  $0,1$   $\text{mg l}^{-1}$  (20 days).

If to compare combined impact of  $^{90}\text{Sr}$  and  $\text{Pb}^{2+}$ , the concentration of metal  $0,01$   $\text{mg l}^{-1}$  practically did not influence on lifetime of shellfish, which was observed in experimental radionuclide concentration. However practically all of  $^{90}\text{Sr}$  activity stimulated increase of daphnia lifetime in a combination with  $\text{Pb}^{2+}$  (in comparison with its separate impact). The exception were daphnia under impact of combined activity of  $^{90}\text{Sr}$   $2\text{E}+05$   $\text{Bq l}^{-1}$  and concentration of  $\text{Pb}^{2+}$   $0,01$   $\text{mg l}^{-1}$ . At the same time the average lifetime of daphnia was about 27 day, that there are less of value for shellfish in similar separate concentration of both agents. The most intensive modifying effect of  $^{90}\text{Sr}$  on impact of  $\text{Pb}^{2+}$  is observed at radionuclide concentration in a range  $2\text{E}+01$  –  $2\text{E}+03$   $\text{Bq l}^{-1}$  (Figure 2).

The analysis of the change of the daphnia linear dimension data during experiment has shown, that the most intensive growth of shellfish in the control was observed within the first 10 day with peak of the maximal value for 5 day (Figure 3). In all experimental concentration there was a displacement of peak of the maximal growth of daphnia: 10th day – for all activity of  $^{90}\text{Sr}$  and concentration of  $\text{Pb}^{2+}$   $0,01$   $\text{mg l}^{-1}$  both joint combinations of the radiation and chemical agents; 15th day – for concentration of  $\text{Pb}^{2+}$   $0,1$   $\text{mg l}^{-1}$ . At the same time if in all experimental concentration the displacement of peak of a growth occurred on a background lower rates of growth (in comparison to the control) at concentration of  $^{90}\text{Sr}$   $2\text{E}+03$   $\text{Bq l}^{-1}$  the maximal growth of daphnia was at a level of control individuals.

Concentration of  $\text{Pb}^{2+}$   $0,01$   $\text{mg l}^{-1}$  practically did not effect on daphnia growth rate at combined impact with  $^{90}\text{Sr}$  activities if to compare the value of this parameter for shellfish in experiment without  $\text{Pb}^{2+}$ . However the positive modifying effect of all  $^{90}\text{Sr}$  activities is authentically registered at combined impact with  $\text{Pb}^{2+}$  shown both in increase of a daphnia growth rate, and in earlier maximal values of this parameter. As well as in a case with average lifetime the most intensive modifying effect of  $^{90}\text{Sr}$  on impact of  $\text{Pb}^{2+}$  is observed at radionuclide concentration in a range  $2\text{E}+01$  –  $2\text{E}+03$   $\text{Bq l}^{-1}$  (Figure 4).

The authentic decrease of daphnia productional parameters, expressed in decrease of number of litter during life (Figure 5), in decrease of average quantity of juvenile in litter (Figure 6) and, accordingly, in decrease of total juvenile (Figure 7) are registered in all experimental concentration. At the same time the increase of  $\text{Pb}^{2+}$  concentration was accompanied by decrease all described productional parameters. As to impact of  $^{90}\text{Sr}$ , it carried some other character. In particular parameter of number of litter for life with increase of radionuclide activity was decreased, while the average juvenile in litter on the contrary – was increased, that can be explained by display of compensatory reactions at impact of ionising radiation. Nevertheless the total number of juvenile during experiment with increase of  $^{90}\text{Sr}$  activity was decreased.

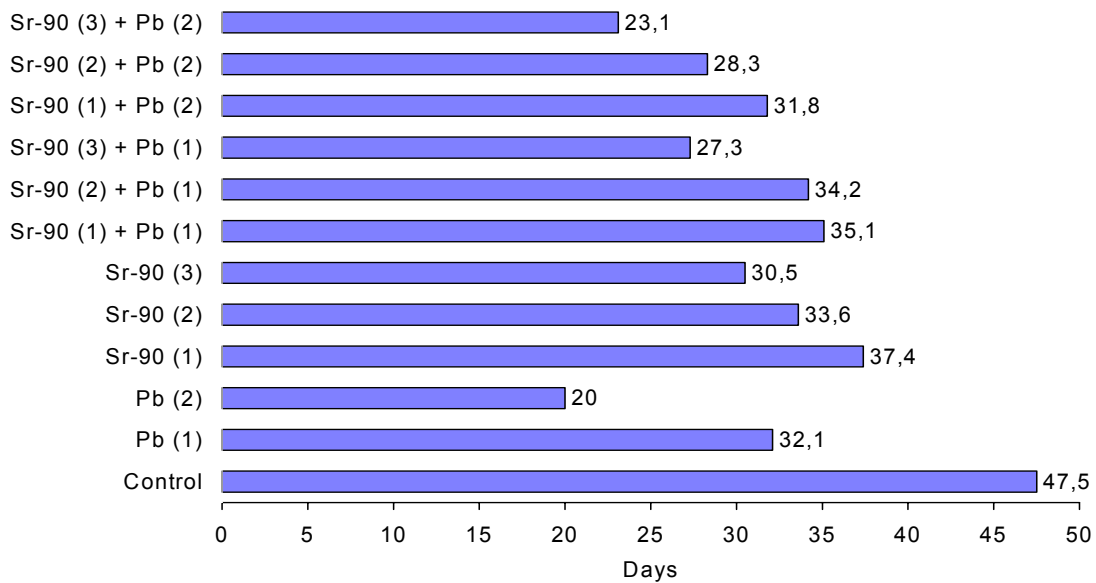


FIG. 1. Average lifetime of daphnia during experiment. Here and in FIG. 5, 6 and 7: Sr-90 (1) –  $^{90}\text{Sr}=2\text{E}+01 \text{ Bq l}^{-1}$ ; Sr-90 (2) –  $^{90}\text{Sr}=2\text{E}+03 \text{ Bq l}^{-1}$ ; Sr-90 (3) –  $^{90}\text{Sr}=2\text{E}+05 \text{ Bq l}^{-1}$ ; Pb (1) –  $\text{Pb}^{2+}=0,01 \text{ mg l}^{-1}$ ; Pb (2) –  $\text{Pb}^{2+}=0,1 \text{ mg l}^{-1}$ .

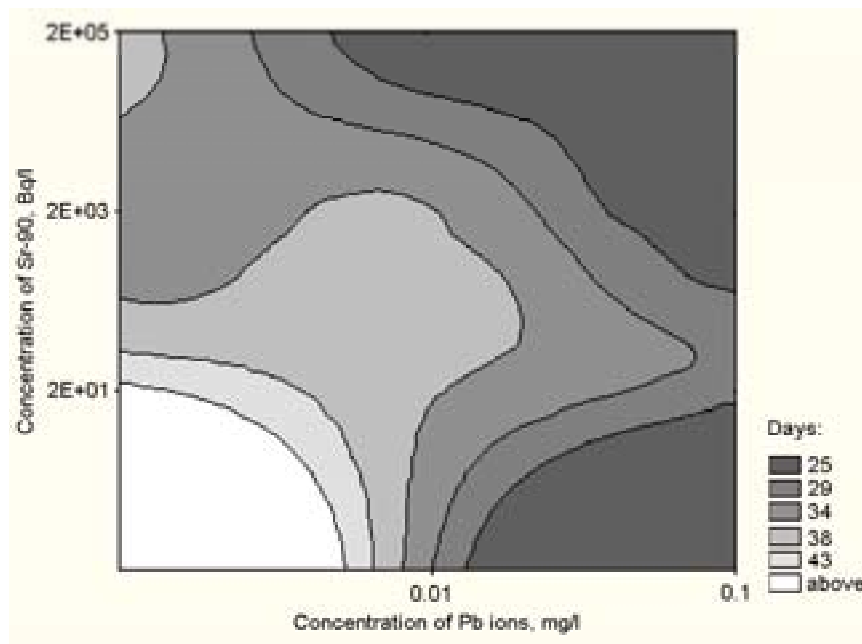


FIG. 2. Dynamics of lifetime of daphnia under combined impact of  $^{90}\text{Sr}$  and  $\text{Pb}^{2+}$ .

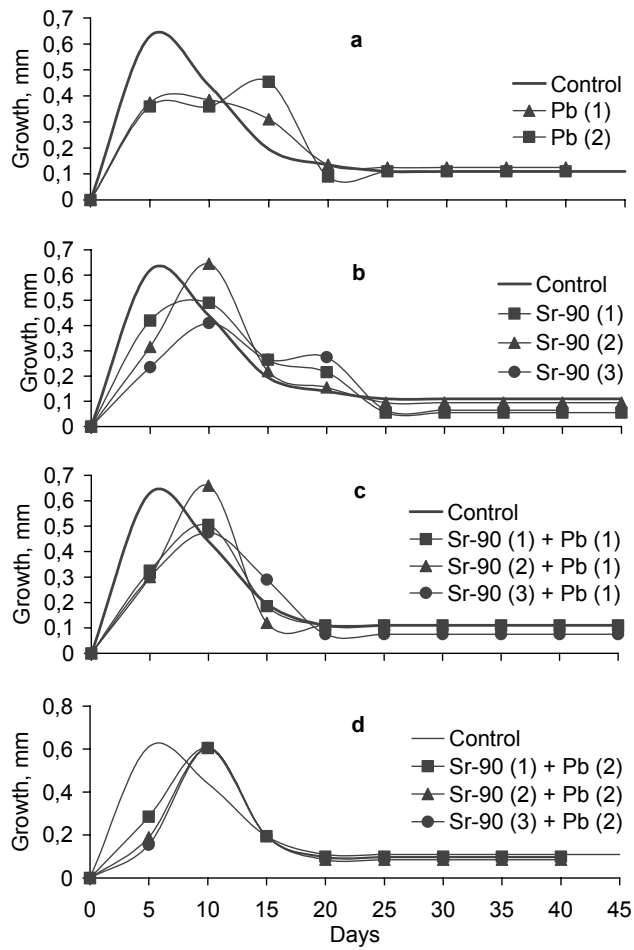


FIG. 3. Impact of  $^{90}\text{Sr}$  and  $\text{Pb}^{2+}$  on daphnia growth dynamics: a – different concentration of  $\text{Pb}^{2+}$ ; b – different  $^{90}\text{Sr}$  activity; c – combined impact of  $^{90}\text{Sr}$  activity and concentration of  $\text{Pb}^{2+}=0,01 \text{ mg l}^{-1}$ ; d – combined impact of  $^{90}\text{Sr}$  activity and concentration of  $\text{Pb}^{2+}=0,1 \text{ mg l}^{-1}$ .

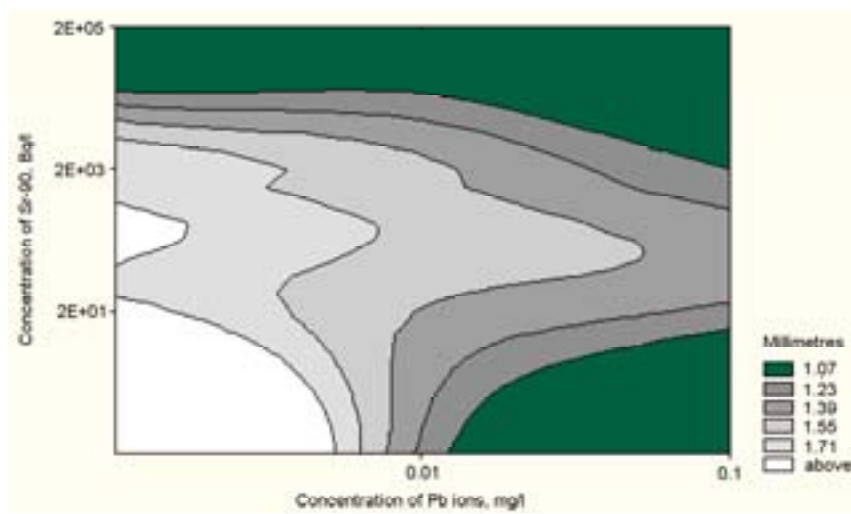


FIG. 4. Dynamics of growth rate of daphnia during life under impact of  $^{90}\text{Sr}$  and  $\text{Pb}^{2+}$ .

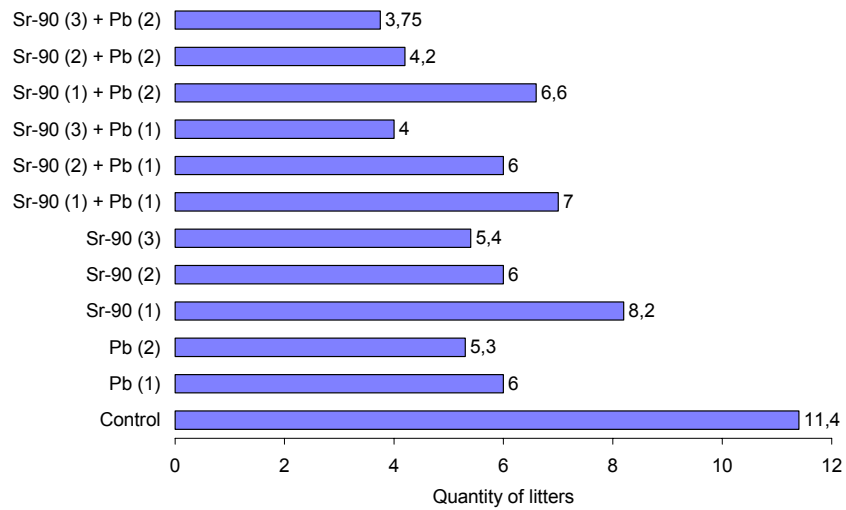


FIG. 5. Average quantity of litters during life.

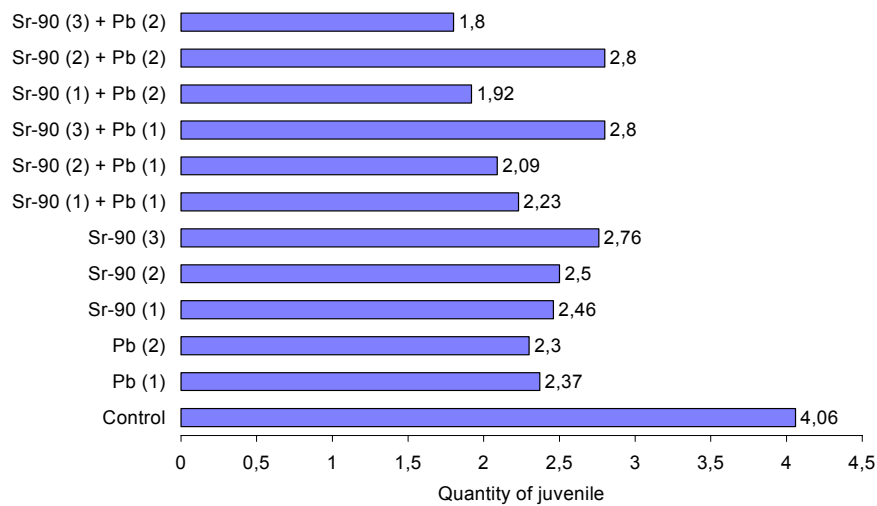


FIG. 6. Average quantity of juvenile in litters.

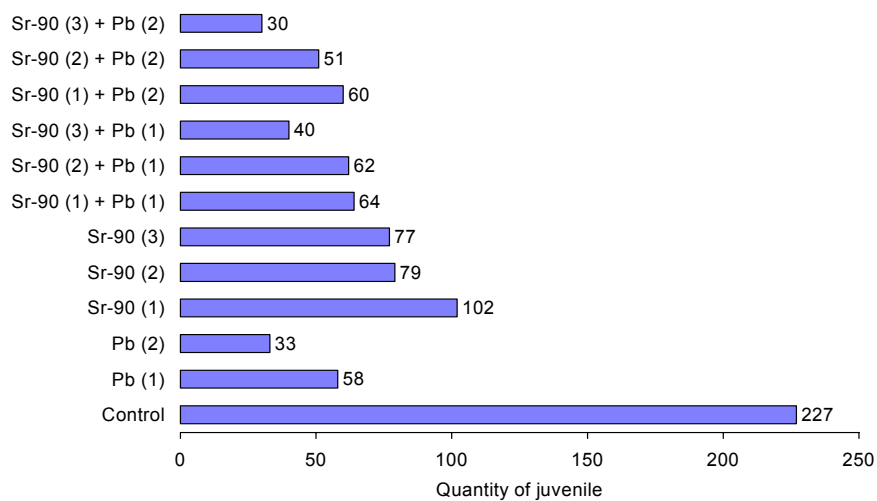


FIG. 7. Total quantity of juvenile during experiment.

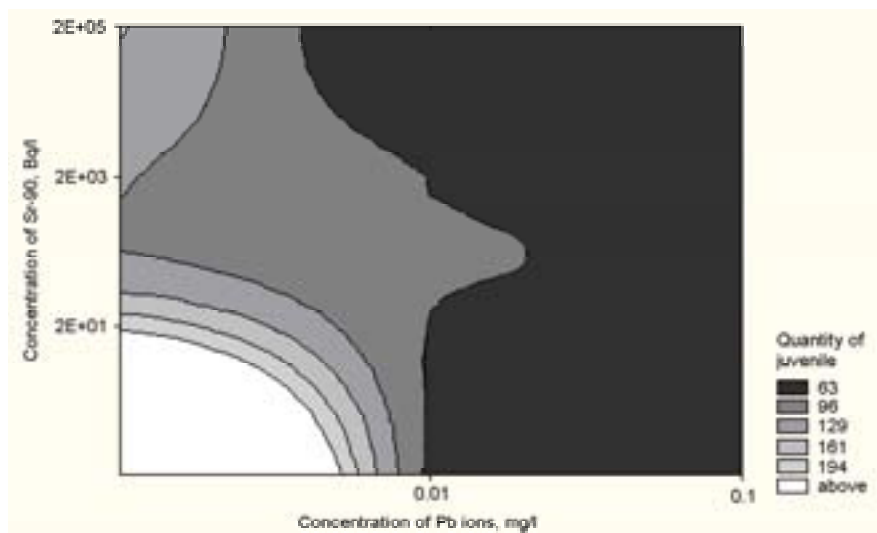


FIG. 8. Dynamics of the total quantity of juvenile during experiment under combined impact of  $^{90}\text{Sr}$  and  $\text{Pb}^{2+}$ .

The combined impact of the chemical and radiation factors on a parameter of average quantity of daphnia litter during life was characterised by the similar tendencies shown at separate influence of the researched agents. At the same time the modifying effect of  $^{90}\text{Sr}$  on  $\text{Pb}^{2+}$  impact did not carry the expressed character. The combined influence of  $\text{Pb}^{2+}$  and  $^{90}\text{Sr}$  had as antagonistic and stimulating character on average quantity of juvenile in litter at different combinations of the agents. However most indicative was the influence of combined impact on total juvenile during experiment. Positive modifying effect of  $^{90}\text{Sr}$  in a range of concentration  $2\text{E}+01 - 2\text{E}+03 \text{ Bq l}^{-1}$  on impact of  $\text{Pb}^{2+}$  here prevailed (Figure 8). The combined impact of the maximal activity of  $^{90}\text{Sr}$  ( $2\text{E}+05 \text{ Bq l}^{-1}$ ) with  $\text{Pb}^{2+}$  concentration rendered the greater negative effect, than separate impact of the researched agents. Authentic differences in terms of daphnia sexual maturity in experimental concentration and in the control (on the average 10 day) are not registered. At the same time it is necessary to note that with increase of  $^{90}\text{Sr}$  concentration the quantity of sterile individuals was grew. Its maximal quantity is observed at activity of  $^{90}\text{Sr}$   $2\text{E}+05 \text{ Bq l}^{-1}$  as separately and together with different concentration of  $\text{Pb}^{2+}$ , that allows to make a conclusion about primary impact of the radiation factor on value of this parameter.

#### 4. CONCLUSION

The  $\text{Pb}^{2+}$  in concentration of 0,01 and 0,1  $\text{mg l}^{-1}$  and  $^{90}\text{Sr}$  in concentration of  $2\text{E}+01$ ,  $2\text{E}+03$  and  $2\text{E}+05 \text{ Bq l}^{-1}$  in conditions of two-factor experiment rendered a negative impact practically on all researched biological parameters of freshwater cladoceran *Daphnia magna* Straus, shown in decrease of the productional characteristics, delay of rates of growth and reduction of daphnia lifetime. The level of toxic effect positively correlated with the contents of the researched agents and their combinations in water.

At combined impact of the radiation and chemical factors the positive modifying effect of radionuclide on toxic influence of  $\text{Pb}^{2+}$  is registered in a range of  $^{90}\text{Sr}$  activity  $2\text{E}+01 - 2\text{E}+03 \text{ Bq l}^{-1}$ .

It is supposed that at impact of ionising radiation on daphnia in a range of doses 7–70 cGy and  $\text{Pb}^{2+}$  in concentration of 0,01–0,1  $\text{mg l}^{-1}$  occurs a stimulation of protective reactions of organism, interfering to realisation of damaging influence of the radiation factor. At the same time one of mechanisms of radioprotection is the activation of the glutathione system and increase of a sulphhydic groups level of endogenous glutathione ( $\gamma$ -glutaminy-cysteinyl-glycine) having the important significance for oxidation-reduction reactions and participating in a fermentative reparation of damages [7–9]. However besides of radioprotective function glutathione takes part in synthesis of proteins-metallotioneins, which by chelation are capable to remove the heavy metals from a biochemical

exchange [1, 3]. Thus metabolic changes, initially directed on realisation of radioprotective effect, at combined impact of the radiation and chemical factors can carry multifunctional character, being shown in positive modification of toxic influence caused by ions of heavy metals.

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# Influence of 17- $\beta$ -estradiol and metals (Cd and Zn) on radionuclide ( $^{134}\text{Cs}$ , $^{57}\text{Co}$ and $^{110\text{m}}\text{Ag}$ ) bioaccumulation by juvenile rainbow trout

C. Adam<sup>a</sup>, O. Ausseil<sup>a</sup>, J. Garnier-Laplace<sup>a</sup>, J-M. Porcher<sup>b</sup>

<sup>a</sup> Department of Environmental Protection, Institute of Radioprotection and Nuclear Safety, Saint-Paul-lez-Durance, France

<sup>b</sup> Ecotoxicological Risks Assessment Unit, National Institute of Industrial Environment and Risks, Verneuil en Halatte, France

**Abstract.** A laboratory experiment was performed in order to characterize the influence of sublethal concentrations of the organic micropollutant 17- $\beta$ -estradiol (E2) and metals (Cd/Zn) on the bioaccumulation of  $^{57}\text{Co}$ ,  $^{134}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  by the rainbow trout, when micropollutants were applied separately or in different mixtures. Different groups of trouts were constituted and exposed to various pollutant mixtures (metal(s) and/or E2). In parallel, the effect of these exposures were explored through biomarker measurements in order to define the health status of the fish and to gain indication on the possible mechanisms involved. After 21 days, plasmatic parameters (vitellogenin and aminotransferases) and liver biomarkers (enzymatic and non enzymatic antioxidants, stress proteins and EROD activity) were assessed. Subsequently, fish were exposed to waterborne  $^{134}\text{Cs}$ ,  $^{57}\text{Co}$  and  $^{110\text{m}}\text{Ag}$  for 3 weeks. The results obtained show that the exposure to organic pollutants lead to an increase of radionuclide bioaccumulation, while the exposure to metals have the opposite effect. For the group exposed concomitantly to metals and E2, the observed effect corresponds to the addition of the two types of effects taken separately.

## 1. INTRODUCTION

In the context of the new European water policy based on the double objective of restricting the release of hazardous substances into aquatic systems and defining quality standards for ecosystems, there is an increasing need to characterize the patterns of artificial radionuclide accumulation under realistic hypotheses. Several questions must be addressed; in particular the eventuality of an interaction between widespread metallic and organic micropollutants and radionuclides has to be considered.

Among the major gamma emitting radionuclides identified in the liquid effluents of Pressurised Water Reactors, the radioisotopes of cesium, cobalt and silver have been largely studied and the characteristics of their bioaccumulation by freshwater organisms is now well known. However, the experiments have been focused on the behaviour of radionuclides taken separately, which was the first step to understand the involved mechanisms. Recently, the work of Sugg *et al.* [1] and Jagoe *et al.* [2] have focused on *in situ* measurements of stable metals (Hg, Pb) and radioactive pollutants ( $^{137}\text{Cs}$ ) in fish in the cooling pond of the Chernobyl power plant without, however, seeking to elucidate any possible interactions. The impact of stable micropollutants on radionuclide uptake by aquatic organisms may be conceptualized into different stages. First of all, the stable micropollutant must be able to cross the biological barriers as a function of its bioavailability. Once inside the cell, different cellular mechanisms may transform the pollutant into an inactive form and eliminate it. With an increasing ecotoxicological dose, defense mechanisms will counteract the toxic action of the pollutants, until they cannot compensate the damages anymore. Molecular and cellular biomarkers are very often used to give an earlier indication of the health status of an organism before an irreversible damage is observed at the individual or population level. In a multipollution context, the main goal for radioecologists is to assess the influence of this health status modified by the presence of stable pollutants on the characteristics of radionuclide bioaccumulation.

Within this framework, some experiments have been performed to test this hypothesis. A laboratory experiment showed that for the bivalve species *Dreissena polymorpha* and *Corbicula fluminea*,  $^{57}\text{Co}$  soft body concentration decreased with increasing zinc and cadmium concentration, whereas the opposite trend was observed for  $^{110\text{m}}\text{Ag}$ . No significant effect could be evidenced in the case of  $^{134}\text{Cs}$  [3]. The same tendency was observed for *C. fluminea* contaminated *in situ* by Cd and Zn downstream



from an old zinc ore treatment facility and by radionuclides in an artificial channel at the nuclear power plant of Golfech (France). No statistically significant difference was evidenced between control and metal polluted groups for  $^{137}\text{Cs}$ . On the contrary, for  $^{58}\text{Co}$ ,  $^{60}\text{Co}$  and  $^{110\text{m}}\text{Ag}$ , specimens exposed at the moderately metal polluted station were less contaminated by radionuclides than those from the control group [4]. As regards fish, rainbow trout have been caged during 4 weeks downstream from the same zinc ore treatment facility and were then brought back to the laboratory to be contaminated by  $^{110\text{m}}\text{Ag}$ ,  $^{57}\text{Co}$  and  $^{134}\text{Cs}$ . A clear trend was observed for the three radionuclides, i.e. fish exposed to metals accumulated smaller amounts of radionuclides. The decrease in radionuclide concentration for these individuals ranged from 20 to 90 % depending on the organ and on the radionuclide [5].

The study presented in this paper aims at evaluating under laboratory controlled conditions, the possible influence of cadmium and zinc when applied in mixture or alone, with or without a co-exposure to 17- $\beta$ -estradiol (E2), on the characteristics of  $^{57}\text{Co}$ ,  $^{134}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  bioaccumulation by the rainbow trout.

## 2. MATERIAL AND METHODS

### 2.1. Fish

Juvenile rainbow trout (*Oncorhynchus mykiss*) of an average body weight of  $10 \pm 0.14$  g w.w. were obtained from the fish farming "Petit Large" of Saumane, France. Throughout the experiment, including during the acclimation phase, the fish were fed with a standard pellet trout food (0.16  $\mu\text{g}$  Cd/g and 2.1  $\mu\text{g}$  Zn/g d.w.), at a rate of 1% per day of the mean fish body weight. They were acclimated for two weeks in 500 l tanks containing oxygenated tap water (pH 7.85, conductivity 421  $\mu\text{S}/\text{cm}$ , 145 mg/l  $\text{HCO}_3^-$ , 68 mg/l  $\text{Ca}^{2+}$ ) at  $12 \pm 0.5$  °C under artificial light reproducing day light on a 16 h-light 8 h-dark cycle.

### 2.2. Experimental design

The experiment was designed in such a way as to simulate the discharge of low-level radioactive liquid effluents into a freshwater ecosystem contaminated chronically with stable pollutants. During a first phase of 21 days (phase I), fish were exposed to stable pollutants. Eight groups of 45 fish were randomly placed in 90 l tanks and exposed to different stable pollutant mixtures. Three groups were contaminated from waterborne metals (Cd alone, Zn alone, mixture of Cd and Zn); one group was contaminated by an intra peritoneal injection of 17- $\beta$ -estradiol (E2) dissolved in corn-oil; one group was contaminated from waterborne Cd and Zn and received an injection of E2. To allow a real comparison between the fish exposed to metals and those exposed to organic micropollutants, an additional group exposed to waterborne Cd and Zn was injected with corn oil alone. Two control groups were used, the first one for the waterborne exposure and the second one where fish were injected with corn oil (carrier control). Cd and/or Zn were added in the water at nominal concentration of 1.2  $\mu\text{g}$  Cd/l and 160  $\mu\text{g}$  Zn/l from stock solutions of  $\text{CdCl}_2$  and  $\text{ZnCl}_2$  (Merck) acidified with 2 % (v/v) of HCl. E2 (Promochem) was dissolved in sterile corn oil and administrated by two successive intraperitoneal injections at the beginning of the experiment and after 14 days. Each injection corresponded to 2  $\mu\text{l}$  of corn-oil per g fish in which E2 was dissolved in order to be administrated at a concentration of 0.25 mg/kg fish. To limit alterations of the chemical characteristics of the water and a decrease in cadmium and zinc concentrations, the water was totally changed three times a week and the metal contamination was renewed.

During a subsequent phase lasting 21 days (phase II), a radioactive contamination was added. The radionuclides were obtained from the Amersham International Radiochemical Centre (UK). They were added to the water respectively in the form of  $^{57}\text{CoCl}_2$ ,  $^{134}\text{CsCl}$  and  $^{110\text{m}}\text{AgNO}_3$ , at a nominal level of 15 Bq/ml for each radionuclide.

### 2.3. Sampling and chemical analyses

Samples of water were taken every day for measurement of stable metals (Cd and Zn) throughout the experiment and radionuclides ( $^{57}\text{Co}$ ,  $^{134}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$ ) during phase II.

In order to measure the metal content in the fish, the following organs and tissues were sampled at day 21: liver, digestive tract, kidney, gills, muscles and blood. Each organ or tissue was mineralised individually in a glass tube with a screw stopper (HNO<sub>3</sub> 65%, 3 hours, 105°C, Blockdigest). The cadmium content was measured by graphite furnace atomic absorption spectrometry (Perkin Elmer 4110 ZL). The detection limit using this technique was 0.1 µg Cd/l. Zinc concentration was determined by flame atomic absorption spectrometry (Varian, Spectro AA 200), with a detection limit of 10 µg/l.

Radioactivity measurements were performed on living fish twice a week during phase II. The fish were taken randomly, weighed and introduced into a tube filled with non-contaminated water kept at 12°C to measure their radioactivity for three minutes. All measurements were carried out on by gamma spectrometry, in a multichannel analyser (SM512, Intertechnics), connected to a sodium iodide well probe. At day 35 and 42 (corresponding respectively to 14 and 21 days of radionuclide exposure), 5 fish per group were dissected. The measurements were performed using a high-purity germanium detector connected to a multichannel analyser. Under these conditions and for a counting time ranging from 1 to 10 hours, the detection limit was of the order of 1 Bq and the maximum relative error on the counting was 10 %. All the results of the measurements were related to the first day of the experiment by correction for the physical decay of the radionuclide.

At the end of phase I, 10 fish were sacrificed for each condition in order to carry out different analyses of the plasma and the liver. First, a blood sample was taken from each specimen. The sample was introduced into a heparinated tube (Lithium heparine plasma microtubes, Sarstedt), then centrifuged (15 min, 3000 g, 4°C). Following this procedure, the supernatant was collected in order to recover the plasma. The samples were then rapidly plunged into liquid nitrogen and kept at -80°C until analysis. On each aliquot of plasma, the following measurements were carried out: enzyme activities such as aspartate (AST), alanine aminotransferase (ALAT) and alkaline phosphatase (ALP). These analyses were measured with a Cobas Fara automated multi-analyser (Roche) by using commercial Roche kits. Vitellogenin (Vtg) levels were measured using a competitive method ELISA, with anti-salmon Vtg antibodies (BN5, Biosence Laboratories) and purified rainbow trout Vtg as standard. Hepatic biomarkers were also measured on the supernatant of homogenated liver centrifuged at 10,000 g for 15 min at 4°C (S9 fraction). Measurements of glutathione (tGSH, GSSG) and GSH redox status were performed. The GSH redox status was expressed as the ratio between GSSG, as GSH equivalent, and tGSH. Antioxidant enzymes such as total glutathione peroxidase (GPx), superoxide dismutase (SOD) and glutathione reductase (GR) were assessed. The total antioxidant activity, Trolox equivalent antioxidant capacity (TEAC) and Heat shock proteins (Hsp70, Usp60 and β-actin contents in the liver) were also determined. All the analyses and methods used for these biomarker measurements are further detailed by Aït-Aïssa *et al.* [6].

#### 2.4. Statistical analyses

For the analyses of variance, the exposure groups are divided into two categories depending on the type of treatment. For fish exposed to waterborne metals, groups were compared to control group, whereas for fish exposed by intra peritoneal injection, they were compared to carrier control group. For metals and radionuclides concentrations measured in the filtered water of the different exposure tanks, the means over the exposure period were compared by one-way ANOVA, followed by a Tukey's test. The same tests were performed to compare metals and radionuclides in fish organs. Interaction effects of exposure conditions were determined by a two-way ANOVA with the different micropollutants concentrations fixed as co-factors. These tests were performed using the Systat v10 software. As regards the biomarkers analyses, some biomarker data were log-transformed (group 1: GSSG, 2GSSG/tGSH, GR, TEAC; group 2: GSSG, GR, HSP70a, HSP70b, HSP60 and EROD) to conform to the normality assumption (Lilliefors test, based on a modification of the Kolmogorov-Smirnov test) and to the homogeneity of the variances (Levene's test). Significant effect of chemicals on the biomarker measurements were then determined by one-way ANOVA, followed by an unilateral Dunnett's test.

TABLE 1. CADMIUM CONCENTRATIONS (ng/g w.w.) IN DIFFERENT ORGANS AND TISSUES AFTER 21 DAYS OF EXPOSURE. CONCENTRATIONS IN THE MUSCLE WERE BELOW THE DETECTION LIMIT. VALUES ARE MEAN  $\pm$  CI<sub>95%</sub> (n=8). (\* AND BOLD CHARACTERS: SIGNIFICANTLY DIFFERENT FROM THE CONTROL, p<0.05)

	Liver	Kidney	Digestive tract	Gills
Control	5 $\pm$ 3	30 $\pm$ 15	10 $\pm$ 7	31 $\pm$ 6
Cd	<b>45 <math>\pm</math> 8 *</b>	<b>105 <math>\pm</math> 18*</b>	<b>28 <math>\pm</math> 6*</b>	<b>550 <math>\pm</math> 80*</b>
Zn	12 $\pm$ 9	20 $\pm$ 6	14 $\pm$ 8	42 $\pm$ 8
CdZn	<b>43 <math>\pm</math> 11*</b>	<b>114 <math>\pm</math> 22*</b>	<b>26 <math>\pm</math> 8*</b>	<b>580 <math>\pm</math> 110*</b>
Carrier control	6 $\pm$ 4	18 $\pm$ 9	12 $\pm$ 9	20 $\pm$ 12
E2	5 $\pm$ 3	31 $\pm$ 10	8 $\pm$ 7	44 $\pm$ 15
CdZn <sub>carrier</sub>	<b>39 <math>\pm</math> 12*</b>	<b>124 <math>\pm</math> 23*</b>	<b>30 <math>\pm</math> 10*</b>	<b>510 <math>\pm</math> 60*</b>
CdZnE2	<b>48 <math>\pm</math> 6*</b>	<b>98 <math>\pm</math> 12*</b>	<b>35 <math>\pm</math> 12*</b>	<b>620 <math>\pm</math> 140*</b>

### 3. RESULTS AND DISCUSSION

#### 3.1. Metals concentrations in water and fish organs

No significant difference could be evidenced between the metal concentrations of the water for the five different metal exposure conditions (p<0.05). As regards organs and tissues (Table 1), the highest discrepancies between Cd exposed and control groups were found in the case of gills and liver, where the Cd concentrations were respectively 30 and 8 fold the amount measured in the control group. In the case of Zn, no significant difference was highlighted between zinc exposed groups and the control. The mean concentrations measured were 160, 60, 37, 30 and 8  $\mu$ g/g (w.w.) respectively for digestive tract, gills, kidney, liver and muscle.

#### 3.2. Biomarkers responses at day 21

The exposure to metallic and organic pollutants did not induce any growth or survival alteration. A mean mortality of 4 to 6 % was observed but did not differ between control and exposed groups. This result indicated that the exposure concentration were really sublethal. This has been confirmed by other experiments [7] where mortalities up to 30 % occurred in the fish groups that had received 10  $\mu$ g Cd/l and/or 1000  $\mu$ g Zn/l, whereas no mortality was observed for the groups that had been exposed to 1.5  $\mu$ g Cd/l and 200  $\mu$ g Zn/l. The liver somatic index measured at day 21, 35 and 42 did not show any effect of the concentrations of Cd and/or Zn used. On the opposite, at day 42, all the groups that had received an intra peritoneal injection were characterized by a significant increase of this parameter, when compared to the control. The injection of corn oil leads to an increase of the liver somatic index, which should be considered when performing this type of experiment. This increase may be linked to the high load of oil administrated, leading to an alteration of liver function. As regards the biochemical analyses of the plasmatic parameters AST, ALAT and AIP, no significant difference was evidenced between control, Cd, Zn and Cd/Zn groups on one hand and on the other hand, between carrier control, E2, CdZn<sub>carrier</sub> and CdZnE2. On the contrary, high levels of Vtg were measured in E2 and CdZnE2 treated groups, with no statistical difference between each other (0.87 $\pm$ 0.26 and 0.80 $\pm$ 0.14 mg/ml). In the other groups, Vtg concentrations were below the detection limit of 300 ng/ml.

Liver biomarkers responses are summarized in Table 2. In the case of fish exposed to waterborne metals, a general increase of antioxidant defences is noted. For Cd alone, non enzymatic (total GSH and TEAC) and enzymatic (SOD) antioxidant defenses are increased. For GR an important but not significant increase is found. In the case of Zn alone, a significant increase of SOD and GR is noted and this trend is also observed for glutathione and TEAC but is not significant. For stress protein, no effect of metal exposure could be highlighted, except a significant induction for Zn. Possible joint effect of Cd and Zn were evaluated. Significant interactions were noted for tGSH (p=0.015), GSSG (p=0.007), GSH redox status (p=0.022), GR (p=0.013) and HSP70a (p=0.047), that indicate an antagonism between Cd and Zn action. Cadmium is well known for its oxidant capacity and its interaction with intracellular thiols, thus leading to the formation of aberrant complexes such as protein-glutathione complexes [8]. The results show that the exposure to Cd and/or zinc lead to an

increase of glutathione forms. On the contrary, no impairment of the GSH redox status nor increase of GR levels were noted. On the whole, the adaptative response to the oxidative action of metals is sufficient to counteract their toxic effects since no sign of toxicity was evidenced (no induction of stress proteins, nor antioxidant depletion and nor GSSG accumulation). As regards EROD activity, a significant induction was observed for the three metal conditions. Such an inductive effect was observed by Lemaire-Gony *et al.* [9] for sea breams exposed to 40 µg Cd/l for 15 days. These authors indicate that this effect probably results from a non specific interaction of metals with cellular membranes in the endoplasmic reticulum. However, this induction remains very low compared to the much higher induction observed during the same experiment (results not shown), where EROD activities in fish exposed to PCB77 was induced up to 30 fold as compared to control fish [6].

In the case of the groups exposed to organic compounds, the fish exposed to E2 alone were characterized by a decrease of antioxidant defense (tGSH, GSSG, TEAC and SOD) and by an increase of GPx activity. The GSH redox status and GR were not altered by the treatment. This antioxidant defences depletion could be explained by the study of Palace *et al.* [10] who showed that during normal vitellogenesis, the vitamins are remobilized from visceral organs to be stored into the gonads before their incorporation into growing oocytes. No effect of E2 was observed on stress protein, but the CdZn<sub>carrier</sub> group was characterized by a significant increase of HSP 70a and b. A similar result was found by Heikkila *et al.* [11] on *Oncorhynchus tshawytscha* hepatocytes exposed to Cd, where a significant decrease of EROD activity was noted. The basal level of EROD activity has been shown to be affected by several biological factors, such as sexual maturation. Therefore, Andersson and Förlin [12] have suggested that estradiol could exert a negative control on cytochrome P4501A1 levels. The most striking result concerns fish exposed concomitantly to waterborne Cd and Zn and to E2 by i.p. injection, in which the biomarker responses seem to compensate themselves. The biomarker responses observed for E2 and CdZn when applied separately were in opposition, whereas they seem to additivate themselves for the CdZnE2 group. Such an effect has been observed for Cd and E2 in rainbow trout by Valencia *et al.* [13]. The authors showed that Cd causes the transcriptional down-regulation of Vtg synthesis in E2 injected fish. This response is correlated with (i) the preferential binding of Cd to non-MT proteins in the liver, (ii) decreased cadmium induction of hepatic MT mRNA and (iii) increased sensitivity of fish to cadmium toxicity.

TABLE 2. EFFECT OF A 21 DAYS EXPOSURE TO DIFFERENT METAL AND ORGANIC MICROPOLLUTANTS ON HEPATIC BIOMARKERS. VALUES ARE RELATIVE TO THE CONTROL LEVEL. BOLD CHARACTERS AND <sup>+</sup>(<sup>-</sup>): SIGNIFICANT INCREASE (DECREASE) OF THE BIOMARKER RESPONSE FOR EXPOSED GROUPS AS COMPARED TO CONTROL GROUP; <sup>+</sup>(<sup>-</sup>): p<0.05; <sup>++</sup>(<sup>-</sup>): p<0.01

	Waterborne metals			Organic compounds		
	Cd	Zn	CdZn	E2	CdZn <sub>carrier</sub>	CdZnE2
tGSH <sup>a</sup>	<b>1.7<sup>++</sup></b>	1.2	<b>1.4<sup>++</sup></b>	<b>0.7<sup>-</sup></b>	1.1	1.0
GSSG <sup>a</sup>	1.2	1.6	0.4	<b>0.7<sup>-</sup></b>	0.7	<b>0.4<sup>-</sup></b>
2xGSSG/tGSH	1	1.6	0.3	1	<b>0.5<sup>-</sup></b>	<b>0.4<sup>-</sup></b>
TEAC <sup>b</sup>	<b>1.3<sup>+</sup></b>	1.3	<b>1.5<sup>+</sup></b>	<b>0.7<sup>-</sup></b>	<b>1.6<sup>++</sup></b>	1.2
SOD <sup>c</sup>	<b>1.5<sup>+</sup></b>	<b>1.6<sup>+</sup></b>	<b>2.0<sup>++</sup></b>	<b>0.7<sup>-</sup></b>	0.9	0.8
GPx <sup>d</sup>	1.0	0.8	0.8	<b>1.9<sup>+</sup></b>	<b>1.7<sup>+</sup></b>	<b>2.0<sup>++</sup></b>
GR <sup>d</sup>	2.0	<b>1.5<sup>+</sup></b>	1.0	0.9	1.4	0.9
HSP 70a <sup>c</sup>	1.2	<b>2.1<sup>+</sup></b>	1.0	0.9 <sup>#</sup>	<b>3.1<sup>+</sup></b>	1.6 <sup>#</sup>
HSP 70b <sup>c</sup>	1.2	1.2	1.1	0.5 <sup>#</sup>	<b>1.5<sup>+</sup></b>	1.2 <sup>#</sup>
HSP 60 <sup>c</sup>	0.8	1.0	0.8	0.8 <sup>#</sup>	1.2	0.9 <sup>#</sup>
EROD <sup>f</sup>	<b>2.0<sup>+</sup></b>	<b>2.4<sup>++</sup></b>	<b>2.7<sup>++</sup></b>	<b>0.4<sup>-</sup></b>	0.7	0.9

<sup>#</sup> mean over 3 replicates.

### 3.3. Radionuclides concentrations in water and in fish (organs/whole body)

The mean radionuclide concentration in the water during the 21 days of the phase II were  $13.6 \pm 1.1$ ,  $10.5 \pm 0.2$ , and  $11.6 \pm 3.9$  Bq/l for  $^{57}\text{Co}$ ,  $^{134}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  respectively. No significant difference could be evidenced between the radionuclide concentrations in each treatment tank.

As regards the  $^{110\text{m}}\text{Ag}$  concentrations in fish organs (Table 3), concentrations in the liver are very high as compared to the other radionuclides or to the other organs, with concentrations up to 9000 Bq/g in the liver of the control group. Similar results were found by Garnier *et al.* [14] after a 57 day waterborne exposure to AgCN in the liver of *Salmo trutta*. The authors showed that  $^{110\text{m}}\text{Ag}$  liver content represented 70 % of whole body content, with a very slow depuration since after a 28 day depuration period, 62 % of  $^{110\text{m}}\text{Ag}$  were still found in that organ. It is now admitted that silver is sequestered in the very stable chemical form  $\text{Ag}_2\text{S}$  in the liver, but also in the gills and digestive tract.

For the groups exposed to Cd and/or Zn, silver concentration is lower in all the organs and at the whole body level as compared to control, with a statistical significance for 15 values over 21. Depending on the organ, this decrease ranges from 2 to 3 fold as compared to control group. An interaction effect of the two metals, corresponding to a synergism, has been found for the liver ( $p < 0.001$ ), the digestive tract ( $p < 0.009$ ), the kidney ( $p < 0.069$ ) and at the whole body level ( $p < 0.001$ ).

As regards the trouts exposed to organic compounds, in the  $\text{CdZn}_{\text{carrier}}$  group (exposed to waterborne Cd and Zn and injected with corn-oil), a similar decrease is observed in the liver, the digestive tract and the whole body, with a decrease up to 2.6 for the liver concentrations. On the contrary, a significant increase is evidenced in the blood compartment for this group, which was also observed for the groups exposed to waterborne metal, but not significantly. For fish exposed to E2, an increase of the accumulated amounts is observed in each organ and at the whole body level. Significant increase is evidenced up to 1.6 in the gills, 1.3 in the liver and 1.2 in the whole body. In the same way as for the biomarker results, the most striking observation concerns the fish group exposed concomitantly to metals and E2. For this group, the effects of metals and estradiol seem to compensate themselves. A significant decrease is found in the liver and at the whole body level, but stands between the increase induced by E2 alone and the decrease observed for the  $\text{CdZn}_{\text{carrier}}$  group. This effect is observed for all the organs except for blood, where the radionuclide concentration is closer to the level measured in the control group. According to the two-way factorial ANOVA, a slight interactive effect between metals and E2 was evidenced for gills ( $p < 0.069$ ) and blood ( $p < 0.001$ ).

$^{134}\text{Cs}$  is distributed in a more homogeneous way (Table 4) in fish organs as compared to silver, which may be linked to the chemical analogy of cesium with potassium, a macroelement widely spread in different organs. As regards fish exposed to waterborne Cd and/or Zn, a similar significant decrease of  $^{134}\text{Cs}$  accumulation is evidenced in every organ and at the whole body level. This decrease is less important than that observed for  $^{110\text{m}}\text{Ag}$ , since the maximal decrease observed for liver is of 1.8 fold as compared to 3.2 fold in the case of silver. An interactive effect of Cd and Zn is significant for all the organs and for the whole body, according to the two-way factorial ANOVA.

As regards the groups exposed to waterborne Cd and Zn and to oil injection, very similar significant results are obtained, with a maximal decrease of 2 fold in the liver as compared to carrier control. Concerning E2 exposed group, an increase of accumulated  $^{134}\text{Cs}$  is noted in every organ, but is only significant at the whole body level ( $p < 0.05$ ). Finally, for the group exposed to waterborne Cd and Zn and injected with E2, the results are similar to those found for  $^{110\text{m}}\text{Ag}$ , particularly in the case of liver and gills where it appears that the effects of E2 and metals compensate themselves. The global result tends to a decrease of accumulated cesium, but this decrease is less marked than in the  $\text{CdZn}$  contaminated group. No interaction could be evidenced between the organic and the metallic pollutants.

TABLE 3. CONCENTRATION OF  $^{110m}\text{Ag}$  (Bq/g w.w.) IN DIFFERENT ORGANS AND IN THE WHOLE BODY OF TROUTS AFTER 42 DAYS OF EXPOSURE TO DIFFERENT METAL AND ORGANIC MICROPOLLUTANTS. VALUES ARE MEAN  $\pm$  SD (n=5). BOLD CHARACTERS AND  $^{+(-)}$ : SIGNIFICANT INCREASE (DECREASE) OF THE CONCENTRATION FOR EXPOSED GROUPS AS COMPARED TO CONTROL GROUP;  $^{+(-)}$ : p<0.05;  $^{++(-)}$ : p<0.001

	Liver	Digestive tract	Kidney	Gills	Muscle	Blood	Whole body
Control	8700 $\pm$ 724	93 $\pm$ 19	564 $\pm$ 74	221 $\pm$ 14	6.1 $\pm$ 1.7	28 $\pm$ 7	166 $\pm$ 13
Cd	<b>3324 <math>\pm</math> 592<sup>-</sup></b>	<b>52 <math>\pm</math> 9<sup>-</sup></b>	<b>304 <math>\pm</math> 57<sup>-</sup></b>	<b>163 <math>\pm</math> 21<sup>-</sup></b>	4.4 $\pm$ 0.9	36 $\pm$ 7	<b>81 <math>\pm</math> 11<sup>-</sup></b>
Zn	<b>3776 <math>\pm</math> 311<sup>-</sup></b>	<b>52 <math>\pm</math> 15<sup>-</sup></b>	<b>409 <math>\pm</math> 90<sup>-</sup></b>	182 $\pm$ 6	4.8 $\pm$ 0.7	39 $\pm$ 6	<b>94 <math>\pm</math> 11<sup>-</sup></b>
CdZn	<b>2723 <math>\pm</math> 391<sup>-</sup></b>	<b>47 <math>\pm</math> 10<sup>-</sup></b>	<b>271 <math>\pm</math> 53<sup>-</sup></b>	<b>169 <math>\pm</math> 47<sup>-</sup></b>	<b>3.7 <math>\pm</math> 0.8<sup>-</sup></b>	31 $\pm$ 12	<b>66 <math>\pm</math> 10<sup>-</sup></b>
Carrier control	7368 $\pm$ 355	72 $\pm$ 18	489 $\pm$ 122	231 $\pm$ 9	5.1 $\pm$ 1.3	21 $\pm$ 3	171 $\pm$ 5
E2	<b>9730 <math>\pm</math> 2096<sup>+</sup></b>	91 $\pm$ 15	656 $\pm$ 238	<b>372 <math>\pm</math> 55<sup>++</sup></b>	8.0 $\pm$ 3.7	29 $\pm$ 2	<b>213 <math>\pm</math> 29<sup>+</sup></b>
CdZn <sub>carrier</sub>	<b>2827 <math>\pm</math> 677<sup>-</sup></b>	<b>47 <math>\pm</math> 4<sup>-</sup></b>	331 $\pm$ 26	191 $\pm$ 18	4.0 $\pm$ 0.6	<b>34 <math>\pm</math> 9<sup>+</sup></b>	<b>74 <math>\pm</math> 10<sup>-</sup></b>
CdZnE2	<b>5126 <math>\pm</math> 365<sup>-</sup></b>	59 $\pm$ 5	595 $\pm$ 88	278 $\pm$ 23	4.3 $\pm$ 0.9	17 $\pm$ 5	<b>118 <math>\pm</math> 10<sup>-</sup></b>

TABLE 4. CONCENTRATION OF  $^{134}\text{Cs}$  (Bq/g w.w.) IN DIFFERENT ORGANS AND IN THE WHOLE BODY OF TROUTS AFTER 42 DAYS OF EXPOSURE TO DIFFERENT METAL AND ORGANIC MICROPOLLUTANTS. VALUES ARE MEAN  $\pm$  SD (n=5). BOLD CHARACTERS AND  $^{+(-)}$ : SIGNIFICANT INCREASE (DECREASE) OF THE CONCENTRATION FOR EXPOSED GROUPS AS COMPARED TO CONTROL GROUP;  $^{+(-)}$ : p<0.05;  $^{++(-)}$ : p<0.001

	Liver	Digestive tract	Kidney	Gills	Muscle	Blood	Whole body
Control	30 $\pm$ 5	43 $\pm$ 6	30 $\pm$ 7	23 $\pm$ 2	22 $\pm$ 3	7.4 $\pm$ 0.7	22 $\pm$ 3
Cd	<b>18 <math>\pm</math> 2<sup>-</sup></b>	<b>26 <math>\pm</math> 2<sup>-</sup></b>	<b>19 <math>\pm</math> 5<sup>-</sup></b>	<b>15 <math>\pm</math> 1<sup>-</sup></b>	<b>13 <math>\pm</math> 1<sup>-</sup></b>	<b>5.0 <math>\pm</math> 0.7<sup>-</sup></b>	<b>14 <math>\pm</math> 0.5<sup>-</sup></b>
Zn	<b>18 <math>\pm</math> 2<sup>-</sup></b>	<b>33 <math>\pm</math> 2<sup>-</sup></b>	22 $\pm$ 4	<b>17 <math>\pm</math> 2<sup>-</sup></b>	<b>15 <math>\pm</math> 1<sup>-</sup></b>	5.9 $\pm$ 1.5	<b>16 <math>\pm</math> 1<sup>-</sup></b>
CdZn	<b>17 <math>\pm</math> 3<sup>-</sup></b>	<b>33 <math>\pm</math> 3<sup>-</sup></b>	21 $\pm$ 4	<b>18 <math>\pm</math> 2<sup>-</sup></b>	<b>13 <math>\pm</math> 2<sup>-</sup></b>	5.5 $\pm$ 1.4	<b>14 <math>\pm</math> 2<sup>-</sup></b>
Carrier control	29 $\pm$ 4	44 $\pm$ 7	35 $\pm$ 4	22 $\pm$ 2	22 $\pm$ 3	6.8 $\pm$ 0.5	23 $\pm$ 2
E2	34 $\pm$ 5	52 $\pm$ 4	38 $\pm$ 7	26 $\pm$ 3	27 $\pm$ 3	8.8 $\pm$ 2.0	<b>28 <math>\pm</math> 3<sup>+</sup></b>
CdZn <sub>carrier</sub>	<b>15 <math>\pm</math> 3<sup>-</sup></b>	<b>33 <math>\pm</math> 2<sup>-</sup></b>	<b>21 <math>\pm</math> 3<sup>-</sup></b>	<b>16 <math>\pm</math> 2<sup>-</sup></b>	<b>14 <math>\pm</math> 2<sup>-</sup></b>	5.2 $\pm$ 1.0	<b>15 <math>\pm</math> 2<sup>-</sup></b>
CdZnE2	22 $\pm$ 5	<b>34 <math>\pm</math> 6<sup>-</sup></b>	<b>21 <math>\pm</math> 7<sup>-</sup></b>	20 $\pm$ 5	<b>16 <math>\pm</math> 3<sup>-</sup></b>	6.1 $\pm$ 1.3	<b>16 <math>\pm</math> 3<sup>-</sup></b>

TABLE 5. CONCENTRATION OF  $^{57}\text{Co}$  (Bq/g w.w.) IN DIFFERENT ORGANS AND IN THE WHOLE BODY OF TROUTS AFTER 42 DAYS OF EXPOSURE TO DIFFERENT METAL AND ORGANIC MICROPOLLUTANTS. VALUES ARE MEAN  $\pm$  SD (n=5). BOLD CHARACTERS AND  $^{+(-)}$ : SIGNIFICANT INCREASE (DECREASE) OF THE CONCENTRATION FOR EXPOSED GROUPS AS COMPARED TO CONTROL GROUP;  $^{+(-)}$ : p<0.05;  $^{++(-)}$ : p<0.001

	Liver	Digestive tract	Kidney	Gills	Muscle	Blood	Whole body
Control	25 $\pm$ 4	40 $\pm$ 4	82 $\pm$ 9	90 $\pm$ 11	6.9 $\pm$ 0.7	101 $\pm$ 9	24 $\pm$ 2
Cd	25 $\pm$ 11	33 $\pm$ 7	71 $\pm$ 17	94 $\pm$ 15	6.2 $\pm$ 0.8	123 $\pm$ 18	23 $\pm$ 5
Zn	21 $\pm$ 4	41 $\pm$ 13	71 $\pm$ 18	87 $\pm$ 17	6.1 $\pm$ 1.1	92 $\pm$ 13	22 $\pm$ 4
CdZn	26 $\pm$ 4	40 $\pm$ 6	75 $\pm$ 22	92 $\pm$ 17	5.7 $\pm$ 0.6	106 $\pm$ 10	22 $\pm$ 2
Carrier control	20 $\pm$ 2	43 $\pm$ 6	74 $\pm$ 10	97 $\pm$ 15	6.6 $\pm$ 1.2	106 $\pm$ 13	25 $\pm$ 1
E2	<b>34 <math>\pm</math> 10<sup>+</sup></b>	64 $\pm$ 20	<b>110 <math>\pm</math> 22<sup>+</sup></b>	<b>147 <math>\pm</math> 22<sup>+</sup></b>	<b>10 <math>\pm</math> 1.3<sup>+</sup></b>	<b>150 <math>\pm</math> 12<sup>++</sup></b>	<b>39 <math>\pm</math> 8<sup>+</sup></b>
CdZn <sub>carrier</sub>	22 $\pm$ 7	40 $\pm$ 10	78 $\pm$ 9	87 $\pm$ 9	6.4 $\pm$ 1.5	107 $\pm$ 8	23 $\pm$ 4
CdZnE2	29 $\pm$ 5	51 $\pm$ 10	102 $\pm$ 23	122 $\pm$ 27	8.1 $\pm$ 1.5	<b>135 <math>\pm</math> 9<sup>+</sup></b>	31 $\pm$ 6

Finally, the results of  $^{57}\text{Co}$  contamination (Table 5) do not allow to conclude to an effect of a concomitant exposure to Cd and Zn on cobalt uptake. No significant difference nor any tendency could be evidenced. The same conclusion can be drawn from  $\text{CdZn}_{\text{carrier}}$  group. On the opposite, a significant increase of cobalt accumulation was observed in groups injected by E2. The maximal increase (1.7 fold the value measured in the carrier control group) was observed in the liver. As for  $^{110\text{m}}\text{Ag}$  and  $^{134}\text{Cs}$ , the groups contaminated by metals and E2 are characterized by a less important increase, only significant in the blood compartment. This result is more surprising in the case of cobalt, since no decreasing effect of Cd and Zn was observed on cobalt accumulation. As for cesium, no interaction between metals and E2 could be evidenced.

#### 4. CONCLUSIONS

Some hypotheses may explain the effect of stable micropollutants on radionuclide uptake by fish. With regard to the biomarkers responses, it appears that a decrease in radionuclides concentrations is linked to an increase of antioxidant mechanisms, as if these defense mechanisms would manage the detoxification of the radionuclides together with Cd and Zn. For organic compounds, the inverse relationship is observed, but the toxicological significance remains to be explained. More general hypotheses may be drawn. For metals, several authors have shown that the exposure to high concentrations of Cd (1 mg/l) and Zn (15mg/l) induces increased mucus production in the gills [15, 16]. This mucus made of glycoproteins acts as a complexing agent for cations, with probably a different affinity for different metals, which could explain why cobalt is not influenced by a Cd and Zn co-exposure. Another hypothesis could be based on an alteration of gill epithelium permeability following Cd exposure, leading to ultrastructural damages of gills. The effect of Cd on  $\text{Na}^+/\text{K}^+$ -ATPase activity have been shown [17] which could be linked to differences in selective permeability of gill epithelium to  $\text{Cs}^+$  and other cations. Moreover, the competitive effect of Cd on calcium channels has been shown [18] and could be applied also to  $\text{Ag}^+$ . This competition hypothesis corroborates with the significant difference in silver distribution in the metal exposed groups as compared to control group (results not shown). In the control group,  $^{110\text{m}}\text{Ag}$  liver content represents 70 % of the total amount in the fish, whereas it represents *c.a.* 60 % in the metal exposed fish, with a concomitant increase in gills and kidney. The competition of the radionuclide with Cd and/or Zn for complexation with binding sites such as MT in the liver may occur. Concerning the exposure to E2, the increase in radionuclide uptake is observed for the three radionuclides, which would indicate that the mechanism governing this effect is not specific. A logical hypothesis would be an increase of the respiratory activity of trouts, which has been shown for several micropollutants. In the particular case of cobalt, an additional hypothesis would be that the radionuclide is remobilized from visceral organs as shown by Palace *et al.* [10] following vitellogenin induction by E2, which could explain the significant increase of cobalt concentration in blood observed for E2 exposed groups. Finally, the results obtained for trouts exposed concomitantly to metals and E2, show that the effects are more or less additive. The main hypothesis to explain this result is based on Valencia *et al.* [13] paper, where the authors showed that (i) Cd causes the down-regulation of Vtg, which could explain a decreased need in oligoelements such as cobalt, (ii) decreased induction of MT mRNA, which would explain a decreased accumulation of silver.

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**Use of genetic markers for ecological risk assessment at the Idaho National Engineering and Environmental Laboratory: Microsatellite mutation rate of burrowing mammals**  
*Genetic markers for ecorisk assessment\**

**A.I. Stormberg<sup>a</sup>, S. Perry<sup>a</sup>, M. Lucid<sup>b</sup>, J.A. Cook<sup>b</sup>**

<sup>a</sup> Idaho National Engineering and Environmental Laboratory, Idaho Falls, ID, United States of America

<sup>b</sup> Idaho State University, Department of Biological Sciences, Pocatello, ID, United States of America

**Abstract.** Radiological and hazardous waste have been disposed at the U.S. Department of Energy's Idaho National Engineering and Environmental Laboratory since 1952. Escape of radionuclides and hazardous constituents from uncontained wastes, and from deterioration of waste containers and waste disposal practices has resulted in contamination of the subsurface soils in the Subsurface Disposal Area (SDA) and at other facilities. To assess the risks to human and ecological health, the potential impacts of contaminant exposure on identified receptors must be determined. Burrowing and excavation of the soil by small mammals, including deer mice (*Peromyscus maniculatus*), is responsible for some radionuclide transport through the SDA environment; however, the genomic effects of exposure to contaminants are not known. This research will evaluate if molecular genetics can be used to determine whether exposure to contaminants affects organisms at the genomic level.

The ratio of microsatellite mutant alleles vs. the non-mutant alleles (m/nm) was used as a direct assessment of mutation, using parent/offspring comparison of allele differences. Preliminary allele scoring was performed with 10 microsatellite markers for females and offspring from two uncontaminated control locations (Burn and Atomic City), and two contaminated test locations (SL-1 and RWMC). Fetal genotypes that gave a single inconsistent genotype with the mother were scored as mutations. The proportion of mutant alleles from each population was compared and tested for significant differences using the Fisher's exact test.

Preliminary data suggest that the contaminated SL-1 site may have higher mutation rates in comparison to at least one of the control sites (Atomic City). The rather small sample size for Burn (N = 6) makes a quantitative estimate of the difference in mutation rate between contaminated and uncontaminated locations approximate at this time. Finally, the mutation rate obtained by combining the contaminated RWMC and SL-1 sites was significantly higher than the uncontaminated Burn and Atomic City sites combined.

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\* Only an abstract is given here as the full paper was not available.

# The use of biomarkers in the assessment of biological damage in the lugworm (*Arenicola marina*) and the lobster (*Homarus gammarus*) due to environmental contamination

J.L. Hingston<sup>a</sup>, D. Copplestone<sup>a</sup>, P. McDonald<sup>b</sup>, T.G. Parker<sup>c</sup>

<sup>a</sup> Environmental Research and Consultancy, University of Liverpool, Birkenhead, United Kingdom

<sup>b</sup> Westlakes Scientific Consulting, Whitehaven, United Kingdom

<sup>c</sup> British Nuclear Fuels Limited, Seascale, United Kingdom

**Abstract.** There is increasing realisation that environmental monitoring practices need to ensure the protection of the environment as a whole. Previous research has put forward a precautionary approach based upon the principle 'to protect the environment is to protect human populations'. A more suitable way to determine if the environment is being protected is to monitor organisms (sentinel species) that are in direct contact with contaminated materials such as sediment, water and air. This paper assesses biological damage in the lobster and the lugworm from several sites round the UK. These sites are influenced with enhanced levels of radionuclides, heavy metals or persistent organic compounds from anthropogenic practices. The biological damage is assessed by the use of biomarker techniques (in particular the comet assay) and analytical results have been obtained for site comparisons. To date biological responses in lugworms, for the reference site and one test site, have been shown to be significantly different. As a result of this, the work lends itself to the current discussion on the identification of suitable endpoints for use in ecological risk assessments. This paper also raises the issue of harmonizing environmental radiation protection with non-radiological environmental protection.

## 1. INTRODUCTION

Biomonitoring techniques and their application to human population studies have been well documented [1–3] but with focus of environmental protection now directed upon non-human biota [4, 5] there is a need to find techniques to demonstrate responses in alternative monitoring (sentinel) species. The species chosen for this work were the lobster (*Homarus gammarus*) and the lugworm (*Arenicola marina*). Species were selected based on criteria put forward by Phillips [6] which included abundance, length of life, ability to accumulate contaminants and suitability of tissues for sampling. More specific criteria concerning dosimetric models and public and political perceptions were also taken into account [7]. The lobster for example has frequently been reported as having one of the highest affinities for <sup>99</sup>Tc in the seafood group and as a result has achieved a high public and political profile [8]. The lugworm is not part of the human foodchain but its use for biomonitoring sediment bound contamination in the intertidal area is now being considered [9]. Radionuclide studies on lugworms however, are rare.

The aim of this work was to determine whether a biomarker technique derived from human biomarker studies can be used to show a response in the lobster and the lugworm. To achieve this the Single Cell Gel Electrophoresis (SCGE or the comet assay) was used, which measures DNA strand breaks in single cells. The technique is inexpensive, relatively simple and quick to perform, does not require dividing cells (unlike most cytogenetic techniques) and has already been developed for the mussel (*Mytilus edulis*) and the ragworm (*Nereis virens*) [10, 11]. DNA strand breakage can be induced by a wide range of contaminants and is not a signature of specific substance exposure [12]. As the comet assay can measure cumulative DNA damage caused by all genotoxic contaminants present in an environment [13] the method could be used to compare individuals from different industrial locations.

This paper assesses biomarker responses in the chosen reference species and demonstrates the application of the comet assay method. Results of a fieldwork sampling programme to study lugworms from four sites around the UK is also reported.

## 2. METHODS

### 2.1. Site descriptions

Field sites were selected after a literature review of the levels of radionuclides, metals and persistent organic pollutants from sites around the UK. Several sites were considered and the following sites were selected. The locations of these sites are depicted in Figure 1.

#### 2.1.1. Drigg and Braystones, Cumbria, United Kingdom

Situated within 2km either side of the main BNFL Sellafield discharge pipeline these sites are subject, under authorization, to discharge effluent containing radionuclides. These sites are predominantly contaminated with anthropogenic radionuclides and are included in the BNFL statutory environmental monitoring programme.

#### 2.1.2. Tamar Estuary, Plymouth Tor Point, Devon, United Kingdom

The Tamar estuary is subject to many industrial discharges, including persistent organic pollutants (POPs), metals, hydrocarbons and organometallic compounds, such as tributyltin.

#### 2.1.3. Seabait Ltd, Ashington, Northumberland, United Kingdom

Seabait Ltd breeds worms for use in sea angling, brood feeds and scientific research. This site was used as a substitute for the preferred reference site when the UK Foot and Mouth crisis of 2001 prevented access to The Rine (5km from Ballyvaughan), County Clare, Republic of Ireland.

### 2.2. Comet assay

A brief summary of the methods are presented here. Full details will be published in Hingston et al. [14].

#### 2.2.1. Lobster

Haemolymph was extracted and prepared cell suspensions were spun in a microcentrifuge at 717 x g before being resuspended in 0.65% low melting agarose and spread onto agarose-coated slides. The slides were immersed in cold lysing solution (2.5M NaCl, 10mM Tris, 0.1mM EDTA, 1% Sarcosyl, 1% Triton X-100 and 10% DMSO, pH 10) at 4°C for 1 hour. The slides were then rinsed in cold unwinding buffer (30mM NaOH, 2mM EDTA at pH 12.5) and left at 4°C for 20 min in fresh unwinding buffer. Electrophoresis was conducted at 4°C for 10 min at 25V (300mA). The slides were rinsed once in distilled water and washed three times for 5 min in neutralization buffer (400mM Tris at pH 7.5) before being stained with ethidium bromide and a coverslip added. Using a fluorescent microscope the DNA was visualized and the parameters, comet moment, tail moment, tail length and percentage of DNA in the tail were recorded using a CCD camera and an image analysis system. A minimum of 50 cells per slide were scored.

#### 2.2.2. Lugworm

Coelomic fluid was extracted from the lugworm and the comet protocol for lobster was applied with the following modifications; the unwinding step takes place for 30 min, electrophoresis for 20 min and after electrophoresis there are two washes with distilled water for 10 min and one with neutralization buffer for 10 min.

### 2.3. Dose responses

Dose responses were carried out to establish that biomarker responses could be detected in the selected species. Samples of haemolymph and coelomic fluid were extracted and exposed, *in vitro*, to doses of 0, 1, 3 and 5Gy using a Seifert isovolt 320 x-ray set. DNA damage was then assessed using the methods described.



*FIG. 1. Map of Great Britain illustrating the position of the field sites.*

### 3. RESULTS

#### 3.1. Biomarker responses

Using image analysis software the tail moment values for cells per individual were obtained. Tail moment represents a measure of tail length against a measure of DNA content in the tail. Tail moment is the measure used to depict dose and field responses in this paper, however due to a general consensus amongst investigators [15] values for the parameter tail length are also reported.

The dose responses and field responses (see Section 3.1.3) are plotted as histograms with the x-axis representing tail moment values in increments of 0.5. The last column ('more') on this axis represents any tail moment value which is above 11. This is necessary as it incorporates infrequent values which range from 11 to 174 in the examples described below.

##### 3.1.1. Dose responses – Lobsters

Only at doses of 3 and 5Gy can a clear visual response be observed in lobster cells (Figure 2). At 0Gy a positively skewed distribution is depicted with the majority of values located in the 0.5 category. At a dose of 1Gy the majority of values were still located in the 0.5 category but values were recorded in the 'more' category but not in the 0 category. At 3 and 5Gy the distribution of tail moment values were more evenly spread between 0.5 and 10.5 with a distinct shift towards higher tail moment values.

##### 3.1.2. Dose responses – Lugworms

A similar trend in response is observed in the lugworms cells (Figure 2). At doses of 0 and 1Gy the majority of values are located in the 0.5 category. At 3 and 5Gy the distribution of tail moment values are more evenly spread. At 5Gy, in particular, there is a shift in distribution towards higher tail moment values with the lowest values recorded in the 2.5 category.

##### 3.1.3. Lugworm field responses

An illustration of typical field responses in lugworms from the Seabait and Plymouth sites is given in Figure 3. Histograms of tail moment values for four individual lugworms from Seabait (reference site) are depicted alongside histograms for four individuals from Plymouth (contaminated site). The majority of the tail moment values for the Seabait individuals range from 0.5 to 3. The individuals from Plymouth, however, show a much broader distribution of tail moment values, with a large number of cell tail moment values in the 'more' category. It is, therefore, apparent that the individuals from the Plymouth site show more damage. The Kolmogorov-Smirnov test [16] was undertaken to determine the statistical significance of the differences in tail moment distribution between the Seabait and Plymouth individuals, depicted in Figure 3. Each of the four individuals from the Seabait site was compared to each of the four individuals from the Plymouth site using the Kolmogorov-Smirnov test ( $p < 0.05$ ) giving a total of sixteen tests. Plymouth lugworms b, c, and d were shown to differ significantly to Seabait lugworms a, b, c and d. The only individual from the Plymouth site not to differ significantly in tail moment distribution was Plymouth lugworm individual a which could not be distinguished from any Seabait individual.

The Seabait and Plymouth samples, however, were processed on separate occasions which leads to the possibility of inter-run experimental variation being introduced. Causes, effects and mitigation of this variation are considered in the discussion. Field responses in lobsters are currently being collated.

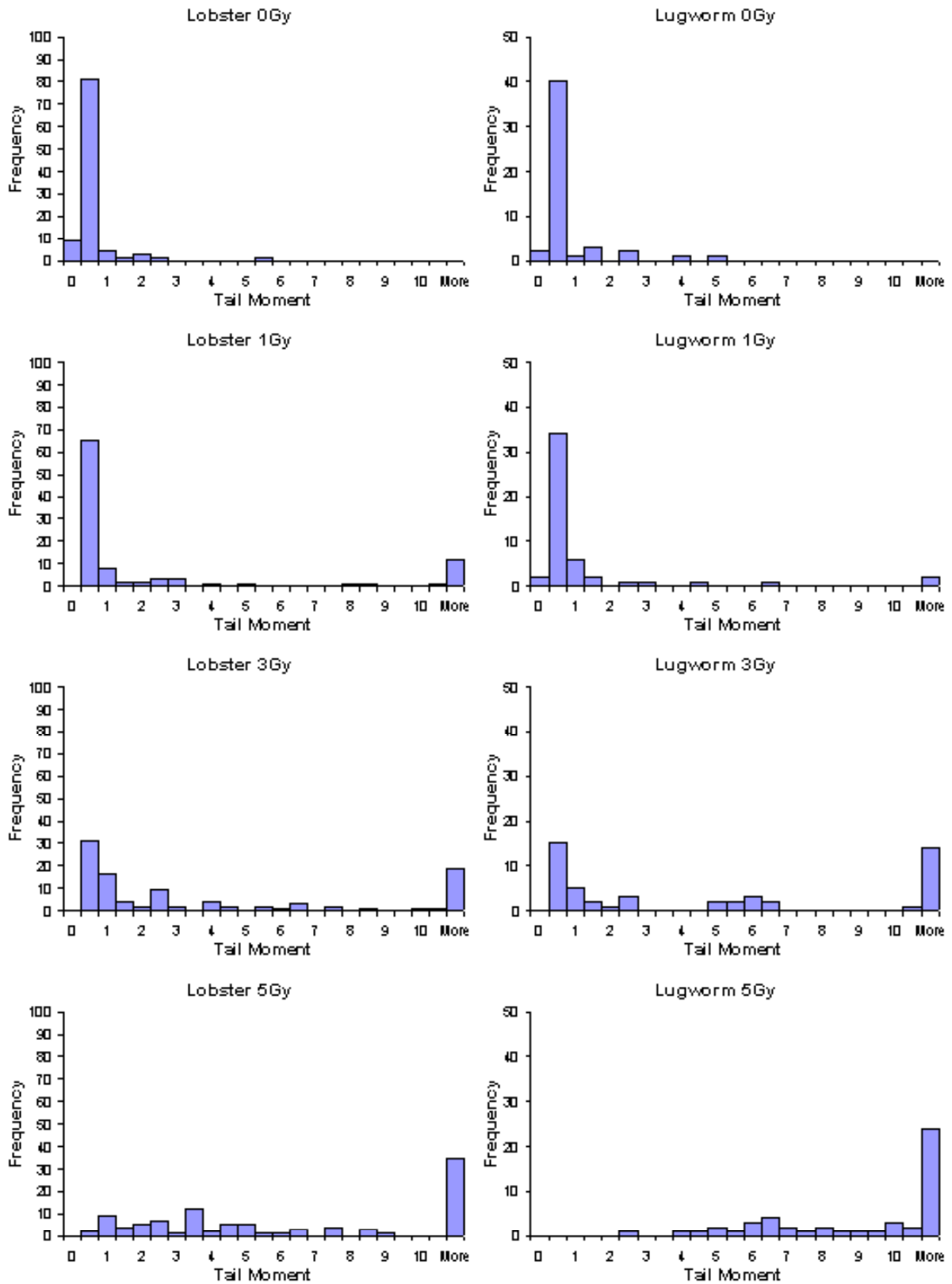


FIG. 2. Dose responses for the lobster ( $n=100$ ) and the lugworm ( $n=50$ ).

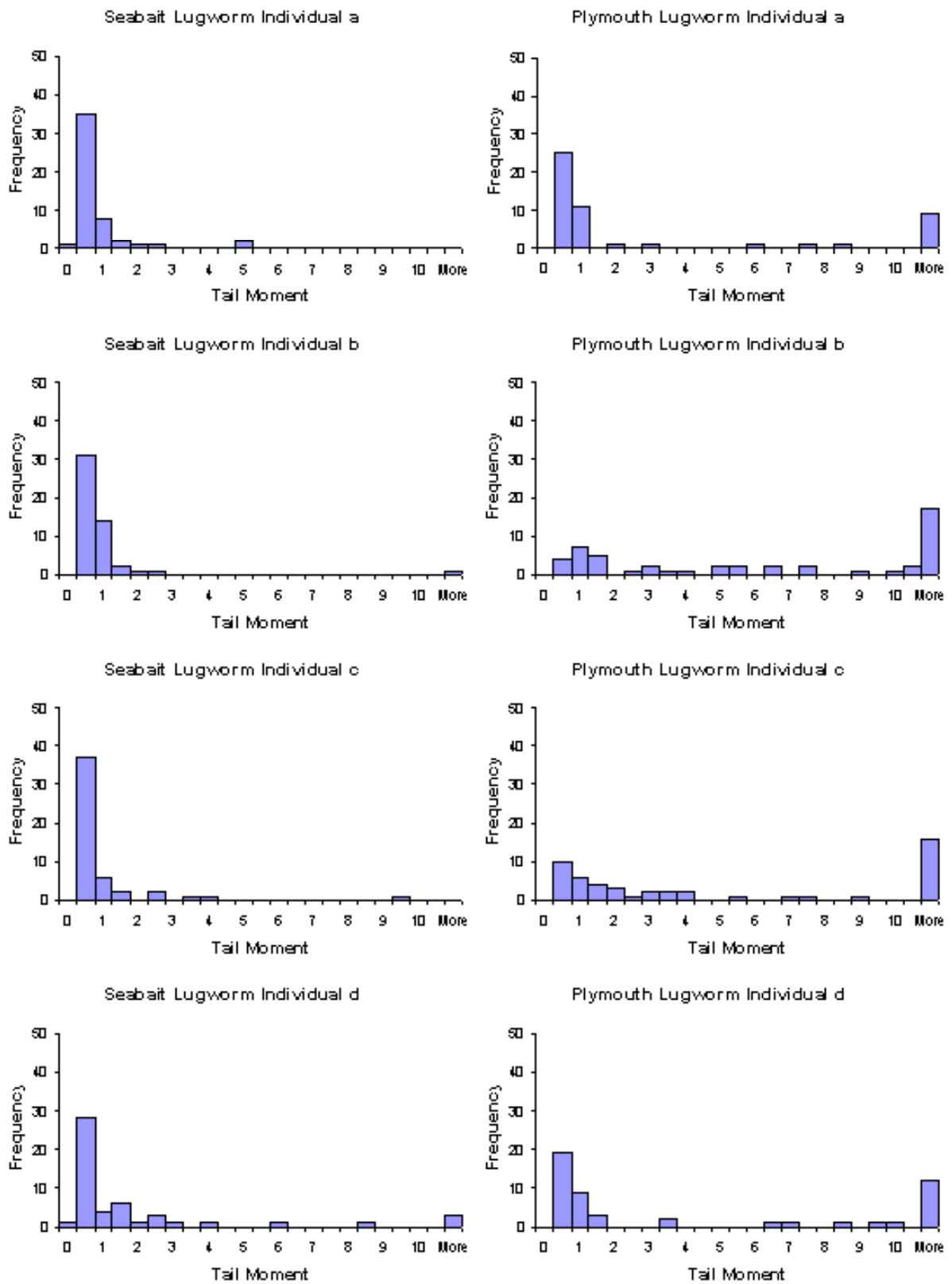


FIG. 3. Distribution of tail moment in cells from field site lugworms ( $n=50$ ).

### 3.2. Comparison of sites with enhanced level of radionuclides or persistent organics – illustrative example

The results presented here form part of a larger study looking at the DNA damage of lugworms from three contaminated sites and one reference site. Four individuals from each site were sampled and 50 of their cells assessed for DNA damage using the parameters tail moment and tail length. For each individual the mean tail moment and mean tail length values were calculated. These values, for each of the lugworms, were plotted against each other to show how the two parameters vary for and between individuals (Figure 4).

From Figure 4, a clear relationship between the two parameters can be observed. Those individuals with high tail moment values generally also have high tail length values. This is perhaps not unexpected, as the tail moment is partly dependent on the tail length. Looking at the variations between sites, it appears the individuals from the reference site (Seabait) have lower values for both parameters. In contrast, individuals from Plymouth show the highest values and individuals from Braystones and Drigg have values between these extremes. However three of the four individuals in both the Seabait and the Braystones groups have very consistent tail moment values within those groups. The tail length values in the Seabait individuals encompass a narrower range than the values observed in contaminated site individuals. In general individuals, from the contaminated sites show a wider variation in response of both tail moment and tail length.

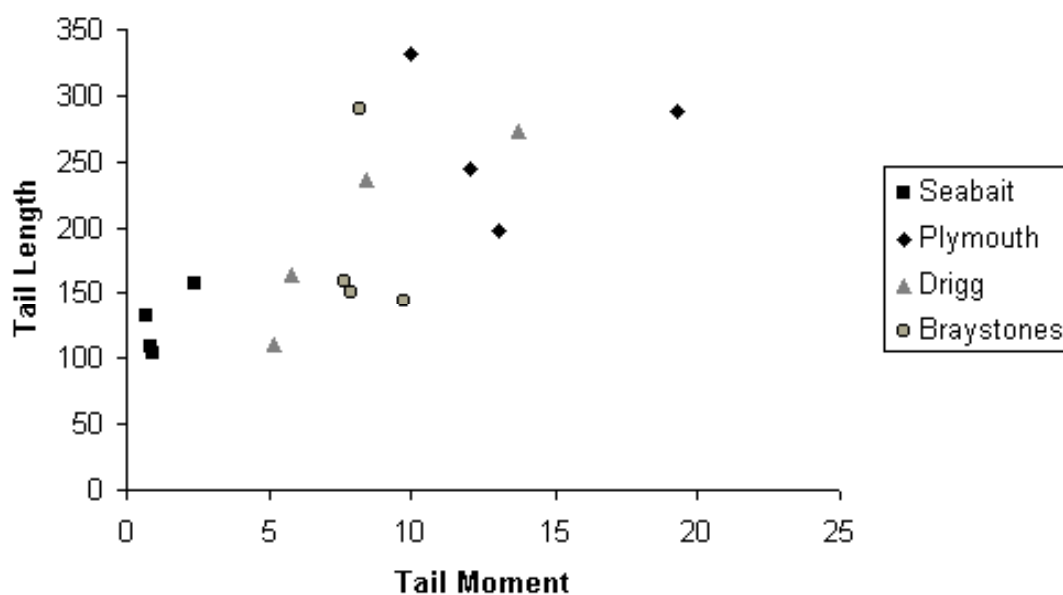


FIG. 4. Average tail moment values versus average tail length values for cells from individual lugworms from sites around the UK.

## 4. DISCUSSION

This work has shown that a biomarker response has been seen in lobster and lugworm cells subjected to SCGE. However in the lobster and the lugworm a clear, definite response is only seen at doses of 3 and 5Gy in vitro (Figure 2). It could be argued that this reflects either the sensitivity of the assay or the increased tolerance of these species to genotoxic insult. In humans however, the comet assay has detected a response at doses of 50mGy [17]. This supports the argument that the lobster and the lugworm are more robust species, capable of withstanding a certain level of environmental stress.



With the lugworm however, the distribution of tail moment values at 3 Gy are similar to the observed field responses in the Plymouth lugworm individuals in Figure 3. It has to be noted that the dose responses were irradiation specific and undertaken within controlled laboratory conditions on worms taken from a controlled environment (Seabait). The field responses, however, reflect the influence of multiple factors, such as contaminant mixes, which need further investigation.

The lugworm field responses depicted in Figure 3, show that levels of DNA damage do appear to differ between the contaminated site (Plymouth) and the reference site. The Kolmogorov-Smirnov test carried out stated that with the exception of Plymouth lugworm individual a, the tail moment distributions obtained for the Plymouth individuals were significantly different than those obtained for the Seabait individuals. This difference in response may only be due to the Seabait worms living in a protected environment without the stresses of, for example, predation found in their natural habitat. Sampling the intended reference site (County Clare) will verify this. The fact that one of the Plymouth individuals did not differ significantly from the Seabait individuals could indicate that the results obtained from populations will overlap. However to allow direct comparison of field site results gained in separate comet assay runs inter-run experimental variation should be accounted for. This variation can be caused, for example, by changes in electric current during the electrophoresis step of the assay. This could affect the rate that DNA migrates in the electrophoresis step. This project is currently investigating the use of an internal standard to account for inter-run experimental variation. By pooling a sample of blood from several reference lugworms/lobsters, separating the sample into different vials and freezing these vials for storage in liquid nitrogen, a sample could be run in every comet assay carried out for that species. To date, however, the freezing process has caused additional DNA damage. Cells from frozen sub samples are unscorable due to severe damage i.e. the cells are broken apart. The issue of variability within the comet assay is the subject of many papers and it has been concluded that more information is required on its sources, not only between experiments but also between cell and cell, and individual and individual [15].

In the site comparison (Figure 4), the tail moment and tail length values for Braystones and Drigg appear to be higher than those for the reference site and lower than those for Plymouth. With Braystones and Drigg primarily contaminated with radionuclides this is one of few studies which puts such sites in context with sites contaminated with other non-radioactive materials. Figure 4 further highlighted the low level responses of the Seabait worms seen in Figure 3 but also revealed a consistency in response in these worms. In contrast the lugworms from contaminated sites show a wider range of responses and these observations are regardless of inter-run experimental variation. Consequently one issue to consider is that of outliers. In cytogenetic studies rogue cells (cells with an abnormally high number of aberrations) have been defined as outliers and are removed from data sets before analysis [18]. Identification of outliers in the comet assay has not been defined but with the 'more' category of the histograms in Figures 2 and 3 containing infrequent numbers of high tail moment values, it is of concern here.

Despite the uncertainty regarding variability of the comet assay, this study has demonstrated that the comet assay can discern DNA damage in individuals from sites with differing levels of environmental contamination. Statistical confidence in the results will be increased as more individuals per site are assessed and an additional site (Milford Haven) is also being incorporated into the next stage of this project to represent a location predominantly contaminated with heavy metals. Contaminant analyses have also been carried out for all sites and initial results have shown that the majority of sites support a mixed inventory of contaminants. As no one site is solely contaminated by one type of contaminant, exposure to a low-level mix of contaminants may have unpredictable effects compared to the effects observed from laboratory exposures to single contaminants [19]. Therefore due to possible synergistic and antagonistic effects this work can, at present, only suggest whether certain contaminants have an effect on the level of DNA strand breakage in biota. To define the effects of a contaminant further, laboratory based exposure studies would have to be employed. Further investigation into contaminants, mixtures and their effects will place biological responses to radiological and non-radiological contaminants in the same context and should become instrumental to environmental protection in the future.

## ACKNOWLEDGEMENTS

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# Statistics of extreme values – comparative bias associated with various estimates of dose to the maximally exposed individual

M.D. Wilson, T.G. Hinton

University of Georgia, Savannah River Ecology Laboratory, Aiken, SC, United States of America

**Abstract.** Protection of the environment from anthropogenic radiation is an on-going international concern. The paradigm currently in use argues that the population is adequately protected if the maximally exposed individual's dose is below a certain limit. Based on data sampled from natural populations, resource managers need to be able to test the hypothesis that the maximally exposed individual's dose is acceptable. Recognizing the difficulty of sampling the maximally exposed individual within a contaminated environment, risk assessors have used various alternative approaches. One statistic currently used is the upper 95% confidence limit on the sample mean. An alternative approach is to make no distributional assumptions and use the sample maximum value as an estimate of the maximally exposed individual. Other managers assume that an increased conservatism, added to the model parameters used to estimate risk, will compensate for the inability to sample the maximally exposed individual. While some risk assessors have changed the internationally accepted paradigm and applied recommended dose limits to representatively, rather than the maximally exposed individuals. We propose an alternative: given a sample, find the maximum likelihood estimates of the assumed population parameters and use the 99<sup>th</sup> percentile as an estimate of the maximally exposed individual. To determine the effectiveness of our proposed alternative, we use computer simulation techniques to generate a "population" of doses with known distributional qualities, and then mathematically "sample" this population and compare the different statistics. The simulation procedure is repeated many times, each time producing a measure of the distance between the estimate and the "true" value. We are thus able to quantify the bias associated with several approaches used to determine compliance with dose limits.

## 1. INTRODUCTION

There is considerable interest in understanding the risks that nonhuman biota experience when exposed to radiation. A consensus, however, as to how to conduct radioecological risk analyses does not exist. The science associated with radioecological risks is actually fairly well developed in that a paradigm of acceptable dose rates for nonhuman biota have been largely agreed upon internationally. For example, a dose rate of 10 mGy d<sup>-1</sup> has been generally accepted as a level below which populations of aquatic animals are thought to remain healthy [1]. Indeed, the U.S. Department of Energy has proposed these same dose limits within a technical standard for all of their facilities. Implementing this criterion, however, is problematic because the criterion states that if the *maximally exposed individual* receives  $\leq 10$  mGy d<sup>-1</sup> then the *population* is adequately protected [2]. When attempting to estimate the dose of the maximally exposed individual in a population several difficulties arise. If the assumed distribution is Normal, there is no population maximum because the tails of the Normal distribution go to infinity. This is the case for many continuous distributions. Hence there is no unbiased statistic for the population maximum. Realistically, how can the dose rate to maximally exposed individuals be determined? What sample statistic should be used to estimate such an extreme value?

Recognizing the difficulty of determining what the maximally exposed individual's dose rate is, risk assessors are considering other approaches and other parametric estimators. Our intention in this paper is to compare some of the popular alternative approaches, as well as to suggest a statistic that we think realistically approximates the criterion as established by the International Atomic Energy Agency [2]. We hope that this paper will stimulate discussion concerning the problem resource managers face when trying to test the hypothesis that the maximally exposed individual's dose is below an established criterion.

## 1.1. Qualitative approaches

Two approaches qualitatively address the problem of identifying maximally exposed individuals and will be briefly discussed here before we proceed with a more rigorous mathematical comparison of other methods. The first qualitative approach is a well-accepted method that purposely uses conservative parameters in a computer simulation to estimate the upper bound maximum dose rate that an organism might be exposed to. This so-called screening model method was first used in human risk analyses [3], but was quickly adapted for ecological risk analyses by both U.S. [4, 5] and European groups [6]. The concept of a screening model is that if the computer predictions fall below the dose rate criterion (e.g. 10 mGy d<sup>-1</sup> for aquatic organisms), then there is high confidence that the actual dose rate received by animals in the contaminated environment are much less. The confidence that the model over-estimates the actual dose rates experienced in the field stems from using upper bound estimates of important model parameters, such as distribution coefficients, concentration ratios, and bioavailability constants. If, however, dose rates based on the screening model are above the dose rate criterion, then a series of steps are invoked that add more realistic, site-specific data to the risk analysis. If failure to comply with the criterion continues, then field samples of the biota are eventually taken and their contaminant body burdens are used to more accurately estimate dose rates. Even though actual body burden data are obtained from a sample of the population, the fundamental problem of determining the dose rate to the maximally exposed individual within that population still remains. Thus, screening levels models are very useful, but only as long as their predictions fall *below* the target criterion.

We also classify the second method as qualitative because it changes the basic paradigm for protecting biota established by the IAEA [2]. At least one group has rationalized that a shift in the paradigm is warranted, and that if a *representative* animal within the population receives < 10 mGy d<sup>-1</sup>, then the population is adequately protected [5]. If the dose of a representative individual is estimated using the sample mean, this change in dose limit interpretation could result in 50% or more of the population receiving dose rates > 10 mGy d<sup>-1</sup>, rather than only maximally exposed individuals as the original guidelines established. Although a change in the paradigm may eventually prove to be correct, we are unaware of peer-reviewed data that support such a drastic shift in the established criteria, and thus consider it an untested, qualitative method.

## 1.2. Quantitative methods

In the remainder of the paper we use Monte Carlo simulations to “sample” from a known, model-generated “population”. We can then compare the ability of various sample statistics to estimate the maximally exposed individual’s dose. (The latter is a known value from our model-generated population, whereas in a field sampling situation complete knowledge of the distribution does not exist). The statistics we tested were:

- (1) the 95<sup>th</sup> percentile of the sample mean, (i.e. the value at which the probability of the mean lying above this value falls to 5%);
- (2) the maximum from a sample of the population;
- (3) the maximum likelihood estimate (MLE) for the 99.99<sup>th</sup> percentile to estimate the maximally exposed individual’s dose; and
- (4) the MLE for the 99.99<sup>th</sup> percentile to estimate the true population 99.99<sup>th</sup> percentile.

The last statistic is proposed as a small change in the current criterion by making the assumption (not validated here) that if 99.99% of the population has received less than some appropriate regulatory limit, then the population is adequately protected. The third and fourth statistics differ in that one is attempting to estimate the population maximum, while the other is attempting to estimate the 99.99<sup>th</sup> percentile of the population.

Although the challenge to a resource manager is in determining if the maximally exposed individual’s dose rate is below an accepted criterion (e.g. 10 mGy d<sup>-1</sup>), in this paper we use the Maximally Exposed Individual’s <sup>137</sup>Cs Activity Concentration (MEIAC) as the endpoint for comparing the various parameters listed above. Activity concentrations are used here because of the lack of appropriate,

field-derived dose data. The use of activity concentrations (i.e.  $^{137}\text{Cs}$  body burdens) are a reasonable surrogate for dose rate in that an organism's internal dose is derived directly from its contaminant burden, and the use of activity concentrations allowed us to use real data in constructing our model simulations. Our conclusions should be very similar to those obtained here, had we had dose rate data to work with.

## 2. METHODS

### 2.1. Field data

Population characteristics used in the model simulations were obtained from  $^{137}\text{Cs}$  contaminated fish sampled in 1996 from a lake located on the Savannah River Site, a former U.S. Department of Energy nuclear production facility located in South Carolina, USA. We used data on the top predatory fish in the lake (Largemouth Bass;  $n = 194$ ;  $\mu = 267 \text{ Bq kg}^{-1}$ ;  $\sigma = 61 \text{ Bq kg}^{-1}$ ), and from a sample of assorted fish species normally preyed upon by bass ( $n = 52$ ;  $\mu = 105 \text{ Bq kg}^{-1}$ ;  $\sigma = 42 \text{ Bq kg}^{-1}$ ). Kolmogorov-Smirnov tests revealed that the bass data were normally distributed ( $p = 0.35$ , Figure 1), but not the prey data ( $p = 0.04$ , Figure 1). A log-transformation of the prey data normalized the distribution ( $p = 0.31$ ). These data sets were chosen so that we could test the four statistics under both normally and log-normally distributed assumptions.

### 2.2. Model simulation

We simulated a population by taking a large, model-derived sample of size 5000, which represented a natural, finite population from approximately the same distribution as our field data. The mean ( $\mu$ ) and standard deviation ( $\sigma$ ) used for the model-simulated populations were the sample mean and sample standard deviation from  $^{137}\text{Cs}$  concentrations in the fish populations described above. From the model-simulated population we then randomly drew a sample ( $n=194$  for the bass simulation and  $n = 52$  for the fish prey simulation) and calculated the four statistics listed above, as well as the model-simulated population maximum. We then calculated the bias by subtracting the statistic of interest from the maximum of the simulated finite population and saved all results. To calculate the bias when estimating the true 99.99<sup>th</sup> population percentile, we used the theoretical normal percentile in place of the population maximum. This procedure of deriving a population of 5000 and randomly sampling from it was repeated 1000 times for each dataset. Histograms were then developed for each of the statistics and compared to the histogram for the model-derived population maximum (i.e. the desired criterion). The simulation and random sub-sampling used a Markov Chain/Monte Carlo (MC/MC) procedure similar to a parametric bootstrap [7]. The parametric bootstrap differs from a regular bootstrap in that, instead of re-sampling the data with replacement, we obtain parameter estimates from the data for the assumed distribution and then simulate a random sample, calculate our statistic, and repeat this procedure a large number of times. Model simulations were necessary in order to compare the population maximum to each proposed statistic, i.e., to calculate the bias. The estimate of the bias associated with each statistic was the mean of all 1000 biases obtained during the simulation procedure. The MC/MC estimate of the 95% confidence interval for the bias was the 25<sup>th</sup> and 975<sup>th</sup> elements of the ranked bias vector. Likewise, the confidence intervals around the Maximally Exposed Individual's Activity Concentration for each statistic were the 25<sup>th</sup> and 975<sup>th</sup> elements of the ranked 1000 statistics obtained during the simulation procedure. All calculations, simulations, and sub-samplings were performed in S plus. The normality tests were performed in SYSTAT.

## 3. RESULTS

### 3.1. Bass data

Monte Carlo estimates of the expected bias, the 95% confidence intervals for the bias and for the statistic are shown in Tables 1 and 1a for the model-simulated data derived from the normally distributed bass population. The second and third columns show the bias and 95% confidence interval for the bias when the MEIAC is the 'parameter' of interest. The fourth column of Table 1 is the estimated 95% confidence interval for the statistic itself. Table 1a shows the bias and confidence interval when the 99.99<sup>th</sup> population percentile is the parameter of interest. The results can be easily visualized in a histogram (Figure 2).

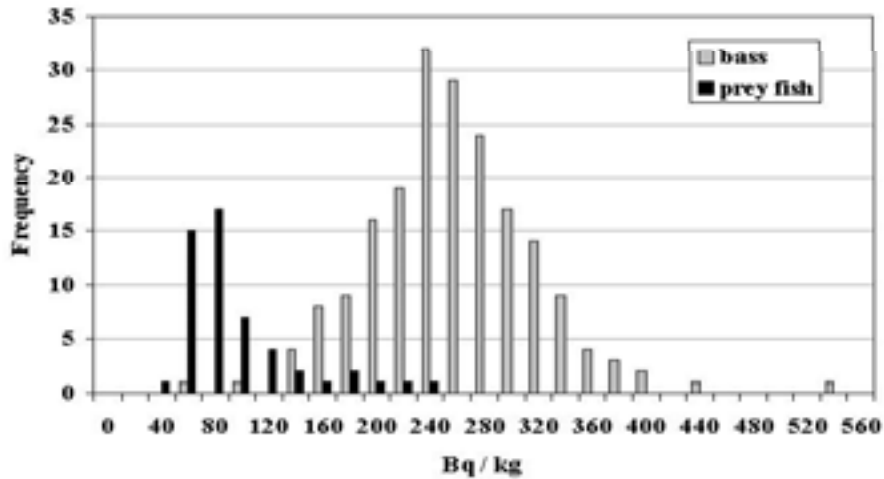


FIG. 1. Histograms of <sup>137</sup>Cs concentrations in bass (n = 194) and an assortment of fish species preyed upon by bass (n = 52). Data are from a 1996 sampling of a Cs-contaminated lake and show the different distributions of the two populations.

TABLE 1. BASS DATA SET—BIAS ESTIMATE FOR THE MAXIMALLY EXPOSED INDIVIDUAL'S ACTIVITY CONCENTRATION (MEIAC) WHEN USING THE 95<sup>th</sup> PERCENTILE OF THE MEAN, THE SAMPLE MAXIMUM, AND THE MLE FOR 99.99<sup>th</sup> PERCENTILE OF THE DISTRIBUTION. BIAS INDICATES THE DIFFERENCE IN Bq kg<sup>-1</sup> OF THE STATISTIC'S ESTIMATE OF THE MEIAC FROM THE POPULATION MEIAC. A NEGATIVE BIAS INDICATES THAT THE STATISTIC OVER-PREDICTED THE MEIAC BY THE NUMBER OF Bq kg<sup>-1</sup> SHOWN

Statistic of Interest	Mean Bias of MEIAC	95% CI of Bias	95% CI of Statistic
95 <sup>th</sup> Percentile of Mean	224	191 to 269	259 to 277
Sample Maximum	57	0 to 118	396 to 494
99.99 <sup>th</sup> Percentile MLE	-2	-43 to 47	470 to 519

TABLE 1A. BASS DATA SET—ESTIMATING THE TRUE POPULATION 99.99<sup>th</sup> PERCENTILE BY THE MLE. UNITS ARE Bq/kg

Mean Bias	95% CI of Bias	95% CI of Statistic
0.3	-25 to 23	470 to 519

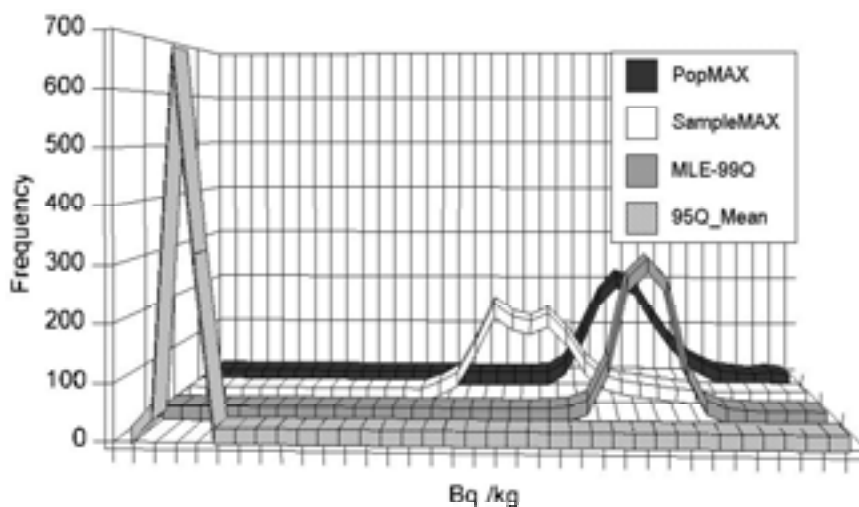


FIG. 2. Histograms of 1000 model simulations derived from a normally distributed population of bass. The maximum likelihood estimate of the 99.99<sup>th</sup> percentile (MLE-99Q), the sample maximum, and the 95<sup>th</sup> percentile of the sample mean (95Q\_Mean) are compared to the endpoint of concern (i.e. the population maximum).

Note that for this dataset, the maximum likelihood estimate (MLE) for the 99.99<sup>th</sup> percentile (Table 1a) has the lowest bias for both the Maximally Exposed Individual’s Activity Concentration (MEIAC) and the true distribution 99.99<sup>th</sup> percentile. It, of course, has a smaller bias when estimating the true 99.99<sup>th</sup> percentile, than when estimating the MEIAC. It also has reasonably short confidence intervals. The first row, third column in Table 1 shows that the bias for the 95<sup>th</sup> percentile of the mean was between 191 and 269 Bq kg<sup>-1</sup> 95% of the time. The sample max will never have a bias less than zero, because the population max will always be greater than or equal to the sample max. Occasionally, we will sample the population maximum or values very close to it, yielding a bias of zero or near zero. The bias for the sample maximum when estimating the population maximum was between 0 and 118 Bq kg<sup>-1</sup> 95% of the time. The bias for the MLE 99.99<sup>th</sup> percentile when estimating MEIAC was between -43 and 47 Bq kg<sup>-1</sup>. Finally, if we seek to estimate the true 99.99<sup>th</sup> percentile, rather than the MEIAC, using the MLE for the 99.99<sup>th</sup> percentile resulted in an average bias of 0.3 Bq kg<sup>-1</sup> and a 95% confidence interval between -25, and 24 Bq kg<sup>-1</sup>. Also note that the confidence interval around the 95<sup>th</sup> percentile of the mean (259 to 277) was of the same order of magnitude and at about the same range as the confidence interval around the bias (191 to 269), showing that this statistic is a very poor estimator for the MEIAC.

### 3.2. Fish species preyed upon by bass

A dataset consisting of mixed prey species (n=52) was analyzed using a log-normal assumption. The log-transformed data passed the KS test (p=0.31), indicating there is insufficient evidence to reject the null hypothesis that the data are log-normally distributed. Tables 2 and 2a show the simulation results for the fish prey species, and the associated histograms are presented in Figure 3. Here again the bias for the MLE for the 99.99<sup>th</sup> percentile was by far the smallest among the statistics tested.

The next set of analyses (Tables 3 and 3a, with associated histograms in Figure 4) were performed to see what would occur if, as sometimes happens in practice, the investigators incorrectly assumed that the data were normally distributed. The 99.99<sup>th</sup> percentile remained the best estimator of the population maximum. However, its bias increased from -6 (Table 2) to 117 Bq kg<sup>-1</sup> (Table 3). This suggests that misidentification of the distribution will decrease your ability to accurately estimate the Maximally Exposed Individual’s Activity Concentration.

TABLE 2. FISH PREY DATA SET—ESTIMATING THE MEIAC USING A LOG-NORMAL ASSUMPTION. BIAS ESTIMATE FOR THE MAXIMALLY EXPOSED INDIVIDUAL’S ACTIVITY CONCENTRATION (MEIAC) WHEN USING THE 95<sup>th</sup> PERCENTILE OF THE MEAN, THE SAMPLE MAXIMUM, AND THE MLE FOR 99.99<sup>th</sup> PERCENTILE OF THE DISTRIBUTION. BIAS INDICATES THE DEVIATION IN Bq kg<sup>-1</sup> OF THE STATISTIC’S ESTIMATE OF THE MEIAC FROM THE POPULATION MEIAC. A NEGATIVE BIAS INDICATES THAT THE STATISTIC OVER-PREDICTED THE MEIAC BY THE NUMBER OF Bq kg<sup>-1</sup> SHOWN

Statistic of Interest	Mean Bias of MEIAC	95% CI of Bias	95% CI of Statistic
95 <sup>th</sup> Percentile of Mean	257	197 to 361	91 to 110
Sample Maximum	137	34 to 248	167 to 299
99.99 <sup>th</sup> Percentile MLE	-6	-135 to 118	282 to 479

TABLE 2A. FISH PREY DATA SET—ESTIMATING THE TRUE POPULATION 99.99<sup>th</sup> PERCENTILE BY THE MLE USING A LOG-NORMAL ASSUMPTION. UNITS ARE Bq/kg

Mean Bias	95% CI of Bias	95% CI of Statistic
-2	-120 to 79	282 to 479

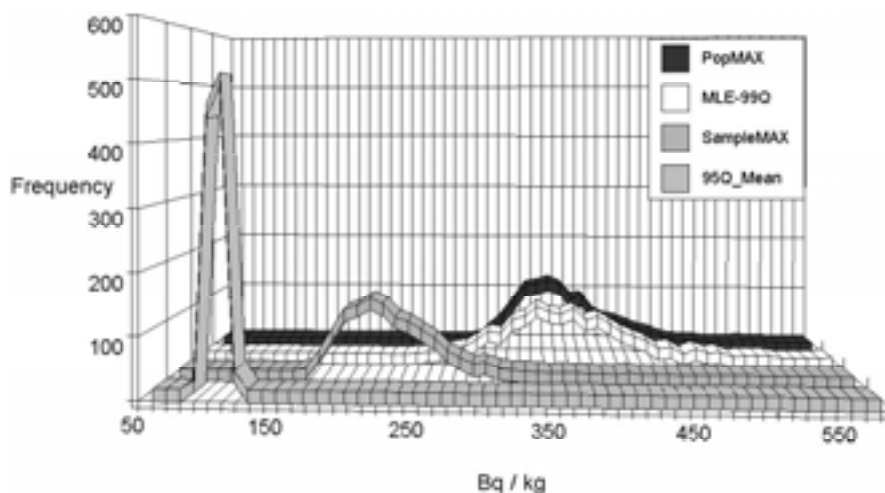


FIG. 3. Histograms of 1000 model simulations derived from a log-normally distributed population of contaminated fish preyed upon by bass. The maximum likelihood estimated of the 99.99<sup>th</sup> percentile, the sample maximum, and the 95<sup>th</sup> percentile of the sample mean are compared to the endpoint of concern (i.e. the population maximum).

TABLE 3. FISH PREY DATA SET—ESTIMATING THE MEIAC USING A NORMAL ASSUMPTION. BIAS ESTIMATE FOR THE MAXIMALLY EXPOSED INDIVIDUAL'S ACTIVITY CONCENTRATION (MEIAC) WHEN USING THE 95<sup>th</sup> PERCENTILE OF THE MEAN, THE SAMPLE MAXIMUM, AND THE MLE FOR THE 99.99<sup>th</sup> PERCENTILE OF THE DISTRIBUTION. BIAS INDICATES THE DIFFERENCE IN Bq kg<sup>-1</sup> OF THE STATISTIC'S ESTIMATE OF THE MEIAC FROM THE POPULATION MEIAC. A NEGATIVE BIAS INDICATES THAT THE STATISTIC OVER-PREDICTED THE MEIAC BY THE NUMBER OF Bq kg<sup>-1</sup> SHOWN

Statistic of Interest	Mean Bias of MEIAC	95% CI of Bias	95% CI of Statistic
95 <sup>th</sup> Percentile of Mean	254	194 to 355	96 to 117
Sample Maximum	142	40 to 263	165 to 309
99.99 <sup>th</sup> Percentile MLE	117	38 to 225	203 to 292

TABLE 3A. FISH PREY DATA SET—ESTIMATING THE TRUE POPULATION 99.99<sup>th</sup> PERCENTILE BY THE MLE USING A NORMAL ASSUMPTION. UNITS ARE Bq/kg

Mean Bias	95% CI of Bias	95% CI of Statistic
84	22 to 153	203 to 292

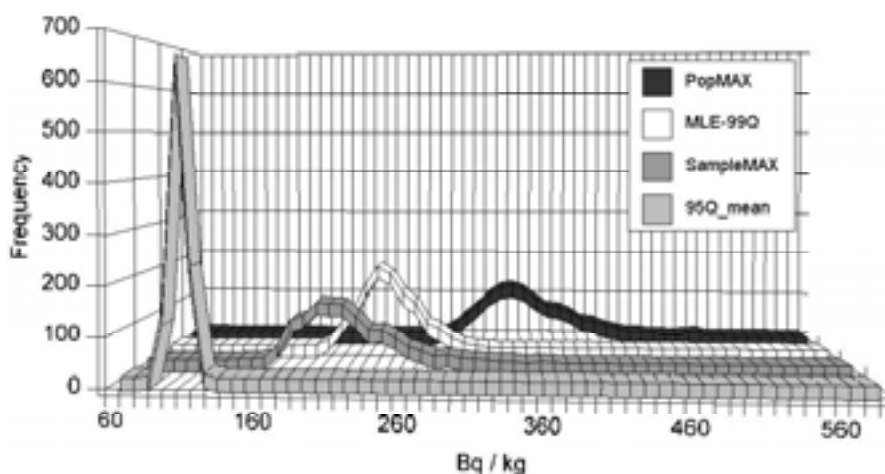


FIG. 4. Histograms of 1000 model simulations assuming a normal distribution of log-normally distributed data (fish prey from Figure 3). The maximum likelihood estimate of the 99.99<sup>th</sup> percentile, the sample maximum, and the 95<sup>th</sup> percentile of the sample mean are compared to the endpoint of concern (i.e. the population maximum).



## 4. DISCUSSION

As mentioned in the Introduction, several difficulties exist when attempting to estimate the dose of maximally exposed individuals in a population. For example, there is no population maximum in a Normal distribution because the tails of the distribution go to infinity. Thus, there is no unbiased statistic for the population maximum. Additionally, the quantiles of the sample mean are highly dependent on sample size and are not necessarily close to the quantiles of the population that the sample came from. The use of the 95<sup>th</sup> percentile of the sample mean could possibly result in the perverse result that an estimate of the MEIAC is below that of an observed sample value.

### 4.1. Implementation issues

For this investigation, we used MC/MC estimates of the 95% confidence intervals around each statistic, using a procedure similar to a parametric bootstrap. These confidence bounds are valid so long as the distributional assumptions hold and can be used to determine the rejection region in a hypothesis test. When the sample size is quite large, the MLE for the 99.99<sup>th</sup> percentile will have an asymptotic distribution that is normal. The confidence interval can then be constructed using the asymptotic standard error and the appropriate quantile from the standard normal distribution, depending on the degree of significance desired. However, because we are estimating an extreme quantile, the convergence to normality is slow, and for small to moderately large samples, the error in the normal approximation will be large. For smaller samples, a true bootstrap could be performed on the data and bootstrap estimates of the confidence bounds obtained. Recent improvements in software have made the bootstrap reasonably simple to perform in such packages as Excel, S plus, and Matlab. In practice, an investigator would use the confidence interval around each statistic to conduct the following test:

- $H_0$ : the maximally exposed individual's dose is less than or equal to the current regulatory limit,
- $H_a$ : the maximally exposed individual's dose is greater than the current regulatory limit, and would reject  $H_0$  if the lower bound of the confidence interval for the statistic being used were greater than the regulatory limit.

It should be noted here that these results apply only to natural populations that have the same distribution as those shown here. The bias and variance of these statistics will vary with sample size and the underlying distribution that the data are drawn from. The 99.99<sup>th</sup> percentile is difficult to interpret for small populations. For example, in a population of size 5000, only about 2 individuals will lie at or above the 99.99<sup>th</sup> percentile. If the population has fewer than 1000 members, the meaning of the 99.99<sup>th</sup> percentile is difficult to interpret. For this reason, we suggest exploring the possibility of using the 99<sup>th</sup> (as opposed to the 99.99<sup>th</sup> percentile), and shifting the regulatory criterion appropriately to argue that if the top 1% (as opposed to the top 0.1%) of the population has a dose less than or equal to the regulatory limit, then the population is adequately protected. The 99<sup>th</sup> percentile was tested (results not shown here) and was found to be a biased statistic for the MEIAC. However, the MLE for the 99<sup>th</sup> percentile is clearly unbiased for the 99<sup>th</sup> percentile of the distribution. So if regulatory limits could be written for the top 1% of the population rather than the maximally exposed individual, the MLE for the 99<sup>th</sup> percentile would be a superior statistic.

## 5. CONCLUSIONS

This work highlights the difficulty resource managers face if tasked with testing whether or not their data set complies with an extreme statistic criterion (i.e. maximally exposed individuals). The data indicate that statistics based on characteristics of the sample mean or sample maximum have considerable bias when attempting to estimate a maximum. For the data sets we tested, the MLE for the 99.99<sup>th</sup> percentile proved to be the best statistic for estimating the Maximally Exposed Individual's Activity Concentration in that it had the smallest bias and the shortest confidence intervals. It was also shown that the MLE for the 99.99<sup>th</sup> percentile was unbiased for the true population percentile. However, its accuracy was reduced if the distribution (normal versus log-normal) was not properly identified. It is our contention that shifting the paradigm from regulating the dose of the maximally exposed individual to regulating the top 0.1% of the population is not a significant change. Therefore, if adherence to the IAEA criteria [2] is desired, we recommend using the MLE for the 99.99<sup>th</sup>

percentile to estimate to true population percentile, rather than to estimate the dose to the maximally exposed individuals. Furthermore, if appropriate regulatory limits for the top 1% of the population can be identified, then we suggest using the 99<sup>th</sup> percentile, rather than the 99.99<sup>th</sup>.

## ACKNOWLEDGMENTS

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# Radiation effects on the environment beyond the level of individuals\*

U. Kautsky<sup>a</sup>, M. Gilek<sup>b</sup>

<sup>a</sup> Swedish Nuclear Fuel and Waste Management Company, Stockholm, Sweden

<sup>b</sup> Södertörns Högskola, Huddinge, Sweden

**Abstract.** Currently there are several initiatives to address radiation effects on the environment. A common standpoint is that the goal is to protect the environment, i.e. the function of ecosystems and maintaining populations. However, the questions of effect are usually studied on the individual, organs, cell or sub-cellular level. This leaves a large gap between where the measurement endpoint (individual and lower levels) and the assessment endpoint (population and ecosystems) which is not addressed sufficiently.

This paper discusses effects and processes above the individual level important for a framework for addressing effect on the environment. First it is realised that there are few examples how radiation-effects can have major disturbance only on level above individuals. All population effects and ecosystem effect are mediated through the individual level. This is contrary some other environmental stressors, e.g. nutrients and highly biomagnifying substances (e.g DDT, PCB). However, even there are effect on the individual level few of them will affect the populations or ecosystem function and usually in a lower extent. The impact of individual changes on the population and ecosystem level is dependent, on population size, total biomass, life history, reproduction investment, generation time, inter- and intra-specific competition, predation and resource exploitation.

Thus for regulatory framework, if the objective is to protect the function of the ecosystem and maintaining populations (in the sense of the Rio declaration), it must be realised that addressing effects on individuals can be a overly pessimistic approach.

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\* Only an abstract is given as the full paper was not available.

# The FASSET radiation effects database

## *A demonstration*

D. Coppelstone<sup>a</sup>, I. Zinger-Gize<sup>b</sup>, D.S. Woodhead<sup>c</sup>

<sup>a</sup> Environmental Research and Consultancy, University of Liverpool, Birkenhead, United Kingdom

<sup>b</sup> Environment Agency, Warrington, United Kingdom

<sup>c</sup> Centre for Environment, Fisheries and Aquaculture Sciences, Lowestoft Laboratory, Lowestoft, United Kingdom

**Abstract.** FASSET (Framework for ASSESSment of Environmental impacT) is an EC funded project, within the 5<sup>th</sup> framework programme (Euratom) in the field of nuclear energy. Its main objective is to create a framework for assessing the impact of radioactive contamination on non-human biota. Effects analysis forms an integral part of the framework, taking account of available data on the biological effects of radiation. For the purpose of FASSET, dose rates need to be estimated, and dose (rate) – effect relationships need to be identified: both will form an important input into the overall framework. This will provide the basis for determining dose rates at which different degrees of effects in the environment may be expected.

In order to start identifying relevant biological effects for different organisms and different environments, a Microsoft<sup>®</sup> Access database is being assembled in which data from the literature for a number of wildlife groups (e.g. birds, plants, fish) are being compiled. This information is grouped under four umbrella effects (morbidity, mortality, reproduction, and mutation). The database also records whether data are suitable for use in deriving RBE (relative biological effectiveness) values for different types of radiation.

The database is divided into two main functions: data entry and query reports. Its aim, under FASSET, is to develop dose-effect and dose-response-relationships, which can be used to judge environmental consequences resulting from exposure to ionizing radiation. Quality assurance exercises have been used for both data entry consistency by the various organisations taking part, and for detecting numerical inaccuracy.

A number of constraints in data entry were necessary in order for the database to be manageable and reasonably focused, recognizing that judgement will need to be applied for the purposes of FASSET.

The structure and use of the database will be demonstrated including examples of its application using the query options. The intention is to publish the database on the FASSET web-site, and consider its expansion beyond the duration of this project. The database will allow the identification of gaps in knowledge and, so, give direction to future research.

## 1. BACKGROUND

The FASSET project's Technical Annex [1] clearly specifies, under its Work Package 3 (WP3) on "Effects", the need to gather scientific information, in order to:

- identify a range of dose rates at which different degrees of effects in the environment would be expected;
- derive dose rate/response relationships for the chosen endpoints;
- determine dose-rate thresholds or minimum dose rates at which effects in the environment are expected to be minimal with a high degree of certainty; and,
- help define the reference organisms for dosimetric purposes, for integration in WP1 "Dosimetry";

and more generally to:

- describe the biological effects of ionizing radiation that are likely to be of significance for protection, at the intended biological level, in an environmental context;
- identify data in the literature that may be of use in determining the relative biological effectiveness (RBE) of the different types of radiation with respect to chosen endpoints; and,

— identify reference organisms, which can be used in a radiation protection framework.

The way in which to gather information was only described in the Technical Annex as:

“The available information will be organised into a format that will indicate the approximate dose rate/response relationships and, therefore, the threshold dose rates at which minor radiation effects can currently be expected to become apparent in the defined biological processes in the selected target organisms.”

Information to be gathered should include data on:

- acute and high dose rate exposures;
- chronic, low-level exposures extending over a significant fraction of the life time of the organism; and,
- endpoints such as morbidity, mortality, reproduction (as fertility and fecundity) and mutation rate.

The estimated dose rates will form an important input to the overall framework. They will also indicate the range likely to arise from controlled radioactive waste management practices, and that are likely to persist in the long-term after an accidental release of radionuclides when remediation activities might be required. This will provide the appropriate context for the collection and organisation of the data obtained from the literature. This has been undertaken in several steps.

## 2. COLLATION OF REFERENCES

A review of the available literature on the effects of radiation on plants and animals, other than humans, was undertaken in context of generating the required tools within FASSET. The workload was divided between six organisations (Swedish Radiation Protection Authority; Environment Agency of England and Wales, UK; Centre for Environment, Fisheries and Aquaculture Sciences, UK; Spanish Research Centre in Energy, Environment and Technology; Norwegian Radiation Protection Authority; and Westlakes Scientific Consulting Ltd, UK). The literature search was based initially on seven categories: mammals, plants, soil fauna, social insects, bacteria, birds, amphibians and reptiles, and aquatic biota.

As papers were identified, the list of categories was widened to include: fungi, moss/lichens, and the aquatic biota group was subdivided into fish, crustaceans, molluscs, zooplankton, aquatic invertebrates and aquatic plants. (It is fully recognised that there is overlap between these categories, but they provide a convenient and practical means for structuring the available data).

As a starting point for the literature searches, existing major reviews were obtained such as by UNSCEAR [2] and IAEA [3]. Using a cascade approach, the references cited within the original reviews were also checked and obtained if relevant. In addition to the cascade search, available electronic search engines were used to supplement the information in particular with respect to more recent publications. Combining these two approaches, literature was obtained for the period 1945 to 2001.

A search of literature abstract sources was also carried out to establish the number of references that have been published overall in the ‘radiation effects’ research field. This was compared with the total number of references to be entered into the database (Table 1).

This helped put into perspective the completeness of papers reviewed under the FASSET radiation effects database. It also partly support the justification for using (or not) a given reference organism based on available data.

TABLE 1. NUMBER OF REFERENCES THROUGH IN ABSTRACT SEARCHES [4, 5], USING “RADIATION EFFECTS” KEYWORDS PER WILDLIFE GROUP, AND THOSE BEING ENTERED IN THE FASSET RADIATION EFFECTS DATABASE

Abstract searches by wildlife groups		References to enter in the FASSET Database <sup>+</sup>	
Amphibians	357		
Reptiles	100	Amphibians & Reptiles	30
Invertebrates	37		
Soil fauna	12		
Bacteria	6 203	Soil fauna and bacteria	74
Birds	1 089	Birds	60
Insects	3 415	Insects	41
Mammals	26 144	Mammals	310
Plants	16 965	Plants	399
Fish	1 531	Fish	200
Crustaceans	217		
Molluscs	194	Aquatic invertebrates	42
		Review papers	46
Total	56 264	Total	1 202

<sup>+</sup> Numbers not yet finalised.

## 2.1. Data requirements

- It was recognised that there is a need to be open and transparent in gathering the literature, and that this process is as important as the information itself. Furthermore, in order to be manageable and reasonably focused within the available time and resources, a pragmatic approach was required in collating the information. Consequently a number of constraints were applied to the literature:
- Data were collated back to 1945 – due to potential problems in accessing earlier literature.
- Searches concentrated on the most relevant papers. For example, there is a vast quantity of published papers on the effects of ionizing radiation on mammals (e.g., radiobiological studies in relation to radiotherapy treatment). Therefore there is a need to use informed judgement on the FASSET aims and requirements in order to select the appropriate literature for inclusion in the database.
- Literature on mammals with data on acute exposure with doses greater than 10 Gy was excluded to limit the volume of data for inclusion. These doses are unlikely to be encountered under normal environmental situations.
- References that couldn’t be accessed were at least recorded. If information did not obviously fit into one of the four umbrella categories e.g. genomic instability or bystander effects (both of these might affect mutation rate by mechanisms not yet understood), the information was retained for discussion.
- Data were collated at species level, with the expectation that it will be condensed to give a generic summary appropriate to the selected reference organisms (the selection of candidate reference organisms appropriate to the needs of FASSET is listed in its Deliverable 1[6]).
- Units given in the original publication were used, but these were converted into ‘standardised’ units for the total dose (Gy) and dose rate ( $\mu\text{Gy h}^{-1}$ ) for interpretation.
- Both laboratory experiments (so as not to miss out potentially important and relevant data) and field studies were included but identified as such.
- Exposures to radiation of high and low linear energy transfer were noted to provide an indication whether suitable data for deriving RBE values for different types of radiation were available.

### 3. DATABASE DESIGN

From the first FASSET workshop in Madrid (February 2001), it was agreed that a structured database should be constructed in which the relevant literature could be assembled. Construction and use of a structured approach should help ensure consistency in data entry and make interpretation of results easier.

Whilst a two dimensional array was originally envisaged, i.e. Microsoft<sup>®</sup> Excel spreadsheet format, all WP3 participants agreed that an Microsoft<sup>®</sup> Access 97 database was the best way forward to collate, and later interrogate, all the relevant effects' literature and data.

During the design phase of the database, it became apparent that there were three important aspects involved. These are to:

- have clear objectives of what data are needed, and to restrict the information gathered to the FASSET requirements, i.e. to relate dose to effects data from the most relevant literature for a range of wildlife groups and endpoints;
- have clear guidance for the data entry in order to ensure consistency in the data; and,
- undertake a series of Quality assurance (QA) exercises to ensure that all participants involved in data entry are aware of the requirements, data entry system and that everyone extracts information in the same manner. Additional QA checks have also been put in place to ensure that the information transcribed into the database is correct.

The first database version, contracted out by the UK Environment Agency, was relatively simple but significantly made use of a series of menu driven options to guide the user to perform the operations required. Furthermore, predefined drop-down lists were extensively used to improve consistency of the data being entered. The first QA exercise was performed, using the first database version, to ensure that all participants involved in gathering and entering the literature were working in unison. For this QA, ten papers were selected, read in conjunction with an initial guidance document, and data entered into the database. Once data entry had been completed, an assessment was performed to highlight erroneous data and identify any problems. Significant difficulties were identified particularly with respect to the data extraction objectives, the format of the database and the level of operator interpretation that should be employed (or not). Both the guidance and database structure were revised to finally produce the "Radiation Effects Database" version 1e.

In the final version 1e, all user options are still selected from a series of menus, which should be self-explanatory, and drop-down menus widely relied upon. An example of the typical menu options is provided in Figure 1.

Additional guidance has now been given at key points within the database and there is a help option, which allows the user to link directly to relevant sections within the Operating Guide (which is in Microsoft<sup>®</sup> Word 97). The operating guide provides information on the use of the database, detailing how the information should be entered, and describes the search/report functions and capabilities of the database using screen shots to illustrate the different screens and discusses the options that can be taken.

There are three levels of access to the database: data entry, basic searches and reports, and restricted maintenance. The password protected security system is needed to secure the integrity of the database. Every user may access the searches and reports but data entry access is restricted to personnel that have participated into the QA exercise to ensure consistency in data entry; and maintenance is reserved to the designer for full access to the underlying tables and queries that form the structure of the database. These options are briefly discussed below.

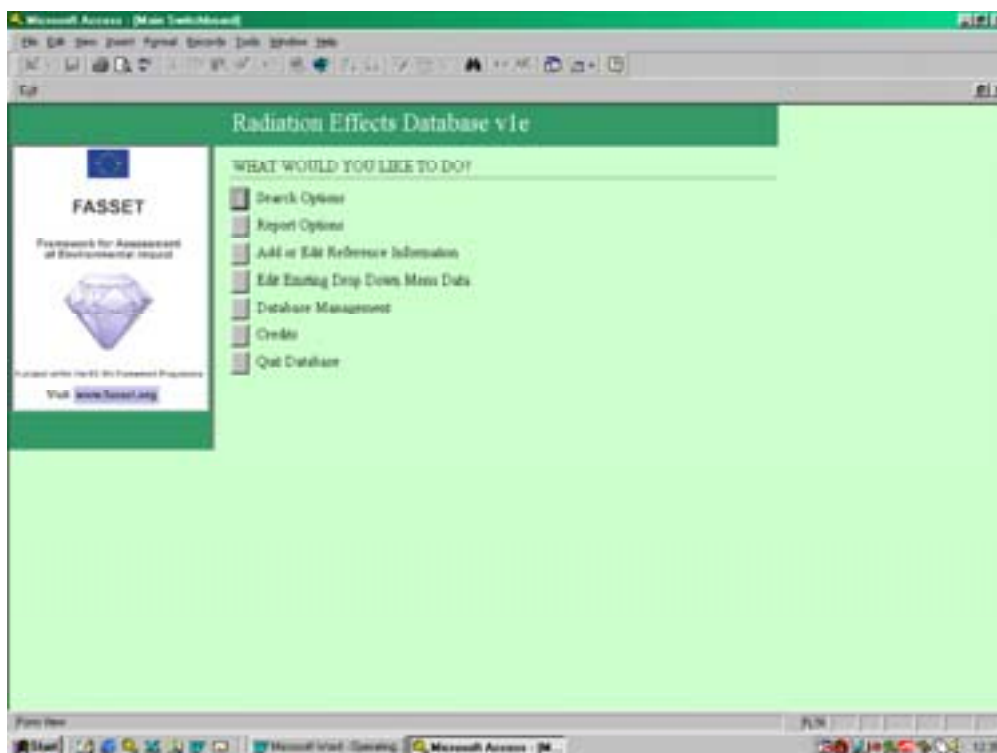


FIG. 1. Example of Menu option within the FASSET Radiation Effects Database v1e.

### 3.1. Data entry

Data entry is divided into two parts: publication details and the effects data extracted from the reference. The publication details include a description of the following:

- Author(s), (year), title, journal, volume number, page numbers;
- Article type, keywords, language; and,
- The person entering the data (note for QA checking purposes the records are time and date stamped on entry).
- As a hard copy of most original papers has been obtained, further information is entered on the institute holding the reference, and the person to contact.

The effects data part consists of recording details of the study, including:

- Laboratory or field study, radionuclide involved, radiation type and whether it is an external or internal exposure, acute, chronic or transitory exposure, ecosystem type (e.g., terrestrial, marine, freshwater or estuarine), species studied and their associated wildlife 'group' e.g., mammal, bird, reptile etc., any activity concentrations reported and a freeform notes box.

The data are also classified against one of four umbrella endpoints, which are described as:

- morbidity: including effects such as reduced immune function, damage to the central nervous system, reduced mobility, reduced growth rate;
- mortality: including change to the death rate, usually as an increase;
- reproduction: including change to gamete production, fertilization rate, embryonic and/or larval survival rates, increases in sterility; and,
- mutation: including increases in DNA damage, genetic effects in both somatic and germ cells.



The effects data are then summarized against a specific endpoint (e.g. egg production or hatching success). The data are recorded as either dose or dose rate data (in the original units), duration of exposure (which may then be used to determine either total dose or dose rate if the information is not reported) before adding a numeric value for the measurement of the specific effect. In addition, there is a series of check boxes which can be used to record information such as the Lowest Observable Effect Dose (or Dose Rate), Highest No Effect Dose (or Dose Rate) and whether the data line is for a control or background exposure level. A comment box also permits the effect of the radiation exposure to be described. Wherever possible such comments are taken from the paper and include such things as whether the effect is significantly different from the control etc. Note the interpretation of the reviewer should be minimal at this point.

It was decided to actively avoid interpretation/manipulation of the source data by the person inputting the data. Thus only raw data are entered into the database. The only 'manipulation' of the data now includes the automatic conversion of the original dose units to the standardised Gy and  $\mu\text{Gyh}^{-1}$ , and the calculation of the dose or dose rate when both corresponding dose rate or dose and duration of exposure were given.

### **3.2. Data searches and reporting options**

Once the database is compiled, then most users will only use the query reports. There are three search options:

- a sequential search on wildlife group, umbrella endpoint and dose or dose rate (with options for viewing and reporting the data after each search);
- a manual search of the database (which allows the user to search the database on any field contained within it but at the moment it is only possible to view the search results from within the database itself; and,
- search for references that may be used to generate RBE information.

The search results can be exported to Microsoft<sup>®</sup> Excel for further interpretation and assessment.

Reports can also be created for listing references, and saved as delimited text files, which can be opened in Microsoft<sup>®</sup> Word 97 or via the Windows Notepad. The reference list can also be viewed from within Microsoft<sup>®</sup> Access.

### **3.3. Database management**

This option is for exclusive use of the database designer and people charged with maintaining the database. It allows access to the underlying data tables and it is possible to carry out some routine tasks for checking data integrity, for example by checking for duplicate reference entries.

## **4. EXAMPLE**

To illustrate the function of the database, an example of the information contained on reproduction in fish has been provided in Table 2. It summarizes the data on chronic exposures for standardised dose rate ( $\mu\text{Gyh}^{-1}$ ) ranges, which will be used in FASSET. For acute exposures, FASSET will consider dose ranges (Gy) of 0 – 0.199; 0.2 – 0.499; 0.5 – 0.999; 1.0 – 1.99; 2.0 – 4.99; and >5.

The information obtained has been collated from a number of references, which have been input into the database, full reference details are available from within the database (e.g. authors, title, journal or report, year and page numbers etc.). Interpretation and assessment of this information was still required to summarize the information into the statements made in the Tables. Therefore, the summarization process should be justified by anyone undertaking such an assessment.

TABLE 2. CHRONIC EFFECTS OF IONIZING RADIATION ON REPRODUCTION IN FISH

Dose Rate Range ( $\mu\text{Gyh}^{-1}$ )	Reproduction	References
0 – 99.9	The majority of values obtained in this group reflect the control or background dose group. Normal cell types observed or normal levels of damage described. Normal mortality rates outlined.	[7, 8] [9, 10, 11] [12, 13, 14]
100 – 199.9	No data available	
200 – 499.9	Reduced spermatogonia and sperm in testes	[15, 14] [8]
500 – 999.9	Delayed spawning Increased proportion of non-germinal cells in testes Reduction in testes mass	[16, 8] [14, 9]
1000 – 1999	Mean lifetime fecundity decreases Early onset of infertility	[13, 14, 9] [8]
2000 – 4999	Reduced number of viable offspring Increased number of embryos with abnormalities Increased number of smolts in which sex was undifferentiated Increases in brood size have been reported Increased mortality of embryos	[13, 9, 8] [7]
5000 – 9999	Reduction in young fish surviving to 1 month Increased vertebral abnormalities	[16, 9] [10, 8]
>10000	Interbrood time tends to decrease with increasing dose rate Significant reduction in neo natal survival Sterility in adult fish Destruction of germ cells within 50 days in Medaka High mortality of fry, germ cells not evident Significant decrease in irradiated male salmon returning to spawn after 3 years. After 4 years, female salmon exhibited significantly reduced fecundity on return.	[16, 17] [9, 8, 13] [10]

In this case, the data have been summarized into statements that reflect the current state of knowledge based on papers from 1945 to 2001. Data extracted reflect a variety of species, although it is recognised that some of them are more radiosensitive than others. The list of species, both freshwater and marine, includes: *Acerina cernua*, ruff; *Alburnus alburnus*, bleak; *Aristichthys nobilis*, Bighead; *Brachydanio rerio*, Zebrafish; *Carassius auratus*, Goldfish; *Carassius carassius*, Crusian carp; *Chasmichthys glosus*, Goby; *Cyprinus carpio*, Carp; *Esox lucius*, Pike; *Fugu niphobles*, Puffer fish; *Fundulus heteroclitus*, Killifish; *Gambusia affinis affinis*, Mosquito fish; *Lebistes reticulatus*, Guppy; *Misgurnus anguillicaudatus*, Loach; *Oncorhynchus kistuch*, Silver salmon; *Oncorhynchus tshawytscha*, Chinook salmon; *Oryzias latipes*, Medaka; *Paralichthys olivaceus*, Flounder; *Pimephales promelas*, Fathead minnow; *Pleuronectes platessa*, Plaice; *Poecilia reticulata*, Guppy; *Rutilus rutilus*, roach; *Salmo gairdnerii*, Rainbow trout; *Salmo trutta*, Brown trout; *Tinca tinca*, Tench.

The types of experiments that have been summarized include assessments on the impact of x-rays, gamma rays (principally external Cs-137 and Co-60 sources) and beta emitting radionuclides (Tritium, Sr-90/Y-90). A range of specific endpoints have been considered: mutation frequency; % hatchability; % abnormalities; survival rates (germ cells, embryos); histopathology of gonads; % sterilization (embryos, adults); size and weight of gonads; and egg production capacity.

The effects were summarized into generic statements of effect by the authors and these are given in Table 2. Note that effects are reported in order of occurrence, therefore once it has been described at a particular dose or dose rate, it is not then reported for higher dose rates although it is expected to occur. In this way, the effects are described only once.

The information was summarized for this paper to provide an indication of the data's availability within the database, and further assessment is required before this information should, or could, be used to derive threshold dose (rate) limits or to guide the selection of reference organisms. At least one

data gap has been identified in the dose rate range of 100 – 199.9  $\mu\text{Gyh}^{-1}$ . It is recommended that consideration be given to experiments, which are designed to help complete the missing information. For the purposes of FASSET it is recommended that these experiments should in the first instance focus on the organisms from which the reference organism concept (see [ ]) has been developed. Furthermore, there is a need to ensure consistency in the data being reported within future publications in order that the relevant information is provided for use in the assessment of the impact of ionizing radiation on wildlife under the umbrella endpoints described.

## 5. CONCLUSIONS

The database contains data compiled from the literature on the effects of ionizing radiation on wildlife. Search options have been used to generate data (as in the example provided here) for deliberations within WP3 of the FASSET programme on the dose or dose rate relationships with respect to four umbrella endpoints of interest in a range of different wildlife groups. This information has been reviewed and will be described in FASSET Deliverable 4 [18].

The completed “Radiation Effects Database v1e” is an addition to the original intended FASSET deliverables, and will be available on [www.fasset.org](http://www.fasset.org), hopefully by September 2002.

In addition to providing the information needed for the requirements of FASSET, the database can also be used to identify gaps in the available information, which may guide further research. It is also hoped to secure the maintenance and update of the database beyond the duration of the current project.

In summary, the database has been created for Work Package 3 (Effects of low dose rate chronic irradiation of native wild organisms) of the FASSET (Framework for the Assessment of Environmental Impact) project. Its main use is to gather literature data to help summarize dose-effect relationships between radiation and selected organisms.

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# Alpha radiation weighting factors for biota

D.B. Chambers, M. Davis, N. Garisto

SENES Consultants Limited, Richmond Hill, Ontario, Canada

**Abstract.** It is well known that the potential for radiation effects on living organisms depends not only on the absorbed dose and dose-rate, in the tissue or organ of interest, but also on the type and energy of the radiation causing the dose among other factors. For example, alpha particles and neutrons can produce observable damage at lower absorbed doses than gamma radiation. This observation has led to the practice of multiplying the absorbed dose by a modifying factor commonly referred to as relative biological effectiveness or RBE, and other terms such as quality factor, radiation weighting factor and for non-human biota, ecodosimetry weighting factor. For human dosimetry, ICRP 60 recommends radiation weighting factors of 1 for beta and gamma rays, 10 for neutrons and 20 for alpha radiation. To distinguish from the radiation weighting factors for humans, we adopt the provisional term “ecodosimetry radiation weighting factor”  $e_R$  for application to non-human biota. The selection of an ecological relevant  $e_R$  must not be considered in isolation but rather, it must be the consequence of an integrated evaluation which includes the selection of the relevant biological endpoint, the approach to calculating relevant dose, and the selection of the most appropriate ecodosimetry radiation weighting factor. This paper reviews the selection of ecologically relevant endpoints for alpha radiation, the corresponding estimation of dose, and the selection of ecodosimetry radiation weighting factors. Overall, a nominal  $e_R$  of about 10 for population relevant deterministic endpoints, with a range from about 5 to about 20 is recommended. Sources of uncertainty in this estimate are discussed.

## 1. INTRODUCTION

The effects of exposure to ionizing radiation depend not only on the adsorbed dose (appropriately averaged over a relevant target volume), but also on the type of radiation. This latter factor has been variously referred to as “radiation weighting factor”, quality factor, and relative biological effectiveness (RBE). For protecting humans, the International Commission on Radiological Protection (ICRP) has developed the concept of a single equivalent dose, which allows the summation of doses from different types of radiation using radiation weighting factors in order to satisfy a single equivalent dose limit. The radiation weighting factors currently recommended for humans by the ICRP are 1 for beta and gamma rays, 10 for neutrons and 20 for alpha particles [1].

To date, there is no similar consensus for the equivalent radiation weighting factors for non-human biota. In view of the current and anticipated future interest in protecting non-human biota (e.g. [2–5]), there is a need to develop a scientifically appropriate yet manageable system for the dosimetry. A biota equivalent of “radiation weighting factor” or “RBE” is an important element of such a system. A recent paper by Trivedi and Gentner [6] has suggested the term “ecodosimetry [radiation] weighting factor ( $e_R$ )” for the non-human biota equivalent of the human “radiation weighting factor”. The term proposed by Trivedi and Gentner is descriptive and distinctive from the term “radiation weighting factor” which is used in human dosimetry; therefore, in the remainder of this paper we adopt the naming convention of Trivedi and Gentner.

Various authors have proposed dosimetry systems to assess the potential dose to non-human biota from both internal and external radiation (including among others [7–13]). There are many sources of uncertainty in the dosimetry, including for example the selection of a biologically relevant endpoint, the implications of the non-uniform distribution of radionuclides; the ecodosimetry weighting factor, and the different radiation sensitivities of tissues, and organs, which for non-human biota are largely unknown. The ecodosimetry weighting factor ( $e_R$ ) is the focus of this paper.

## 2. RBE AND $e_R$

The concept of RBE can be understood as the “inverse ratio of absorbed doses of different quality radiations, delivered to the same locus of interest, that produce the same degree of a given biological effect in a given organism, organ or tissue” all other factors being equal [15]. Gamma rays from  $^{137}\text{Cs}$  or  $^{60}\text{Co}$  and 250 kVp x-rays have been used as the “standard” or “reference” radiations. It is important to understand, in looking at the literature, that  $^{60}\text{Co}$  gamma rays are less effective than 250 kVp x-rays. At high doses, the difference is small (RBE = 0.86 for  $^{60}\text{Co}$  relative to x-ray as the standard); however, the difference is larger at lower dose rates (15). Overall, the difference in the relative effectiveness of  $^{60}\text{Co}$  gamma rays and 250 kVp x-rays is about a factor of 2 [15–17].

RBE depends on many factors among them, the type of cell or tissue irradiated, dose and dose-rate, the distribution of LET or lineal energy, the endpoint (effect) of interest, and other factors (e.g. [18]) Barendsen [19] indicates that in addition to cellular processes, RBE depends on other factors such as “multicellular interactions, immunological, hormonal, and possibly other system factors”. Amongst the other factors, RBE depends on Linear Energy Transfer (LET) which is the amount of energy absorbed by the target tissue per unit path length. Low LET radiations such as x-rays, gamma rays or electrons of any energy have an average LET of about 3.5 keV/ $\mu\text{m}$  (of water) or less [15, 17]. In many systems, the RBE increases with increasing LET until the LET reaches about 100 keV/ $\mu\text{m}$  and then begins to decline. This phenomenon is shown for example in the impairment of regenerative capacity of cultured human cells inactivated by monoenergetic particles (e.g. Figure 4 of [18]). The peaking of the RBE at an LET of about 100 keV/ $\mu\text{m}$  can perhaps be explained by noting that it only requires a few tens of keV of energy to break a single stand of DNA and that a single particle with a LET of 100 keV/ $\mu\text{m}$  is sufficient to produce a double strand break which is prone to imperfect repair and may result in the death of the cell. Thus at LETs greater than 100 keV/ $\mu\text{m}$ , there is sufficient energy to ensure a double strand break in target DNA and additional energy is simply wasted. ICRU 40 [16] discusses shortcomings to the use of LET as a measure of biological effect since LET is not simply related to energy deposition in a given irradiated volume and suggests that lineal energy ( $y$ ), in essence the ratio of the energy absorbed in a volume of interest divided by the (mean) chord length of that volume is a better metric for biological effect. Issues of microdosimetry are reviewed in [20].

The selection of the relevant (target) tissue is an important consideration. For example, for radiation induced bone cancer in beagles, the RBEs for bone seeking radionuclides relative to Ra-226 for  $^{231}\text{Pu}$ ,  $^{225}\text{Th}$ ,  $^{228}\text{Ra}$  and  $^{90}\text{Sr}$  respectively have been estimated as 6, 8, 2.5 and 0.07 to 0.24. This is explained by the different pattern of energy deposition of the radionuclides which leads to different irradiation of sensitive tissues. For alpha emitters, “surface seekers” are more toxic than “volume seekers” and  $\beta$  radiation is less effective than alpha radiation in inducing bone cancer [21].

A considerable range of RBE values have been reported in the literature. For example, Rao et al., [22] report studies of sperm head abnormalities from in vivo studies of mice following the injection of various radiochemicals including  $^{210}\text{Po}$  citrate. The authors raise the question of whether it is appropriate to compare the effect of a uniform dose (120 kVp x-rays delivered acutely) to the effect of very localized doses arising from internal alpha particles. There is also the question of which is the most relevant endpoint, induction of sperm head abnormalities with a reported RBE of 245 or survival with a reported RBE of 6.7.

There is also the question of the role of repair in estimating RBE. Petin and Kabakova [23] in commenting on their experiment with wild and radiosensitive mutants of yeast note a high correlation between the RBE of high LET radiation and cell repair capacity. They observed that RBEs for alpha particles, relative to gamma radiation, were high in cells capable of repair and that RBEs were lower in cells deficient in repair capacity. Jenner et al [24] reporting on the induction and rejoining of double strand breaks (dsbs) in DNA from mammalian cells discuss the increased efficiency of high LET radiation, such as alpha particles, at inducing inactivation of repair mechanisms. The RBE for induction of dsbs by alpha particles was  $<1$ . However, the rejoining of dsb damage was found to depend on radiation quality. For cell survival, the authors found alpha RBEs (at 10% survival) of about 5.3 and 11.8 for aerobic and hypoxic conditions respectively.

Consider the two dose response curves show in Figure 1 for a standard reference radiation A (assumed low LET) and the radiation of interest B (assumed high LET).

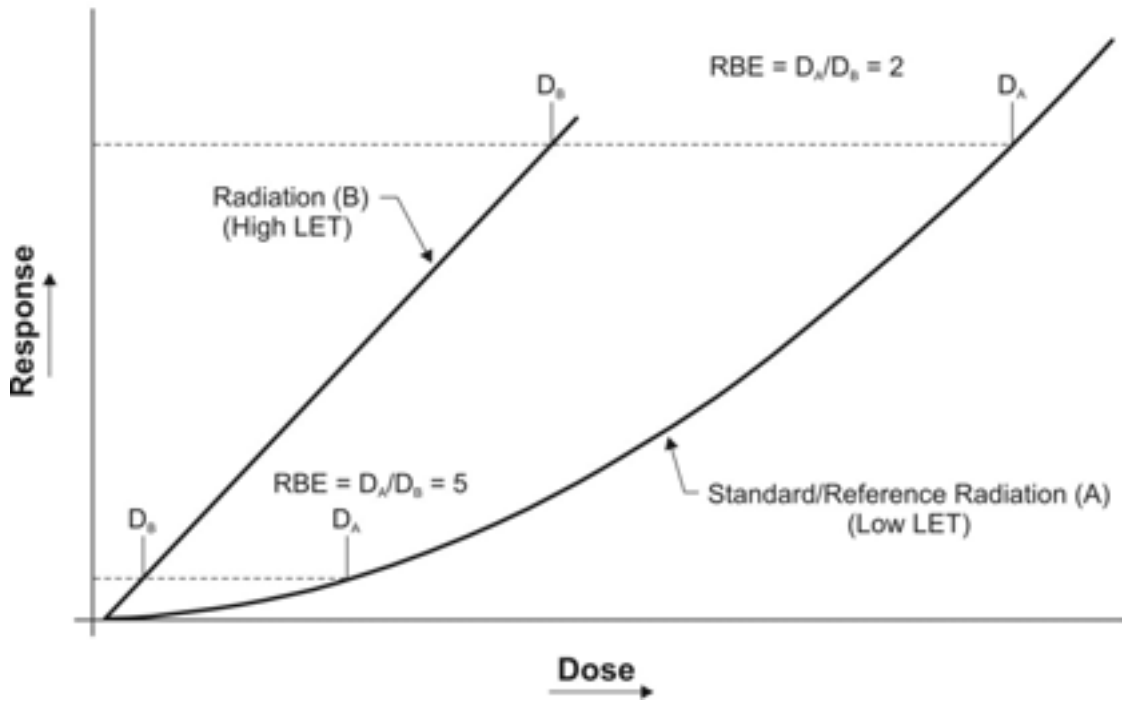


FIG. 1. Dose Response and Relative Biological Effectiveness (RBE).

In this instance, we assume the reference radiation follows a linear-quadratic dose-response relation of the form “Effect =  $\alpha_A D + \beta_A D^2$ ” when  $D$  is the absorbed dose and that radiation B follows a linear dose response relation of the form “effect (E) =  $\alpha_B D$ ” in this instance, the RBE is defined as;

$$RBE_B = \frac{D_A(E)}{D_B(E)}$$

Where  $D_A(E)$  and  $D_B(E)$  are the absorbed doses of radiations A and B which cause the same (probability) of effect E. It is clear from the figure that the calculated RBE depends on both the level of effect and the absorbed dose of the two radiations needed to produce that effect. In general terms, RBE increases with decreasing dose and reaches a maximum value which, following ICRP practice, is referred to as  $RBE_M$  for stochastic effects and  $RBE_m$  for deterministic effects [18]. At the origin, the RBE is equal to the initial slopes of the two radiation (i.e.  $\alpha_A/\alpha_B$ ).

In discussing the factors which affect the quantification of harm to cells and tissues, Feinendegen et al [25] argue that there is a need to assess risk at the cellular and tissue levels without using the “rather inaccurate” RBE. Irrespective of the validity of this position, it is very evident that the concept of RBE is complicated and selection and use of RBES, or in this instance  $e_R$ , must be carefully thought out.

### 3. ECODOSIMETRY WEIGHTING FACTOR ( $e_R$ )

According to the ICRP, deterministic effects arise from the “collective injury to substantial numbers or relatively large proportions of cells in effected tissue” (18). In addition for deterministic effects, the dose-response relation for many specific endpoints shows a threshold below which there is either no effect or the effect is so small it is undetectable. The ICRP also indicates that RBE values for high LET radiation at doses below the threshold for deterministic effects are necessary to assess the effects of exposure to mixed high and low LET radiation. The ICRP further note these  $RBE_m$  can be estimated by extrapolation from information at higher doses (18). Nevertheless, questions remain about the interpretation of  $RBE_m$  for doses below the threshold for the endpoint of interest.

A number of authors have reported data and evaluations of RBE as discussed below. For present purposes, we adopt the view that the goal of ecological protection is to protect the viability of the population of organisms of interest even though some individual organisms might be affected. Although we recognize this view is the subject of current and ongoing discussion, such a view is generally consistent with current practices and views previously expressed by various authors (e.g. [2–4, 7, 8, 27]). While there are considerable data on the effects of ionizing radiation on individual biota of various types, information relating to effects on populations is limited. Thus, in utilizing the available data it is important to understand that the presence of detectable radiation effects in individuals would not necessarily have a significant effect on the population of organisms. For non-human biota, it is therefore appropriate to focus on population relevant endpoints such as reduced fecundity or reproductive capacity.

In the authors' view, the use of mechanistic effects (such as morphology of sperm head aberrations, peripheral blood lymphocytes, mutations in marker genes etc.) could potentially provide an “early warning” of potential harm but their relevance to the whole organism is not yet well understood in most cases. Similarly, stochastic effects which are important to individuals are unlikely to represent significant risk to the population unless a quite large fraction of the population is affected.

Much of the currently available data has been summarized by various authors including the ICRP [18], the NCRP [17], Kocher and Trabalka [27], Trivedi and Gentner [6], Environment Canada/Health Canada [4], and the (former) Advisor Committee on Radiation Protection [2] and the U.K. Environment Agency [13] among others.

#### **ICRP Report No. 58 (18) and NCRP Report No. 104 (17)**

The available data on deterministic RBE in these documents can be summarized as follows:

- for 1–5 MeV neutrons,  $RBE_m$  ranges from about 4 to 12 with an average of about 7;
- for 5–50 MeV neutrons,  $RBE_m$  ranges from about 3 to 10;
- for heavy ions,  $RBE_m$  ranges from about 2 to 5;
- limited data on alpha radiation in the lung suggest RBE's in the range of 7–10 (for  $\alpha$ -particles relative to  $\beta$  radiation); and
- the ICRP did not consider data for plants.

#### **Kocher and Trabalka (27)**

These authors reviewed the available data on RBEs for alpha radiation in non-human biota. They concluded that, the relevant endpoints for non-human biota are deterministic, and that the available data on RBEs for deterministic effects suggest the  $RBE_m$  should be in the range of about 5–10.

#### **Trivedi and Gentner (6)**

These authors provide a comprehensive review of the available data on RBEs for non-human biota, discuss various factors which affect the selection of an “RBE” for non-human biota and suggest:

- the use of the term ecodosimetry weighting factor ( $e_R$ ) for non-human biota;
- an  $e_R$  for alpha radiation of 5 for deterministic endpoints related to survival and fitness;
- an  $e_R$  for alpha radiation of 10 for deterministic effects associated with reproductive endpoints; and
- an  $e_R$  for alpha radiation of 20 for stochastic endpoints.

#### **Environment Canada/Health Canada (4)**

These authors reviewed published data on RBE and noted that there is no current consensus as to the appropriate value of RBE for alpha radiation in non-human biota. Amongst other citations, these authors refer to UNSCEAR 1996 [26] which suggests an RBE of 5 for protection of non-human biota, and to Pentreath and Woodhead [14] who suggest a “provisional RBE of 40 until” a more securely based set of values had been obtained. These authors have adapted an RBE of 40 on the basis that it was considered not to “*underestimate potential effects and yet not be very conservative*”.



## U.K. Environment Agency (13)

In their review of dosimetry for non-human biota, the U.S. Environment Agency discuss the concept of RBE and application to the dosimetry of non-human biota. The authors recommend an alpha weighting factor of 20 for biota noting that “*the value of 20 is likely to be conservative in respect of deterministic effect*”. They also note that, in part, the lower effectiveness of the reference radiation at low dose rates may contribute to higher reported RBE values for alpha radiations.

## ACRP 22 (2)

The (former) Advisory Committee in Radiation Protection (ACRP), reviewed available data concerning the protection of non-human biota from ionizing radiation. These authors concluded that deterministic endpoints are most relevant for assessing population effects. With respect to the RBE issue, the authors reviewed the underlying concept of RBE, the available data from which the RBEs for biota could be estimated, and critiqued (largely on the basis of dose and dose distribution) several *in vivo* studies in animals which reported much higher RBEs than those derived from studies of cultured cells. Based on their evaluation of the data, the ACRP recommended a nominal radiation weighting factor for alpha radiation in non-human biota of about 10 with a range of 5 to 20.

## 4. CONCLUSIONS

As discussed in the foregoing paragraphs, there are many factors which affect the calculation of an  $e_R$  for a biologically relevant population endpoint for a particular target organism. In addition, the available data show a considerable range of possible values. Thus some judgment is required in assigning values of  $e_R$ . In the authors’ opinion, for deterministic population relevant endpoints, RBE values in the order of 5 to 10 seem reasonable with an upper value of about 20. Uncertainty has to be acknowledged in these estimates. In the future, it may be possible to develop probability distribution functions (pdf’s) to capture (some of) the uncertainty in  $e_R$ . Such pdf’s would need to be developed for specific biological endpoints.

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# Recommended RBE weighting factor for the ecological assessment of alpha-emitting radionuclides

P.A. Thompson<sup>a</sup>, C.R. Macdonald<sup>b</sup>, F. Harrison<sup>c</sup>

<sup>a</sup> Canadian Nuclear Safety Commission, Ottawa, Canada

<sup>b</sup> Northern Environmental Consulting and Analysis, Pinawa, Manitoba, Canada

<sup>c</sup> Lawrence Livermore Laboratories, Washington, United States of America

**Abstract.** Over 90% of the radiation dose received by biota exposed to radionuclides released from uranium mines and mills in Canada is from internally deposited alpha-emitters. Therefore, to properly characterize the potential environmental effects of ionizing radiation resulting from such exposures, it is necessary to take into account the greater relative biological effectiveness (RBE) of alpha particles. There is currently no consensus on the value of a radiation weighting factor to be used in calculating doses to non-human biota from exposures to alpha-particles. Values on the RBE of alpha particles have been derived in a broad range of experiments. Experimental results suggest that RBE values tend to be higher at the lower dose rates and when the irradiation is from radionuclides incorporated into tissue rather than external radionuclides. Therefore, to obtain an RBE value that has the greatest ecological relevance, the available data were reviewed and studies conducted in whole organisms at relatively low dose rates were given the most weight. Primary studies using endpoints such as mortality, reproduction and immune function provided the following RBE values: 40–50 (life shortening in mice); 47–377 (best estimate = 80) (oocyte mortality); 130–180 (hemopoiesis- acute gamma reference); 250–360 (hemopoiesis- chronic gamma reference). Based on the available data, we recommend a RBE value of 40 for the ecological risk assessment of biota chronically exposed to alpha-emitting radionuclides. This value is representative of the RBE data on life shortening in mice and is in the low range of values obtained for other endpoints. A value of 40 for chronic exposures is realistic and is not considered overly conservative particularly because there are little data on the biological effects of alpha emitters on a wide range of aquatic and terrestrial species.

## 1. INTRODUCTION

The assessment of ecological risk from radiation exposure is conducted by comparing the EEV (estimated exposure value) expressed as a dose of radiation (internal + external) to the ENEV (estimated no effects value), the dose at which effects are not expected. Estimated no effects values have generally been derived from experimental data obtained by exposing whole organisms to gamma sources (e.g., <sup>60</sup>Co or <sup>137</sup>Cs) because the vast majority of the radiation effects data has been obtained through experiments with external gamma sources [1].

Since assessments focus on determining whether or not the radionuclides in effluents released from nuclear facilities may have an adverse effect on the environment, biota residing in habitats contaminated with radionuclides will be exposed to radiation (in excess of natural background radiation) over their entire life cycle, and perhaps for several generations. Therefore, the doses (and dose rates) of interest in ecological risk assessments represent chronic rather than acute exposures. In addition, exposures are mainly internal from alpha, beta and gamma emitters incorporated into the tissues of organisms. In the case of biota exposed to releases from U mines and mills over 90% of the dose is from alpha-emitters deposited internally. Therefore, to properly characterize ecological risk, the greater effectiveness of alpha particles in producing biological damage must be taken into account. This will allow an appropriate comparison of an EEV resulting from an internal dose of radiation from alpha emitters with an ENEV derived from an exposure to an external source of gamma radiation.

There is currently no consensus on the value of a radiation weighting factor to be used in calculating doses to non-human biota from exposures to alpha particles. Some investigators have not modified the calculated absorbed dose (in Gy) due to alpha particles [2] because the weighting factor of 20 used in human radiation protection may not be appropriate for non-human biota. Others [3] state that for non-human biota weighting factors are required to modify the calculated absorbed dose and thus give a measure of the biologically effective dose in aquatic organisms. NCRP, Blaylock et al., and

Copplestone et al., [3–5] suggested the use of a quality factor of 20 to the absorbed dose from alpha-particles. UNSCEAR [6] has taken a different view stating that the experimental data for animals indicate that a lower weighting factor of 5 for alpha radiation would be more appropriate and that the weighting factors for beta and gamma radiation would remain unity. More recently Kocher and Trabalka [7] have expressed the view that an appropriate radiation weighting factor for alpha particles for use in protection of biota probably lies in the range of about 5–10. This is based on the assumption that deterministic effects (e.g., cell killing) are the primary concern in protection of biota. Finally, Pentreath and Woodhead [8, 9] have recommended a value of about 40 for provisional application.

Studies conducted on non-human species for a variety of endpoints have reported increased biological effectiveness of alpha particles (range from 1 to over 300). The purpose of this review is to derive a RBE weighting factor for non-human biota. The value for a RBE for ERA purposes should be representative of experimentally-determined RBE values for a specific type of radiation with respect to biological endpoints and dose-rates most relevant for the protection of non-human biota. The following sections summarize the approach taken in recommending a RBE weighting factor for alpha-emitting radionuclides for the purpose of calculating EEVs.

## 2. METHOD

Endpoints that are the most relevant to the assessment of ecological effects are those that provide a measure of change in the ability of organisms to reproduce. Some endpoints such as reduction in the number of gametes produced, gamete death, increased abnormalities, and mortality at early life stages affect fertility or sterility directly. Other endpoints such as alterations in genetic material may cause indirect effects on reproductive potential [6, 10]. For example, Blöcher [11] reports that residual double strand breaks are a major cause for loss of cellular reproductive capacity after irradiation with both x-rays and alpha particles. Harrison and Knezovich [10] reviewed the literature on whole organism irradiation experiments and found that many of the radiation effects on fertility can be attributed to damage to chromosomal material. Radiation effects on hemopoiesis are also relevant because hemopoiesis is linked to immune function and organism fitness.

The extensive database on RBEs presents a wide range of values derived for different endpoints and using different radiation exposures. Therefore, when recommending a RBE value for alpha emitters, experimentally derived RBE values should be weighted according to the ecological relevance of the endpoint, how the dose was delivered (external or tissue incorporated radionuclides) and the type of exposure (acute or chronic).

Studies on RBE can be divided into *in vitro* studies, in which external exposures to cell cultures were used primarily to determine an effect or mechanism of toxicity, and *in vivo* studies, in which internal or external exposures of whole organs or entire organisms were performed. For this assessment we reviewed experimental data summarized in NCRP [12] and more recent literature on experiments conducted *in vivo* using environmentally relevant dose rates. The *in vivo* studies retained for this review are those in which the irradiation protocol and internal distribution of the alpha-emitter varied between 100% in the cytoplasm to 100% in the nucleus of the irradiated cells. The studies in which the alpha-emitter was primarily found in the cellular cytoplasm were retained because experimental data have shown that irradiation of only the cytoplasm can induce mutations in the nucleus of the target cells by a process involving oxy-radicals [13].

Our analysis suggests that RBE values tend to be higher at lower dose rates and when the irradiation is from tissue incorporated radionuclides rather than external radionuclides. To obtain a RBE weighting factor that has the greatest ecological relevance studies conducted in whole animals at relatively low dose rates were given the most weight. After these primary studies, cell culture experiments performed at doses ranging from about 10 mGy to over 150 Gy were considered for supporting information with more weight being accorded to experiments conducted at the lower doses.

### 3. LITERATURE REVIEW

#### 3.1. Damage caused by alpha particle irradiation

The amount and type of biological damage produced by radiation is related to the type of radiation exposure [14, 15]. The efficiency with which the radiation causes damage is the Relative Biological Effectiveness (RBE) of the radiation and it is generally expressed relative to a low energy reference radiation whose characteristics in a medium (i.e., water, air or tissue) are well known. High linear energy transfer (LET) radiations such as alpha particles and neutrons have a higher effectiveness for producing effects than low LET radiation at the same absorbed dose. In some studies, the higher energy associated with high LET radiation has been shown to produce unique types of damage which are not observed with low LET radiations [11, 16–18].

The original model of radiation damage suggested that damage to cells occurs mainly as a result of direct damage to DNA and as sub-lesions combine to cause the death of the cell. Subsequent experiments indicated that damage to the cell was caused by ionization tracks and clusters of ionization [16]. Thus, increased damage from high LET radiation is caused by higher numbers of ionizations in the region of an alpha track. Also, the damage from low LET and high LET radiations is qualitatively different [16]. Cell lethality studies indicated approximately 10 ionizations for high LET and 3 ionizations for low LET radiations. For high LET mutagenic damage, the increase in RBE with LET is due to clusters of 15–20 ionizations. This model is used to explain RBE values of about 10 for cell lethality.

Barendsen [19, 20] suggested that RBE is strongly dependent on LET for some types of damage and that the damage from alpha-emitters is not as readily repaired as that from x-rays or gamma rays. The RBE value increases after a cellular repair period [11]. This is explained by the fact that double strand breaks induced by alpha particles are repaired more slowly and a higher fraction remains unrepaired. More effective damage repair at lower gamma doses contributes to higher RBE values at low doses.

There is also evidence that the damage caused by alpha radiation is fundamentally different from that of low LET radiations [16]. This is because of the presence of large clusters of physical damage from the alpha particle impact, which the cell cannot repair. Recent studies also indicate that irradiated tissue cells which have not been hit by an alpha particle contribute significantly to the response. For example, Zhou et al., [13] report that irradiation of 10%–20% of a cell population resulted in a mutagenic yield that was similar to when 100% of the cells in a population were hit. This response, termed the “bystander effect” may be mediated through gap function cell-cell communication. These effects, which are not observed under exposure to low LET radiations, indicate that the RBE of alpha-emitters can be very large.

#### 3.2. Relative biological effectiveness of alpha particles

##### 3.2.1. Summary of experimental data reviewed in NCRP [12]

NCRP [12] concluded that data are generally of insufficient quantity and quality to permit accurate estimation of the RBE at the low dose and dose rates relevant to radiation protection. One of the difficulties in accurately estimating the RBE of alpha particles is that for most endpoints examined, RBE values increase as the dose rate and dose decrease and the majority of the studies used high doses and dose rates.

From their review, NCRP [12] concluded that a nominal RBE value of 50 with a range of 30 to 70 would accommodate the majority of the data on chromosome aberrations in spermatogonia. In the case of specific locus mutations in oocytes, there is a wide range of RBE values (14 to 70) probably attributable to differences in ages of the females studied and the differing sensitivities of the maturing oocyte stages.

NCRP [12] also reviewed experimental data on life shortening in mice from all causes of death obtained from long-term studies. The RBE of internally incorporated alpha particles varied between 15 and 20 when compared to weekly fractionated gamma ray exposures and ranged up to 40 to 50 when compared with continuous gamma ray exposures.

Studies conducted using plant systems also show that low doses of radiation yield large RBE values while high doses yield lower values. For example, exposure of plant systems to high doses and high dose rates resulted in an average RBE value of  $12 \pm 3$  (average of 16 studies) while exposure to a low dose administered at any dose rate resulted in an average RBE value of  $65 \pm 5$  (average of 23 studies) [12]. NCRP [12] also concluded that plants with either greater DNA content per cell or larger nuclear volumes than mammalian cells, the RBE values tend to be larger than those observed in mammalian test systems by a factor of two or more.

### 3.2.2. Summary of *In vivo* Studies

There are few studies on the RBEs of alpha-emitting radionuclides administered to test animals in which sufficient time was permitted to allow the incorporation of the radionuclides into tissue. Nine such *in vivo* studies were reviewed. Endpoints included hemopoiesis and reproduction in male mice, mortality of oocytes in mice, lens opacification, and mortality and general injury in the rat. Relatively high RBE values are generally evident (Table 1). Stannard and Cassaret [21] report a RBE value of 24 for mortality and general injury in the rat. The RBE value for oocyte mortality in mice ranges between 47 and 377, with a best estimate of about 80 to account for the uncertainty in the dose to the tissue due to the assumption of uniform distribution. The RBE for spermhead survival is much lower (6.4 and 6.7). In comparison, the RBE value for spermhead abnormality is about 245.

The RBE value for effects on hemopoiesis was also investigated *in vivo*. Of special concern are radiation effects on hemopoietic stem cells whose propagation is important in combating diseases and the deleterious effects of environmental contaminants. The results indicate higher RBE values when the reference radiation was administered continuously (RBE between 250 and 360) than when the gamma exposure was acute (RBE between 130 and 180) (Table 1).

TABLE 1. RBEs DETERMINED FOR TISSUE-INCORPORATED ALPHA-EMITTING RADIONUCLIDES IN RODENTS

Test system	RBE	Alpha Emitter RBE	Dose	Reference
Spermhead survival	$7.4 \pm 2.4$	<sup>148</sup> Gd-citrate	$90 \pm 29$ mGy	Howell et al. [22]
Spermhead survival	$5.4 \pm 0.9$	<sup>223</sup> Ra-citrate	$124 \pm 20$ mGy	Howell et al. [22]
Spermhead survival	4.7	<sup>223</sup> Pb/Bi/Po	$143 \pm 14$ mGy	Howell et al. [22]
Spermhead survival	$6.7 \pm 1.4$	<sup>210</sup> Po-citrate		Rao et al. [23]
Abnormal Spermhead	$245 \pm 23$	<sup>210</sup> Po-citrate	1 to 150 mGy	Rao et al. [24]
Hemopoiesis	130–180 *	<sup>239</sup> Pu	10 to 14 mGy	Jiang et al. [25]
Hemopoiesis	250–360**	<sup>239</sup> Pu	10 to 14 mGy	Jiang et al. [25]
Hemopoiesis	150*	<sup>239</sup> Pu	10 to 14 mGy	Lord and Mason [26]
Oocyte mortality	47–377	<sup>210</sup> PoCl <sub>3</sub>	0.11 to 1.8 mGy	Samuels [27]
Lens opacification	4–8		250 mGy	Brenner et al. [28]
Lens opacification	10–40		50 mGy	Brenner et al. [28]
Lens opacification	50–100		10 mGy	Brenner et al. [28]
Lethality, general injury	24	<sup>210</sup> Po		Stannard and Cassarett [21]
Spermhead, genetic damage	22–24	<sup>239</sup> Pu	0.9 mGy/day	Searle et al. [29]

\* Acute <sup>60</sup>Co gamma reference radiation.

\*\* Chronic <sup>60</sup>Co gamma reference radiation.

In a study of *in utero* hemopoietic sensitivity to an alpha-emitter (Pu-239) in mice administered at 100 kBq·kg<sup>-1</sup> bw, Kozlowski et al., [30] estimated an RBE value of 50–100 for the induction of chromosomal aberrations in bone marrow cells. The doses to the critical tissue in offspring were estimated to be about 10 mGy (administration on day 6) and 5 mGy (administration on day 13), whereas dose to maternal bone varied between 420 and 840 mGy. However, despite the apparent *in utero* sensitivity of hemopoietic tissue to alpha irradiation, this was not always reflected in incidences of acute myeloid leukemia. Kozlowski et al., [30] report two studies conducted at doses of 10 to 85 kBq·kg<sup>-1</sup> in which no cases of acute myeloid leukemia were observed in offspring and two other studies conducted at doses of 5 to 25 kBq·kg<sup>-1</sup> in which significant incidences of acute myeloid leukemia were reported in adults. These studies on the incidence of acute myeloid leukemia in mice at different ages exposed to alpha-emitters suggest that the RBE for acute myeloid leukemia may not always be as high as 50–100 found when using the *in utero* chromosomal aberration endpoint. The ambiguity in the data on incidences of acute myeloid leukemia suggests, however, that there is uncertainty in this regard.

Lens opacification was investigated at a range of doses (10 mGy to 250 mGy) and again RBE values increased with decreasing dose and ranged from 4–8 at 250 mGy to 50–100 at 10 mGy.

The data summarized above were generally obtained in experiments where animals were exposed to environmentally relevant doses ranging from about 1 mGy to 150 mGy. The reported RBE values ranged from 6.4 to ~360 and are considered most relevant to the derivation of a RBE weighting factor because they cover endpoints that are directly affecting reproductive capacity and potential fitness of the animal (immune function).

### 3.2.3. Summary of *In vitro* Studies

*In vitro* studies have generally been conducted at high dose rates using external radionuclides (i.e. not incorporated into cells). Of the studies summarized in Table 2, only one study [31] appears to have been conducted at doses below 1 Gy. In this study doses in the 10–500 mGy range were used and an RBE of 7 is reported for embryo cell mortality, whereas a value of 60 is reported for morphological transformations of these cells. Results from cell culture tests at doses above 1 Gy using death of cells as the endpoint indicate RBE values for alpha particles are in the range of 2 to 10 (Table 2).

The RBE values for genetic endpoints generally have a broader range, from less than 1 for double strand breaks in hamster ovary cells [32] to >100 for sister chromatid exchange in V79-4 hamster cells [33] (Table 2). These values are consistent with the view that RBEs for mutation are higher than that for cell survival observed in other studies [31].

## 4. RECOMMENDED RBE WEIGHTING FACTOR FOR ALPHA-EMITTERS

Two significant features are evident from the *in vivo* studies above. First, these studies are *in vivo* tests at relatively low doses and dose rates, and hence are much closer to environmental exposure conditions than *in vitro* tests, which use very high doses and dose rates. Second, the endpoints used are critical from the standpoint of the maintenance of a population of organisms (effects on oocytes, sperm and immune system health). Based on the data presented above, and considering the wide range of RBE values (<1 to 300) reported in studies using several endpoints and species, it is important to recommend a weighting factor which will not underestimate potential effects and yet not be overly conservative. The majority of studies report RBE values <10. However, for ecologically significant endpoints and at environmentally relevant doses and dose rates higher RBE values of >100 are reported. It is these environmentally relevant RBEs that should be used for weighting doses from alpha emitters. The RBE for alpha-emitters from low-dose whole organism and low-dose cell-culture studies is log-normally distributed with a geometric mean (GM) of 40 and a geometric standard deviation of 5.2.

TABLE 2. RBE VALUES FOR ALPHA RADIATION FOR IN VITRO TEST SYSTEMS USING SEVERAL BIOLOGICAL ENDPOINTS

Test System	Endpoint	Alpha Emitter	RBE	Reference
Human Diploid Fibroblasts	chromosome breaks	Pu-238	2.16 ± 0.13	Bedford and Goodhead [34]
Erlich ascites tumor cells	double strand breaks	Am-241	2.7 ± 0.4 3.8 ± 1.2 (10 Gy)	Blöcher [11]
Rat lung fibroblasts	binucleated cells; micronuclei	Radon	65.2 ± 8.4 *	Brooks et al. [35]
Human fibroblasts	cell mortality	Pu-238	5.2 RBE = 13.3 and 18 (mutation frequency)	Chen et al. [36]
Human peripheral lymphocytes	chromosomal aberrations		15	Schmid et al. [37]
C3H 10T1/2 cells	cell death		4.5 5.1 (at 80% cell survival)	Durante et al. [38]
V79-4 Chinese Hamster cells	double strand breaks	Pu-238	1.19 ± 0.18 1.16 ± 0.16 (23 keV·µm <sup>-1</sup> )	Jenner et al. [39]
V79-4 Chinese Hamster cells	10% cell survival	Pu-238	5.3 anaerobic = 11.8 ± 0.5	Jenner et al. [32]
V79-4 Chinese Hamster cells	double strand breaks	Pu-238	0.68 ± 0.12 anaerobic = 3.0	Jenner et al. [32]
SV40 – transformed Chinese hamster embryo cells	gene sequences	Pu-238	6 RBE = 1 (Day 6)	Lücke-Huhle et al. [40]
Syrian Hamster embryo cells	10% cell survival	Radon progeny	7 to 12	Martin et al. [31]
Syrian Hamster embryo cells	Morphological transformation	Radon progeny	60 to 90	Martin et al. [31]
C3H 10T1/2 cells	cell survival	Pu-238	4.6 to 7.9	Roberts and Goodhead [41]
Chinese Hamster ovary cells	Chromosome damage		15 to 20	Brooks [14]

\* *in vivo* exposure, *in vitro* measurement

Another consideration is that very few studies have been conducted to investigate the RBE value of alpha emitters in organisms other than mammals. Information does exist, however, on DNA repair in some aquatic organisms. The results of these experiments indicate that DNA strand breaks are repaired but that the time course of repair is slower than in mammals (i.e. days rather than hours) [10]. This has been attributed to the lower metabolic rates of poikilotherms, such as fish and invertebrates, at low temperatures [42]. In addition, NCRP [12] concluded that for many plant systems with either greater DNA content per cell or larger nuclear volumes than mammalian cells, the RBE values tend to be larger than those observed in mammalian test systems by a factor of two or more. For example, for cytogenetic effects, the average RBE for plants exposed to low doses of radiation was reported to be  $65 \pm 5$  (average of 23 studies) [12]. These results suggest that a level of conservatism (or safety factor) is needed in deriving a RBE weighting factor for all biota based on data from mammalian studies.

Given these considerations and that RBE values are higher both at environmentally relevant dose rates and for ecologically significant endpoints, the GM RBE value of 40 is recommended as the weighting factor for the assessment of biota chronically exposed to alpha-emitters. This is consistent with Pentreath and Woodhead [8, 9] who have suggested for provisional application the adoption of a weighting factor for alpha emitters of about 40. The proposed weighting factor of 40 also falls within the range of RBE values (40 to 50) reported for life shortening in mice from all causes of death when the gamma ray dose is administered through continuous exposure [12].



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## **2. FRAMEWORKS FOR ENVIRONMENTAL RADIATION PROTECTION**

# Radiological protection of the environment

L.-E. Holm

Swedish Radiation Protection Authority, Stockholm, Sweden

**Abstract.** Protection of the environment is developing rapidly at the national and international level, but there are still no internationally agreed recommendations as to how radiological protection of the environment should be carried out. The International Commission on Radiological Protection (ICRP) is currently reviewing its existing recommendations for human protection. It has set up a Task Group with the aim of developing a protection policy for, and suggesting a framework of, protection of the environment that could feed into its recommendations for the beginning of the 21<sup>st</sup> century.

Although the Task Group has not yet finally decided on the objectives for the environment, these might be to *safeguard the environment* by preventing or reducing the frequency of effects likely to cause early mortality, reduced reproductive success, or the occurrence of scorable DNA damage in individual fauna and flora to a level where they would have a negligible impact on conservation of species, maintenance of biodiversity, or the health and status of natural habitats or communities. To achieve these objectives, a set of reference dose models, reference dose per unit intake and external exposure values will be required, plus reference data sets of doses and effects for both man and the environment. This paper provides a progress report of the work of the Task Group.

## 1. INTRODUCTION

Environmental protection generally has made substantial progress in philosophy and guidance since ICRP's recommendations on radiological protection were published in 1991 [1]. The increasing public concern about environmental hazards has led to a great number of international conventions and a variety of national statutory requirements to protect the environment. Radiological protection of the environment has over the last decade attracted increasing attention. The need to protect the environment in order to safeguard the future well being of man is one of the cornerstones of the Rio Declaration [2]. There is today a generally held view that explicit protection should be provided for non-human organisms and ecosystems also from harmful effects of ionising radiation, and there are specific legal requirements to do so in many countries.

There is rapid progress of the development of approaches to protect the environment, driven to a large extent by the needs of national regulators and of international organisations as part of their initiatives for a sustainable development. ICRP has up till now not explicitly dealt with protection of the environment, except in those situations where radionuclide levels in non-human organisms were of relevance for the protection of man. The current view of the ICRP is that "*the standards of environmental control needed to protect man to the degree currently thought desirable will ensure that other species are not put at risk*" [1]. Hence, there is little ICRP guidance as to how radiological protection of the environment directly should be carried out, or why. There are also clearly situations where ICRP's view is insufficient to protect the environment, e.g., where humans are not present or have been removed, or situations where the distribution of radionuclides in the environment is such that the exposure to humans would be minimal, but other organisms in the environment could be considerably exposed.

The Sintra Declaration of the OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic [3] emphasises a need to "*prevent pollution of the maritime area from ionising radiation through progressive and substantial reductions of discharges, emissions, and losses of radioactive substances with the ultimate aim of concentrations in the environment near background levels for naturally occurring radioactive substances and close to zero for artificial radioactive substances*". It has been agreed that environmental quality criteria should be developed in order to do this, and progress in this area to be reported by the year 2003. The International Atomic Energy Agency (IAEA) has addressed environmental protection in several documents, and has recently published a report on ethical considerations in protecting the environment from the effects of ionising radiation [4]. The Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development has pointed out the need to clarify ICRP's current view on environmental protection [5].

NEA recently arranged an international forum on protection of the environment, and is planning to organise another two meeting over the next two years. The International Union of Radioecology (IUR) organised a consensus conference on the protection of the environment in 2001. The consensus statement included the following principles: *Humans are an integral part of the environment, and whilst it can be argued that it is ethically justified to regard human dignity and needs as privileged, it is also necessary to provide adequate protection of the environment. In addition to science, policy making for environmental protection must include social, philosophical, ethical (including the fair distribution of harms/benefits), political and economic considerations. The development of such policy should be conducted in an open, transparent and participatory manner. The same general principles for protection of the environment should apply to all contaminants.*” [6]. At a national level, authorities are introducing legislation to protect the environment from harmful radiation effects. There is thus a risk that this may lead to different scientific and social approaches and make harmonisation with other systems used for environmental protection difficult.

ICRP has set up a Task Group with the aim of developing a protection policy for, and suggesting a framework of, environmental protection in order to feed into the Commission’s recommendations for the beginning of the 21<sup>st</sup> century [7]. The Task Group consists of six persons from Canada, Norway, Russian Federation, Sweden, UK, and USA. In addition, there are 21 corresponding members from 12 countries, European Union, UNSCEAR (FN:s strålningskommitté) and Greenpeace (Table 1).

TABLE 1. MEMBERS AND CORRESPONDING MEMBERS OF THE ICRP TASK GROUP ON PROTECTION OF THE ENVIRONMENT

<b>Members:</b>	<b>Corresponding Members:</b>	
L.-E. Holm (chairman), Sweden	F. Brechignac, France	C.-M. Larsson, Sweden
R. Alexakhin, Russian Federation	D. Cancio, Spain	I. Likhtarev, Ukraine
J. Pentreath, UK	S. Carroll, Greenpeace	C. Mothersill, Ireland
K. Shrader-Frechette, USA	International	C. Robinson, IAEA
P. Strand, Norway	M. E. Clark, USA	S. Sadasivan, India
P.-A. Thompson, Canada	S. Domotor, USA	S. Saint-Pierre, France
	F. Fry, UK	R. Saxén, Finland
	K. Fujimoto, Japan	A. Shpyth, Canada
	N. Gentner, UNSCEAR	S. Sundell-Bergman, Sweden
	G. Hunter, EU	D. S. Woodhead, UK
	A. Janssens, EU	H. Yang, China
		A. Zapantis, Australia

## 2. WHY DO WE NEED A SYSTEM TO PROTECT THE ENVIRONMENT?

The increasing public concern over environmental hazards has led to a variety of national statutory requirements for environmental protection, and the importance of explicit radiological protection of other living organisms is also acknowledged. The human habitat has probably been afforded a fairly high level of protection through the application of ICRP’s current system of protection. However, it is impossible to convincingly demonstrate that the environment will be adequately protected in different circumstances, since there are no explicit sets of agreed assessment approach, criteria, standards or guidelines with international authority or endorsement. This is leading to different national approaches and makes harmonisation with other systems used for environmental protection difficult.

Different approaches have also been made to address directly in a wider context the many questions raised with respect to the application of the Commission’s current statement on environmental protection. These include:

- Arguments that because humans are protected, therefore all other environmental components are protected;

- Calculations to demonstrate that, in hypothetical situations and if radionuclide concentrations in the environment are such that humans would not receive more than  $1\text{mSv a}^{-1}$ , other organisms would receive dose-rates that are lower than those likely to cause harm at the population level [8];
- Derived environmental concentrations in a tiered approach, based on environmental dose rates considered safe [9];
- Target dose rates that have been developed for biota based on an ecotoxicological approach including application of safety factors [10];
- The development of a hierarchical system for protection to provide ‘derived consideration levels’ that could be used to help decision making in different situations [11,12]; and,
- Attempts to produce systematic frameworks for assessing environmental impact of radiation in specific ecosystems, including the EC projects Framework for Assessment of Environmental Impact (FASSET) and Environmental Protection from Ionising Contamination in the Arctic (EPIC) [13].

The question whether or not radiation experts want to protect individuals, populations, or ecosystems is becoming less important, because a growing number of animals, plants, areas, and habitats are already afforded legal protection from harm from all kinds of activities, including radiation. Many species are also protected at the individual level. At the national level there is also often the requirement to address environmental protection transparently through environmental impact assessments, in order to demonstrate compliance with existing legislation.

There are several reasons why ICRP needs to consider its future involvement with regard to protection of the environment. These include:

- the need to demonstrate that the principles of radiological protection are consistent with existing international principles and recognise the inter-dependence of humans and the environment;
- the need for operators and regulators to demonstrate compliance with existing environmental requirements;
- the need to provide advice with respect to intervention situations; particularly where the potential for human exposure is minimal or where action to protect people has already been undertaken;
- the need to demonstrate explicitly how knowledge of the potential extent of radiation effects on the environment can be used to inform stakeholders.

It is not for ICRP itself to derive an ethic upon which environmental protection should be based, as others have already done this on behalf of society as a whole. There is sufficient evidence to indicate the level of interface required between general knowledge of radiation effects and the requirements of environmental protection. There also exists overwhelming demand and sufficient information for this to begin now.

A system of protection is necessary to enable frequent reviews of what we know and do not know about radiation doses and effects on different organisms. It should be applicable to all situations and allow a systematic approach to the derivation and revision of the different parameters that it contains [11,12]. Some basic elements of a system for the protection of the environment are:

- a clear set of objectives and principles;
- an agreed set of quantities and units;
- a reference set of dose models for a number of reference fauna and flora;
- a reference set of values to estimate radiation exposure;
- a basic knowledge of radiation effects;
- a means of demonstrating compliance; and
- regular reviews and revisions as new knowledge develops.

### 3. A COMMON APPROACH FOR HUMANS AND THE ENVIRONMENT

It will not be possible to provide a general assessment of the radiation effects on the environment as a whole. Instead, the ICRP Task Group uses the approach proposed by Pentreath [11,12], which is based on reference sets of *dosimetric models* and *environmental geometries*, applied to one or more *reference sets of fauna and flora*. This would allow judgements to be made about the probability and severity of radiation effects, as well as an assessment of the likely consequences for either individuals, the population, or for the local environment.

The Task Group recommends that the radiation-induced biological effects in non-human organisms be summarised into three broad categories: early mortality, reduced reproductive success, and scorable DNA damage (e.g., mutations, aberrations, etc.) in order to simplify and enable the development of a management framework. These categories comprise many different and overlapping effects and recognise the limitations of our current knowledge of such effects.

Calculations of radiation dose require a consistent set of reference values to describe the anatomical and physiological characteristics of an exposed individual. These reference values for tissues and organs define a reference individual. For humans, Reference Man [14]) has been the primary reference for dose assessments for several decades, supported by secondary sets of data for a foetus, a child etc. For environmental protection, a set of primary Reference Fauna and Flora has proposed that could serve as typical ‘hypothetical’ representatives of animals and plants [11]. A possible set of reference dose models and environmental geometries for them has also been suggested [14], along with the concept that secondary and less complete data sets could also be compiled to reflect different fauna and flora of relevance for specific environmental situations. The Task Group supports this approach.

Reference organisms are not intended to describe an ‘average’ individual of a specified group. The purpose is rather to create a standard and a point of reference for the procedure of dose estimation. A reference fauna and flora contains various components, and the selection criteria for these *primary* reference organisms will include many scientific considerations [12]. The reference organisms should be ‘typical’ of different habitats and have public or political recognition. It contains reference dose models for different terrestrial and aquatic animals and plants, and reference dose per unit intake (look-up tables), a few ‘effect’ end points, ‘Derived Consideration Levels’, and primary and secondary reference fauna and flora. Such a system would make best use of existing data on doses and effects in other organisms and would also identify data gaps for further research.

The ICRP Task Group intends to propose a system for radiological protection of the environment, that is harmonised with the principles for the radiological protection of humans along the lines described above. This system will be designed so that it can be integrated with methods that are already in use in some countries. The objectives of a combined approach to the radiological protection of humans and the environment might be to:

— **to safeguard human health by:**

- preventing the occurrence of deterministic effects;
- limiting stochastic effects in individuals and minimising them in populations; and

— **to safeguard the environment by:**

- preventing or reducing the frequency of effects likely to cause early mortality, reduced reproductive success, or the scorable DNA damage in individual fauna and flora to a level where they would have a negligible impact on
- conservation of species, maintenance of biodiversity, or the health and status of natural habitats or communities.

A common approach to the achievement of these objectives could be centred on a set of reference dose models, reference dose per unit intake and external exposure values, plus reference data sets of doses and effects for both humans and the environment. The variety of dose models needed for such reference organisms will depend upon the biological effects, and a hierarchy of dose model complexity has been suggested by Pentreath & Woodhead [15]. Such models have already been used and form the basis of the current studies in the EC-project FASSET [13].

#### 4. DOSE CONSIDERATION LEVELS

ICRP is currently discussing an approach to protect humans based on *Bands of Concern and Protective Action Levels* with reference to background dose rates (6). This idea is similar to the concept of *Derived Consideration Levels* that has been proposed for fauna and flora to guide the *consideration* of different management options [11,12]. There are only two factors upon which to assess the potential consequences for fauna and flora: natural background dose rates, and dose rates known to have specific biological effects on individuals. Bands of Derived Consideration Levels for reference fauna and flora could be compiled by combining information on logarithmic bands of dose rates relative to normal natural background dose rates plus information on dose rates that are known to have an adverse biological effect.

Other factors would also have to be taken into account, particularly with regard to the scale of the area affected in terms of elevated dose rates, and the specific nature of the fauna and flora that lived within it. This would also define the boundary between radiation protection expertise and the need for advice that would be provided by others, depending upon the circumstances of the situation. An example of what the Derived Consideration Levels might be for a reference organism is shown in Table 2.

TABLE 2. DERIVED CONSIDERATION LEVELS FOR A REFERENCE ORGANISM (BASED ON [12])

Relative Dose Rate	Likely Effect on Individuals	Aspects of concern
>1000 normal	Early mortality	Possible remedial action considered
> 100 normal	Reduced reproductive success	Concern dependent on what fauna and flora, and their numbers, likely to be affected
>10 normal	Scorable DNA damage	Consider action? Depend on faunal type?
Normal background		No action considered
< Normal	Low-trivial	No action considered

Whereas dose rates higher than background would be of increasing concern. One advantage of this approach is that for any given spatial and temporal distribution of radionuclides, one should be able to estimate both the relevant bands of concern with respect to both members of the public (based on Reference Man or 'secondary' data sets) and the environment (based on primary or secondary Reference Fauna and Flora). These two concepts would be independent of each other, although derived in a complementary manner, and they would each be related to the same concentration of a specific radionuclide, within a specific environmental material, at any particular site (Figure 1).

#### 5. DISCUSSION

A systematic approach is needed in order to provide high-level advice and guidance for protection of the environment. This should ideally include a clear set of principles and objectives; an agreed terminology (particularly with regard to quantities and units); reference dose models and related data sets to estimate exposures; biological end points (radiation effects); data relevant to the needs of environmental protection; guidance on the practical application of the system; plus clear ownership and management of review and revision processes in the light of new data and interpretations.

Given the speed with which radiological protection of the environment is developing, and the lack of any systematic and structured approaches that have wide support, there are strong expectations from many quarters on the ICRP to act. No doubt, ICRP is the organisation that is best placed for providing guidance that could be globally accepted and that could combine radiological protection of humans and of the environment into a coherent framework. This would require that the ICRP system for assessments be expanded to incorporate the environment, that a system for evaluation of environmental risks is developed, and that a new system for managing environmental risks be formed. In the latter case, the ICRP must recognize its appreciation of developments in other fields.



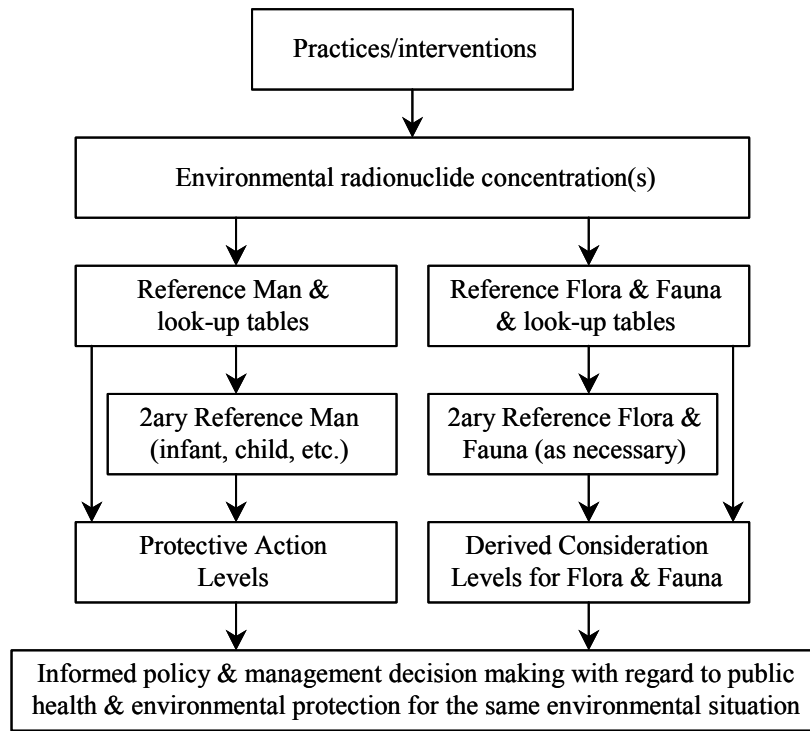


FIG. 1. Developing a common approach for the radiological protection of man and the environment (based on [12]).

If the ICRP were to widen its scope of activities and address directly radiological protection of all living organisms, international and national authorities would welcome such a commitment. ICRP has a long experience on which such a system of protection could be built. This would add credibility both to the environmental protection among those working with radiation and to ICRP among those working with other sectors of environmental protection. For the regulators and the industry, an ICRP initiative would increase the possibilities of demonstrating compliance with environmental requirements relating to release of radionuclides into the environment. It would also provide advice in intervention situations on how to deal with questions relating to certain segments of the environment. Finally, it could also be used to inform stakeholders and help bring the radiological protection more in line with the regulation of other potentially damaging industrial practices or of other contaminants associated with practices of interest to ICRP.

For ICRP, another consequence would be the need to demonstrate its commitment for environmental protection. This commitment must be reflected in the organisation of the Commission's work and in the composition of experts. It would require expertise in radioecology, radiobiology, ecotoxicology, dosimetry to deal with issues relating to the environment, such as estimating exposures, identifying data on biological effects and developing dose models for reference organisms.

ICRP's system of protection has evolved over time as new evidence has become available and as our understanding of underlying mechanisms has increased. Consequently the Commission's risk estimates have been revised regularly, and substantial revisions are made at intervals of about 10–15 years. It is therefore likely that any system designed for the radiological protection of the environment would also need time to develop, and similarly be subject to revision as new information is obtained and experience gained in putting it into practice.

## ACKNOWLEDGEMENTS

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# Development of an international framework for the protection of the environment from the effects of ionizing radiation

C.A. Robinson

Division of Radiation and Waste Safety, International Atomic Energy Agency, Vienna

**Abstract.** The International Atomic Energy Agency (IAEA) has established a programme of work to develop safety standards related to the protection of the environment from the effects of ionizing radiation, in cooperation with other national and international organizations undertaking related work. The focus for the IAEA work has been on the identification of the ethical basis and the corresponding protection principles for environmental protection in general and with application to radiation in particular. This paper will provide a summary of progress.

Three representative ethical viewpoints have been considered that reflect the spectrum of views on the environment and man's interaction with it — anthropocentric, ecocentric and biocentric ethics. Five broad principles have been identified that are incorporated in international legal instruments, and thus represent a consensus reached by signatories from many different cultural and ethical backgrounds. These are: sustainability, maintenance of biodiversity, conservation, environmental justice, human dignity. The identified protection principles are discussed in the light of the representative ethical viewpoints.

In order to develop a practical framework for assessing the impact of ionizing radiation on the environment, it is necessary to link measurable components of the five principles with scientific information relating to radiation-induced changes. Four types of effect have been considered (morbidity, mortality, reduction in reproductive success and scorable cytogenetic effects), all of which are relevant to each of the above protection principles. In order to assess the impact of radiation on the environment, and to make decisions about its protection, it is necessary to relate these effects to measurable environmental quantities and this is the subject of on-going research by other organizations.

The implications of both the identified principles and current scientific knowledge on the effect of radiation on living tissue for the development of radiation protection philosophy is discussed. Factors affecting the choice of measurable endpoints and approaches to develop a framework to aid decision-making environmental radiation protection are explored.

## 1. INTRODUCTION

The International Atomic Energy Agency (IAEA) has had a programme of work on the assessment of the effects of ionizing radiation on species other than man for a number of years. In 1979, IAEA published, in its Technical Reports Series No. 190, *A methodology for assessing impacts of radioactivity on aquatic ecosystems* [1]. A subsequent report, IAEA Technical Report Series No. 288 (1988) on *Assessing the Impact of Deep Sea Disposal of Low Level Radioactive Waste on Living Organisms* [2] discussed the doses to a number of 'typical' marine species living at or near the sea floor. Both of these reports were prepared to provide guidance on setting limits on the practice of dumping at sea, which has since been abandoned. However, significant elements of the procedures and data adopted remain valid. In 1992, IAEA published its Technical Reports Series No. 332, on *Effects of Ionizing Radiation on Plants and Animals at Levels Implied by Current Radiation Protection Standards* [3], based on a similar approach, which considered the impacts of radionuclide releases on terrestrial and freshwater environments.

During the last few years, there has been an increasing awareness of environmental issues, evidenced by the growing number of national and international legal instruments that relate to environmental protection. The Rio Declaration of 1992 [4] was a particularly significant step in this regard; it contains a total of 27 Principles, many of which relate explicitly to environmental protection. This change of attitude is also reflected in the IAEA *Safety Fundamentals for Radioactive Waste* [5], published in 1995, which includes the principle: "Radioactive waste shall be managed in such a way as to provide an acceptable level of protection of the environment". As a consequence of the need to provide additional guidance on the practical application of this principle, the IAEA has been working

towards the development of an international environmental protection framework, that will form the basis of additional safety standards.

In 1999, IAEA-TECDOC-1091, *Protection of the Environment from the Effects of Ionizing Radiation* [6], was published, with the aim of stimulating a discussion of the issues that will need to be resolved in developing guidance on this subject. This report provided the basis for discussion at the IAEA Specialists Meeting, held in 2000. In parallel, a series of expert meetings were held to develop draft guidance, paying particular attention to an elaboration of the ethics and principles underlying environmental protection, and their implications for the development of an approach for radiation. This material was discussed at a second Specialists Meeting, held in November 2001, and provided the basis for IAEA-TECDOC-1270, *Ethical Considerations in Protecting the Environment from the Effects of Ionizing Radiation* (2002) [7].

This paper provides a status report on the development of IAEA Safety Standards on protection of the environment from the effects of ionizing radiation. The ethical bases and the protection principles, discussed in Reference [7], are presented. The way in which these issues may be incorporated within developing future guidance is then discussed, drawing particular attention to the results of the discussions at the IAEA Specialists Meetings, held in the years 2000 and 2001.

## 2. IAEA-TECDOC-1091

IAEA-TECDOC-1091 [6] was published in 1999, and provided the first step towards the development of an internationally accepted philosophy and methodology for protecting the environment against ionizing radiation. The report reviewed various related issues and examined possible approaches to establishing criteria. It was intended to stimulate discussion on the subject in Member States. The following general conclusions were drawn:

- (1) Although the ICRP approach is used in many countries for protection of the environment, several countries recognize the need to develop guidance and criteria to explicitly demonstrate that the environment is protected;
- (2) There is, as yet, no clear consensus on what guidelines, endpoints or targets may be used as a basis for environmental protection, but a number of ideas were put forward in Reference [6];
- (3) The extent of knowledge on the effects of radiation on organisms other than man is considered to be sufficient to move forward on this subject;
- (4) Approaches and criteria for the protection of the environment from the effects of ionizing radiation should be developed to take account of approaches taken for other environmental pollutants;
- (5) In order to reduce uncertainties and achieve greater confidence that criteria will provide the desired level of protection, improved knowledge is required in certain areas.

## 3. DISCUSSIONS AT THE IAEA SPECIALISTS MEETING, 2000

The issues raised in the above report were discussed further at the IAEA Specialists Meeting, which took place at the IAEA Headquarters in Vienna, 29 August – 1 September 2000 [8]. Around 60 participants attended this meeting from 22 IAEA Member States and 2 international organizations. It provided an opportunity for information exchange, on work in progress, and for discussion of key issues, and represented a significant step towards the development of an international consensus. The following detailed agreements were reached:

- It was agreed that high priority should be given to developing, in a systematic way, an international system of protection for the environment. In addition, the participants agreed that the objectives of environmental protection are to minimize unnecessary impacts on the environment and to ensure that biodiversity was maintained and ecosystem function sustained.
- Standards and criteria should be developed which provide a transparent basis for determining whether environmental protection is adequate. It was accepted that protection of the population is generally the main focus for environmental protection, recognizing that the overall objective is to protect the ecosystem or environment. However, effects at the population or higher levels

of organization are the result of effects on the individual. Thus, it may be reasonable to consider population and individual effects as two measures of protection.

- Absorbed dose was regarded as the best measure of impact. Environmental concentrations, while valuable as secondary measurable quantities, do not allow the relative impacts of different radionuclides to be taken into account. The development of generic or reference organisms, for dosimetry purposes, was supported. It was stressed that the level of conservatism implied by the models should correspond to the level of analysis.

#### 4. IAEA-TECDOC-1270

Following the above conclusions of IAEA-TECDOC-1091 [6], and the discussions and recommendations of the Specialists Meeting, IAEA work on this subject continued by focusing on the ethics and principles underlying environmental protection. The objective of this work has been twofold; to provide a firm basis on which to develop a practical protection system and, by exploring the implications of a range of ethical views, to determine the extent to which a universal approach would be viable. The results of this work are outlined in IAEA-TECDOC-1270 [7], and the key features that have an influence on the future development of a protection system are outlined below.

##### 4.1. Scientific background

It is known that detrimental effects on biota can be observed at radiation doses and dose rates considerably above those that occur naturally. Indeed, much of the current basic knowledge about the molecular and cellular mechanisms of radiation damage has come from studies with both animals and plants. Some detrimental effects (termed deterministic effects) are manifest in individuals when the radiation dose absorbed by the organism exceeds some threshold — cell killing and resultant tissue damage, for example. Other detrimental effects (termed stochastic effects) are manifest by an increase in the frequency of their occurrence, in a population, with increasing dose. Examples of such effects are the development of cancer in some animals, or mutation in the genome. A consequence of both deterministic and stochastic effects is that the lifetime of some organisms will be shortened, reproductive ability may be reduced, and the genome may be adversely affected. Were sufficient numbers of organisms in a given species to be affected in these ways, changes in populations could be manifest, and any given ecosystem perturbed.

Reviews of environmental radiation levels and radionuclide concentrations, and of the effects of radiation on biota, exist, notably by the United Nations Scientific Committee on the Effects of Atomic Radiation [9, 10] However, uncertainty remains about the actual radiation doses experienced by natural biota throughout their lifetimes, due partly to insufficiencies in the present dosimetric methods. There is also no coherent model for predicting the likelihood of deterministic and stochastic effects that can be applied in a generic way for individual members of species, for populations, or whole ecosystems. The current situation is that assessments of the significance for health, competitive capabilities, and reproductive success of biota, resulting from anthropogenic additions to their radiation exposure, rely on compendia of empirical data on the relationships between deterministic or stochastic effects and radiation doses. There would be value in developing a more systematic and internationally agreed procedure.

##### 4.2. Ethical diversity

The IAEA is an inter-governmental organization comprising over 130 Member States, from all parts of the world. The IAEA membership thus represent a wide variety of cultural backgrounds that will affect the way in which nature is viewed, both by individuals and society, and the way in which ‘protection’ is interpreted and implemented. In an effort to understand the impact of these differences, an IAEA expert group undertook to identify a representative range of environmental ethics and to explore the relationship of these different ethics to ‘norms’ included in legal instruments related to environmental protection.

The most fundamental ethical divergence is between anthropocentrism and non-anthropocentrism. Proponents of anthropocentrism regard human beings, their life and experiences as the only or main thing of moral standing. In this case, environmental protection is considered to be important only in so far as it affects humans [11, 12]. Advocates of non-anthropocentrism reject the assertion that moral value can be derived and justified only in terms of human interests, and offer a variety of alternative bases for moral value. Two different were identified to represent the range of non-anthropocentric outlooks were discussed — biocentrism and ecocentrism [13].

Biocentrism (literally ‘life-centered’) has been broadly defined as an ethical outlook in which it is asserted that moral standing can be derived from a particular biological characteristic of individual members of a species. A necessary consequence of all biocentric outlooks is a recognition that individual life forms other than humans can have value in themselves, and should be respected for what they are. Since biocentrism is focused on individuals rather than the diversity of species, these outlooks have also been described as an ‘individualistic’ environmental ethic [13–15].

Proponents of ecocentrism reject the assumption that morally-relevant value can be derived only from some biological attribute of individual organisms. Ecocentrists regard diversity of species, ecosystems, rivers, mountains and landscapes as having value in themselves, even if they do not affect the welfare of humans or other individual members of non-human species [13, 16, 17]. All ecocentrists attach particular value to the diversity, dynamics and interactions within a healthy ecosystem, but differ in their views on the cause of and solutions to modern environmental problems. The general concern for the biotic and abiotic community as a whole [18], leads to the alternative classification of the ecocentric outlook as an ‘holistic’ environmental ethic [13, 15].

#### 4.3. Environmental protection ‘norms’

There is now a greater concern for other forms of life, for nature and the environment in general, evidenced by a number of recent international legal instruments, of which the Rio Declaration on Environment and Development of 1992 [4] is the most obvious. Thus, while there exists a diversity of ethical outlooks, international agreement has been reached on a number of ‘norms’ that relate to the environment and its protection. A number of such ‘norms’ or principles are identified, in Reference [7], which would be of relevance in developing consistent principles for the protection of the environment from ionizing radiation, as discussed below.

Early concern relating to the natural world centred around the loss of individual species or of unique natural areas, making **conservation** and preservation the major focus of concern. This is still an environmental protection principle, particularly for rare or threatened species or areas. However, a greater understanding of the highly complex inter-dependence of wildlife has led to recognition that conserving individual species is not sufficient in itself, and an additional focus for protection has evolved; biological diversity or ‘biodiversity’. There are three elements to biodiversity: the diversity of habitats (or of ecological complexes); the diversity of species; and the diversity of the genetic variability within each species. The Rio Declaration on Environment and Development in 1992 [4] includes the **maintenance of biodiversity**, and the status of this concept as a central principle of environmental protection is confirmed by the existence of a specific legal instrument; the Convention on Biological Diversity [19].

The Rio Declaration [4] also incorporates the idea of **sustainable development**, of which environmental protection is said to constitute an integral part. It commits States to ‘*cooperate to...conserve, protect and restore the health and integrity of earth’s ecosystem*’, which may be considered as an implicit inclusion of the concept of maintenance of biodiversity. Furthermore, development must be fulfilled to meet, equitably, “... *developmental and environmental needs of present and future generations.*” — this emphasises the importance of inter-generational equity.

Sustainable development and the Rio Declaration [4] also address aspects of human rights. In environmental protection decision-making, such issues are important. For example, some human interests will be in opposition to the need to protect other species, bringing human rights and ‘norms’ related to justice into play. For example, liability and compensation for environmental damage or stress, is also included in the principles of the 1992 Rio Declaration [4], developed into the concept of ‘**environmental justice**’ in IAEA-TECDOC-1270 [7]. The Rio Declaration also requires that ‘...*each*

*individual shall have appropriate access to information concerning the environment that is held by public authorities...'. More generally, human rights are a cornerstone of the Charter of the United Nations [20], and the concept of informed consent, and the implied need to account for different views when making decisions is discussed in Reference [7] under a general heading of 'human dignity'.*

#### **4.4. Application of protection 'norms' or principles**

The impact of ionizing radiation on biota may be assessed in terms of the first of the three 'norms' or principles discussed above [7]; conservation, maintenance of biodiversity and sustainable development. The 'development' aspects of sustainable development are, however, beyond the scope of radiation protection. For this purpose, those aspects that deal with biodiversity and inter-generational equity are discussed under a heading of 'sustainability' [7]. The principle of sustainability thus implies that impacts on future generations and productivity are of particular concern and that the quality of the environment should not be diminished over time. While maintaining biodiversity and conservation (or habitat protection) are important considerations in their own right, they are also essential features of the application of the principle of sustainability.

These principles can be used in combination with scientific information, relating to deleterious effects of ionizing radiation and the scale of such effects (exposure or dose rate), as part of an assessment framework to evaluate the possible impact of radiation on the environment or biota within it. Under normal circumstances, the primary impact of ionizing radiation is on living tissue. Thus, the main focus for protection of the environment from the effects of ionizing radiation is likely to be on the protection of biota. Furthermore, it is concluded in Reference [7] and elsewhere [21, 22] that the principal biological impacts that may have an impact on these principles are: radiation-induced early mortality, increased morbidity, reduced reproductive success and possible deleterious hereditary effects.

In making environmental management decisions, there is a need to balance various interests. The two other principles — those related to human dignity and to environmental justice — inform such judgements. Human dignity provides support for preference to be given to human interests, relative to those of biota, but it also acts to support the idea that those affected should be involved in the decision-making process (informed consent). The principle of environmental justice allows both the distribution of benefit and impact to be taken into account, and for potential compensation for environmental damage incurred.

Although included in international legal instruments, the ideas encompassed by the above principles will clearly be given a different priority, depending on cultural and ethical backgrounds. An anthropocentric would, presumably, place a high weight on human dignity and environmental justice, and while the interests of sustainability may be of central interest in developing a protection approach, the primary motivation — of ultimately protecting humans — will be different from those with an ecocentric viewpoint. These differences may lead to differences in the detailed application of the principles, particularly in trade-off situations.

## **5. DISCUSSIONS AT THE IAEA SPECIALISTS MEETING, 2001**

This meeting was of a similar format to the previous one and again provided an opportunity for both information exchange and detailed discussion. It was attended by around 60 experts, this time drawn from 19 Member States and 6 international organizations. Three Working Groups were established and the main conclusions of each group is outlined [23].

### **5.1. General environmental protection principles**

It was agreed that the ethics and principles, outlined above and in Reference [7], are a reasonable basis for developing a framework for protection of the environment, although it was recognized that there was a need to distinguish the concept of the protection of biota from that of the environment, which includes abiotic components. However, it was agreed that the initial focus for environmental radiation protection should be on biota.

Harmonization of the approach adopted to protect the environment from radiation with those for chemicals was seen as being desirable. It was also suggested that the concepts and tools (such as the precautionary approach and ALARA), incorporated in different protection systems, should be considered so that those best suited to a framework for protection of biota from radiation could be identified and incorporated as appropriate.

## **5.2. Application and specification of endpoints**

In order to develop a practical framework for assessing the impact of ionizing radiation on the environment, it is necessary to link measurable components of the five principles with scientific information relating to radiation-induced effects in biota. It was agreed that the four effects or protection endpoint categories discussed above (early mortality, morbidity, reduced reproductive success and deleterious hereditary effects) provided a useful basis. Further, it was recognised that an assessment of the potential impact of levels of radionuclides on biota also requires the effects data to be related to measurable environmental quantities. The most directly useful quantity is likely to be an environmental dose rate, but the need for other measurable quantities is also recognized for a number of reasons, for example for research or for early-warning purposes.

In developing standards and criteria for compliance, it is important to allow for the fact that scientific information is only one input into decision-making, socio-political issues related to 'acceptability' are also relevant.

For biota, the concept of dose is not yet fully developed; radiation weighting factors and tissue weighting factors for the relevant protection endpoints are necessary. An international consensus on this issue would be valuable.

Although the focus for consideration was on the effects of ionizing radiation, it was recognized that exposures to other stressors might modify the response to radiation, and therefore the combined effects of radiation in the presence of other stressors should ideally be taken into account.

## **5.3. Selection of reference biota**

The choice of biota and habitats is, to some extent, determined by environmental protection principles, and the ecological hierarchy that is considered to be the focus for protection. This may also be defined by specific legislative commitments. It was agreed that the reference organism approach is a reasonable basis on which to develop a generic framework to protect biota from the effects of ionizing radiation. In accepting this, it was recognized that effects on higher levels of organization (e.g. populations) occur only if individual organisms are affected, and that effects data are generally available for individuals rather than higher levels of organization. The need for further information on the extrapolation of individual effects to higher levels of organization was also identified.

The general features of the ecosystem and organism, that are of importance in defining the target for assessment, may be identified. A list of criteria for selecting reference organisms was developed, which addressed societal demands, ecological characteristics, and technical aspects [23].

It was recognized that a reference organism approach can be integrated within a tiered system, for example to demonstrate regulatory compliance, although the way in which effects data are incorporated in the compliance assessment may vary between different countries. A tiered approach provides flexibility; ability to iterate through the evaluation process, and to address multiple environmental assessment scenarios and user needs.

## **6. FURTHER DEVELOPMENT OF A PROTECTION APPROACH**

Following on from these meetings and publications, work has continued towards the specification of the components of a protection framework. In doing so, it is recognized that scientific information is only one input into decision making, and the acceptability of 'risk' is essentially a socio-political decision. The overall approach should therefore provide a transparent scientific assessment that is input to decision-making and management. The way in which criteria are included in such a system is, at present, open to question. A preliminary illustration of a step-wise process is demonstrated in Figure 1.



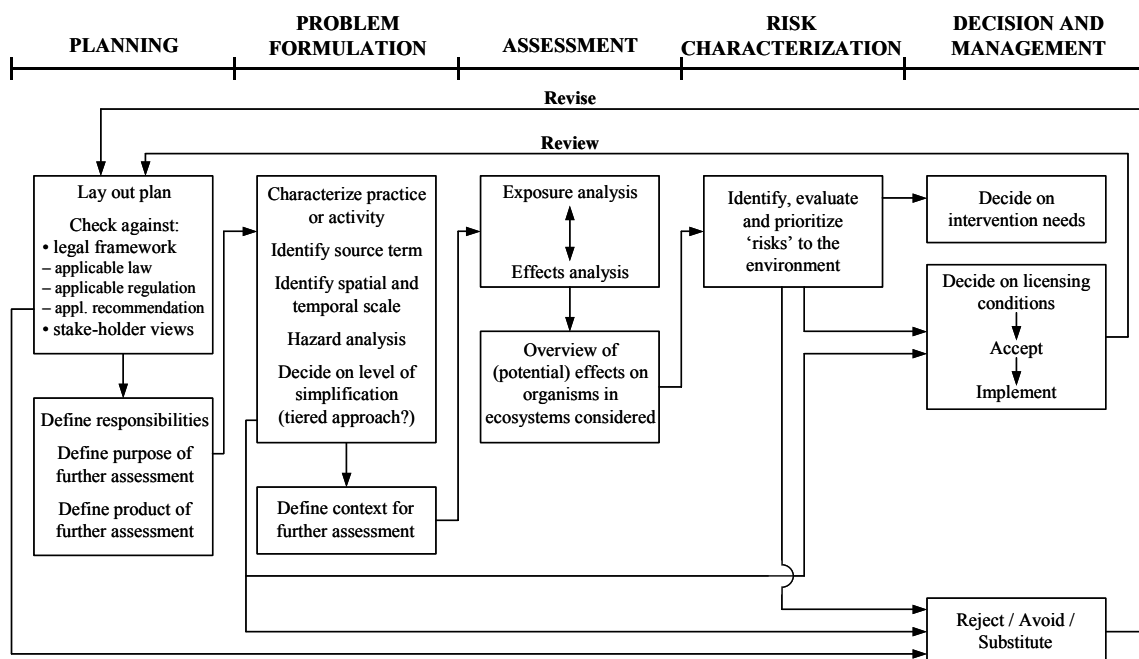


FIG. 1. Preliminary illustration of the components of an approach for environmental radiation protection.

The first step, planning, entails the identification of the legal and technical basis for further assessment. It is at this stage that the purpose of the assessment is characterised — for example, whether the assessment is to support pollution control decisions or in another context, perhaps in regard to nature conservation legislation. Different requirements may yield different assessment endpoints.

The processes involved in undertaking an impact assessment may be characterised by three basic steps; problem formulation, assessment and risk characterization. The first of these steps is largely an information gathering exercise, on the basis of which it is possible to scope the scale of assessment needed, determine whether additional research is required to address the assessment requirements, and to allow more detailed planning of the assessment. Issues considered here may include a consideration of whether an assessment based on a generic reference organism approach is sufficient for the purpose, or whether a more site and environment-specific approach is called for.

The next stage in this process is the assessment itself, which involves an analysis of exposures and possible effects. Exposure analysis includes taking account of environmental transfers and an evaluation of the doses to representative organisms, defined during the previous stages. The effects analysis stage takes account of the information that relates exposure and dose to effects. It would generally be expected that these effects would relate to the principles outlined above. The final stage of the assessment process is termed 'risk characterization', which entails the presentation of the results within the assessment context. It may involve, for example, a prediction of the dose rate to a certain organism, together with information about the uncertainties and limitations implied by the assessment. Alternatively, different protection options may be categorised in terms of the potential effects on a range of organisms. Thus, this stage could include comparison with a criterion or standard, but a judgement on the acceptability or otherwise of the assessed impact, forms part of the management stage, together with choices about the appropriate actions to take as a result.

The final stage in this process is decision and management. The issue of acceptability is related to the five principles described above. The relative importance of each of these principles, and the way in which they are incorporated in national legislation and regulations will vary as a result of the prevailing cultural influences, as discussed above. Criteria, used as a measure of acceptability, may also arise from different types of requirement. Two main categories of requirement can be identified; pollution control and nature conservation. In the first, the criteria will relate to inputs into the environment, while in the later case, specific requirements related to the environmental status or the total impact of man's activities on particular species may be called for.

## 7. CONCLUSIONS

It is necessary to develop an international approach to address the protection of the environment (or biotic components of it) from the effects of ionizing radiation. This development should take account of, but should not be restricted by, the current state of knowledge. It is recognized that, under normal circumstances, the main direct impact of ionizing radiation on environmental media is due to its interaction with living tissue. Thus, the assessment of effects on biota is likely to be the primary focus of a framework for protection of the environment from ionizing radiation.

There is a consensus emerging that the principles discussed above provide a reasonable basis for the development of a framework for environmental protection [23, 24]. Although not mutually exclusive, they incorporate quite distinctive features that are important to retain. In order to develop a practical framework for assessing the impact of ionizing radiation on biota, it is necessary to link the five principles with scientific information relating to radiation-induced changes. Four types of effect have been considered to be relevant (morbidity, early mortality, reduced reproductive success and deleterious hereditary effects [7, 21, 22]).

The challenge now is to build on this ethical base and set of protection principles, the scientific and management framework that will guide Member States in implementing programmes to assure adequate protection of biota. Progress is being made in all these areas by the IAEA, as demonstrated, and by other organizations as discussed in more detail elsewhere [7, 25–29].

This development will need to take account not only of the scientific information on the effects of ionizing radiation, but also allow for the ethical diversity existing amongst the Member States of the IAEA and elsewhere. The development of a systematic framework that separates assessment and management aspects of the protection process may be of value in ensuring that national and regional differences may be accommodated, while still allowing an international standard that will allow protection professionals to communicate on the same basis. To further facilitate this process, it is important to consider developments in environmental protection from other pollutants and assumptions and limitations inherent in the approach adopted should be clearly identified.

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## **From human to environmental radioprotection: Some crucial issues worth considering**

**F. Bréchnac, J.C. Barescut**

Institute for Radioprotection and Nuclear Safety, Department for Environmental Protection, Saint-Paul-lez-Durance, France

**Abstract.** The need to establish a system capable of ensuring adequate protection of the environment from the harmful effects of ionising radiation is at present particularly challenged. This comes both from a restrictive consideration of the environment in the so far existing system for human radioprotection, and the planetary-wide growing concerns about man's technogenic influence on his environment which have yielded "sustainability" and "precaution" as guiding principles for environmental protection. For the sake of protection, the environment is traditionally addressed through its biota since these are the sensitive components of ecosystems. Similarities between man and biotas root from the ubiquitous mechanistic effects of radiation on life which disrupt molecules. However, important differences also arise in a number of perspectives, from the vast biodiversity of species with a large spectrum of radio-sensitivities to their hierarchical self-organisation as interacting populations within ecosystems. Altogether, these aspects are prone to promote complex arrays of different responses to stress which lie beyond the scope of human radioprotection which only considers individuals of one single species. The focus on individuals in a bottom-up approach, due to its easier amenability to quantification, has prompted the development of current ecotoxicological methods as a scientific foundation to regulating environmental protection. Exclusive basement of Ecological Risk Assessment on this reductionist approach, however, is currently questioned by the most recent ecological theories which call for additional consideration of more holistic, top-down, approaches. In moving from man to environment radioprotection, these current challenges are discussed by highlighting some crucial issues linked to setting up dose limits in chronic exposure, weighting them according to radiation types (RBE), identifying appropriate effect endpoints (stochastic/deterministic, individual/population- or ecosystem-relevant), and taking due account of other concomitant contaminants (synergies/antagonisms) which call for filling critical gaps in knowledge.

### 1. INTRODUCTION

The development of a system capable of ensuring adequate protection of the environment from the harmful effects of ionizing radiation is currently particularly challenged. This need evolves first from a re-examination of the ICRP position which postulates that respecting rules and standards set for human radioprotection will implicitly ensure adequate protection of the other living beings, therefore of the environment [1, 2]. This first approach has been founded on the observation that man is one of the most radio-sensitive species among the life kingdom. However, human radioprotection has considered the environment only in the perspective of a sanitary impact on individuals of the *Homo sapiens* species, an approach that does not provide explicit protection of all other biotas. For example, habitats and biota not closely associated to humans (as a source of exposure) have not been considered. Also, the respect of the dose limit in man, as the endpoint of a given food chain, does not prove that biota situated earlier in this food chain are not exposed to toxic levels. This shows that moving from the radioprotection of man only to that of the environment necessitates a widened view. This is re-enforced by the recent planetary-wide concern on the technogenic influence of man on the environment which calls for concepts such as "sustainable development" and "precaution" as principles guiding the general philosophy of environmental protection [3].

In terms of radioprotection, the environment is often reduced to fauna and flora because they constitute the sensitive component of ecosystems. Similarities which gather man and other biotas are rooting from the ubiquitous effects of ionising radiation on life in altering the integrity of organic molecules. However, important differences need to be acknowledged, from the wide spectrum of very different species inhabiting the environment to their highly hierarchical self-organisation within ecosystems where inter-population interactions are dominating [4]. As such, the environmental response to radiological stress largely exceeds the only domain of man radioprotection which is, per definition, restricted to individuals of one single species.

## 2. CURRENT APPROACH TO RADIOPROTECTION OF THE ENVIRONMENT

### 2.1. The proposal of dose limits from existing knowledge

Several extensive reviews on the effects of ionising radiation on biotas have been promoted during the past two decades, often upon the incentive of international organisations. The most recent key documents are the reviews from NCRP [5], IAEA [6] and UNSCEAR [7], the essential conclusions of which having been recently summarized [8]. In brief, the extensive literature reviewed indicated that there were no convincing evidence that animal and plant populations would be affected at dose rates less than 1 mGy.d<sup>-1</sup> (terrestrial animals) or 10 mGy.d<sup>-1</sup> (aquatic animals and terrestrial plants) of low LET radiation (photons), even if such dose rates were noted to promote visible effects at the scale of individuals.

This conclusion has prompted the development of regulations based on dose limits, as for the American DOE which has proposed to include these (Table 1) in the Code for Federal Regulation (10 CFR 834) aimed at the “radiological protection of the public and the environment”. Guidelines and method recommendations have been elaborated during the past years, and fully detailed in a Technical Standard which describes a “graded” approach [9]. Recently, the UK Environment Agency recommended its authorities to adopt these dose limit levels, while acknowledging that recent studies in the Russian contaminated territories revealed noticeable effects at lower dose rates on individuals [10]. This recommendation was illustrated by an impact assessment of the Sellafield installations over the surrounding ecosystems which concluded that the local biotas were exposed to levels situated at several orders of magnitude below these limits.

Although still based on the concept of dose limits, the approaches pursued in Canada and Russia are rather different, and also yield different limit values. The CNSC recommendations are fully compliant with the Ecological Risk Assessment methodology currently used for chemicals which is based on ecotoxicology [11]. It uses “Environmental No Effect Values” which are derived from toxicological effects on individuals (obtained in the laboratory) divided by a safety factor (typically 10 to 1000) to account for extrapolations to real conditions. This different interpretation of the literature data leads to lower dose limits (or ENEV values) ranging from 0.2 to 2.5 mGy.d<sup>-1</sup> (Table 1). Finally, Russia is considering to set dose limits with respect to the lethal dose such as not to exceed 1 % of the LD<sub>50</sub> for marine organisms, for example [12]. This leads to an even lower range of dose limits (0.07 to 1 mGy.d<sup>-1</sup>, Table 1), which are considered as “primary”, i.e. corresponding to an ideal situation where only the radiation-induced stress is involved. Indeed, most dose-effect relationships knowledge has been obtained in the laboratory in such an ideal context. In a second step, “secondary” (local) dose limits are established by applying correction coefficients to the primary sets, which take into account the combined action of additional stress (climatic, other pollutants, anthropogenic pressure).

It is important to note at this stage that this dose-based approach complies with the “bottom up” alternative of the Ecological Risk Assessment procedure set up for non-radioactive pollutants which uses the methods of ecotoxicology. This reductionist alternative, in contrast to the other “top-down” which follows an ecosystem approach [4, 13], relies on dose-effect relationships knowledge derived for individuals of a few representative species. This is next extrapolated to higher levels of organisation, communities and ecosystems whose protection is aimed at.

Not considering the question of the extrapolation to higher organisational levels, which is not relevant for human protection, it is also worthwhile noting that this dose-based approach also complies with the current philosophy of man radioprotection set up by ICRP, as it is also aimed at the protection of individuals. This initial similarity for man and biota has been acknowledged and further exploited by some authors in the proposal to consider a common approach for man and biota which would ensure an overall protection coverage of both man and the environment [14]. This is done in particular by mimicking the “reference man” concept to yield “reference fauna and flora” [15]. There is an optimisation interest in this approach in that it makes maximum use of past experience gained through dealing with man radioprotection, hence minimising the need for novelty and effort, as it simply applies to each considered biota (with some relevant adjustments) what is currently done for man.

TABLE 1. PROPOSED DOSE LIMITS FOR PROTECTING FAUNA AND FLORA AGAINST DELETERIOUS EFFECTS OF IONISING RADIATION

	Classes of biotas considered	Dose limit or environmental no-effect value (ENEV) (mGy.d <sup>-1</sup> )
UNSCEAR [7]	Terrestrial plants	10
US DOE [9]	Aquatic animals	10
UK EA [10]	Terrestrial animals	1
Canada [11]	Algae, macrophytes	2.5
	Terrestrial plants, invertebrates	2.5
	Benthic invertebrates	1.6
	Small mammals	1
	Fish	0.5
Russia [12]	Amphibians	0.2
	Plants, invertebrates	1
	Poikilotherm animals	0.3
	Hematotherm animals (life time < 5 years)	0.14
	Hematotherm animals (life time > 5 years)	0.07
ICRP	<i>Man</i>	<i>0.0027</i>

## 2.2. Robustness of the corresponding scientific foundation

There are strong limitations to the relevance of the scientific knowledge from which the above limits have been derived. The existing literature essentially addresses short-term exposure to high doses of external  $\gamma$  radiation on individuals, whereas the vast majority of current environmental concerns are linked to long-term chronic exposure effects of low doses to populations and ecosystems [8]. In order to secure a robust radioprotection system rooted within a sound scientific foundation, it is crucial to gain better knowledge and understanding on the following issues.

There is poor knowledge currently on the effects which may appear in the **long-term**, i.e. those which, farther than the scale of individuals may, through their reproduction, appear later on at the scale of populations. This is particular crucial for **chronic exposure** situations (several generations) to **low dose** rates generated internally through **bioaccumulation** processes [16]. In this context, the most critical radionuclides are  $\alpha$  and  $\beta$  emitters susceptible to bioaccumulation. If reproduction-related criteria are recognised as being the most radio-sensitive, one must bear in mind that genetic damages are promoted at even lower dose rates, and that the long-term implications of such an observation is not known. One can question if doses too weak to affect reproduction could nevertheless promote in the long-term a mutational pool in the population sufficient to alter the course of its evolution [17].

Present knowledge is even poorer on the dose-effect relationships at the **supra-individual scales** (population, community and ecosystem), and this is a general limitation for all sources of stress, not only the particular field of radiation. A growing number of studies driven *in situ* on contaminated natural environments have been reported recently, but the strength of their conclusions is suffering from the lack of controls inherent to this descriptive approach [18]. The scientific basis for a well mastered understanding of the effects of radiations on biota at the higher levels of organisation is not yet acquired. Located at one end of the complexity scale of living entities, ecosystems are complex integrators, and quantifying the stress they suffer from the introduction of toxicants within their looped processes of energy and matter transformation is still an unresolved, although central, issue [19, 20].

Finally, it is worth recalling that real situations of environment contamination usually consist of the concomitant occurrence of **multiple pollutants**. In particular, the transfer of radionuclides within, and in-between biota through food chains, has been recently demonstrated to exhibit synergies or antagonisms with the occurrence of other chemicals, a feature which can no longer be ignored [21].

TABLE 2. RBE VALUES FOR  $\alpha$  PARTICLES RECOMMENDED FOR ESTIMATING DOSES TO BIOTA

RBE values	Author
1	Amiro, [23]
5	UNSCEAR, [7]
5–10	Kocher and Trabalka, [24]
20	NCRP, [5]; Blaylock et al., [25], Copplestone et al., [10]
40	Pentreath and Woodhead, [26]; CNSC, [11]

### 3. WHAT RBES FOR BIOTAS?

In human radioprotection, accounting for the different efficiencies with which high and low-LET radiations promote biological harm has been achieved through the use of a correction coefficient (Relative Biological Effectiveness, RBE), an approach which allows for dose additivity, and hence, total dose calculation. For man, the RBE values suitable for dose calculations are well documented, they concern one single species (*Homo sapiens*) and a given effect endpoint (cancer induction). As an example, this has resulted in a consensual value of 20 to be used for  $\alpha$  particles. The situation is far less clear for fauna and flora where a large array of different RBE values have been reported (up to 300 and more), which vary not only with the species, but also with the effect endpoints considered. Also, some studies have indicated that RBE values tended to be higher at low dose rates and for internal contamination, and to be lower for deterministic endpoints. Therefore, still for the example of  $\alpha$  particles, different recommendations have arisen ranging from values of 1 up to 40 (Table 2). This is an issue of importance which generates a significant uncertainty, especially in the case of chronic exposure situations, where  $\alpha$  and  $\beta$  particles prone to bioaccumulation phenomena play a predominant role. A good illustration of this problem has been recently provided [22, 13] in showing that the contribution of  $\alpha$  particles in the total dose estimation to biota around Sellafield raised from 30 to 90 % when taking a RBE value of 20 instead of 1, with a potential increase of the total dose up to one order of magnitude.

Another problem concomitant to selecting RBE values is the identification of relevant endpoints which would be appropriate for assessing ecological effects [11]. The current approach largely relies on endpoints which provide a measure of deterministic effects susceptible to change the ability of biota to reproduce, effects which are thought to have the largest ecological impact at population level and higher. These are for example gamete death or reduction in production, increased abnormalities and early mortality. However, chromosome alterations may also affect reproduction, and even further, the general metabolism (through protein synthesis) and haemopoiesis linked to immune fitness. Due to the poor knowledge on their long-term ecological significance, especially at the population level and higher, such cytogenetic effects are currently still largely ignored.

### 4. DOSE COMPARISONS OF INTEREST

Currently, the dose limit in use for protecting the mammal species *Homo sapiens* is set at 1 mSv.y<sup>-1</sup> following ICRP recommendation. When assuming, for simplification, this dose rate to be promoted by low LET radiation, this dose limit falls within the order of magnitude of the natural background (a few mGy.y<sup>-1</sup>). For other non-human mammal species, the most often proposed dose rate limit, 1 mGy.d<sup>-1</sup>, lies approximately 2 orders of magnitude above the natural background (Figure 1).

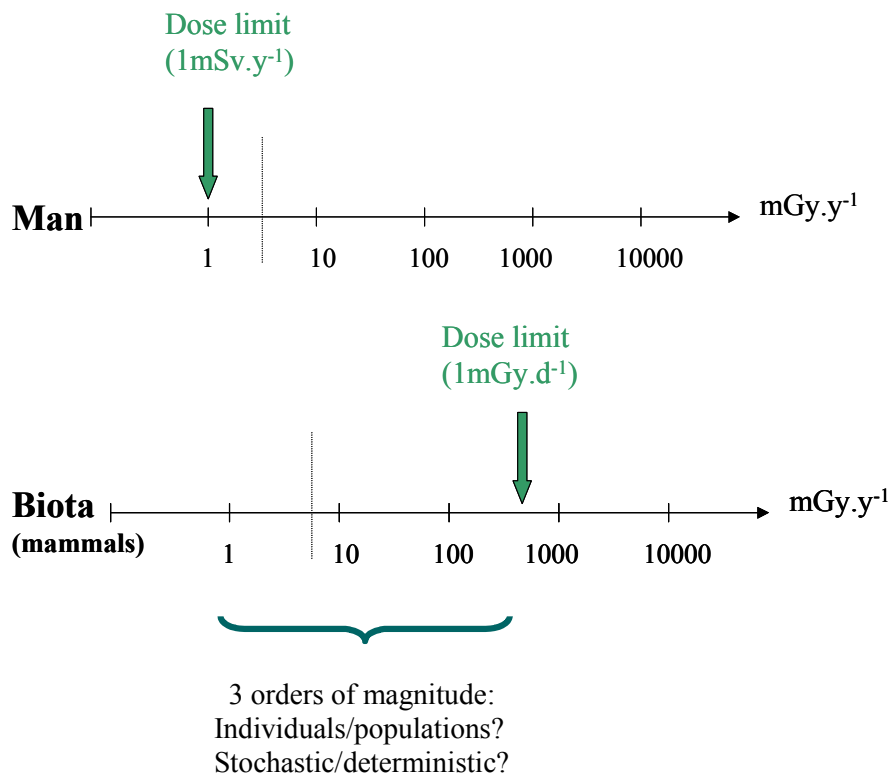


FIG. 1. Comparison between man and non-human mammals of currently proposed dose limits in chronic exposure.

It is therefore quite clear that, whilst belonging to the same family where radiation effects should not *a priori* differ greatly, the protective dose limits proposed are quite different for man and the other mammals, the most severe limit applying to the former. The question next comes: is this 3 orders of magnitude difference justified? Some preliminary elements of explanation can be put forward. First, there is a clear difference in approach in that man protection concerns the individual whereas protection of biota usually addresses the population. Several authors have claimed the occurrence of compensation mechanisms acting at the higher organisation levels due to redundancy [27, 28], but it is difficult to make sure that under a given stress ecosystems will always be less sensitive than individuals, especially bearing in mind that only a few individuals will ever be tested (therefore questioning the notion of the “most sensitive species”), and that ecosystem response may be driven indirectly (i.e. not necessarily via a direct effect of radiations). Furthermore, this individual/population justification for the difference cannot be invoked anymore in the case of endangered species for which the level of protection is also the individual. A second element of explanation comes from the fact that the effect endpoints considered in both cases are different, stochastic for man (cancer induction) and most often deterministic for biota (reproduction attributes, believed to be more relevant), and it is known that stochastic effects are usually induced at lower doses than deterministic ones. But this explanation would require a demonstration that the radio-sensitivity difference between stochastic and deterministic effects is indeed covering 3 orders of magnitude.

Recently, some authors attempted to compare the 1 mGy.d<sup>-1</sup> dose rate level to the kind of levels to which biota are currently exposed in the present existing environments [29–31]. They showed that such a level was only encountered in areas which had been previously contaminated, the non-contaminated environments promoting background dose rates to biota quite lower. From this observation, these authors questioned the acceptability of such a dose rate as a limit for protecting biota.



## 5. ON THE PERTINENCE OF THE CURRENT APPROACH

It is worth highlighting that any system of protection, irrespective of its limits and uncertainties, has the first merit to be existing. A non-perfect protection is always better than no protection at all. But conversely, a protection system is necessarily limited and eventually turning out to be false as the knowledge improves, and care must be taken that its only existence does not lead to the comfortable feeling of no risk. Such an attitude would certainly drive to crude disillusion. It is therefore of paramount importance to always keep in mind the limits of the system, in such a way as to always remain capable of reducing them. Hence, a good protection system is that which co-evolves continuously with the concomitant effort of improving the knowledge on which it is settled. Reinforcing the pertinence of the radioprotection system of the environment as it is currently evolving requires therefore to address and fill the gaps of knowledge mentioned above (effects of chronic low doses, in a multipollution context, at population/ecosystem level, [20]).

Meanwhile, it is of paramount importance to ensure that the future system of radioprotection of the environment will remain compatible with the rules of the ERA methodology which is currently in use for chemical toxicants [32]. It is the essential condition to avoid any fatal divergence that would drive it away from the general philosophy of environmental protection as it is setting up nowadays in all domains. The radiological stress is only but one of many other stresses, and must therefore be treated in a way coherent with the whole sum of stresses. The domain of low doses also implies that radiations do not constitute anymore a dominant source of stress, but only one component of a multipollutant-promoted stress that needs to be tackled seriously since several combinations have been demonstrated to present synergistic or antagonistic effects [21, 33–36]. Ideally, the domain of low mixed doses should drive to the consideration of a unique system of protection, able to cope with chemical toxicity as well as radiation toxicity.

One observes next that the dose limits currently proposed are not consensual. It is probably worthwhile noting that the most recent recommendations (Russia, Canada) are more severe, with a difference of approximately one order of magnitude. This comes from different appreciations of the doses promoting risk, a limited knowledge on the radiation effects to low dose in chronic exposure and uncertainties inherent to how such radiation effects, whilst evaluated on individuals, should be extrapolated to higher levels. Currently, an important effort is undertaken on elucidating bioaccumulation in individuals whose influence on repartition and effects is particularly relevant in situation of chronic exposure to low dose rates [16]. However, one central source of uncertainty, the recurrent problem of extrapolation of effects from individuals to higher organisational levels, still remains to be appropriately tackled, both in terms of fundamental concepts and practical aspects.

## 6. EXTRAPOLATION TO HIGHER LEVELS

Currently, both the scientific foundation and the regulatory approaches to environment protection are based upon the response of the individual. Nevertheless, there is today a large consensus in that environmental protection is not primarily aimed at the protection of individuals of biota species, but rather at the protection of the system as a whole, i.e. populations and their interactions within ecosystems. This discrepancy essentially roots from a reductionist pressure on the regulation side which favours the individual due to its easier amenability to measurement and quantification through ecotoxicological methods. A growing pool of environmental specialists are recommending that, since this hinders the success of Ecological Risk Assessment [37, 38], efforts shall be devoted to develop a complementary “ecosystem approach”.

Any ecosystem is more than a simple collection of individuals: its understanding cannot be reduced to the only description of its constituting individuals. Indeed, toxicants impact different processes at these two levels of organisation, the ecosystem level bringing additional constraints to the system (predation, competition, ...) which are prone to yield system dynamics quite different from its subsystems. This is because the ecosystem associates various populations of different species playing different, but complementary (symbiotic-like), functional roles where important features are hierarchical self-organisation and matter-energy cycling. At this level, interdependence between populations often overrides the single responses of individuals, a feature which can promote a loss of predictability from subsystems to system behaviour [39–41].

Illustration of such counter-intuitive ecological responses have been shown regarding radiation impact. Short-term, single-species, ultraviolet-B irradiation of a benthic diatoms population demonstrated reduced photosynthesis and growth, but long-term, multi-species level, tests demonstrated an increased standing crop of diatoms because the radiation also inhibited chironomids larvae, which are algal consumers [42].  $\gamma$  irradiation of a simplified model ecosystem assembling populations of an algal producer (*Euglena*), a protozoan consumer (*Tetrahymena*) and a bacterial decomposer (*E. coli*) also showed that a collapse of the ecosystem balance could be promoted via trophic restrictions indirectly mediated by radiation effects, through differences in species sensitivities [43, 44]. Predation and competition are known ecosystem-level processes which, through balancing inter-population relationships, act on ecosystem structure and resilience.

With respect to extrapolation to ecosystems, which form the level of concern most relevant (but often not yet applicable), the challenge here is to be able to move from the blind application of a “safety factor” to some methods grounded on sounder science. It is within this context that model ecosystem experimental approaches have been proposed [20], and are currently emerging [43–45]. In non-radiological environmental science, this has already proved to be a powerful method for evaluating and unravelling impacts of physical, chemical and biological perturbations [46].

## 7. CONCLUSION

Environmental radioprotection faces the challenge to be located at the crossroad of several pathways which all need to be assembled together: 1) a large toxicological knowledge and experience, but focused on chemical stress for biotas, as formalised under ERA, and more focused on man with respect to radiation impact; 2) a limited environmental radiotoxicology understanding facing the need for a regulation system to be based upon it; 3) a current knowledge essentially based upon the individual response whereas the protection aims at higher organisation levels.

It is this context which drives the current european framework for environmental radioprotection to evolve along a present approach guided by practicability: a focus on individual responses (concept of reference organisms) with a rather flexible choice of endpoints (concept of umbrella endpoints) to be selected as with the best relevance as possible to ecological effects at higher levels of organisation [47]. But meanwhile, the limitations in relevance of this approach, are identified, acknowledged and recommendations for further work put forward to promote future improvements and better relevance. In particular, recent ecological theories founded on complex systems are promoting a more realistic understanding of ecosystems which points to the danger of basing Ecological Risk Assessment exclusively on a reductionist approach [48].

Shifting emphasis from the individual to the ecosystem is prone to resolve the recurrent debate on the different possible views on environment protection, from anthropocentric to biocentric and even sometimes ecocentric. As the smallest functional and structural unit of the environment, the ecosystem embeds all, men, biotas and their abiotic biotopes, and acknowledges this complex, but not random, interactive assemblage as being the basis which supports all forms of life, including that of man.

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# Radiation protection in the 21<sup>st</sup> century: Ethical, philosophical and environmental issues

## *The Oslo Consensus Conference on Protection of the Environment*

D. Oughton<sup>a</sup>, P. Strand<sup>b</sup>

<sup>a</sup> Department of Environmental Science, Agricultural University of Norway, Aas, Norway

<sup>b</sup> Norwegian Radiation Protection Authority, Østerås, Norway

**Abstract.** A number of international organisations are focussing on a revision of radiation protection policy from the existing system which addresses only effects on man, to one which also addresses effects on the wider environment. These developments are expected to effect a wide range of stakeholders, including industry, regulators, scientists, users and the public. With this in mind a “Consensus Conference on Protection of the Environment” was arranged as part of an International Seminar on “*Radiation Protection in the 21<sup>st</sup> Century: Ethical, Philosophical and Environmental Issues*” held at the Norwegian Academy of Science and Letters. The conference attracted 45 international experts representing various disciplines and affiliations including Environmental Science, Health Physics, Radioecology, Ethics and Philosophy and a wide spectrum of perspectives bearing on the question of radiation protection of the environment. The conference was novel in that the participants were professionals rather than laypersons, and the purpose of the consensus procedure was to identify areas of agreement as an input to the ongoing regulatory developments. The success and innovation of the model is reflected in the significant areas of agreement identified in the final consensus statement, and the subsequent interest at an international level. Participants also noted the need for furthering the debate through ongoing work. Notable issues were the harmonisation of standards for radiation with other environmental stressors, guidance for balancing different interests and values within practical management, and the need for assessment criteria.

### 1. INTRODUCTION

Traditionally, radiological protection frameworks have focused almost exclusively on the protection of man, relying on the tenet that “*if man is adequately protected then other living things are also likely to be sufficiently protected*” [1]. The flaws in such a philosophy have been increasingly recognized, from both a scientific and risk management point of view. There is a growing awareness that radiation protection needs to explicitly address the question of effects on the environment [2–4], and the issue is now on the agenda for the majority of international organisations concerned with nuclear issues (e.g., ICRP, IAEA, OECD/NEA, EU) [5–11]. Some form of international legislation and regulation is anticipated within the next 3–4 years.

Interestingly, many organizations have identified a need to clarify the ethical and philosophical basis of any framework of environmental protection (IAEA, ICRP, IUR) [7, 8, 12, 13]. Developing a framework of protection raises a number of philosophical issues and dilemmas [14]. A number of the issues are already familiar (if still controversial) within environmental ethics, such as valuing the environment, animal rights, environmental risk, the precautionary principle, and differing cultural and social attitudes towards nature [15]. Practical management questions for radiation protection include the definition of harm, genetic change, the level at which damage is occurring (individual, species, ecosystem), and comparison of natural and man-made radiation. Other relevant issues are the public’s perception of radiation risks and similarities between attitudes towards biotechnology and nuclear technology. Finally, authorities need to consider the increased public awareness and concern for environmental issues in general, and from the evolving integration of environmental protection into international convention and legislation, such as the Rio declaration, OSPAR, the Aarhus convention, and the recent WSSD) [16–19].

It is clear that developments in radiation protection of the environment will affect a wide range of stakeholders, including industry, regulators, scientists, users and the public. With this in mind a recent “Consensus Conference” was arranged as part of an International Seminar on “*Radiation Protection in the 21<sup>st</sup> Century: Ethical, Philosophical and Environmental Issues*” held at the Norwegian Academy

of Science and Letters, Oslo, 22–25<sup>th</sup> October 2001 [20, 21]. The conference attracted more than 50 international experts representing various disciplines and affiliations including Environmental Science, Health Physics, Radioecology, Ethics and Philosophy, and a wide spectrum of perspectives bearing on the question of radiation protection of the environment.

## 2. CONSENSUS CONFERENCE MODELS

Since “procedure is often as important as outcome” in ethical decision-making, it is necessary to be clear about the aims, formats and organisation of the Oslo consensus conference. Different procedures or models for consensus conferences exist world wide, and there are different interpretations of what one might mean by “consensus conference” [22–23]. Essentially, as for any participatory decision-making or evaluation process, the “model” can be described according to:

- (1) the overall aims and objectives (i.e., consultancy, actual decision making, input to an ongoing process, satisfaction of free informed consent);
- (2) the participants (i.e. professionals, specialists, “experts”, lay persons, ethical committees, stakeholders) and their representative status (i.e., themselves, organisations, unions, or some stakeholder group who cannot “speak for themselves” such as future generations or animals); and
- (3) the procedure (i.e., closed or by invitation only, open, media access, accountability).

There is no one “best” or “most ethical” consensus conference model, and the design of the process will depend on the issue in question. But the success and value of the outcome of any process will hinge primarily on openness about the overall procedure from the very start, and transparency and accountability are fundamental in this respect. Failure is almost guaranteed if participants suspect that they are taking part in a process where the decision has already been made [24]. If the eventual decision goes against the views of some participants, the decision-maker should take care to justify and rationalise why those views were overridden. Although, it may seem that “open shop” should be preferable to “closed shop”, there are situations where closed shop can be justified [25, 26]. This is often the case when the aim is consensus on an individual level, which can be compromised in an open forum, since individuals would have to “tow the party line”. Lay participation in consensus conferences on highly contentious matters, where there is disagreement between experts and where policy-making is not the aim, often does little to promote public respect and trust in science. However, open procedures, at least for part of the time, are almost always to be preferred in cases of actual societal decision making.

Despite the many variables, the two most common perceptions of consensus conferences are the so-called “American” and “Danish” Models. The American model derived primarily from the “science courts” in the 1980’s where groups of experts or professionals were responsible for reaching consensus on some actual area of policy or decision. These were particularly common within medical ethics. Alternatively, the Danish model is often used to describe conferences where participants are primarily laypersons, with the objective of providing an input to decision-making. Other examples include citizen’s juries; round table conferences; stakeholder consultation, and value workshops [23, 28].

## 3. THE OSLO CONSENSUS CONFERENCE

The Oslo conference might be best described as a mixture of the two models [26]. The participants were professionals, having a broad range of disciplines, but predominantly although not exclusively with some expertise or background in radiation protection. The two participants from NGOs specialised in radiation issues. The intention was that individuals should represent themselves rather than their organisations, hence badges stated only name and country, although their affiliation was available from the participant list and the background of speakers was made clear to the audience. The level of “prior contact” was rather low. A rough estimate by the two chairs of the number of persons known to each participant gave a mean of 5.3 in a total of 45 participants.

The conference aims were to provide a forum for discussion of current issues in radiation protection, to have an input to international developments related to protection of the environment, and to encourage wider participation in the debate. The purpose of the consensus procedure was to identify areas of agreement as an input to the ongoing regulatory developments. Some form of consent was a main goal, but not a requisite. Implementing the consensus procedure at the start, rather than at the end of the development of legislation, gave stakeholders the opportunity to influence the ongoing procedure, without constraints that the consensus has to be reached at a legislative level.

The participants were there primarily by invitation, however the conference was open to any member of the NKS or IUR, space permitting. Other people who heard of the conference and expressed an interest in “dropping-in”, were permitted (e.g. a member of the American embassy). But these only attended for a single day or session, had no active input and have not been included in the participant tally.

4. LECTURES AND WORKING GROUP DISCUSSIONS

The approach to consensus was carried out by having invited plenary lectures giving an overview of a particular area, followed by discussions in small working groups (Figure 1). The subject areas were divided into: i) Risk and public perception; ii) General values; ii) Management criteria; iv) Precautionary principle. The first day was intended to act as a “trial day” in order that participants could become familiar with the procedure. But the outcome of this day was deemed important to the issue and to some extent reflected in the eventual statement. There were three groups, all with mixed representation. Each group elected a chairperson and secretary, and although people had been asked by the organisers to volunteer in advance, it was made clear to the group that the eventual choice was in their hands, and re-elections were possible after the trial. Speakers, chairs, organisers and participants all took part in discussion groups. Each group had an independent facilitator. The facilitators had no background in radiation protection, but did have expertise in facilitating at lay person consensus conferences on scientific issues. There was interaction between the facilitators, organisers and chairpersons prior to the conference and the facilitators were fully informed of the background, aims and objectives to the conference.

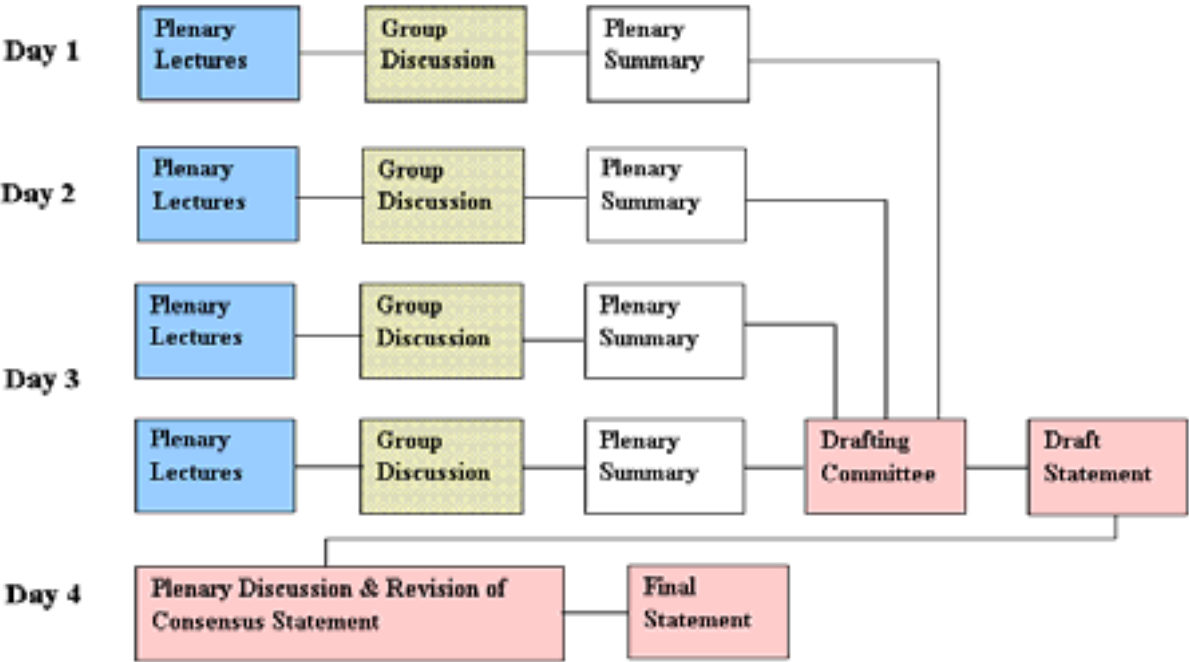


FIG. 1. Overview of the conference procedure [20].



At the start of the conference, each participant was given a set of statements, four per session, related to the topic under discussion. The 16 statements were carefully chosen to be deliberately provocative, covering science and value statements, and “extreme” and “generally accepted” viewpoints. It was certainly not the aim to draft a collection of statements that the organisers thought the participants might agree on. A selection of statements, one from each session is given below.

#### Example Discussion Statements

- The general public does not have the level of scientific competence needed for engagement in policy making.
- The standard of environmental control needed to protect man to the degree currently thought desirable will ensure that other species are not put at risk
- Radioecology needs a dose effect unit for flora and fauna, other than the Gray.
- Our present level of knowledge allows setting quantified limits and constraints that will protect man, future generations and the environment sufficiently

The groups could accept reject or amend the statements, in all cases giving reasons for their decisions. If there was no consensus it was important to have an idea of *why* there was not consensus. The facilitators helped the chairman to steer the discussion process, were aware of the reasons behind the choice of the various statements, and in some cases helped formulate the discussion statements.

At the end of each group session the participants reconvened in plenary and the secretary and/or chairman of each group presented the outcome. Participants were able to comment and question these outcomes. A brief summary of each group’s response to the discussion statements was typed up, distributed to the whole conference and these were intended to form the basis for consensus (Figure 2). The intention was that areas where different groups came to similar conclusions independently would enhance the chance of consent in the whole group. Obviously there were areas where consent was not so extensive, particularly in the precautionary principle session. But overall, the degree of agreement was surprisingly consistent.

#### 5. DRAFTING COMMITTEE AND CONSENSUS STATEMENT

At the end of the third day, a drafting committee consisting of the chairman and secretary from each group meet to draft a consensus statement, aided by one of the facilitators. Although the brief summaries formed the main input for the drafting committee, the members had access to their more detailed background notes from discussion. These proved helpful in supporting and clarifying the areas of agreement and controversy. To decrease the perception of an undue influence from the two conference chairs on the eventual statement, they were effectively “removed” from the active consensus procedure and participated as observers, but were allowed to respond to questions from the committee.

The draft statement was distributed to the remaining participants in the morning of the day 4, and the whole group met in plenum to discuss and revise the statement. This was aided by having a computer projection of the statement on screen, so that participants could follow all revisions. The statement was scrutinised, commented on and edited line-by-line, with active participation from all parties (excepting the two chairs). By lunchtime the final statement was available. Of the 26 participants remaining at the final session, all gave consent to the statement. 19 of the early departees gave consent *ex post*. 1 person has not given consent (not replied to emails). Apart from addition of the missing signatories, the statement has not been altered in any way since distribution at the final session (even the typographical error!).

The general public does not have the level of scientific competence needed for engagement in policy making

- **α-group:**
  - The general public has the right to accessible understandable scientific information  
For some members of the public this may facilitate engagement in policy making
- **β-group:**
  - Scientific competence?
  - But they bring common sense, expertise in other areas, and generally have something to say that is relevant.
- **γ-group**
  - Wrong - demonstration of scientific arrogance and not acceptable in 21 century
  - Science is one (not necessarily primary) input into the decision making process; the public have a right to take part in this process, irrespective of their scientific competence;
  - to achieve this they have the right to best available info. Presented in an appropriate manner

Radioecology needs a dose effect unit for flora and fauna, other than the Gray.

- **α-group:**
  - To assess the impact on the environment there is a need to take into account radiation type, type of organism, and biological end points in developing quantities and units with the intent to avoid unnecessary complexity.
- **β-group:**
  - The authoritative body (or bodies) should give serious consideration to the development of quantities and units for flora and fauna that are equivalent to the Sievert for humans. This should be in addition to the continued use of the Gray.
  - Holding back the development of the subject
  - Should be undertaken as soon as possible
- **γ-group:**
  - Absorbed dose is a necessary but not a sufficient quantity; we should have units to reflect the different types of impact/biological effects.
  - Concern about weighting factors, avoid the difficulties of the past, but we do want to relate exposure to effects.

FIG. 2. Examples of group summaries for discussion statements.

## 6. CONCLUSION

The success and innovation of the model is reflected in degree of agreement achieved in the final consensus statement (Figure 3), the response and ongoing interest of the original participants, and the interest at an international level. The statement has been referred to in most of the international documents and international meetings pertaining to protection of the environment [7,8,29,30,31]. Although the final consensus statement identified significant areas of agreement on protection of the environment from ionising radiation, participants also noted the need for furthering the debate through ongoing work. Notable issues were the harmonisation of standards for radiation with other environmental stressors, guidance for balancing different interests and values within practical management, and the needs for assessment criteria.

### Guiding Principles

Humans are an integral part of the environment, and whilst it can be argued that it is ethically justified to regard human dignity and needs as privileged, it is also necessary to provide adequate protection of the environment.

In addition to science, policy making for environmental protection must include social, philosophical, ethical (including the fair distribution of harms/benefits), political and economic considerations. The development of such policy should be conducted in an open, transparent and participatory manner.

The same general principles for protection of the environment should apply to all contaminants.

### Statements

1. As part of the effort to revise and simplify the current system of radiological protection for humans, there is a need to address specifically radiological protection of the environment.
2. There are several reasons to protect the environment including ethical values, sustainable development, conservation (species and habitat) and biodiversity.
3. Our present level of knowledge should allow the development of a system that can be used to logically and transparently assess protection of the environment using appropriate end points. The development of the system ought to identify knowledge gaps and uncertainties that can be used to direct research to improve the system.
4. The best available technology including consideration of economic costs and environmental benefits should be applied to control any release of radionuclides into the environment in a balanced manner with respect to other insults to the environment.
5. When a product or activity may cause serious harm to the human population or to the environment, and significant uncertainties exist about the probability of harm, precautionary measures to reduce the potential risk within reasonable cost constraints should be applied. In making such assessments and decisions, an improved mechanism for incorporating developing scientific knowledge needs to be established.
6. To assess the impact on the environment there is a need to take into account *inter alia* radiation type, type of organism, and biological endpoints (impact-related). In order to improve the transparency of assessing environmental impacts, the authoritative bodies should consequently give consideration to the development of quantities and units for biota, with the intent to avoid unnecessary complexity."

FIG. 3. Excerpt from the Oslo Consensus Statement [21].

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# **Is there a role for comparative radiobiology in the development of a policy to protect the environment from the effects of ionizing radiation?**

## ***Comparative radiobiology and radiation protection***

**C. Mothersill, C. Seymour**

Radiation and Environmental Science Centre, Dublin Institute of Technology, Dublin, Ireland

**Abstract.** The last few years has seen what people are now referring to as a “shifting paradigm” in our way of thinking about radiation effects on biological systems. The concept of the central role of DNA damage due to double strand breaks induced by a radiation “hit” has been itself hit by many studies showing persistent effects in the distant progeny of radiation exposed cells. This phenomenon is known as radiation induced genomic instability. More recently evidence has been accumulating that not even the parent cell need be exposed to radiation (the bystander effect), and that the bystander cells can demonstrate genomic instability and effects at low doses which are inconsistent with a mechanism based on DNA hits as important targets at low doses. The new paradigm suggests that cellular stress responses or damage signalling through a range of signal transduction pathways are involved. Cell-cell contact or secretion of damage signalling molecules can induce responses in undamaged and unirradiated cells. Are these new effects relevant to risk assessment, or does it matter how radiation affects cells if we have good epidemiological evidence on which to base our risk estimates? If DNA based dose responses are not so important at environmentally relevant doses, then it is not logical to base our environmental protection system on consideration of radiation dose as if this is in some way unique and not affected by the presence of other environmental stressors. The aim of this paper is to review the new concepts and to consider reasons why they might alter our methods of risk estimation. In particular the paper considers the impact of the new concepts on environmental protection and discusses the need for research in the field of comparative radiobiology if we are to develop policies which can adequately protect biodiversity.

### 1. INTRODUCTION

Until very recently, Radiation protection standards have been set using two pillars of “truth”; 1. Our knowledge of the effects of acute high doses on man allows us to extrapolate using the LNT hypothesis to determine the effects of chronic exposure in the low dose region and 2. Our knowledge of the effects of ionizing radiation on man is enough to allow us to set standards for protection of the environment. Both of these statements can be challenged. The first can be disputed because of the epidemiological data emerging from studies of Chernobyl and because of the considerable progress in the elucidation of mechanisms underlying low dose responses. These suggest that genetically predetermined biological responses to cellular stress induced by radiation but also many chemical pollutants, are involved in the final expression of cellular damage following exposure. Cellular signaling mechanisms appear to induce a tissue level or organism level response through systemic coordination. Data for humans and genetically characterized mice suggest that protective responses for an organism may appear at the cellular level as death or terminal differentiation (i.e. removal of the damaged or mutated cell). On the other hand, damaged organisms with for example cancer or mutations causing disease, may contain cells which escaped an active cell killing response to the insult. The generation of sensible policies for radiation protection, require insights into the mechanisms involved in level of selection decisions by organisms and into the role of genetics and specific genes involved in sensitivity and resistance. A study of resistant and sensitive genomes throughout the animal and plant kingdoms can clearly offer potential approaches to the discovery of important mechanisms and genes. In regard to the second “truth”, there is a very fundamental difference in our requirements for protection of man versus protection of “the environment”. Protection of man is concerned with protection of individual human lives, and mainly with prevention of cancer. Protection of “environments” is concerned with protection of populations of species, where the individual life is relatively insignificant. Key issues for radiation protection policy for the environment are whether radiation exposure makes the population, or elements within it, less fit to deal with the other stressors (chemical, physical or biological). This would make it less competitive.

Are there mechanistic thresholds where a substance in isolation is not a problem but because it shares a common mechanism with other stressors, it pushes the species beyond a mechanistic tolerance level? What are relative adaptive responses like between different species? Other concerns must be relative effects on dominant species, which could allow the emergence of new dominants. Central to all this is a basic understanding of the comparative effects of radiation on species which could lead to modelling some of the above effects. A final point is that a clear distinction needs to be made between preservation of the status quo and protection through understanding of an ecosystem which is evolving. The contribution of comparative radiobiology to these questions will be discussed with particular reference to newly appreciated low dose effects including genomic instability and bystander effects.

## 2. GENOMIC INSTABILITY

While cancer is clearly the most worrying consequence of exposure of people to low dose radiation, reproductive failure is of much more consequence for environmental radiation protection. We are always assured by the radiation protection agencies that the doses to the human population from the nuclear industry, medical exposures and the natural environment are below the level that causes cancer but in the last ten to fifteen years it has become apparent that low doses of radiation can cause subtle effects in cells surviving the dose, which may not become apparent for many, many cell generations (See major reviews cited) and which may damage the reproductive integrity of progeny from both sexual and asexual reproduction. These effects have been shown in human cells and in mice and also by our group in prawns and trout, to result in multiple mutations and excess cell death for at least 50 and up to 400 generations after the initial exposure of the parent cell to radiation [1–4]. A characteristic of this type of damage is that it appears in the progeny of cells, which had previously appeared to have survived the radiation dose unharmed. It is known from human and animal work that certain genetic subtypes are more susceptible to genomic instability than others [2–4]. The mechanism by which radiation causes these changes in the genetic material is unknown but may be related to oxidative stress induced by the initial radiation exposure, which then predisposes the surviving cells to sustain mutations more easily [5, 6]. What is clear from all the work in this field is that the mutations, whether lethal or not are non-clonal and random. That means they are not induced as a distinct mutational event in the parent cell at the time of irradiation. If they were then the same mutation would be passed to all the cells, which come from the parent. What happens is that all daughter cells in the irradiated population have an increased probability of getting a totally unpredictable and random mutation somewhere in the DNA. Some may be lethal, some non lethal and some may occur in oncogenes implicated in carcinogenesis. This latter type of mutation is the one most relevant to radiation protection of man but subtle loss of reproductive success at the cellular level may be much more relevant for species other than man, where population survival rather than individual survival is critical.

## 3. GENOMIC INSTABILITY AND BYSTANDER EFFECTS IN SPECIES OTHER THAN MAN

Among the considerable literature on environmental effects of radiation, there are very few studies of comparative radiobiology. Most investigations are ecological in nature and study biodiversity or environmental health of high background habitats. While these are very valuable for monitoring effects of radiation on ecosystems and charting the recovery of populations, they do not contribute to our knowledge of cellular mechanisms by which different species deal with radiation exposure. This means that while we may know precisely what effect radiation exposure has on a habitat or population we do not know why. There is also a long-term problem, given our emerging knowledge of delayed effects such as genomic instability, of ensuring that vulnerable or sentinel species are adequately protected by blanket protection legislation.

Actual studies of comparative radiobiology, (other than those relating to man or models used to study selected aspects of human radiobiology, for example, Nematode, fruit fly and yeast) tend to be very limited. They are mainly found in old literature, and they use extremely high doses, which are irrelevant to environmental conditions. The endpoint is usually death of the irradiated animal in the laboratory, which really precludes any mechanistic or long-term studies. The current state of the art was reviewed in two major reviews by our laboratory [M1 and 2] and shows that in the old literature using death of the animal in the laboratory as the main endpoint, species other than man are extremely

radioresistant. This work needs to be challenged now in the light of studies by our group and others [7–13] showing that the delayed effects discussed above do occur in environmentally relevant species and that the loss of reproductive success at the cellular level may result in a cumulative loss of success at the organism level and ultimately at the population level. The key facts leading us to this conclusion are the relative sensitivity of haematopoietic tissues across species [13, plus Lyng et al and Otwell et al., this meeting], and the demonstration of delayed reproductive death due to bystander effects in these tissues. Both these findings mean that dose and effect cannot be linked simply in a direct relationship and provide a possible mechanism whereby very low doses exposure at one point in time might lead to reproductive failure and immune compromise at distant time points. A further complication arises from the consideration that other environmental pollutants can cause delayed reproductive death [7, 9, 10]. Thus we can assume no simple relationship between dose and effect and need to consider multifactorial aetiologies over time. The challenge will be to ascribe a relative importance to radiation among other environmental agents or to develop an integrated environmental protection policy which is mechanism driven and which measures population *response* over time rather than cumulative *dose*.

#### 4. IMPLICATIONS FOR RADIATION RISK ASSESSMENT

Some of the implications of these considerations for risk assessment and for development of new protection policies are listed below:

- At present we know that radiation induces the phenomena of genomic instability and bystander response, described above, in man, other mammals, fish and crustaceans. This covers all classes in which an effect has been sought.
- The induction dose required is low, the effect is fully expressed at acute doses of 3–5mSv for sparsely ionising radiation and one track through a population of cells for densely ionising radiation. Action limits for workers in the radiation industry are in this range but this is for yearly exposures. It is important to realise that no one has tested lower and maximum expression of delayed effects is already seen at the relatively low doses tested.
- There does not appear to be an increasing effect with increasing dose so the effect is relatively more significant as a risk factor at low doses than at high doses.
- Delayed reproductive death (lethal mutations) is a common occurrence in progeny which survive irradiation. This cell loss is probably an important factor in determining long-term reproductive fitness at the population level.
- Immune system components are very sensitive to these delayed effects.
- Cancer is a multistep process and requires several mutations to establish a tumour, therefore any process which increases the mutation frequency of a cell population must, by definition, increase the risk of a carcinogenic mutation. Given the fact that these effects lead to an increased tolerance for mutation, the possibility of the involvement of genomic instability or bystander mechanisms in other disease aetiologies cannot be ruled out.
- We know that in medical conditions where genomic instability is a part of a recognised genetic syndrome (Bloom’s syndrome, Fanconi’s anaemia), cancer frequency is high.
- We have strong evidence from human in vitro experiments and mouse in vivo and in vitro experiments that there is a genetic basis for instability and therefore we suspect that some species/individuals will be more likely to become genetically unstable after exposure to radiation than others.
- We do **not** know how radiation induces instability or what the mechanism of the bystander effect might be. Such knowledge might enable us to prevent its induction.
- We do **not** know if there is a natural mechanism for controlling or preventing the establishment of cells carrying instability. Research is needed to investigate this and to determine mechanisms underlying the control of survival and death post irradiation.



- We do **not** know what underlies the apparent genetic or species basis for instability effects. It is particularly important to determine whether there are different subpopulations of humans and animals and whether these can be identified by simple screening tests. Again this information would provide possible avenues for protection of exposed populations such as the use of sensitivity scaling factors.
- We do **not** know whether there is a low dose threshold for genomic instability or the bystander effect. The lowest doses tested showed, in most cases, maximum expression of the effect. These doses are at the upper limit of environmental relevance. What happens at lower doses? The concept of “background levels of mutation” does not apply here since the radiation effect appears to be to raise the background or intrinsic mutation rate for the whole population of cells for as long as has been measured.
- We do **not** know how other pollutants such as mutagenic chemicals affect unstable cell populations. It is reasonable to expect higher levels of mutations following chemical exposure if the population already has a higher susceptibility to mutation induction. Research to clarify this might help to explain why studies in one area show evidence of “radiation” induced cancer while studies in another area do not. Radiation may just be facilitating the mutagenicity of another factor. Again knowledge of the mechanisms and interactions would aid development of logical and effective protection strategies.

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# The application of an ecological risk assessment approach to define environmental impact of ionizing radiation

I. Zinger-Gize<sup>a</sup>, C-M. Larsson<sup>b</sup>, C. Jones<sup>c</sup>

<sup>a</sup> Environment Agency, Warrington, United Kingdom

<sup>b</sup> Swedish Radiation Protection Authority, Stockholm, Sweden

<sup>c</sup> Kemakta Konsult AB, Stockholm, Sweden

**Abstract.** Frameworks for assessing and managing environmental risks are normally organized in three different stages: 1) the problem formulation stage where the purpose of the assessment is determined, based on considerations for hazard identification and characterization, temporal and spatial scales, as well as levels of simplification and conservatism; 2) the assessment stage, where the most suitable methodology, including the exposure and effect analyses, is selected and employed, resulting in a characterization of the risk; and 3) the risk management stage involving methodologies to eliminate, mitigate and/or prevent environmental consequences.

No international guidance is available on a methodology for assessments and management of environmental risks resulting from radioactive contaminants. Whereas the current ICRP system addresses both the problem formulation and assessment stages, it is not concerned with the environment per se, and the usefulness of the management principles (justification, optimization and dose limits) in environmental radiological protection is debatable.

The present paper explores approaches, commonalities, differences and applicability of recent developed methodologies, notably the European Commission funded FASSET (Framework for ASSESSment of Environmental impacT) project, and the BIOMASS (BIOsphere Modelling and ASSESSment) project supported by the International Atomic Energy Agency. Furthermore, reference is given to the regulatory guidance in England and Wales to protect wildlife from ionizing radiation. The paper considers, *inter alia*, the following elements:

- methods for selection of radionuclides, based on, e.g. toxicity, persistence, particle-reactivity, dispersion, and type of source;
- the need for generalizations, e.g. reference ecosystems, reference organisms and stylized dosimetric models, and the applicability of probabilistic approaches; and
- the selection of biological effects, covering the range from acute to chronic effects.

## 1. INTRODUCTION

The current system for radiological protection, as outlined by the International Commission on Radiological Protection (ICRP) in its Publication 60 [1] makes no reference to environmental protection, although the view is held that the environment indirectly is afforded adequate protection through application of the system. This indirect approach is nowadays, however, generally considered inadequate or even inappropriate. This may not only apply in situations where man is absent or not exposed, but may also be doubted on more scientific grounds. Consequently, there is much international effort in development of new assessment and management systems focusing on protection of the environment or even trying to link the systems for protection of humans and the environment, respectively, together.

Ecological risk assessment has been developed in the non-radiological field. It may be defined as: “an evaluation of the likelihood of adverse effects on organisms, populations and communities from chemicals in the environment” [2]. The word “chemicals” may be replaced by “stressors”. As the ecological risk assessment approach is meant to be flexible to be adapted to different situation, its application to the radiological field is a natural progression, radionuclides being just another stressor [3].

Ongoing international activities to establish frameworks for radiological impact assessments focusing on biota and ecosystems, and to various extents incorporating elements of frameworks created for non-radiological assessments, include those of the International Atomic Energy Agency [4], the ongoing revision of the ICRP recommendations, and EC (FASSET – Framework for ASSESSment of Environmental impacT [5]; EPIC – Environmental Protection from Ionizing Contamination in the Arctic). On a national level, the US Department of Energy has developed a tiered approach to demonstrate compliance to certain derived environmental nuclide concentration standards [6]; the approach to assessment based on exposure and effects analysis developed by the Environment Agency of England and Wales in collaboration with English Nature [7]; and the Canadian Nuclear Safety Commission’s initiatives [8].

A related activity has recently been concluded in the form of the IAEA BIOMASS (BIOSphere Modelling and ASSESSment) project [9]. The BIOMASS programme aimed to develop and apply a methodology for defining biospheres for practical radiological assessment of releases from radioactive waste disposal, as simplified in Figure 1.

The assessment context under BIOMASS is a systematic analysis of the background, purpose and expected outcome of the assessment. The present paper analyses how FASSET has applied elements of the BIOMASS assessment context.

## 2. FASSET PROBLEM FORMULATION IN RELATION TO BIOMASS

The ecological risk assessment involves considerations to be made during *problem formulation*, development of relevant *methodologies for assessments*, and development of the relevant *management methods*, as shown in Table 1. The general structure of existing frameworks for environmental risk assessment has been considered to be appropriate to FASSET.

The use of ecological risk assessment is also favoured from the simpler environmental impact assessment approach, as assumptions and uncertainties are explicitly considered and examined throughout the risk assessment [3].

From Table 1, it is obvious that important choices need to be made at the problem formulation stage, and that these choices will guide the methodologies applied during assessments. Furthermore, the problem formulation will be the foundation for which managerial measures that can be taken on the basis of the assessment.

In order to guide assessors, the BIOMASS project has reviewed and systematically analyzed the different options that can be considered during the problem formulation stage, and their implications for the subsequent assessment. In this section, the FASSET approach is described for the problem formulation elements identified in BIOMASS. An overview of the FASSET approach is given in Table 2, whereas some of the elements are expanded on in Sections 2.1 – 2.7. Full justification will be detailed in the Deliverable D2 “Existing programmes for the assessment of risks to the environment from ionizing radiation and hazardous chemicals; their relevance to FASSET”, to be published in November 2002, on the FASSET web-site [www.fasset.org](http://www.fasset.org).

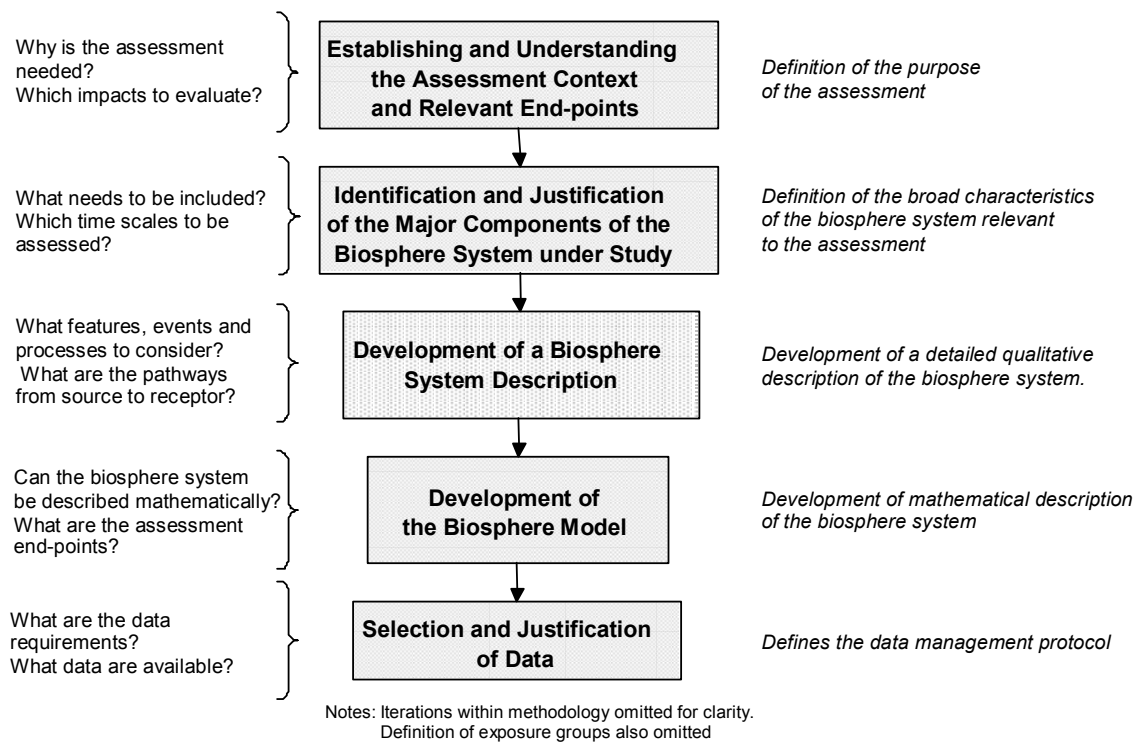


FIG. 1. Simplified Stages of the BIOMASS Methodology [10].

TABLE 1. GENERIC FRAMEWORK FOR ASSESSMENT AND MANAGEMENT OF ENVIRONMENTAL RISKS FROM IONIZING CONTAMINATION

Problem formulation	Assessment	Management
Description and definition of the assessment context:	Methodologies relevant to:	Strategies and methodologies relevant to:
Purpose	– Exposure analysis	– Prevention, mitigation and elimination of environmental consequences
– Rationale and Approach	– Effects analysis	
– Spatial and temporal considerations	– Risk characterization and consequence analysis	
– Source characterization and hazard identification		
– Identification of effects and objects for protection		
– Data requirements		
– Treatment of background		

TABLE 2. PROBLEM FORMULATION ELEMENTS CONSIDERED IN THE FASSET FRAMEWORK – EXAMPLES

Formulation Element	Relevance to FASSET
Purpose of the assessment	<ul style="list-style-type: none"> <li>– FASSET focuses on impact assessment on biota and ecosystems, not primarily on management.</li> <li>– FASSET should be capable of guiding a wide range of stakeholders.</li> <li>– FASSET should deal with all effects of radioactive substances; Multi-contaminant effects will be considered, though they will be dealt with as uncertainties.</li> </ul>
Rationale and approach to assessment	<ul style="list-style-type: none"> <li>– Realistic estimates will be made, on the basis of which a precautionary approach can be built.</li> <li>– Effects analysis included within the assessment</li> <li>– Framework to be user friendly and understandable, but based on sound science.</li> </ul>
Spatial and temporal considerations	<ul style="list-style-type: none"> <li>– The framework is to consider both accidental and chronic situations.</li> <li>– Some general guidance about the definition of the assessment area will be given.</li> <li>– No specific consideration of changes in the biosphere with time, or of the transition between one biosphere state and another, is made.</li> </ul>
Source characterization and hazard identification	<ul style="list-style-type: none"> <li>– Source term characterization requires information to be supplied by the assessor (list to be provided).</li> <li>– A preliminary list of radionuclides will be included in the framework.</li> <li>– Method for hazard identification will rely on biological and chemical properties of the radionuclides.</li> </ul>
Identification of effects and objects of protection	<ul style="list-style-type: none"> <li>– Input to FASSET is given in terms of the radionuclide concentration, deposition, or flow in environmental media.</li> <li>– Dose/dose rate is the appropriate quantity to work with in order to characterize the effects related to a given exposure.</li> <li>– Dose-effect data will be collected and summarized for each reference organism and four groups of effect.</li> <li>– Populations and ecosystems are the targets for protection, whereas individuals are the targets for assessment.</li> <li>– Seven major European ecosystems considered.</li> <li>– Reference organisms will be used as a basis for modelling.</li> <li>– Justification given for the identified 31 candidate reference organisms[11].</li> </ul>
Data requirements	<ul style="list-style-type: none"> <li>– FASSET is based on the use of generalized empirical data from traceable sources for European ecosystems. Quality checks are being carried out on the data.</li> <li>– Where data are insufficient, data gaps will be filled if possible by analogy. A reasonable degree of caution will be adopted, accompanied by clear statements about the assumptions made and the introduced uncertainties.</li> </ul>
Treatment of background	<ul style="list-style-type: none"> <li>– The environmental effect of radionuclides in the environment is related to the total dose, which includes both the natural background, anthropogenic background and the dose increment from the source being assessed.</li> <li>– The framework will give guidance to the assessor about how to measure or derive the background levels, including the consideration of all sources into a given receiving environment.</li> </ul>

## 2.1. Purpose

The FASSET framework will centre on the scientific dimension of an impact assessment framework for biota and ecosystems, and will be appropriate for varying formulated problems, e.g. FASSET will consider:

- Ongoing, past and future releases – the framework should enable for assessment of actual effects on the basis of measurements and direct observations in the environment, which makes it relevant.

- Chronic and acute effects – the framework should allow for assessment of effects of chronic low dose rate radiation as one extreme and acute high doses following e.g. accidental releases as the other extreme. Thus, the range of biological effects as well as environmental dose rates considered has to be wide.
- Be appropriate for various purposes, e.g. licensing, demonstration of compliance, assessments of accidents, decisions concerning remediation – the framework has to come up with as realistic data as possible to support decision-making.

Ethical considerations that drive the need for the assessment, for example cultural or legal dimensions, are not incorporated within FASSET. These issues pre-determined by the assessor's societal requirements, and make part of the planning process that precedes the formulation of the assessment. The risk management stage also lies outside the scope of the FASSET project, as it is not for FASSET to prescribe management decisions. The emphasis of FASSET will rather help the development of tools and data for the assessment phase.

## 2.2. Rationale and approach

BIOMASS distinguishes between the 'equitable' and the 'cautions' approaches. FASSET sets out to provide guidance to regulators, implementers and stakeholders in general by providing *realistic* estimates; a cautious approach can be developed on the basis of a realistic assessment e.g. through application in the managerial stage. The approach also needs to be flexible and manageable in order to take into account the various risk management options, the capability of implementers and regulators to adopt the methodology, as well as societal concern defined in the formulation stage.

A consequence of this is that the effect analysis becomes integrated into the assessment, and is not carried out separately. An important part of the FASSET project thus becomes the methodology for assessing effects (see effects and objects of protection below).

Within the generic ecological risk approach, a tiered approach is often embedded into the assessment for screening purposes, e.g. in regulatory UK [12] and USA [3]. FASSET will not use such a tiered approach since it is not a compliance tool, targeted to predefined dose or concentration thresholds. A generic screening assessment using reference organisms is an appropriate first step of an assessment. However, the FASSET framework recognizes:

- that there sometimes will be a need to follow up a screening stage with more specific assessments (e.g. when the margin of safety in the screening assessment was insufficient);
- that the screening methodology should be designed in such a way that it facilitates rather than hinders subsequent specific assessments (e.g. by using similar basic criteria for choice of endpoints); and
- that several plausible contamination scenarios are site-specific by nature (e.g. nuclear facilities and waste repositories) which implies that there will be a strong demand on the risk assessment of such facilities to generate estimates of risks to valued components of the surrounding ecosystem (i.e. a site-specific assessment).

## 2.3. Spatial and temporal considerations

FASSET will consider both accidental and chronic situations. For accidental situations, discussion is required concerning assumptions of equilibrium in the exposure models, and the interface of FASSET with dispersion and transport models, both in terms of time and space, particularly with respect to modelling a point source.

No specific consideration of changes in the biosphere with time, or of the transition between one biosphere state and another, is presently made within FASSET. FASSET should however, be applicable to future biosphere states (assumed/predicted) or past situations.

Spatial considerations may be very specific to a particular assessment. Again, the interface with dispersion/transport models is important as these models will provide the basis for the definition of the area of assessment. General guidance about the definition of the assessment area would involve e.g., a check to ensure that areas of high concentration/accumulation outside the defined assessment area do not occur.

## 2.4. Source characterization and hazard identification

Source term characterization requires amongst others the following information, which will need to be supplied by the assessor:

- total release of radioactivity and the relative contribution of each isotope;
- distribution of release over time;
- change with time in relative contribution of each isotope;
- isotopic dilution of radionuclides in the receptor ecosystems;
- physical parameters of radionuclides (i.e. half-lives, type and energy of radiation);
- chemical form of the radionuclides; and
- origin of radionuclides; the way in which radionuclides reach the receptor ecosystem, e.g. from below ground, as release directly to surface water, deposition to land or water surfaces.

Hazard analysis should consider:

- the environmental behaviour, either known (e.g. known sorption behaviour), or calculated (e.g. from chemical equilibrium calculations);
- the biological activity, either known (e.g. the uptake of a radionuclide in a range of biota), or calculated (tendency to hydrolyze, form biological ligands *etc*);
- chemical toxicity, either known, or based on classification into two groups; elements with no known biological function and elements which have a known biological function (e.g. macronutrient, micronutrient) or analogous elements; and
- bioaccumulation and biomagnification, expressed in units which allow comparison of radionuclides, e.g.  $\text{g C}^{-1}$  or  $\text{g dw}^{-1}$ , as well as on a fresh weight basis.

## 2.5. Identification of effects and object of protection

A total of 31 candidate reference organisms have been identified so far within FASSET, being representative of the seven chosen European ecosystems. They serve as a basis for exposure analysis, taking into account radionuclide concentrations (external and internal) per unit deposition or unit flux, and dose conversion factors that consider geometric factors. Input into exposure analysis is given in terms of the radionuclide concentration, deposition or flux in environmental media.

Dose and dose rate are the appropriate quantities to characterize the effects related to a given exposure (i.e. to express the dose-effect relationships for the four groups of effects being considered). Dose-effect data, measured or observed in individuals, will be collected and summarized for a number of wildlife group. The information will be divided into four groups of effect (morbidity, mortality, reproductive effects, cytogenetic effects), which are considered to be important to the performance of the population.

## 2.6. Data requirements

FASSET has formulated the data requirement as a part of the problem formulation stage, rather than as part of the assessment stage (cf. BIOMASS methodology). This is partly because of the pre-selection of seven European ecosystems.

The level of uncertainty depends on whether the assessment is probabilistic or deterministic, and whether it is cautious/conservative or realistic. FASSET will take an equitable approach, realistic approach (worst case is not considered). However, the definition of equitable will need to be redefined from the BIOMASS philosophy to be applicable to the FASSET framework.

Consistency within the choices of data is important from the assessment context through to the development of the models. However, data are often supplied from a number of sources, which can lead to uncertainty in the results. Quality checks are being carried out on the data, and use of data is being maximized by pooling the available data. Where data are insufficient, a reasonable degree of



caution will be applied, accompanied by clear statements about the assumptions made and the introduced uncertainties. Where data are missing or incorrect, it is important to investigate ways of identifying the correct data. BIOMASS developed a structured data management system to link data types, data availability and data requirements.

## **2.7. Treatment of background**

The environmental effect of radionuclides in the environment is related to the total dose, which includes both the natural background, anthropogenic background and the dose increment from the source being assessed. Background can be either directly measured, or based on estimates from reference areas. Geological criteria (and others) may be defined to help the assessor in the appropriate selection of background values from a compiled list.

## **3. FASSET ASSESSMENT METHODOLOGY IN RELATION TO BIOMASS**

Carrying out the assessment itself will need to consider a wide range of scientific knowledge. FASSET's "Effects" group is assembling a Radiation Effects Database that will gather literature data to help summarize dose-effect relationships between radiation and selected wildlife groups. The "Dosimetry" and "Exposure" groups will provide tabular parameter values, but coupled with guidance on how to use them, together with data uncertainty and limits of applicability. The overall framework will develop further each of those components needed to complete the assessment methodology, to be published in October 2003.

Questions to be addressed include the selection of key elements (e.g. features, events, processes, reference organisms, radionuclides), as well as the level of simplification to be introduced in assessment methods. The application of the BIOMASS methodology to FASSET will ensure that only relevant parameters are considered within each given assessment, based on the assessor's chosen purpose. The main benefits will consist of following the BIOMASS step by step approach, using interaction matrices and checklist Tables at critical stages of the assessment. For example, FASSET will be able to refer to existing interaction matrices for certain ecosystems, and complement others by using the BIOMASS format as a template.

## **4. INTERACTIONS BETWEEN FASSET AND OTHER EXISTING INITIATIVES**

The BIOMASS approach has also been shown to be applicable to the UK's approach dealing with impact assessment of ionizing radiation on wildlife [13]. The study investigated dose rate calculations and impact assessment of ionizing radiation on target organism. To do this, the assessor's requirements were: a radionuclide source, exposure pathways and ecological parameters. Many assumptions were then made about total absorbed dose and compared with known radiological effects. The result was a comprehensive set of spreadsheets that are able to calculate concentrations in different organisms within an ecosystem, as illustrated in Figure 2 [7]. As a result of the study, some findings have fed into FASSET, and further developed into regulatory guidance in England and Wales to protect wildlife from ionizing radiation [14].

In order to benefit from the views of other organizations that have an interest in environmental radiation protection, the FASSET consortium arranged an External Forum in April 2002, to which representatives from these organizations have been invited. The event also coincided with the project's mid-term review, and provided the opportunity to consult with international views. A number of issues and recommendations were raised and taken on board by FASSET, where appropriate. The outcome of the Forum is posted on the FASSET web-site.

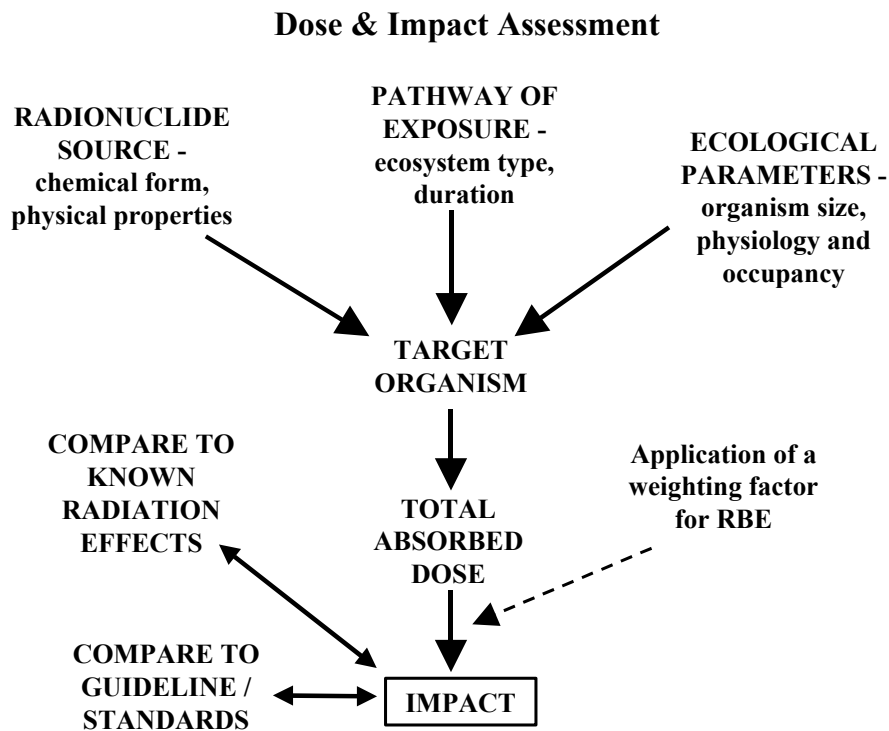


FIG. 2. Impact assessment approach used in R&D Publication 128 [7].

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# Ethical aspects of the protection of animals

L. Koblinger, I. Vigh

Hungarian Atomic Energy Authority, Budapest, Hungary

**Abstract.** Many of the studies dealing with the protection of animal lives against the harmful effects of ionising radiation concentrate on the technical aspects of the question, mainly on the comparison of the radio-sensitivities of various species. In this paper we intend to address rather the basic ethical question: Why should we protect other species than man? What kind of philosophical arguments of the protection can be listed?

First, we discuss the intentional irradiation of animals. From the very beginning of radiation studies series of irradiation of animals were carried out intentionally, partly for the purpose of extrapolation modelling. Furthermore, there were attempts to find radio-protective chemicals to be used as a certain kind of preventive medicine for men. Another branch of intentional exposure is food preservation by irradiation, which is considered by the World Health Organization as “safe and wholesome”. By the application of this technique, lives of millions of human beings can be saved in the developing world. Similarly beneficial is the sterilisation of syringes and other medical instruments.

The second basic philosophical question discussed in our presentation is whether animals should be *per se* protected? While killing a human being is considered as a crime in all civilised societies and the ethical background is given in all ethical religions, the picture is not so homogeneous in respect of the animals. There seems to be a clear difference between the teachings of the Eastern and Western religions.

If we do not protect the animals for themselves, we still may need to protect them in our (human) interest. For the application of a “sake of mankind” philosophy, animals should be distributed into groups of “beneficial” and “damaging” for the present and future life of mankind. Do we have enough knowledge to decide about it nowadays?

Finally, special attention is given to questions on biodiversity. Animals disappeared “continuously” during the biological history of the Earth. This spontaneous change in the spectrum of the fauna has been a fundamental factor of the evolution. Several thousand years ago man started to influence the fate of several kinds of animals. Conservation of the diversity, in our reading, means mainly the conservation of variations much more than conservation of each variant.

## 1. INTRODUCTION

Very soon after the discovery and first applications of the x-rays (Röntgen, 1895) and radioactivity (Becquerel, 1896) scientists took notice of the harmful health effects of ionising radiation.

In 1902 W. H. Rollins reported experimentally induced fatalities in guinea pigs exposed to x-rays [1] i.e. he demonstrated that ionising radiation could kill higher life forms, and from our point of interest it is worth mentioning that this observation was derived from results of animal experiments.

Regulatory systems of radiation protection up to now, however, have been restricted to the protection of men, the protection of other species has not explicitly been included. The International Commission on Radiological Protection (ICRP) stated in its Report No. 26 [2] that “*The Commission therefore believes that if man is adequately protected then other living things are also likely to be sufficiently protected*” and basically this concept is preserved in the newest ICRP recommendations [3]: “*The Commission believes that the standard of environmental control needed to protect man to the degree currently thought desirable will ensure that other species are not put at risk*”.

Quite recently a change of this approach is taking place: the question of protection of the environment against ionising radiation is increasingly discussed. One of the concerns behind is that if radiation is controlled exclusively via the doses received by human beings, no restrictions can be set for releases at non-inhabited areas. Moreover, there is recently a general increase of interest in the protection of our environment, in the conservation of the other forms of life around us.

A more or less systematic investigation of the question should start with the definition of the term “environment”. According to the definition given in an International Atomic Energy Agency report [11] *environment* includes “*all layers of the lithosphere, hydrosphere and atmosphere containing life forms*”. It is practically impossible to scan all these areas in a single presentation, the discussion in this paper is limited to the protection of one kind of life forms, to the protection of animals.

Many of the studies dealing with the protection of animal lives against the harmful effects of ionising radiation concentrate on more technical aspects of the question, mainly on the comparison of the radio-sensitivities of various species. In this paper we intend to address rather the basic ethical question: Why should we protect other species than man? What kind of philosophical arguments of the protection can be listed?

The present discussion is more or less restricted to an exploration of the subject, no formulation of definite answers is aimed.

## 2. INTENTIONAL IRRADIATION OF ANIMALS

From the very beginning series of exposures of animals were carried out intentionally, for studying the biological (somatic and genetic) effects of ionising radiation, partly for the purpose of extrapolation modelling with the hope that some of the results can be applied for human beings. Though such experiments are executed even nowadays, the validity of extrapolation is very questionable, especially in the case of genetic consequences. Whereas severe changes in offspring of irradiated animals have been detected, no such effects are found in human populations.

Furthermore, there were attempts to find radio-protective chemicals to be used as a certain kind of preventive medicine for men [1] mainly for military purposes. Though no really effective radio-protective chemicals have been discovered, exposures of animals are still executed in biological research studies.

The ethical questions of such intentional exposures are out of the scope of this presentation, the rightfulness and restriction of animal experiments in a broader sense, i.e. not exclusively for irradiation, are laid down in international recommendations [5] and national legal means (e.g. [6]).

Another branch of intentional exposure is food preservation by irradiation, which is considered by the World Health Organization as a “safe and wholesome” method. By the application of this technique, i.e. by destroying the micro-organisms and parasites that cause food-borne illness and death, lives of millions can be saved in the developing world. (It is interesting to note here that even in an advanced country like the U.S.A. food-derived pathogenic bacteria and other parasites claim an estimated 10,000 lives annually [7].)

Similarly beneficial is, for example, the wide-spread technique of sterilisation of syringes and other medical instruments. During these processes vast numbers of living organisms are killed intentionally whereas millions of human lives are saved. In contrast to the restrictions against animal experiments there are only negligible protests for refusing preservation or sterilisation just from the point of prevention of microorganisms and bacteria.

After these brief introductory remarks on intentional irradiation, hereafter we shall further restrict the discussion to non-intentional irradiation, irradiation caused by by-products of human activities, mainly irradiation due to environmental releases.

## 3. SHOULD ANIMALS BE *PER SE* PROTECTED?

There is a more or less universal agreement that people should not cause harm to each other. Killing a human being is considered as a crime in all civilised societies and the ethical background is given in all ethical religions.

The picture is not so homogeneous in respect of the animals. There seems to be a clear difference between the teachings of the Eastern (Hinduism and Buddhism) and Western (Judaism, Christianity, Islam) religions. With some simplification, religions protect all creatures having “soul”, however, in the Western religions soul is attributed exclusively to human beings, whereas in the Eastern religions animals (or even plants) are assumed to have it.

According to the Bible [8] human beings have dominion over all other living creatures: “*Let us make man in our image, after our likeness; and let them rule over the fish of the sea and the birds of the air, over the livestock, and over all the earth, and over all the creatures that move along the ground.*” (Gen 1,26). Animals have been created to nourish man: “*Every thing that lives and moves shall be food for you.*” (Gen 9, 3). In a similar manner, Quran [9] leads people “*eat (for yourselves) and pasture your cattle*” (Q 20, 54).

Needless to say, in such a framework animals are never considered as a group of individual beings and animals are not to be protected for themselves.

As a consequence of this way of thinking prominent medieval Christian scholars, like St. Thomas Aquinas, argued that after the Last Judgement people shall not drink and eat, bodies shall have no physiological functions, therefore, there shall be no flora and fauna in New Jerusalem [4]. Things not needed for human beings are not needed at all.

On the contrary, in ancient beliefs there were no such sharp distinctions between men and animals. In Mesopotamian, Egyptian or Greek mythologies quite many animals were worshiped, or divinities manifest (or disguise) themselves in the forms of animals. Similarly, in Hinduism, Vishnu appears in forms of various animals, e.g. fish, turtle, boar, lion [4].

In Buddhism slaughter of animals is considered as a kind of basic ignorance. The Four Noble Truth says: “*We respect the lives of other creatures, even the lives of insects and creatures we do not like. Nobody is ever going to like mosquitoes and ants, but we can reflect on the fact that they have a right to live.*” [10]. Here, human beings are considered as just one singular type of animals having no special thing like ‘soul’. Consequently, all types of animals should be protected in the same manner as human beings.

On the contrary, the actual approach of people living in developed western countries is based rather on emotions than on philosophical backgrounds. Several people consider their pets as individual personalities. For these people, any harm caused to their beloved pets is considered in a similar manner as harm of their friend. On the extremity, some of them argue that they trust more animals than men, because they have never been disappointed by animals. In most cases, these people are much more concerned about mammalian than lower level species. People who are ready to demonstrate for the seals or tigers, kill, without any hesitation, mosquitoes or other insects.

#### 4. PROTECTION OF THE ANIMALS FOR THE BENEFIT OF MANKIND

Even if we do not protect the animals for themselves, we still may need to protect them in our (human) interest. Animals make our life more convenient, or even possible at all, in various ways. The absolute necessity or importance of their presence is questionable in many cases and in some instances the value of their contribution decreases as man-made technologies develop and take over the functions of animals.

One of the most typical beneficial uses of animals is the consumption of their meat or their products (e.g. milk). It is not an absolute necessity, vegetarian people live normal lives throughout the world and millions of people grow up without milk consumption.

The next example of use of animals or animal products is clothing. Tens of thousands of years ago men’s bodies were protected against cold by furs of animals and luxurious dresses were made from the secretion of silkworms. Though artificial products replace natural materials in many instances, in good quality clothes and shoes still furs, wool and skins of animals are used as raw materials.

Animals were extensively used for transport and heavy works. Though motorization released most of the animals of this “type of work” especially in the technically developed world, in less industrialized areas animals still take part in e.g. in transportation of heavy loads.

Parallel to these tendencies there is an opponent trend in highly developed countries, more and more animals are kept just for the fun of men, e.g. horses are bred for sport activities, pets are found in many households.

In several cases governments have to interfere to protect species, to stop the unnecessary slaughter of animals just for satisfying luxury demands, e.g. to control hunting of elephants for ivory or baby seals for their furs.

The arguments written above are directly valid mainly for mammals (exceptions are e.g. birds and fish kept as pets). The benefits of insects are sometimes less obvious but not at all less important. Just to give one example, bees are necessary for pollination of flowers and blossoms.

The whole set of mankind-animals-plants-microorganisms forms a very complicated non-linear system built up of thousands of elements, with thousands of interactions and feedbacks. Even with our present level of biological knowledge it is very difficult, if not impossible, to estimate which elements can be neglected from the system, which can be replaced by others and the removal of which species may lead to a collapse of the whole system.

The extinction of certain species may change the spectrum of living creatures, or may disturb the dynamical biological balance on Earth in such a way that endangers human life.

In this respect though animals should not necessarily be protected individually, species must be conserved. This approach is outlined in the ICRP recommendations when they state that: *“protection of all human individuals is thought likely to be adequate to protect other species, although not necessarily individual members of those species”* [2] and *“Occasionally, individual members of non-human species might be harmed, but not to the extent of endangering whole species or creating imbalance between species”* [3].

If the philosophy of protection is drifted from the point of “the sake of the animals” towards “the sake of mankind” criteria should be found for distributing the animals into groups of “beneficial” and “damaging” for the present and future life of mankind. Do we have enough knowledge to decide about it nowadays?

Let us consider simple examples. Encephalitis and Lyme disease are serious, quite frequently lethal, illnesses. Both are propagated by ticks. Do the human victims serve us with a strong enough argument for killing of the species? Can the millions of people dying from malaria each year give a base for killing Anopheles mosquitoes? Is the evaluation of the life of a bear the same for those people who just see them in zoos and buy those lovely teddies for their children and for those whose relatives were killed by bears in the forest?

There are many non-linear mechanisms governing the development of flora and fauna. Propagation of one species and extinction of another one is sometimes interrelated. Functions of species disappearing from the stage are sometimes taken over by others. We are not able to judge what kind of changes lead to irreversible, non-correctable modifications in the chain of evolution.

Unfortunately, there are no generally accepted answers to these questions.

## 5. QUESTIONS ON BIODIVERSITY

Animals have disappeared “continuously” during the evolution, 99 % of all the animal species ever lived on earth are extinct. This spontaneous change in the spectrum of the fauna has been a fundamental factor of evolution. The extinction of several species seems to be inevitable for the presence of others, one can hardly imagine people living in civilised societies on Earth – together with e.g. Tyrannosaurus Rex.

The non-linear system of mankind-animals-plant-microorganisms is not a statistic one, elements disappear, new ones come into scene, the roles and functions of species change. The dynamics of the system has played an inevitable role during the whole process of natural history. Life on Earth today differs significantly from that of hundreds of thousands of years ago, which differed a lot from that of millions of years before.

For most of the time these changes were ruled by natural laws and, especially by the mechanisms of evolution, and no consciousness was involved.

Several thousand years ago man started to influence the life of several kinds of animals. Hunting resulted in extinction of certain species. Human activity (mainly hunting) accounts for the extinction of the aurochs, the dodo, the huia, and many others.

On the other hand, breeding and domestication lead to modification of the genome of some species.

The spontaneous process has been artificially modified and accelerated. And these types of modifications can impose a danger on nature, since we cannot predict what types of changes may turn to be fatal in the future. Recognising the value of biodiversity the governments of the world have developed the Convention on Biological Diversity.

Conservation of the diversity, in our reading, means mainly the conservation of variations much more than conservation of each variant or the conservation of the present spectrum.

Let us refer to the dynamics of the living system. Changes have always happened and, most probably, an artificial ‘freezing’ of the present state (i.e. removing the spontaneous dynamics) might cause harm in the same manner as the introduction of too many artificial changes.

The question is even more exciting with ionizing radiation that can kill higher forms of life but, at the same time, induce mutations, i.e. modify genomes, create new species. The artificial creation of new species may turn to be either dangerous or beneficial, but increases biodiversity, anyhow!

One point is clear: we must be very cautious to induce any changes with unpredictable consequences, let the changes be introduced either by exposure to ionizing radiation or in any other way. Special care should be taken with the speed of the changes, since there is an obvious danger that human activities can accelerate (or decelerate) the natural processes. Cautiousness, however, may not lead to a refusal of all changes. Especially not, when the basic ethical background of the goals of the conservation and protection is not clearly set and universally accepted.

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# Development of a Framework for ASSESSING the Environmental impact of ionising radiation on European ecosystems – FASSET

C.-M. Larsson<sup>a</sup>, G. Pröhl<sup>b</sup>, P. Strand<sup>c</sup>, D. Woodhead<sup>d</sup>

<sup>a</sup> Swedish Radiation Protection Authority, Stockholm, Sweden

<sup>b</sup> German National Centre for Environment and Health, Institute for Radiation Protection, Neuherberg, Germany

<sup>c</sup> Norwegian Radiation Protection Authority, Østerås, Norway

<sup>d</sup> Centre for Environment, Fisheries and Aquaculture Sciences, Lowestoft Laboratory, Lowestoft, United Kingdom

**Abstract.** A total of 15 organisations (including regulators, research institutes and industry) in seven European countries (Finland, France, Germany, Norway, Spain, Sweden and UK) are collaborating on creating a Framework for ASSESSMENT of Environmental impact (FASSET) of ionising radiation. The project aims at developing approaches and tools for assessing impact on biota and ecosystems, and to support efforts to protect the environment from harmful effects of radiation. The project is supported by the European Union through the EC 5th Framework Programme, and is due to end in October 2003.

The Project is divided into four work packages. In WP2, seven European ecosystems are considered in the assessments; three aquatic (marine, brackish, freshwater) and four terrestrial (seminatural ecosystems including pasture, agricultural ecosystems, wetlands and forests). A list of candidate generic reference organisms has been drawn up on the basis of expert judgement of exposure situations in the selected ecosystems. These organisms are considered references or ‘surrogates’ for existing biota in the natural habitat. They serve as starting points for development of dosimetric models, being developed in WP 1, and for pooling available information on ecological relevance and biological effects. Further analysis of the candidate reference organisms is performed to justify their choice and assess their applicability in different situations, taking into account modelling of radionuclide transfer, estimates of internal and external dose rates, ecological significance and biological effects.

WP3 considers general ‘umbrella’ effects that, when manifested in an individual, may have an impact at population level or at higher levels of the organisational hierarchy. The four categories are: morbidity (fitness or well-being), mortality (death directly attributable to radiation), reproductive success (changed number of offspring) and scorable cytogenetic effects (molecular actions, aberrations, etc.). A database is being assembled, compiling data from the literature for a number of organism categories for each of these four umbrella effects. The database also considers the suitability of data to derive RBE for different types of radiation.

The work from the three WPs on exposure, dosimetry and effects will be organised into a framework for impact assessments, which will take into account experience from application of ecotoxicological approaches in assessing effects of other hazardous substances (carried out in WP 4). Characterisation of risks will be performed in a way that attempts to make the framework useful to regulators, for demonstration of compliance, and for communication with the public and decision-makers.

The latest development in the project will be presented. The progress of the project can be followed on its website, [www.fasset.org](http://www.fasset.org).

## 1. INTRODUCTION – OBJECTIVES OF THE FASSET PROJECT

The requirement for assessments of the environmental effects of radiation is increasing due to growing public concern for environmental protection issues and integration of environmental impact assessments into the regulatory process. Thus, there is a strong need to establish a framework for the assessment of environmental impact of ionising radiation, as well as a system for protection of the environment from ionising radiation. These ambitions are reflected in a number of international efforts and various ‘systems’ have been proposed or are under development [1–7].

The FASSET (Framework for Assessment of Environmental Impact) project is an EC 5<sup>th</sup> Framework Programme project, comprising 15 partners in seven European Countries (Finland, France, Germany, Norway, Spain, Sweden and UK). The FASSET project aims at providing a formal framework for the assessment of radiation effects on biota and ecosystems, aimed at assisting decision-makers and stakeholders when judging the environmental impact of ionising contaminants from past, present or planned sources. The project started on 1 November 2000 for a duration of 36 months, i.e. is to end by 31 October 2003.

The project is organised in four different sub-projects or Work Packages (WP). WP 1 considers environmental dosimetry, WP 2 radionuclide transfer in ecosystems, WP 3 the biological effects of ionising radiation, and WP 4 the creation of the framework for assessments. There is substantial interaction between the WPs, and all interact in the creation of the framework.

The project has the following practical objectives:

- To provide a set of reference organisms relevant to different exposure situations. The identification of reference organisms must take into account the environmental fate of radionuclide releases, exposure pathways, dosimetry and biological effects.
- To provide a set of models for the reference organisms, including models for environmental transport of radionuclides, exposure, dosimetry and biological effects.
- To critically examine reported data on biological effects on individual, population and ecosystem levels, as a point of departure for characterising the environmental consequences of, e.g., a source releasing radioactive substances into the environment.
- To review existing frameworks for environmental assessment used in different environmental management or protection programmes. This review will extend outside the field of radiation protection, and consider, *inter alia*, frameworks for managing risks from genotoxic chemicals. The resulting FASSET framework will thus be set in a wider context of assessments of environmental effects and management of risks to the environment.

In order to communicate the project results, a website has been created, [www.fasset.org](http://www.fasset.org), where the project status can be followed. Also, stake-holders' views have been considered by organising a FASSET External Forum, where a number of invited organisations were recently provided the opportunity to offer guidance and critique on the approaches and further development of the project.

The following description is a brief summary of the project's initial achievements, which also illustrate the project's general direction.

## 2. THE REFERENCE ORGANISM CONCEPT

A special feature within FASSET is the focus on reference organisms. This approach is analogous to the reference man concept that has been adopted within radiological protection to provide a standard set of models and datasets. The project's working definition of the reference organism is:

*“a series of entities that provide a basis for the estimation of radiation dose rate to a range of organisms which are typical, or representative, of a contaminated environment. These estimates, in turn, would provide a basis for assessing the likelihood and degree of radiation effects”.*

An initial step in the construction of the framework is thus the selection of appropriate reference organisms. The final choice of reference organisms for consideration within the FASSET framework will be an iterative process taking into account dosimetry as well as radioecological criteria and radiosensitivity.

The initial work on exposure in different ecosystems was concerned with the identification of candidate reference organisms from the point of view of radioecological sensitivity. The factors determining radioecological sensitivity are:

- whether the habitat or feeding habits of the organism are likely to maximise its potential exposure to radionuclides, based on an understanding of the distribution of the different radionuclides within the ecosystem;

- whether the organism exhibits radionuclide-specific bioconcentration which is likely to maximise internal radionuclide exposures in particular circumstances;
- whether the position of the organism within the foodchain (e.g., top predator) is such that biomagnification of radionuclides up the foodchain may lead to enhanced accumulation.

In order to identify the candidate reference organisms, major European ecosystems have been characterised in terms of their ecological characteristics, important pathways and radionuclide transfer processes. The ecosystems considered are: for the terrestrial environment, semi-natural ecosystems including pastures, agricultural ecosystems, wetlands and forests; and, for the aquatic environment, fresh-water, marine and brackish ecosystems.

Based upon our knowledge of the distribution of radionuclides within the environment, a simplified compartmentalisation has been used: soil, herbaceous layer and canopy for terrestrial ecosystems; bed sediment and water column for aquatic ecosystems. Some organisms may be present in different compartments, most notably the roots and above ground parts of plants. The project considered simplified ecological niches/organism groupings within the selection process, to ensure that candidate reference organisms will be sufficient to protect the environment as a whole within any assessment. In this selection the availability of data for an organism, or the ability in the future to obtain the required data, are also considered.

The approach taken towards their selection should ensure that suitable reference organisms are available for a range of scenarios (chronic and acute exposure) and the different European ecosystems. In total, 31 candidate reference organisms have been suggested. The complete list (Table 1) and the reasoning behind the selection is found in project Deliverable 1 [8], also available on [www.fasset.org](http://www.fasset.org).

TABLE 1. CANDIDATE REFERENCE ORGANISMS IDENTIFIED FROM AN EXPOSURE PATHWAYS ANALYSIS

Terrestrial ecosystems	Aquatic ecosystems
<b>Soil</b>	<b>Sediment</b>
Soil micro-organisms	Benthic bacteria
Soil invertebrates, ‘worms’	Benthic invertebrates, ‘worm’
Plants and fungi	Molluscs
Burrowing mammals	Crustaceans
	Vascular plants
<b>Herbaceous layer</b>	Amphibians
Bryophytes	Fish
Grasses, herbs and crops	Fish eggs
Shrubs	Wading birds
Above ground invertebrates	Sea mammals
Herbivorous mammals	
Carnivorous mammals	<b>Water column</b>
Reptiles	Phytoplankton
Vertebrate eggs	Zooplankton
Amphibians	Macroalgae
Birds	Fish
	Sea mammals
<b>Canopy</b>	
Trees	
Invertebrates	

### 3. EXPOSURE ANALYSIS FOR VARIOUS REFERENCE ORGANISMS

A number of radionuclide transfer models developed for different ecosystems will be used for tabulation of external and internal radionuclide concentrations. Furthermore, tabulations will be made to allow conversion of concentrations in the environmental media, and internally, to absorbed dose (rates); this will include consideration of the relative biological efficiency (RBE) of different radiation types in the development of appropriate radiation weighting factors ( $w_r$ ) for the organisms, endpoints and dose rates of concern. The radionuclide transfer models start with unit deposition for acute exposure and for steady state, and with radionuclide flux for dynamic models developed for, e.g., waste repositories. This work has commenced and is due to be finalised at the termination of the project in the autumn of 2003.

#### 3.1. External exposure

In the first phase of the project, the estimation of external exposure focussed on organisms in the terrestrial environment. The recently applied analytical approaches, such as the point-source-dose-distribution function, provide sufficiently accurate results in media with relatively homogeneous densities as is the case for aquatic environments. However, in terrestrial habitats with pronounced inhomogeneities in materials and densities, analytical approaches are associated with considerable uncertainties.

In order to estimate the external exposure, Monte-Carlo calculations have been made for various reference organisms. Details on the assumed exposure conditions are given in Table 2. In all cases, relatively simplified geometries for the target organisms as cylinders and ellipsoids were assumed. The fur and the outer layers of the skin consist of non-active tissue and, therefore, causes a shielding effect for the living organism. However, this effect has only a significant influence on the external exposure for  $\alpha$ -,  $\beta$ - and low-energy  $\gamma$ -emitters.

In order estimate the impact of the distribution of the radiation source, calculations have been made for various distributions of the radioactivity in soil. Planar sources on the top of the soil, at depths of 5 cm and 20 cm, as well as a homogeneous volume source to a depth of 50 cm have been considered. The calculations have been made for monoenergetic  $\gamma$ -energies of 50 keV, 300 keV, 662 keV, 1 MeV and 3 MeV.

As an example, Figure 1 presents the dose conversion factor for a mole which is exposed to a planar  $\gamma$ -source on top of the soil. The DCF increases in proportion to the  $\gamma$ -energy; it decreases with increasing depth of the target due to the increasing shielding effect of the overlying soil layer. The differences in shielding are more pronounced for low energies.

TABLE 2. CHARACTERISTIC OF REFERENCE ORGANISMS FOR THE ESTIMATION OF EXTERNAL EXPOSURES

Targets	Example	Shape	Length, cm	Diameter, cm	Location relative to soil surface, cm	Shielding layer, cm
Soil invertebrate	earthworm	cylinder		0,5	0, -5, -20	0
Small burrowing mammal	mole	ellipsoid	10	5	0, -15, -25, -35	0,1
	mouse			3	0, -10, -25	
Reptile	snake	cylinder	100		0, -25	0
Herbivorous mammal	rabbit		30	12	0	0,1
	roe deer		60	27	40	0,3
	cattle		150	70	50	
Carnivorous mammal	fox	ellipsoid	30	12	30	0,1
	wolf		60	27	20	
Herbivorous bird	pigeon		10	3	300	0,3
Carnivorous bird	hawk		30	12	1000	

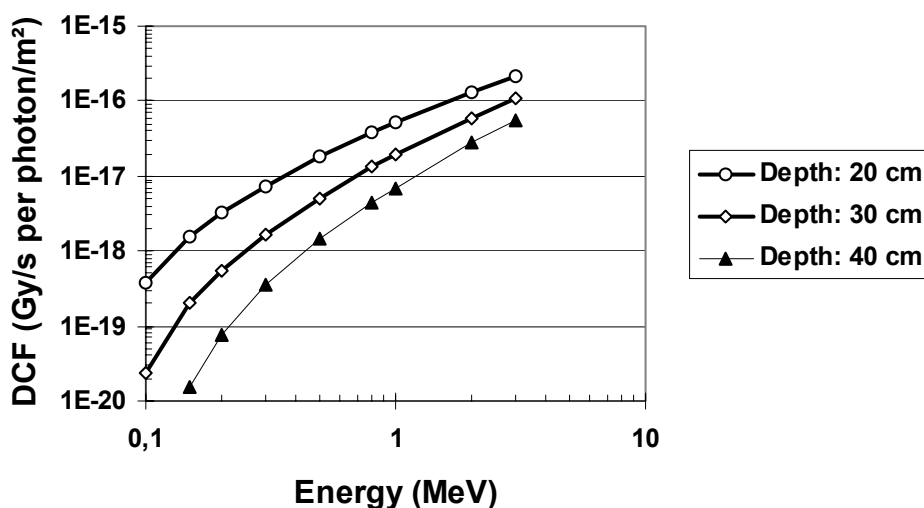


FIG. 1. Comparison of the DCF for a mole as function of the  $\gamma$ -energy and the depth of the target for a homogeneous planar source at the top of the soil.

### 3.2. Internal exposure

For estimating internal exposures to biota, a set of organisms, sizes and energies were defined that allow the assessment of exposures to a wide range of possible species. The most important quantity to assess internal exposures is the fraction of energy absorbed in the organism; this depends on the radiation type, the energy and the size and geometry of the reference organism. As a first step, a homogeneous distribution in the reference organisms will be considered. Additionally, radionuclide accumulations in specific organs, e.g., the thyroid and the gonads, will be simulated. Then, the dose for this specified organ will be calculated. Details on the calculations for internal exposure are given in Table 3.

For the reference plants defined as herbaceous vegetation, shrub and tree, the exposure conditions are specified in Table 4. The exposure will be calculated for the meristem and the buds. These organs are characterised by very intensive cell division, which may cause high radiosensitivity.

For the distribution of the radionuclides in the canopy, a distinction is made between  $\alpha$ -,  $\beta$ -, and  $\gamma$ -radiation due to their different ranges. For  $\gamma$ -radiation the whole canopy is considered to be a homogeneously contaminated source of radiation. For high energy  $\beta$ -radiation, the irradiation of the target is also assumed to occur from a homogeneously contaminated canopy. However, due to the much shorter range of  $\alpha$ - and low energy  $\beta$ -radiation, the irradiation from the external or internal contamination of the target organ has to be considered explicitly. For  $\alpha$ -radiation, due to the very short range of a few centimetres in air, only the exposure from the external or internal contamination of the target has to be taken into account. These assumptions are summarised below (Table 5).

### 3.3. Background exposure

In order to enable a comparison of exposures to biota from radionuclides released from nuclear installations with the background in the specific habitats of the reference organisms, data on the levels of natural radionuclides in different environmental compartment such as marine waters, freshwaters and soils have been collected. Special emphasis is given to the radionuclides  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$ ,  $^{210}\text{Po}$  and  $^{40}\text{K}$ . These data are used to estimate natural background exposures to the biota.

TABLE 3. ENERGY AND GEOMETRY SPECIFICATIONS FOR CALCULATIONS OF INTERNAL EXPOSURES IN ANIMALS

Radiation type	Energy range	Target size range	Geometry
$\gamma$	20 keV – 3 MeV	0.01–1 m	ellipsoids
$\beta$	5 keV – 4 MeV	$10^{-5}$ –0.03 m	ellipsoids
$\alpha$	3–10 MeV	$10^{-5}$ – $10^{-3}$ m	Spheres

TABLE 4. GEOMETRIES FOR CALCULATION OF EXPOSURES IN PLANTS

Plant type	Height	Target organ	Height of plant part considered
Herb	0–0.1 m	Meristem	at the ground (0m)
Shrub	0.1–1 m	Bud, meristem	in middle of canopy (0.55 m)
Tree	1–10 m	Bud, meristem	in middle of canopy (5.5 m)

TABLE 5. RADIATION SOURCES AND CONSIDERED ENDPOINTS FOR CALCULATION OF EXPOSURE OF PLANTS

Radiation type	Source	Endpoint
$\gamma$	homogeneously distributed in the canopy	Average dose rate in the canopy
$\beta$	homogeneously distributed in the canopy, activity on/in the target organ	Average dose rate in the target
$\alpha$	activity on/in the target organ	Average dose rate in the target

#### 4. BIOLOGICAL EFFECTS OF IONISING RADIATION

##### 4.1. Effects and umbrella effects

There is a large number of effects that have been used to describe radiation impact and construct dose-response relationships. Many of the earlier studies have been on the determination of LD 50 values for comparative radiosensitivity purposes, i.e., acute radiation exposures (usually in seconds or minutes) were employed to determine the resulting short term mortality (usually within 30 days). Experimental studies on the effects of low dose rate, chronic radiation exposure, have provided data not only on mortality (frequently a relatively minor effect), but also on fertility, fecundity (or their combination as total reproductive performance), growth rate, somatic and germ cell mutation rates, and so on. Because all the effects that have been observed at the individual level could be presumed to have some possible consequence at the population level, it was decided that FASSET would concentrate on four umbrella effects that have significance at the population level:

- morbidity (including growth rate, effects on the immune system, and the behavioural consequences of damage to the central nervous system from radiation exposure in the developing embryo);
- mortality (including stochastic effect of somatic mutation and its possible consequence of cancer induction, as well as deterministic effects in particular tissues or organs that would change the age-dependent death rate);
- reduced reproductive success (including fertility – the production of functional gametes, and fecundity – the survival of the embryo through development to a reproductive entity separate from its parents);
- cytogenetic effects (i.e. indicator of mutation induction in germ and somatic cells).

It is recognised that these four categories of effect are not mutually exclusive – e.g., effects leading to changes in morbidity may result in a change in the age-dependent death rate, and an increase in mutation rate may lead to changes in reproductive success. They simply provide a convenient means of summarising the available information in a structured way that is meaningful within the objectives of the FASSET project.

In part, the four ‘umbrella’ effects cover the range of relative radiosensitivities within an organism and suggest targets that might be of significance for the purpose of dosimetry:

- the whole body if there is no information on the differential distribution of radionuclides within the organism (this would be relevant for mortality – including stochastic mutation rates in somatic tissues – and morbidity);
- the gonads (fertility and heritable mutations) and the meristems in plants (both for mortality – damage to growth potential – and the gamete bearing tissues);
- externally developing embryos and seeds;
- specific tissues or organs if data are available.

#### **4.2. Radiation effects database**

In order to organise the available information on biological effects in a useful way for the framework, a database is being built; it is aimed at relating dose or dose rate to effects for the specific purpose of FASSET. The collection of data for the database is currently ongoing. The radiation effects database is presented in detail in a separate article in this volume [9].

### **5. FRAMEWORK**

The general structure of existing frameworks for environmental risk assessment has been considered to be appropriate also for FASSET; i.e., a division of the assessment into three stages: problem formulation, risk assessment, and risk management.

The risk management stage lies outside the scope of the FASSET project. The emphasis of FASSET is the development of tools and data for the risk assessment phase of this ecological risk assessment and management process. However, the construction of the FASSET framework must be flexible in order to take into account the various risk management options, as well as societal concern defined in the formulation stage, as these influence (and ultimately must make use of) the way in which a risk assessment is carried out.

The way in which problem formulation is carried out in the different assessment systems studied differ, depending mainly on the different aims and philosophies of the assessments. The FASSET framework must be appropriate for problems of varying formulation, e.g., FASSET must:

- be able to take into account ongoing, past and future releases;
- be able to take into account chronic and acute effects;
- be appropriate for assessments carried out for various purposes, e.g., licensing, demonstration of compliance, assessment of accidents, and decisions concerning remediation.

Some of the elements of other frameworks will be included and appropriately adapted within FASSET, together with a justification for the approach taken. This information will be presented in Deliverable 2 of the project, due by the end of 2002, and will be publicly available at the project website.

Factors to be considered in assembling a framework, and comparisons to other assessment frameworks, are considered in a separate article in this volume [10].

### **6. FOLLOWING FASSET PROGRESS**

#### **6.1. Deliverables**

The project has so far delivered one out of six planned reports. Deliverable 1 consists of one main report and two appendices, and are available at the website:

- Report: Identification of candidate reference organisms from a radiation exposure pathways perspective (Eds: Strand, Beresford, Avila, Jones and Larsson. Contributors: Agüero, Barnett, Brown, Gilek, Howard, Ilus, Kautsky, Kumblad, Näslund, Patton, Robles, Sanchez, Saxén, Stensrud, Suañez, Wright).
- Appendix 1. Ecological characteristics of European terrestrial ecosystems: Overview of radiation exposure pathways relevant to the identification of candidate reference organisms.
- Appendix 2. Ecological characteristics of European aquatic ecosystems: Overview of radiation exposure pathways relevant to the identification of candidate reference organisms.

## **6.2. Communicating FASSET to wider audiences**

The Consortium acknowledged at an early stage the need for the project to be open and transparent, and to allow anyone interested to follow its development. A web-site, [www.fasset.org](http://www.fasset.org), was, therefore, created, and an information leaflet produced to announce the project and to promote its web-site.

The web-site is divided into a public domain and a members' area (password-protected). The organisation of the public domain of the web-site and its material is as follows:

- FASSET leaflet;
- Final version of deliverables;
- List of publications supported by FASSET project;
- Progress reports/Mid term reports;
- Technical annex;
- Databases (e.g., on radiation effects)\*

In order to respond to the interest shown by 'external' stakeholders, and to gain important information, the Consortium organised an External Forum in Bath, UK, 8–9 April 2002. The minutes, including issues raised, recommendations made, and FASSET responses to these recommendations, are available at the FASSET website.

## **6.3. FASSET/BIOMASS workshop**

The International Atomic Energy Agency (IAEA) is concluding its project on Biosphere Modelling and Assessment (BIOMASS). The project has clear relevance to FASSET as regards, for example, problem formulation, but also other aspects resulting from the problem formulation, such as the selection of the biosphere system.

A workshop was held, 30–31 October in Stockholm, to identify which BIOMASS issues are relevant to FASSET, and how FASSET can continue developing concepts within the BIOMASS methodology. The workshop was attended by eight FASSET partners and by IAEA, and also by ANDRA (France) to exchange information between FASSET and the on-going 5th Framework Programme Project BIOCLIM (Modelling Sequential Biosphere Systems under Climate Change for Radioactive Waste Disposal). Again, the minutes are available at the FASSET website.

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\* Members area initially. Public area, when problems concerning database protection/input quality have been resolved.



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# Development of a national environmental monitoring programme for radionuclides – Sweden

P.J. Wallberg, L.M. Hubbard

Swedish Radiation Protection Authority, Stockholm, Sweden

**Abstract.** In parallel with the enforcement of the Environmental Code in 1999, the Swedish parliament adopted 15 national environmental quality objectives that aim towards a sustainable development for the country. The government's primary environmental objective is to hand over a society to the next generation in which the major environmental problems have been solved. One of the quality objectives is "A Safe Radiation Environment" of which the Swedish Radiation Protection Authority (SSI) is the responsible authority. In order to follow the progress towards this objective SSI is currently developing a national environmental monitoring and assessment programme for radionuclides. Many countries have monitoring programmes in the vicinity of nuclear power plants and nuclear industries, as Sweden has also had for many years. The current Swedish effort is a development beyond the local monitoring programmes to incorporate radiation assessment at a national level. This includes long-term issues such as identification of ecological processes that can concentrate radionuclides, and assessment of activities other than nuclear industries that lead to radioactive releases. One of the expected results of this monitoring programme is an improved framework for assessing the dynamics and impact of radionuclide transfer and containment in different ecosystems. This paper will focus on the development and implementation of the framework for a national monitoring programme, include some examples of environments that have been identified as areas of particular concern, and describe an approach to protect species with different ecological prerequisites.

## 1. BACKGROUND

In parallel with the enforcement of the Swedish Environmental Code in 1999, the parliament unanimously adopted 15 environmental quality objectives [1]. These measures were a direct follow-up of the Rio conference in 1992 and aim at achieving a sustainable development for the country. The government's overall goal is to hand over a society to the next generation in which the major environmental problems have been solved. Concrete proposals of interim targets, measures and strategies for how the objectives will be achieved was suggested by the government and subsequently approved by the parliament in 2001 [2].

The Environmental Code and the environmental quality objectives support one another in the task of achieving sustainable development. The Environmental Code is the legal instrument used for achieving the environmental quality objectives. Although the objectives have no formal legal status, they serve as guidelines for public authorities and bodies. Furthermore, the environmental quality objectives can be used as a general basis for assessments in connection with application of the legislation.

## 2. FRAMEWORK FOR THE ENVIRONMENTAL QUALITY OBJECTIVES

The environmental quality objectives define the state of the Swedish environment that the environmental activities should aim to achieve, and visualize the ecological dimension of sustainable development (Table 1). Central agencies are responsible for each of the 15 environmental quality objectives. The quality objective for radiation, A Safe Radiation Environment, concerns both ionizing and non-ionizing radiation, and states:

*"Human health and biological diversity must be protected against the harmful effects of radiation in the external environment".*

TABLE 1. SWEDEN'S 15 ENVIRONMENTAL QUALITY OBJECTIVES

1. Reduced Climate Impact	9. Good-Quality Ground Water
2. Clean Air	10. A Balanced Marine Environment, Flourishing Coastal Areas and Archipelagos
3. Natural Acidification Only	11. Thriving Wetlands
4. A Non-Toxic Environment	12. Sustainable Forests
5. A Protective Ozone Layer	13. A Varied Agricultural Landscape
6. A Safe Radiation Environment	14. A Magnificent Mountain Landscape
7. Zero Eutrophication	15. A Good Building Environment
8. Flourishing Lakes and Streams	

The Swedish Radiation Protection Authority (SSI) is the responsible authority for coordinating the follow-up of this environmental quality objective. Specific for each environmental quality objective, interim targets specify the direction of and the time frame for ongoing concrete environmental measures. There are three interim targets for the objective A Safe Radiation Environment:

- (1) By the year 2010, the concentrations of radioactive substances in the environment emitted from all human activities will be so low that they represent no threat to human health or biological diversity. The annual additional individual dose from each activity to members of the public will be lower than 0.01 mSv per person.
- (2) By the year 2020, the annual incidence of skin cancer caused by UV-radiation should not be greater than that in 2000.
- (3) Studies will continue to be made of the possible risks associated with electromagnetic fields (EMF) and necessary measures will be taken if potential risks are identified.

In order to follow the development within each interim target a number of indicators have been selected, for instance:

- The radiation level in the environment in different regions.
- The yearly radiation dose to individuals from all sources in different geographical regions.
- Attitude studies of the population regarding the risk of UV exposures.
- Grants for research on EMF.

Every year, starting in 2002, the government will present a brief report to the parliament on the progress made towards achievement of the environmental quality objectives. In addition to this, an in-depth evaluation of the progress made will be carried out every four years in order to establish whether the procedures used or the objectives themselves need to be revised.

### 3. A NATIONAL ENVIRONMENTAL MONITORING PROGRAMME

One prerequisite for the follow-up of the first interim target, concerning the concentrations of radioactive substances in the environment, is a national environmental monitoring programme for radionuclides. SSI was given a task to start developing the programme in 2001, which is an effort beyond the monitoring in the vicinity of nuclear installations that Sweden, like many other countries, have had for many years [3]. The national environmental monitoring programme is developing with two long-term aims:

- the total radiation dose to the human population from all sources shall be assessed;
- radiological protection criteria for the environment shall be developed and monitored.

As a first step, the programme is being expanded to monitor and assess the geographical and ecological differences in the radiation environment. The monitoring is carried out within ten programme areas; air, coast and sea, lakes and rivers, wetlands, mountains, forests, agriculture, urban, landscape, and human health. Within these programme areas monitoring will be conducted at different time intervals depending on the purpose of the monitoring (emergency preparedness or environmental monitoring), the source of radiation (natural or anthropogenic sources), the timescales characterizing different ecosystems, or the organisms in question (Table 2). Warning systems, such as gamma or air-filter monitoring stations for accidental releases of radioactive substances, are measured at close time intervals (days or weeks) whilst natural radiation levels are measured only occasionally. The combustion of biofuels from areas that received a high deposition of Cs-137 after the Chernobyl accident is one example of activities that can produce local point sources depending on the handling of the ashes. Monitoring in the vicinity of such point sources can be carried out occasionally by airborne gamma radiometric measurements. In contrast, monitoring in the vicinity of large point sources, i.e., nuclear power plants, is intensive and includes a large number of different bioindicators and different radionuclides that are measured at different time intervals.

Monitoring of organisms and specific ecosystems can be performed as time series, occasionally, as research projects or by using models. For instance, reindeer are monitored annually due to their characteristic of accumulating high levels of Cs-137. An example of an ecosystem where monitoring has been carried out occasionally is an urban area, the city of Gävle, where a high deposition of Cs-137 occurred after the Chernobyl accident. One example of a research project that is closely interconnected with the environmental monitoring programme is presently ongoing in a wetland area in the northeast area of Sweden where considerable enrichment of Cs-137 has occurred [4]. The project aims to clarify when the accumulation of Cs-137 occurred, the processes behind the accumulation and to estimate doses to different organisms in relation to their behaviour and life cycle.

One outcome of this research project will be models for estimation of doses to organisms that cannot be sampled due to their size, behaviour or because they are an endangered species. Another outcome will be knowledge enabling the identification and understanding of other areas where redistribution and concentration of radionuclides can occur.

Monitoring and research go hand in hand. Monitoring defines the state of the environment with respect to radionuclides in terms of concentrations in biota and abiota, time trends and the geographical distribution. Furthermore, monitoring also generates data that can be used to assess doses to biota. However, in order to answer questions of potential effects and to understand the mechanisms and processes behind observed time trends research is needed. Research can result in models for calculating dose and describe processes. Models can also suggest appropriate timescales for monitoring and models for predictions can thereafter be confirmed or rejected by the results of monitoring.

TABLE 2. OVERVIEW OF MONITORING EFFORTS WITHIN THE NATIONAL MONITORING PROGRAMME FOR RADIONUCLIDES

	Time series	Occasionally	Intensive	Research/ Models
Warning systems	X			
Natural radiation		X		
Small point sources		X		
Large point sources			X	
<sup>a</sup> Ecosystems/Organisms	X	X		X

<sup>a</sup> Alternative efforts depending on the ecosystem or organism.

#### 4. CRITERIA FOR BIOINDICATORS

The largest difficulty in designing a new monitoring programme is the selection of bioindicators, because the number of species that can be monitored is by far fewer than the total number of species present in an ecosystem. Thus, bioindicators have to be chosen with care. Generally, several criteria are used for the selection of indicator species.

- (1) **Degree of bioaccumulation and sensitivity.** Indicator species should be exposed to the substance of concern, bioaccumulate the substance and be sensitive to high exposure during parts of or the whole of their life cycle. Differences in the degree of bioaccumulation for different substances must be considered for the selection of indicator species [5]. For a substance that does not bioaccumulate to a high extent, (e.g. Ag-110m), a bioindicator at a lower trophic level should be used, whilst top predators can be used for substances that do bioaccumulate (e.g. Cs-137 and Sr-90).
- (2) **Area of distribution.** If the contaminated area is restricted, the indicator should be stationary, whilst an indicator for a larger contaminated area should have a large home range.
- (3) **Practical restrictions.** The indicator species should be easy to collect, large enough (particularly if individual organs are going to be selected) and abundant to prevent that the monitoring programme causes extinction of a species in an ecosystem. Furthermore, physiology, behaviour and the lifecycle of the indicator species must be well known.
- (4) **Societal relevance and values.** In order to protect a species or an ecosystem the purpose must be understood and accepted by the public and decision makers [6]. Furthermore, the public has concerns to which the authorities must be responsive. For instance, one important aspect of monitoring in the vicinity of nuclear power plants is to provide information to the public about the concentration of radionuclides in foodstuff that generally have very low concentrations. Thus, there are objectives that cannot be met by only using bioindicators with the highest bioaccumulation.

#### 5. PROTECTION OF THE ENVIRONMENT

Operations of activities, such as nuclear industries, are regulated in Sweden so that under normal operation human health and the environment are protected. Today the nuclear industry in Sweden already fulfills the interim target of an individual dose to members of the public of less than 0.01 mSv per person per year, although the interim target is not legally binding. However, there are activities other than nuclear industries that can lead to radioactive releases, such as improper handling of radioactive sources, and the burning of biofuels or peat for energy. In the latter case, dispersion mechanisms and exposure pathways are not thoroughly documented or understood.

The largest concern regarding protection of the environment does not result from regulated activities but from consequences following accidents involving nuclear technology. High concentrations in the environment may occur because of both a primary dispersion of radionuclides as well as a secondary redistribution that can result in accumulation in different ecosystems, such as wetlands and sediments, or as a consequence of human practices. In Sweden the uptake of Cs-137 in trees that in turn are burnt as biofuels and the ashes used as fertilizers in forests or deposited in landfills has recently been identified as a practice that has to be regulated.

It is well known that the greatest radiosensitivity occurs during rapid cell division for either renewal (e.g., spermatogenesis) or growth (e.g., during embryonic and juvenile stages and in plant meristems). It follows that the primary end-point of concern for protection of the environment is reduction in fertility or fecundity [7]. A set of criteria related to the behaviours and lifecycles of animals can be used as a tool to identify the most sensitive animal species in an area with high concentrations of radionuclides (Figure 1). The first criterion in order to identify the most sensitive species is the size of the contaminated area in relation to the home range. The second criterion is the expected time span above an accepted dose limit in relation to the life span of the species. The species that are most likely to get the highest exposure are those for which the contaminated area is larger than their home range and the time span above a given dose limit is longer than their expected life span. The potential effect on the reproduction of the most exposed species is then evaluated based on the total dose over their

lifetime in relation to their behaviour and lifecycle. In the case that the evaluations suggest that the reproductive capacity is significantly repaired, the likelihood of migration of new individuals into the area should be evaluated. If it is likely that new individuals can migrate into the area when the total lifetime dose is below the detrimental level, the need for measures is not immediate. In contrast, if the population is isolated or there are endangered species within the area, measures should be considered.

One possible measure could be to introduce new individuals of the species to the area when the total lifetime dose is below the detrimental level. However, if the species is endangered, it may be necessary to move individuals of the population to uncontaminated areas in order to ensure the possibility that individuals can be retransferred back when the total lifetime dose is below the detrimental level. The given criteria apply for animals but a similar structure to identify the most sensitive plant species can be used. For instance, instead of home range the area of distribution can be used and instead of migration the probability of recolonization by, e.g., spreading of seeds can be considered.

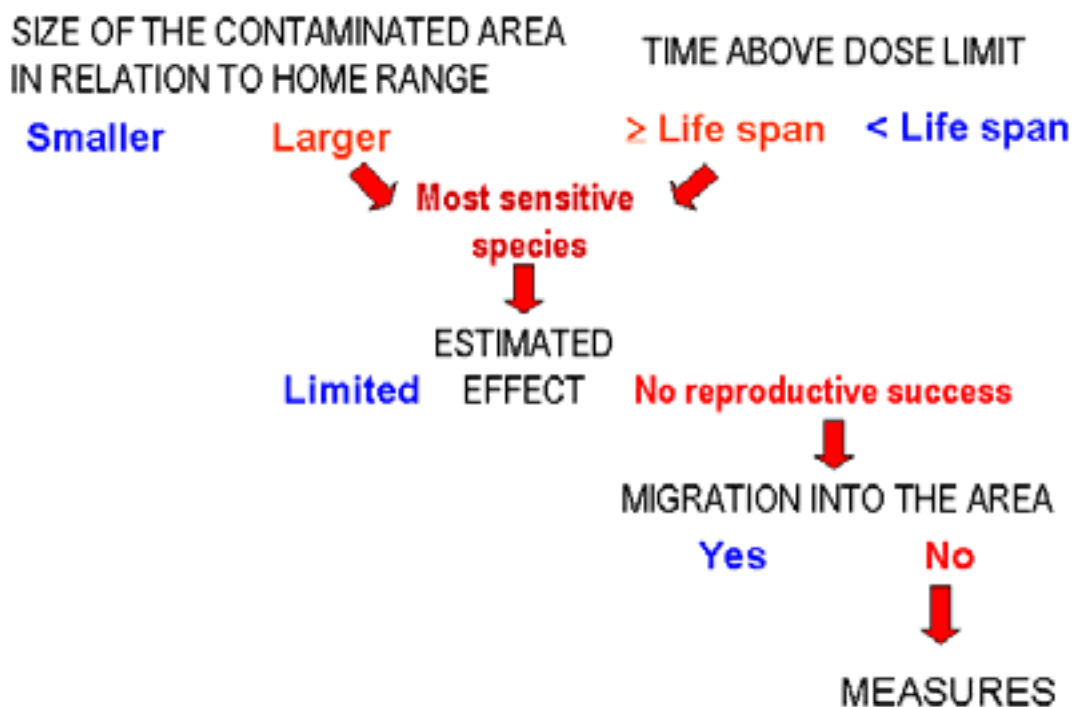


FIG. 1. Criteria to identify the most sensitive animal species in a contaminated area.

## 6. CONCLUSIONS

The fundamental support for the achievement of the environmental quality goals is the fact that the government's proposal was approved unanimously by the parliament. This enables a more long-term perspective since the process is not dependent on the political party in power. At present SSI is working on locating practices that concentrate or spread radionuclides that are not regulated today, and identifying and locating ecosystems that accumulate radioactive substances. In parallel, organisms are being identified that may be exposed to high concentrations or accumulate radionuclides in these ecosystems or in the vicinity of unregulated practices.

SSI has good knowledge on national averages for internal and external doses to humans. One of the most important outcomes of the environmental monitoring programme is to get a better overview of how the radiation dose from different sources varies between regions as well as within regions depending on lifecycle (biota) or lifestyle (humans). However, in the case of biota there is no established methodology to make a weighted estimation of the exposure dose. Furthermore, there are no established dose-effect relationships for non-human species. International work is in progress, for example, within the International Atomic Energy Agency (IAEA) and the International Union of Radioecology (IUR). The outcomes of the EC programme Framework for Assessment of Environmental Impact (FASSET) can also be expected to provide a basis to establish national dose limits for protection of the environment [8].

Due to the limited measures that can be done when high concentrations of radionuclides as well as other pollutants are released into the environment, activities should be restricted so that under normal operation both human health and the environment are protected. Generally, in case of an accident we can only hope that the environment will recover by itself, for instance through long-term burial of contaminants in sediments. One crucial factor for the survival of a population with severely impaired reproduction is migration (animals) or recolonization (plants) of new individuals. Thus, criterion for protection of the environment should be set to indicate when measures are needed to sustain isolated populations in case of an accidental release of radionuclides.

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# The U.S. Department of Energy's graded approach for evaluating radiation doses to aquatic and terrestrial biota

S.L. Domotor<sup>a</sup>, A. Wallo III<sup>a</sup>, H.T. Peterson, Jr.<sup>a</sup>, K.A. Higley<sup>b</sup>

<sup>a</sup> United States Department of Energy, Office of Environmental Policy and Guidance, Washington, DC, United States of America

<sup>b</sup> Oregon State University, Nuclear Engineering and Radiation Health Physics, Corvallis, OR, United States of America

**Abstract.** The United States Department of Energy (DOE) currently has in place a radiation dose limit for the protection of aquatic organisms, and has considered dose limits for terrestrial biota. These limits are: 10 mGy/d for aquatic animals, 10 mGy/d for terrestrial plants, and 1 mGy/d for terrestrial animals. Guidance on suitable approaches to implementation of these and other proposed limits for protection of biota is needed. In response to this need, we have developed methods, models and guidance within a *graded approach* for evaluating radiation doses to biota. DOE's multi-tiered process is described in a technical standard document. Methods are encoded in a series of electronic spreadsheets termed the RAD-BCG Calculator to assist the user in progressing through the evaluation process. A key component of the graded approach is a screening methodology that provides limiting concentrations of radionuclides, termed Biota Concentration Guides (BCGs), for use in screening water, sediment, and soil media to determine if dose limits for biota are likely to be exceeded. The graded approach provides flexibility and the ability to iterate through the evaluation process. User-selected biota dose limits can be entered in place of default dose limits. Parameter values, radiation weighting factors, and organism residence times can be modified within site-specific screening and site-specific analysis phases of the graded approach. The methodology, available since 2000, was developed using an interdisciplinary team approach that included both "developers" and "users" through the Department's Biota Dose Assessment Committee (BDAC). DOE's graded approach framework provides a practical and effective tool for demonstrating protection of biota relative to Dose Rate Guidelines, and for conducting ecological screening assessments of radiological impact. It provides a needed evaluation tool that can be employed within an international framework for protection of the environment.

## 1. INTRODUCTION

There is growing national and international interest concerning the explicit protection of the environment (biota and ecosystems) from the effects of ionizing radiation. The use of human radiation protection criteria to infer ecological protection from the effects of ionizing radiation is being revisited and scientifically challenged. Increasing regulator and stakeholder interest in demonstrating protection of biota from the effects of ionizing radiation will need to be considered in actions regarding the remediation, decontamination and decommissioning (D&D), and long-term maintenance, surveillance, and monitoring of contaminated sites.

The United States Department of Energy (DOE) has been active in the area of requirements and guidance for radiological protection of the environment since the late 1980's. DOE currently has in place a dose limit of 10 mGy/d for native aquatic organisms [1], and has proposed dose limits for terrestrial plants (10 mGy/d) and animals (1 mGy/d) [2]. These dose limits represent expected safe levels of exposure, and are consensus No Adverse Effects Levels (NOAELS) for effects on population-relevant attributes (i.e., using reproduction as the critical endpoint of concern) in natural populations of biota [3–5]. Recommendations from the public comment process on DOE's proposed biota dose limits highlighted the need for standardized evaluation approaches for demonstrating compliance, cost-effective methods that employ screening concepts, and flexibility to apply site-specific information. In the U.S. and internationally, no standardized methods have been adopted for evaluating radiation doses and demonstrating protection of plants and animals from potential radiation effects. In response to this need, DOE has developed methods, models and guidance within a *graded approach* for evaluating radiation doses to biota. An objective of this initiative was to advance the inclusion of biota dose evaluation as a routine part of site radiological and environmental surveillance programs, and the inclusion of biota dose evaluation results in site annual environmental reports.



## 2. DOE'S GRADED APPROACH

### 2.1. A multi-tiered framework

The graded approach was developed using an interdisciplinary team approach through a DOE-sponsored Biota Dose Assessment Committee (BDAC). The BDAC, formed in 1998, has broad representation from DOE sites and facilities, national laboratories, universities, and the private sector. A guiding principle for the BDAC was that both model "developers" and "users" be part of the methods development process.

DOE's graded approach to biota dose evaluation consists of a three-tiered process which is designed to guide a user from an initial, prudently conservative general screening phase to, if needed, a more rigorous analysis using site- and receptor-specific information (Figure 1). The three-tiered process includes: (1) a data assembly phase in which the evaluation area and its characteristics are defined, and radionuclide concentration data for water, sediment and soil are assembled for subsequent screening; (2) an easy-to-use general screening methodology that provides limiting radionuclide concentrations (termed Biota Concentration Guides, BCGs) in soil, sediment, and water such that the dose limits for protection of biota are not exceeded; and (3) an analysis phase containing three increasingly more detailed steps comprised of site-specific screening, site-specific analysis, and site-specific biota dose assessment (Table 1). Any of the three phases of the graded approach may be used at any time, but the general screening tool will usually be the simplest, most cost-effective, and least time consuming.

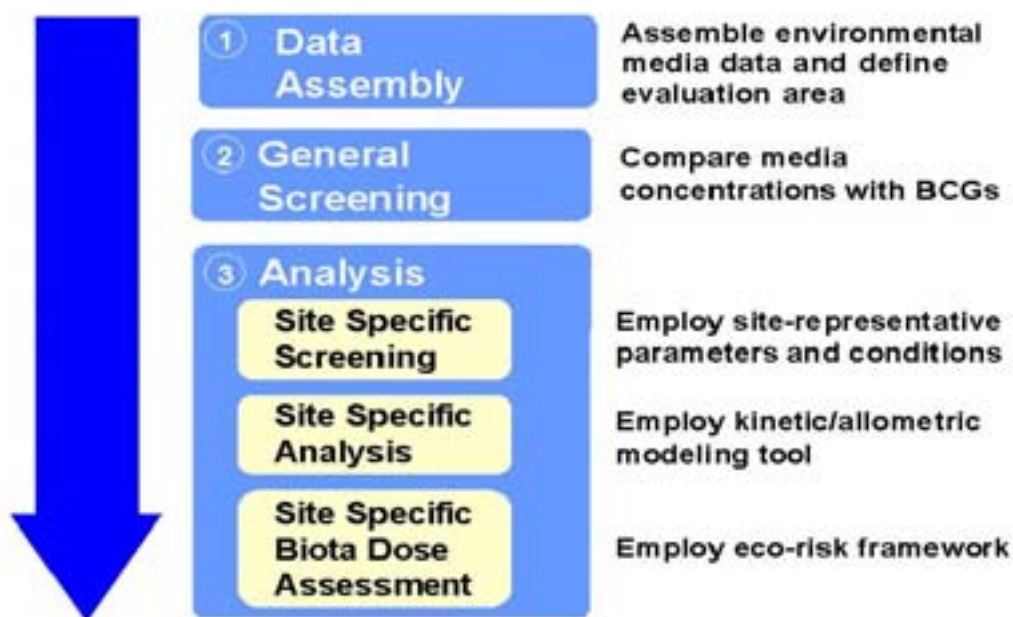


FIG. 1. DOE's Graded Approach for Evaluating Radiation Doses to Biota.

TABLE 1. SUMMARY OF DOE'S THREE-TIERED PROCESS EMPLOYED IN THE GRADED APPROACH

ASSEMBLY	General knowledge of sources, receptors, and routes of exposure for the area to be evaluated is summarized. Measured radionuclide concentrations in water, sediment, and soil are assembled for subsequent screening.
GENERAL SCREENING	Maximum measured radionuclide concentrations in an environmental medium (i.e., water, sediment, soil) are compared with a set of Biota Concentration Guides (BCGs). Each radionuclide-specific BCG represents the limiting radionuclide concentration in an environmental medium which would not result in the specified biota dose limits to be exceeded.
ANALYSIS	This phase consists of three increasingly more detailed steps of analysis:
<i>Site-Specific Screening</i>	Site-specific screening, using more realistic site-representative lumped parameters (e.g., concentration factors) in place of conservative default parameters. Use of mean radionuclide concentrations in place of maximum values, taking into account time dependence and spatial extent of contamination, may be considered.
<i>Site-Specific Analysis</i>	Site-specific analysis employing a kinetic modeling tool (applicable to riparian and terrestrial animal organisms) provided as part of the graded approach methodology. Parameters representing contributions to the organism's internal dose (e.g., body mass, food consumption rate, inhalation rate, lifespan, biological elimination rate) can be modified to represent site- and organism-specific characteristics. The kinetic model employs allometric equations relating body mass to these internal dose parameters. Correction factors for the fraction of time contamination is present in an evaluation area, and for the fraction of time an organism resides in the contaminated area, can be applied to all organism types.
<i>Site-Specific Biota Dose Assessment</i>	An actual site-specific biota dose assessment involving the collection and radiological analysis of biota samples, using ecological risk assessment protocols.

## 2.2. Derivation of Biota Concentration Guides (BCGs)

The technical basis for the general screening methodology within the graded approach is based on the fact that biota dose is a function of the contaminant concentration in the environment, and is the sum of internal and external contributions of dose to the organism. It is possible, given a unit concentration (i.e., 1 Bq kg<sup>-1</sup>) of a contaminant in a single medium (e.g., soil), to estimate the potential dose rate to an organism from both internal and external exposures. Once the dose rate from this unit concentration of contaminant has been calculated, it can be used to back calculate a concentration of the contaminant that will generate a dose rate at *any* specified limit, such as DOE's existing and proposed biota dose limits. This radionuclide- and media-specific limiting concentration is termed a Biota Concentration Guide (BCG). When multiple radionuclides are present in multiple environmental media, the sum of fractions rule is applied to account for all sources of exposure. The derivation of the BCGs and their default assumptions and parameters is discussed in detail elsewhere [6–9]. Key elements of the technical approach are highlighted below.

- Four reference organism types (aquatic animals, riparian animals, terrestrial plants, terrestrial animals) were selected as the basis for methods development. Internal and external sources of dose (and their contributing exposure pathways) are incorporated in the derivation of the graded approach methodology. Sufficient prudence was exercised in the development of each of the assumptions and default parameter values to ensure that the resulting BCGs are appropriately conservative for screening purposes.
- The source medium to which the organisms are continuously exposed is assumed to contain uniform time-invariant concentrations of radionuclides.
- Internal doses were calculated as the product of media concentration, concentration factor(s), and dose conversion factors. Kinetic and allometric techniques were used to fill data gaps in predicting radionuclide concentration factors across a large range of terrestrial and riparian species of animals. Estimates of the contribution to dose from internal sources of radioactive

material were conservatively made assuming that all of the decay energy is retained in the tissue of a very large organism (i.e., 100% absorption), and to include progeny of chain-decaying radionuclides. External doses were calculated based on the assumption of immersion of the organism in soil, sediment, or water.

- Estimates of the contribution to dose from external sources of radioactive material were made assuming that all of the ionizing radiation was deposited in the organism (i.e., no pass-through and no self-shielding). This is conservative, and is tantamount to assuming that the radiosensitive tissues of concern (the reproductive tissues) lie on the surface of a very small organism.

### **2.3. DOE technical standard and RAD-BCG Calculator**

The technical standard document and the RAD-BCG Calculator are discussed below. Both products can be downloaded from the BDAC web site (<http://homer.ornl.gov/oepa/public/bdac>).

#### *2.3.1. DOE technical standard*

The graded approach to biota dose evaluation and associated guidance for its implementation is documented in a DOE technical standard [9]. The technical standard is designed to be user-friendly and applications-oriented. It is organized into three principal modules for ease of use.

- Module 1, “Principles and Applications” – Provides user-friendly guidance and instructions to include: (1) an overview of the evaluation process; (2) application considerations; (3) look-up tables containing screening values; (4) step-by-step implementation guidance; and (5) examples using actual site data and scenarios.
- Module 2, “Detailed Guidance” – Provides specific guidance by topic to support implementation of the graded approach, to include: (1) a primer on radiological ecological risk assessment; (2) defining sources, receptors, and routes of exposure; (3) spatial and temporal averaging of contaminants and doses; (4) methods for representative sampling of soil and biota; (5) selection of a radiation weighting factor for alpha particles; and (6) evaluating doses to individual organisms.
- Module 3, “Methods Derivation” – Provides the technical basis used to derive the BCGs, to include: (1) assumptions and conceptual models used to derive BCGs for each reference organism type; (2) internal and external dose conversion factors; (3) all equations and models; (4) the kinetic-allometric approach used to derive organism bioconcentration factors where limited empirical data was available; and (5) all default parameters and their sources.

#### *2.3.2. RAD-BCG Calculator*

The “RAD-BCG Calculator” provides a set of user-friendly electronic spreadsheets for performing a semi-automated evaluation of biota dose employing the methods contained in the technical standard. The RAD-BCG Calculator provides a wide range of user flexibility, including the ability to: (1) apply a user- or regulatory agency-specified biota dose limit in place of DOE’s limits; (2) modify the radiation weighting factor for alpha emitters; (3) de-select inclusion of progeny in the calculation of internal dose factors; (4) use site- and receptor-specific environmental transfer factor parameters (e.g., sediment and soil distribution coefficients; bioconcentration factors) in place of default values; and (5) apply correction factors for the fraction of time contamination is present in an evaluation area, and for the fraction of time an organism resides in the contaminated area.

### **2.4. Progress resulting from an initial trial use period**

The graded approach was made available to DOE field and program elements for a trial use period beginning in July 2000 through an interim technical standard document. The graded approach methodology also received interest from other national and international organizations during this period. An independent external technical peer review of the methodology and associated guidance contained in the technical standard was also performed and several papers on the graded approach were submitted and accepted for publication in peer-reviewed journals [6–8, 10, 11].

Comments and suggestions resulting from this trial use period and the technical peer reviews were used to refine and make improvements to the methods and guidance. For example: (1) several radionuclide-specific BCGs were re-evaluated and modified; (2) guidance on the relationship between the graded approach and the ecological risk assessment framework (ERA) typically used for the evaluation of chemical stressors was added, along with guidance on specific technical issues inherent in evaluating radiation that are different from those encountered when evaluating chemical stressors to the environment; and (3) guidance was added regarding how to utilize the graded approach in support of other types of environmental assessments.

There was a noticeable increase (e.g., from 10% in 2000, to 50% in 2002) in the evaluation of doses to biota as reported in site annual environmental reports received by DOE's Office of Environmental Policy and Guidance. This steady increase is attributable to the awareness, availability and use of the graded approach to biota dose evaluation at DOE sites, and the standardized but flexible screening and analysis methods contained within the graded approach framework.

### 3. NATIONAL COORDINATION AND PARTNERSHIPS

The Department of Energy is taking a proactive national approach that is providing opportunities for methods acceptance, testing, and improvement. Two examples of projects and partnerships at a national level that are employing the graded approach methodology are highlighted below.

#### 3.1. RESRAD-BIOTA dose evaluation code

Since 2001 DOE has been working in partnership with offices of the U.S. Environmental Protection Agency (EPA) and the U.S. Nuclear Regulatory Commission (NRC) to develop a "next generation" evaluation tool, "RESRAD-BIOTA." The general screening methodology and successive analysis tiers of the graded approach methodology are being used as the starting point for this new code. Additional analysis capabilities above and beyond the current methodology are being incorporated into the code to be responsive to the broader evaluation needs anticipated by each agency. Examples of additional capabilities identified for development or already provided include: (1) ability to apply dose conversion factors based on geometries for a variety of finite-sized organisms; (2) uncertainty and sensitivity analysis capabilities for biota dose estimates and associated parameters; (3) improved tabular and graphical presentation of results; and (4) capabilities to link and import environmental radionuclide concentration data generated by other radionuclide transport codes for subsequent use in evaluating doses to biota within RESRAD-BIOTA. Refer to the paper by Yu et. al. in this Symposium Proceedings for additional details on RESRAD-BIOTA code development.

#### 3.2. Long-term stewardship pilot project

Long-term stewardship program and planning documents at the national and local level indicate that protection of natural resources is integral to a successful long-term stewardship program (i.e., activities to protect human health and the environment from contamination that may remain at sites following site cleanup). A pilot project is underway at the Oak Ridge Site (Oak Ridge, TN, USA) to validate and test the application of the graded approach methodology as a practical and cost-effective environmental surveillance tool for assessing potential impacts to biota for long-term stewardship site applications.

### 4. INTERNATIONAL CO-ORDINATION

The Department of Energy continues to cooperate in discussions regarding concepts for the development of an international framework for protection of the environment from the effects of ionizing radiation. This has included participation in Specialists' Meetings held in 2001 and 2002 by the International Atomic Energy Agency (IAEA), participation as a corresponding member to the International Commission on Radiological Protection's (ICRP) current "Task Group on Protection of the Environment," and participation in the Nuclear Energy Agency's "Forum on Radiological Protection of the Environment: The Path Forward to a New Policy?" (2002). DOE is also a contributing sponsor of the "Third International Symposium on the Protection of the Environment from Ionizing Radiation" (2002).

The DOE's graded approach methodology has the potential for application within an international framework for radiological protection of the environment, as indicated in the following summary statement included in an IAEA Specialists' Meeting Summary Report [12]: "A variety of models continue to be developed along these lines. The U.S. Department of Energy has developed a generic reference organism screening model (contained in their graded approach methodology) and generic/reference organism models are being developed as part of the FASSET programme. It was agreed that these approaches are, more or less, complementary and that they could provide the basis for and agreed methodology within an international framework." DOE will continue to work with international organizations to: (1) harmonize the different biota dose evaluation approaches in use or under development within individual countries and organizations; and (2) foster the inclusion of practical approaches for evaluating doses to biota in concepts for an international protection framework on this topic.

## 5. SUMMARY AND RECOMMENDATIONS

The Department of Energy's development and implementation of the graded approach framework and our participation in international discussions regarding the need for a radiological protection framework for the environment has resulted in the following lessons learned and recommendations.

- The availability of the graded approach to biota dose evaluation is effecting change within DOE. Sites are increasingly assessing potential radiological impacts to biota as a result of the availability of the graded approach methodology. This is in large part because of the design of the graded approach framework. The framework provides users with "a place to start" and "an analysis path forward" where needed. The BCGs are not stand-alone. Exceedance of the BCGs leads the user to the more-detailed tiers of analysis as needed in a stepwise manner. These linkages are an integral part of the graded approach framework and are "built-in" to the RAD-BCG Calculator.
- DOE's existing and recommended biota dose limits are not intended to be a "bright line" that, if exceeded, would trigger a mandatory regulatory or remedial action. Rather, they are applied by DOE more as "Dose Rate Guidelines" that provide an indication that populations of plants and animals could be impacted from ionizing radiation and that further investigation and action is likely necessary. We recommend that the term "Dose Rate Guidelines" be carried forward when establishing national or international dose rates corresponding to effects endpoints.
- Screening is vital as a compliance tool, and as a first step in ecological risk assessments of radiological impact. Screening uses conservative, simple models. It provides a practical approach that is usually cost- and time-effective. As such, it should be part of a multi-tiered process for evaluating the impacts of radiation within any proposed international framework.
- "Standardized" approaches have benefits, but users must have the ability to select alternative approaches. One method or model may not fit all users and application scenarios.
- There are numerous ancillary issues that need to be considered and for which guidance needs to be provided. Topics for additional guidance would include: (1) space and time averaging of contaminant concentrations and dose rates; (2) dealing with high levels of natural background; (3) addressing data gaps in environmental transfer factor parameters necessary for calculation of realistic doses to biota; and (4) how to proceed if biota dose limits or effects levels are exceeded. DOE's technical standard document provides guidance on many of these implementation issues.
- Future international efforts should first concentrate on high-level "umbrella" policy and guidance that: (1) clarifies and re-affirms as appropriate the current ICRP assumptions concerning those exposure scenarios where "if man is protected, then biota is also sufficiently protected"; (2) provides additional policy if there are exposure scenarios where explicit evaluations to demonstrate that biota are protected may be warranted; and (3) provides recommendations on acceptable effects and assessment endpoints for protection of biota.

- Further consideration should be given to the need for a flexible and performance-based evaluation framework that would allow users to work with existing evaluation methods and models, and dose effects data matched to the purpose and data quality objectives of their assessment. There are many biota dose evaluation methods and models that already exist or are under development in the U.S. and internationally that could be directly applied in such a performance-based framework.
- The ecological risk assessment framework typically applied in the evaluation of chemical stressors is general in nature and could serve as a performance-based framework (i.e., allowing for the use of different dose evaluation methods) for evaluating radiation as a stressor to the environment. The DOE graded approach is consistent with the ecological risk assessment framework, but with a particular emphasis on ionizing radiation.
- New international guidelines and approaches must offer practical means of implementation if they are to be widely adopted within the regulatory community.

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## **Expectations for the protection of the environment: *Greenpeace perspectives\****

**S. Carroll**

Political and Science Department, Greenpeace International, Amsterdam,  
The Netherlands

**Abstract.** In seeking to develop and apply a system of radiation protection of the environment, there is a need for the provision of high-quality scientific information. It is essential also to take into account information provided through other disciplines and the experience gained through study of non-radioactive environmental stressors. The policy development and management approaches developed in response to other environmental contaminants have a significant role to play in this process.

Greenpeace advocates use of a precautionary approach for protection of the environment centred upon the following core principles:

- damage should be prevented;
- decisions should be informed by scientific information, whilst recognising the limitations and uncertainties in the available knowledge; and
- all activities should meet the requirement for progressive reduction of the **presence** of environmental stressors.

In developing a policy and in determining a management approach to environmental releases of radioactive materials, it is suggested that it is essential to consider, *inter alia*, that:

- the effects of continued releases of radioactive materials into the environment are unpredictable in the long term, particularly where the physical or chemical form of the radioactive material may make it liable to accumulate and where the radioactivity may be long-lived;
- any accumulation of persistent presence in the environment may be practically difficult to reverse; and
- environmental/ecosystem effects may be undetectable at an early stage which could mean that later intervention to reduce the overall impact may be “too little, too late”.

To meet these and other concerns, the prime management objective should be the cessation of environmental releases of radioactive materials in order to reduce their levels in the environment to the lowest level practically possible given historic releases. In this context, the assessment of risk to non-human species fulfils the purpose of aiding in the determination of how the objective can be achieved and in identifying specifically the various sources, routes and pathways to the environment. In the management phase, the risk assessment will facilitate and inform the decisions on which programmes and measures will be most effective and which should be prioritised and, in addition, provide useful inputs to decisions on remedial measures for addressing historic contamination.

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\* Only an abstract is given as the full paper was not available.

# Uranium mining in Australia: Environmental impact, radiation releases and rehabilitation

G.M. Mudd

Environment Centre of the Northern Territory Inc. (ECNT) (on behalf of), Jabiru, NT, Australia

**Abstract.** The mining and export of uranium and the impacts (and risks) of the nuclear industry have long been a contentious issue in Australia. The ongoing debate primarily relates to the established and potential dangers of ionizing radiation released to the environment from nuclear facilities, such as uranium mines or research reactors. By 2002, three uranium milling projects are operating with a further seven having been operated in the past 50 years, including numerous smaller mines. A critical aspect of the operation of a mine and/or mill is the ionizing radiation rates before, during and after a project has ceased. A number of estimates have previously been made of the environmental releases from Australian projects, such as environmental impact statements and the UNSCEAR 1993 data (although there is some controversy as to the quality of the data and assumptions used in this analysis). In order to assess the environmental impacts of ionizing radiation releases from nuclear facilities, such as radon gas or soil and water quality, it is necessary to compile and quantify these changes based on measured data from the various sites around Australia. This paper presents a brief review, based on more comprehensive studies in progress, of the changes in ionizing radiation rates, radionuclide releases and ongoing issues from some operational and former uranium projects in Australia, allowing a more accurate assessment of the measures required for protection of the environment from potentially harmful situations. The importance of detailed field radiation measurements before, during and after rehabilitation is stressed, followed by a discussion of Non-Government Organisation (NGO) views of the implications for uranium mining and milling.

## 1. URANIUM MINING AND MILLING IN AUSTRALIA

The history of uranium mining and milling in Australia spans the 20<sup>th</sup> century, beginning with radium mining in the early years and expanding to large scale uranium projects over the last 50 years (Figure 1).

The first uranium deposits in Australia were discovered at Radium Hill and Mt Painter in north-eastern South Australia in 1906 and 1910, respectively. Between 1906 to 1932 intermittent mining and milling occurred to extract radium with uranium as a by-product, based on mining of ~3,200 t of ore (0.2–20% U<sub>3</sub>O<sub>8</sub>) giving ~1.8 g of radium and up to 7 t U<sub>3</sub>O<sub>8</sub> [1]. The various sites were abandoned by 1932, including the radium refineries at Hunters Hill in Sydney, NSW, and at Dry Creek in Adelaide, SA.

A new phase of uranium exploration was begun for the Manhattan Project over 1944–45 (World War II), with extensive exploration undertaken by governments, prospectors and mining companies following the war with a view to securing uranium for nuclear weapons and reactor programs. By the late 1950s, there were six uranium mills operating in the Northern Territory, South Australia and Queensland, supported by numerous smaller uranium mines. This phase ended with the closure of Rum Jungle in 1971 following the total production of ~2,500 t U<sub>3</sub>O<sub>8</sub> for nuclear weapons programs of the USA and UK, 4,800 t U<sub>3</sub>O<sub>8</sub> for the UK's nuclear reactor program, plus a national stockpile of ~2,100 t U<sub>3</sub>O<sub>8</sub> [1]. The environmental management of these sites was generally poor or minimal.

The late 1960s saw the eventual emergence of nuclear reactors on a commercial scale and a rapid increase in the intensity of uranium exploration across Australia. The success was virtually instant and by the early 1970s new uranium provinces had been identified in the Alligator Rivers Region of the NT, central Western Australia as well as other uranium deposits of varying significance.

The 1970s coincided with increasing public knowledge and debate about the impacts of the nuclear industry, centred around nuclear weapons, reactor safety, intractable nuclear waste and the dangers of ionizing radiation. Further concerns included indigenous land rights and environmental conservation.





FIG. 1. Location of uranium mining and milling sites (and deposits) in Australia [1].

The Ranger Uranium Environmental Inquiry was instituted in July 1975 to investigate potential Australian involvement in the nuclear industry, principally through mining and export of Australia's uranium deposits. The inquiry presented its first report in October 1976 on the nuclear industry and its second report in May 1977 on uranium mining, land rights and national park issues in the Alligator Rivers Region [2]. The two reports essentially urged caution on all sides while arguing that the potential impacts of ionizing radiation releases from the nuclear industry, especially uranium mining, were within acceptable levels compared to background radiation. The second report also supported indigenous land rights and the creation of a large national park to be called Kakadu, with the Ranger, Jabiluka and Koongarra uranium projects deliberately excised but surrounded by Kakadu. For the Ranger project, a number of important recommendations were made with a view to minimising the environmental releases and potentially harmful impacts of radionuclides and heavy metals.

Between the adoption of most of the Ranger Inquiry recommendations in the late 1970s and the present, there have been three uranium projects at Ranger, Nabarlek (now closed) and Olympic Dam, with the Beverley acid leach mine beginning operation in late 2000. The Mary Kathleen uranium project was re-opened for six years (1976–82) plus numerous trial uranium mines and/or mills were also attempted. A thorough compilation of project data to March 2002 is given in Table 1.

There has been no comprehensive scientific analysis of radionuclide releases from uranium projects in Australia since the Ranger Inquiry. This is now more pertinent than ever, given the data now available from former, current and potential projects and the continued push for increased nuclear power in the future. This paper will present a brief analysis, based on more comprehensive studies [1, 7], of the radionuclide releases from Australian uranium projects, followed by a discussion of the implications for the long-term radiological impacts of uranium mining and rehabilitation requirements.

TABLE 1. URANIUM MINING AND MILLING DATA IN AUSTRALIA TO 31 MARCH 2002 [1]

		t Ore Milled	%U <sub>3</sub> O <sub>8</sub>	t U <sub>3</sub> O <sub>8</sub>	t LGO & WR
Olympic Dam, SA	1988–	51,793,182	0.081%	27,060.2	~6,030,790
Ranger, NT	1981–	22,906,600	0.319%	65,467.6	>88,678,000
Nabarlek, NT <sup>(M)</sup>	1980–88	597,957	1.84%		
Nabarlek, NT <sup>(HL)</sup>	1985–88	157,000	~0.05%	10,955	2,330,000
Beverley, SA	2000–	ISL †	–	578	–
Honeymoon, SA	2003(?)–	ISL †	–	~ 60 <sup>P</sup>	–
Mary Kathleen, QLD	1976–82	6,200,000	0.10%	4,801	17,571,000
Trial Mines	1978–	various		>> 12	>> 150,000
Moline, NT	1956–64	135,444	0.46%	716.0	??
Rockhole, NT	1959–62	13,155	1.11%	139.7	??
Mary Kathleen, QLD	1958–63	2,710,483	0.156%	4,091.76	4,429,764
Radium Hill, SA	1954–61	817,000	~0.005%		
Port Pirie, SA	1955–62	152,300 <sup>C</sup>	~0.8	852.3	??
Rum Jungle, NT	1954–71	1,496,641	0.35%	3,530	14,283,000
Trial Mines <sup>RJ</sup>	1953–62	9,224.9 <sup>RJ</sup>	0.92%	– <sup>RJ</sup>	??
Radium Hill, SA	1906–31	~2,130 t	1.4% ??	<7	??
Mt Painter, SA	1910–32	~933 t	~2.1%	??	??
Total		86,980,025 t	0.168%	118,264 t	>133,472,000 t

<sup>P</sup> Pilot scale mining/milling; <sup>RJ</sup> Milled at Rum Jungle (not included in sub-totals); † In situ leach; <sup>M/HL</sup> Mill (M) or heap leach (HL); <sup>C</sup> Radium Hill concentrate; LGO – Low grade ore; WR – Waste rock.

## 2. URANIUM MINING AND MILLING AND ENVIRONMENTAL RADIOACTIVITY

A brief summary of the environmental radioactivity issues with regards to uranium mining and milling is required before analysing Australian projects. An important aspect of this is the natural or 'background' ionizing radiation that exists prior to any development works on a site.

### 2.1. Environmental radioactivity

Uranium consists of two principal decay chains, <sup>238</sup>U (99.3%) and <sup>235</sup>U (0.7%), each with their own radioactive decay sequence and half-lives (a minor amount of <sup>234</sup>U is in the <sup>238</sup>U chain). The various elements, such as thorium (<sup>230/234</sup>Th), radium (<sup>226</sup>Ra) and radon (<sup>222</sup>Rn), have varying physical and chemical properties important in their environmental behaviour. For example, <sup>230</sup>Th is insoluble while <sup>226</sup>Ra is moderately soluble, compared to <sup>222</sup>Rn which is a noble gas. The many isotopes also decay differently through alpha or beta decay, with most isotopes releasing significant gamma radiation.

The principal radionuclide sources from uranium mining and milling are waste rock, low grade ore and tailings. The radon flux or gamma dose rate from a particular waste will be primarily determined by its uranium content (or specifically radium activity, plus moisture for radon flux). The transport of radionuclides in surface water and groundwater is an important source of environmental radioactivity and is a pivotal issue in water management at, and potential releases from, uranium mines and mills.

### 2.2. Background ionizing radiation

The environment has a general level of natural or 'background' ionizing radiation from the decay of U, Th or other radioactive isotopes. In Australia, background ionizing radiation is typically within global norms and primarily consists of cosmogenic and terrestrial sources (mostly gamma and some radon) [3]. The average <sup>222</sup>Rn flux from Australian soils is about 25 ± 5 mBq/m<sup>2</sup>/s [4], similar to the global average of 15 to 23 mBq/m<sup>2</sup>/s [5]. A typical gamma dose rate for Australia is about 0.02–0.1 µGy/hr [1]. The concentration of radionuclides such as uranium, radium and radon is generally low in surface waters, with subtle variation due to geological sources within a catchment area. The situation is similar for groundwater, again related to radionuclide content in the local geological formation.

One of the principal concerns with uranium mining, excluding broader concerns about weapons, reactors and wastes, are that it could lead to increased radionuclide releases into the environment (plus the potential for accidents), altering the generally low background levels prior to mining. Whether projects are legally surrounded by World-Heritage listed Kakadu National Park or poorly managed arid lands, the environment movement opposes, on principle, any rehabilitation standards which allow permanent increases to ionizing radiation rates or radionuclide loads in the environment.

### 3. RADON FLUXES AND LOADS

The release of radon ( $^{222}\text{Rn}$ ) gas and its decay products is a critical part of assessing ionizing radiation doses for uranium workers and the general public, though it would appear that much less is understood about the environmental behaviour of the products and their cycling through the environment. Through the mining of waste rock and ore and the creation of finely ground tailings, the physical (and chemical) nature of the dominant radon sources is considerably altered after mining compared with the geology beforehand. At some sites, it may be possible that mining and rehabilitation may indeed decrease the radon flux and load after rehabilitation while at others the data is less convincing.

The UNSCEAR 1993 report [6] uses limited operational data or optimistic company estimates of ideally rehabilitated tailings sites at the Olympic Dam, Ranger and Nabarlek projects. Their approach is based on tailings being the principal source of radon, and to a lesser the mill. This is clearly limited since waste rock, low grade ore and  $^{226}\text{Ra}$ -contaminated areas can also be major sources. A compilation of radon fluxes and loads from uranium project sites in Australia are presented in Tables 2 and 3, with more detailed data and estimates presented for the Ranger project in Tables 4 and 5.

As can be seen from the various sites there is high variability in both the radon fluxes from different sources as well as predicted loads. For example, the predicted radon load from various configurations of Ranger tailings management have varied from  $<0.37$  to  $4,440$  GBq/day [7]. Before rehabilitation, Nabarlek was predicted to have a radon flux some  $10^{22}$  lower than pre-mining values (due to the thick layer of waste rock above the tailings), although as the data in Table 2 shows, the post-rehabilitation radon flux is less than 100 (or  $10^2$ ) times lower [1]. The UNSCEAR radon data for Nabarlek, apparently unrehabilitated, is an overestimate of actual post-rehabilitation by a factor of about two.

Another issue of importance is that of water covers for uranium mill tailings, especially at Ranger and Nabarlek. When covered by up to 2 m of water or more, the radon load derived from uranium tailings is regularly stated to be negligible, though no field data has been presented to substantiate this claim.

Based on the laboratory work of [9], the radon flux from water-covered tailings was measurably higher than due to diffusion alone, considered likely to be related to thermal and/or advective processes. After modifying radon flux equations to account for water-covered and/or variably-saturated tailings, a new model was presented to estimate radon fluxes from tailings dams, provided online by [10]. Using this model, the current radon flux and load from Ranger, for example, can be estimated as  $0.75$  Bq/m<sup>2</sup>/s or  $73.8$  GBq/day from the above ground dam (water depth of 1.3 m) and  $0.08$  Bq/m<sup>2</sup>/s or  $3.3$  GBq/day from the tailings repository in Pit #1 (water depth of  $>10$  m) [7].

Although this analysis is brief, it demonstrates that radon fluxes and loads are highly variable and claims about a particular environmental regime need to be supported by actual field measurements. For Port Pirie, the radon flux is likely to be higher than pre-milling, even after rehabilitation of the dams, since the concentrate was imported from Radium Hill. For Nabarlek, the radon flux appears to be lower, though whether the site-wide flux and load is lower remains unclear. Waste rock dumps are evidently an important source of radon, as demonstrated by the low grade ore and waste rock dump sites at Nabarlek, Rum Jungle and Ranger (sometimes  $<0.02$  %U<sub>3</sub>O<sub>8</sub>). The evidence of changes in radon fluxes at different Australian uranium mine and mill sites does not allow a consistent picture to emerge, due mainly to the paucity of pre-mining and post-project field measurements. The inadequacy of the UNSCEAR approach, which assumes tailings as the primary source of radon, is limited and needs to be expanded to include other sources such as waste rock and contaminated areas.

TABLE 2. RADON FLUXES AND LOADS FROM SELECT URANIUM MINING AND MILLING SITES IN AUSTRALIA [1, 7]

	Waste Type ‡	Area (ha)	Uranium (%U <sub>3</sub> O <sub>8</sub> )	<sup>222</sup> Rn Flux (Bq/m <sup>2</sup> /s)	<sup>222</sup> Rn Load (GBq/day)
Rum Jungle, NT	T	35	~0.086%	~2.9	88
White's (Rum Jungle), NT	WR	26.4	0.01%	1.1	25
Rum Jungle Creek South, NT	WR	21.9	0.054%	2.7	51
Rum Jungle, NT	R <sup>(P)</sup>	~500	–	0.14	–
Rockhole, NT (average)	T	~2	0.048%	<5–21.1 (~6)	10.4
Moline, NT (average)	T	~18	0.066%	<1–17.9 (~2)	31
Port Pirie, SA	T	~30	~0.24%	5	130
Port Pirie, SA	R <sup>(D)</sup>	–	–	0.12	3
Jabiluka, NT (Mine Valley)	PM	–	–	0.046	–
Jabiluka, NT (Proposed Haul Road)	PM	–	–	0.025	–
Nabarlek, NT	PM	~5	–	3.7–44.0	–
Nabarlek, NT	R <sup>(D)</sup>	–	–	1.03 ± 0.80	4.5
Nabarlek, NT	U-T	–	–	2.1	9.1
Ranger, NT	PM	245	–	1.78	377
Ranger, NT	U-T	–	–	0.9 <sup>AE</sup> / 0.1 <sup>AQ</sup>	–
Ranger, NT	R <sup>(P)</sup>	–	–	'0'	'0'
Koongarra 1, NT (Koongarra 2)	PM	12.53	–	2.43 (<0.05)	26.3
Olympic Dam, SA	PM	–	–	0.025	–
Olympic Dam, SA	Mill & T	~400	–	–	260–290
Olympic Dam, SA	U-T	75	–	1.6	103.7
Olympic Dam, SA	U-T <sup>(P)</sup>	720	–	0.2	124.4
Honeymoon, SA	PM	–	–	0.035	–

‡ U-T – UNSCEAR 1993 assumed tailings (T) data, <sup>AE</sup> – sub-aerial, <sup>AQ</sup> – sub-aqueous; WR – waste rock; PM – pre-mine (generally above ore zones); R – rehabilitated site (proposed <sup>(P)</sup> or done <sup>(D)</sup>).

TABLE 3. MEASURED RADON FLUX PROPERTIES AT RANGER AND NABARLEK [7]

Mine/Mill Site Ore / Tailings	%U <sub>3</sub> O <sub>8</sub>	<sup>226</sup> Ra Bq/kg	<sup>222</sup> Rn Flux Bq/m <sup>2</sup> /s
Ranger waste rock (dry / wet)	–	–	1.2 <sup>§</sup> / 0.47
Ranger waste rock	–	–	0.52
Ranger very low grade ore	0.03%	3,112	1.3
Ranger tailings (dry)	0.033%	22,100	10.4
Ranger tailings dam wall	0.012%	1,245	0.21
Nabarlek tailings	0.034%	190,853	4.710
Nabarlek waste rock	0.013%	1,348	0.26

§ Calculated based on a measured radon-in-air concentration profile.

TABLE 4. PRE-MINING CALCULATED RADON FLUXES AND LOADS FROM THE RANGER ORE ZONES [8]

Region	Radon Flux	Area	Radon Load
Orebody #1	4.1 Bq/m <sup>2</sup> /s	44 ha	155.8 GBq/day
South of #1	1.0 Bq/m <sup>2</sup> /s	27 ha	23.3 GBq/day
Orebody #3	2.5 Bq/m <sup>2</sup> /s	66 ha	142.6 GBq/day
North of #3	1.0 Bq/m <sup>2</sup> /s	27 ha	23.3 GBq/day
Strip #1–#3	1.0 Bq/m <sup>2</sup> /s	27 ha	23.3 GBq/day
East of Strip	0.13 Bq/m <sup>2</sup> /s	27 ha <sup>†</sup>	3.0 GBq/day
West of Strip	0.23 Bq/m <sup>2</sup> /s	27 ha <sup>†</sup>	5.4 GBq/day
Total	1.78 Bq/m <sup>2</sup> /s	245 ha	376.7 GBq/day

<sup>†</sup> No area given, value assumed.

TABLE 5. PROGRESSIVE ESTIMATES (GBQ/DAY) OF COMBINED RADON LOADS FROM THE RANGER PROJECT [7]

Year	T. Dam Type	Plant	Ore Stockpiles	Waste Rock	Pits	Tailings	Total
Pre-mine		0	0	0	371.8	~4.92 ‡	376.7
1975 <sup>(2)</sup>	>2 m WC *	44.0	19.2 §	–	32.2	<0.37	95.5
1977 <sup>(3)</sup>		20.0–148.0	~96.2 §	–	20.0–281.2	1.44–14.4	137.6–539.8
1981	Dry	–	–	–	–	3,990	–
1980's	sub-aq. †	–	–	–	–	196.8	–
1992	sub-aerial	146.9	318.0	7.6 <sup>(4)</sup>	43.9	96.2	612.6
1993	sub-aerial	149.5	324.9	15.1	25.9	94.2	609.5
1990's	mixed WC *	–	–	–	–	77.1	–

‡ Assuming a pre-dam flux of 0.05 Bq/m<sup>2</sup>/s. § Includes waste rock. † Sub-aqueous. \* water cover.

#### 4. GAMMA RADIATION

An important aspect of uranium project rehabilitation is residual gamma radiation dose rates. Some uranium deposits have been discovered in Australia by searching for small, localised areas of gamma radiation which indicate potential uranium mineralization (eg. Ranger, Yeelirrie, etc). On the other hand, many uranium deposits lie buried beneath a sedimentary cover or other geological formation and there is no elevated gamma dose rate to signify the presence of uranium. There is some pre-project data available for select uranium sites on gamma dose rates (or simply 'counts per second', cps), compiled in Table 6, though it is not as comprehensive as desired.

The data shows that for most uranium deposits in the Alligator Rivers Region, there is no significant or elevated gamma radiation dose rate noticeable, although some sites have small and localised areas (with Ranger being an obvious, rare exception to this). For many Australian uranium deposits a similar table could be demonstrated (eg. Olympic Dam, Beverley, Manyingee) with some deposits showing geologically localised areas of elevated gamma dose rates (eg. Yeelirrie, Kintyre, Mt Painter).

TABLE 6. BACKGROUND RADIATION COUNTS AT SELECT URANIUM SITES WITHIN THE ALLIGATOR RIVERS REGION [7]

Uranium Deposit	Aerial Radiometric Surveys				Ground Surveys	
	Total Count (cps)	x Back-ground ‡	No. § Lines	Area	x Back-ground ‡	Area
Koongarra	345	~6	1	100 m	10	90×90 m
Ranger 1 & 3	1,460->4,000	~30->80	4	6.5x1.5 km	30–250	6×0.5 km
Ranger 9 / 68	–	1	None	–	1 / (radon) †	–
Jabiluka 1	–	1	None	–	2	105×45 m
Jabiluka 1	–	1	None	–	1.5	80×40 m
Jabiluka 2	–	1	None	–	1	–
Nabarlek	700–1,960	~20–65	2	0.5x1.8 km	50	0.15×1.5 km
Coronation Hill	–	–	–	–	2–3	0.4 ha
El Sherana	–	–	–	–	1.5–10	2 ha

‡ Ratio of anomaly to background count (~20–100 cps; exact figure used is often not quoted, which will depend on the survey height, equipment used, etc). § Number of flight lines. † 'Radon' anomaly.

The process of uranium mining and milling leads to the dispersal and changed nature of many radionuclide sources, thus posing a particular challenge for rehabilitation. Some examples include [1]:

- (1) Nabarlek – mineral exploration and environmental surveys were used to estimate an average pre-mine gamma dose rate of about 0.18  $\mu\text{Gy/hr}$  [11]. Detailed post-rehabilitation surveys have been undertaken, based on correlation of aerial and ground radiometric surveys by [11], with the average gamma dose rate being derived at 0.27  $\mu\text{Gy/hr}$  [11]. The gamma dose rate above the former ore zone has been decreased by about half, however, over the 97.6 ha of the project area the gamma dose rate has therefore been increased by 50%.
- (2) Rum Jungle – a ‘radioactive anomaly’ can be traced downstream in the Finnis River for many kilometres. There is no published aerial or ground gamma surveys for Rum Jungle, especially after rehabilitation, and based on geology and site operations, it is highly likely that an increase similar to or perhaps higher than Nabarlek has also occurred, but over a much larger area.
- (3) Hunter’s Hill (Sydney, NSW) – the site of the radium refinery for Radium Hill ore between 1911–15. In the late 1970’s it was discovered to contain high gamma dose rates (as well as radon) ranging from 0.14 to 1.4  $\mu\text{Gy/hr}$ , derived from the 3,000 t of radium mill tailings [1, 12].
- (4) Rockhole (South Alligator Valley, NT) – the poor management of uranium mill tailings (as well as partially effective recent ‘hazard reduction’ works) has seen the surrounding areas reach gamma dose rates ranging from 0.33 to 6.0  $\mu\text{Gy/hr}$  through further erosion and dispersal.
- (5) Moline (near the South Alligator Valley, NT) – due to the erosion and dispersal of about 63,000 t of mixed uranium-base metal tailings, gamma dose rates 1 km downstream were around 0.25 to 1.0  $\mu\text{Gy/hr}$ , higher than the measured background of about 0.02  $\mu\text{Gy/hr}$ .

Although many of the gamma dose rate examples quoted do not represent acute or dangerous situations, from an environmental perspective these ‘chronic’ and perhaps permanent increases are of legitimate concern. Changes in gamma dose rates clearly need to be given greater consideration in the long-term assessment of ionizing radiation and radionuclide loads released by uranium projects.

## 5. ER QUALITY

The management of water and associated (or potential) impacts is often the most publicised aspect of radionuclide releases from uranium facilities in Australia. Historically, this is related to the serious water quality and environmental impacts from Rum Jungle, concerns over mining and national parks (eg. Ranger), seepage from tailings management facilities (eg. Olympic Dam) as well as impacts on groundwater from in situ leach mines (eg. Beverley, Honeymoon). A summary of the radionuclide issues from these various sites and their associated environmental issues is presented.

### 5.1. Surface water

The Ranger Inquiry [2] made strong recommendations that uranium projects in the Alligator Rivers Region operate a ‘no-release’ water management system. Initially the Jabiluka, Koongarra and Nabarlek projects accepted this approach, though Ranger fought to maintain the legal right to release contaminated minesite waters under certain intense wet season conditions (eg. Magela Creek flow  $>20 \text{ m}^3/\text{s}$ ). The attention which the Ranger Inquiry placed on water was a combination of national park and indigenous concerns and the lasting impacts from Rum Jungle, where poor waste management and acid mine drainage had led to widespread contamination of the Finnis River for some 100  $\text{km}^2$  [1].

The NGO movement continues to oppose the discharge of radionuclides to surface water ecosystems, and, in general, believes all wastes from mining should be safely contained within a project area.

#### 5.1.1. Rum Jungle

A detailed study and analysis of the impacts from Rum Jungle is given in [1], with a concise summary in [13]. The principal points concerning environmental radionuclides are:

- The discharge of 1 ML/day of acidic liquid wastes and gradual erosion of tailings deposited on lowlands adjacent and into creeks which flowed into the Finnis River led to some 17 TBq of radium ( $^{226}\text{Ra}$ ) entering the environment. Accounting for the radium has been extremely poor,

with very little focus on radium uptake in the environment or current levels leaching from the site. Monitoring of radium activities in the Finnis River was stopped in 1988, shortly after rehabilitation, with annual loads still being of the order of 0.4 to 1.6 GBq per wet season.

- Despite uranium being highly soluble in the acidic, oxidising geochemical environments prevailing within wastes at Rum Jungle, there was no U concentrations or load data published in studies in the 1970's, with the only data available being for the 1992/93 wet season (Table 7).

### 5.1.2. Alligator Rivers Region

The confluence of Aboriginal land rights, uranium mining and environmental conservation have always made scientific debate about Ranger and nearby projects highly contentious, with water (and tailings) management often at the top of the list of concerns. This section is based on [1, 7, 14].

After considerable debate, the Ranger uranium mine was forced to accept a 'no-release' water management system by the mid-1980's. However, poor data and understanding of evaporation and rainfall in the region led to the accumulation of contaminated waters at Ranger and Nabarlek [1].

To overcome this, the typical approach has been to temporarily remove contaminants from water through irrigation onto nearby pristine lands, or more recently, the use of artificial wetlands. The principal mechanisms suggested to remove radionuclides (U, <sup>226</sup>Ra) from the water include adsorption on soils and plant uptake. The less reactive contaminants (eg. Mg, NH<sub>4</sub>, SO<sub>4</sub>) are often left to reach groundwater and adjacent creeks. This clear divergence from 'no-release' at Ranger, Nabarlek and again at Jabiluka is of significant legitimate concern to the environment movement, especially as long-term sink and uptake issues are poorly addressed in ongoing project management and regulation.

Throughout its operation, the Ranger project has had to meet specific downstream water quality in the Magela Creek (at gauging station 'GS8210009'), near the boundary with Kakadu National Park. The debate again flared in early 1995 when Ranger applied to discharge contaminated 'Restricted Release Zone' (RRZ) water to the Magela, and, although winning the court case against the traditional owners (who were clearly opposed), Ranger withdrew the application.

In recent years a new system has been implemented to assess the impacts on water quality downstream of Ranger, with a similar regime also in place for the stalled Jabiluka project. The system is based on the use of three trigger levels to assess water quality, rather than specific concentrations and loads. The triggers are termed 'focus', 'action' and 'limit', with focus suggesting that heightened vigilance over environmental data is necessary, action requires investigation and limit suggests a failure of management systems onsite (that is, potentially unacceptable environmental impacts). The levels are derived using methodology in the revised Australian Water Quality Guidelines [15]. In general, the aim is to prevent water quality deviating significantly from background (or upstream) by deriving the trigger values based on statistical variation or using ecotoxicological data. The criteria for Ranger and Jabiluka are summarised in Table 8, including typical background values.

In general, there is a strong trend of increased Mg-SO<sub>4</sub> concentrations in the Magela Creek due to Ranger, though the data for metals and radionuclides is less consistent. Some of the principal concerns (among many) relate to the high U concentrations allowed (5.8 µg/L) over background, especially for Jabiluka with a background of <0.01 µg/L, the creep of operations into previously pristine areas (eg. RP1), the high concentrations of leaks or failures, and the continual focus solely on Kakadu while downplaying the potential environmental impacts within the Ranger and Jabiluka project areas.

TABLE 7. FINNISS RIVER WATER QUALITY, DOWNSTREAM OF RUM JUNGLE, 1992/93 WET SEASON (µG/L) [13]

( <sup>†</sup> mg/L)	Al <sup>†</sup>	Ca <sup>†</sup>	Fe <sup>†</sup>	As	Ba	Co	Cr	Cu	Ni	Pb	Th	U
Average	3.6	9.9	1.71	4.1	37	176	5	485	169	76	3.3	33
Minimum	0.21	4.2	0.096	0.6	21	53	0.7	180	53	2	0.02	6
Maximum	9	29	14	41	120	480	33	1,100	430	880	26	63

TABLE 8. ANNUAL DOWNSTREAM WATER QUALITY SUMMARY FOR RANGER AND JABILUKA [1, 7, 14, 16, 17, 19]

			pH ‡	EC ‡	Mg ‡	SO <sub>4</sub> ‡	NO <sub>3</sub>	Mn	<sup>226</sup> Ra	U
			–	µS/cm	mg/L	mg/L	mg/L	µg/L	mBq/L	µg/L
1979–01	Ranger	Crit. †	ND	ND	10	19	0.6	24	13 §	3.8
2001–	Ranger	Focus	5.84–6.50	22	(use	(use	ND	11	>10	0.30
Present		Action	5.51–6.83	30	EC)	EC)		19	>10 <sup>1</sup>	1.90
		Limit	5.18–7.16	43				37	>10 <sup>2</sup>	5.8
2000/01	MC-U	Ave-	5.93	10	0.48	0.28	ND	4.96	ND	<0.1
Wet	MC-D	rage	6.02	12	0.72	0.73		4.35	3–20	0.1
2001–	Jabi-	Focus	4.61–5.31	15	0.37	0.60	0.30	ND	ND	0.02
Present	luka	Action	4.27–5.65	18	0.50	0.91	0.63			0.03
		Limit	3.92–6.00	21	0.76	1.50	1.26			5.8
2000/01	SC-U	Ave-	4.70	9	0.30	0.60	0.15	4.11	3–9	0.014
Wet	SC-D	rage	4.98	12	0.25	<0.1	0.07	2.43	<3–16	0.022

§ GBq/yr. <sup>1/2</sup> >10 mBq/L above upstream for 90 consecutive days / annual average. ‡ Guideline only. MC / SC – Magela / Swift Creek; U/D – Up- / Downstream; ND – No data. † Load limits also applied.

## 5.2. Groundwater

The protection of groundwater is widely recognised as a fundamental environmental issue, especially for the 21<sup>ST</sup> century. The experience in Australia, however, suggests that the attention by regulators and companies is clearly not in step with community expectations [1, 7, 13, 14, 18]. Some of the many complex issues include the long-term impacts on groundwater quality (eg. redox state, metals), potential for contaminant migration through fractures (or other permeable pathways such as carbonate units) and potential hydraulic connections between groundwater and surface water ecosystems.

## 6. DISCUSSION AND CONCLUSIONS

This paper has presented a concise analysis, based on more comprehensive studies in progress (which governments have failed to undertake properly), of the impacts of the uranium industry in Australia, thereby illustrating particular issues around ionizing radiation and the protection of the environment. There are many important issues raised by the work which the environment movement sees as pivotal.

It is clear that the release of radionuclides into the environment or changes in ionizing radiation rates are still poorly quantified from uranium mining and milling, despite some improvements in recent years. Critical issues such as radon flux and loads, gamma dose rates and impacts on groundwater need to be more rigorously monitored and assessed. While surface water and tailings receive most of the attention, the downstream water quality standards in the NT allow for substantive increases in uranium. For example, at Jabiluka, the ‘limit’ value is some 580 times higher than background. It is not merely an academic exercise – even approaching a quarter of 5.8 µg/L shows that significant environmental impact (not just change) has or is occurring due to the increase over background. This issue remains of deep concern to Aboriginal people and the environment movement.

There are many complex issues which fail to be taken into proper account when examining questions of ecotoxicology and the potential impacts of ionizing radiation and radionuclides in the environment:

- the ultimate capacity of sinks, such as wetlands, soils and plants, to retain limited quantities of contaminants such as U, Mn, <sup>226</sup>Ra, etc;
- the cycling of radionuclides through the environment, between soils, plants, insects, aquatic species, mammals, etc (ie. both macro and micro scales);
- the radionuclide transfer factors (or bioaccumulation factors) between these components of the environment in different climates (eg. <sup>226</sup>Ra uptake is higher in the tropics than arid lands);
- the inability to focus on ‘low-dose, long-term’ exposure to radionuclides which cause chronic, sub-lethal effects, non-fatal diseases, chemical toxicity and/or genetic damage (as opposed to the traditional approach of ‘fatalities’ in most current ecotoxicological testing regimes);



- the lack of a truly long-term approach to assessing and regulating uranium operations;
- the rehabilitation standards to try and minimise the long-term release rates; etc.

The available evidence from uranium project sites around Australia shows that, in general, ionizing radiation rates and radionuclide are generally within normal background prior to development. At many of these sites, the operations appear to have led to deterioration from the pre-project situation. The increased radiation rates are also cumulative in their impacts over all project sites. Rehabilitation is proving more difficult than predicted. It is well documented that radionuclide uptake and internal exposure to ionizing radiation is dangerous. The absence of being able to prove harm at low doses should not be a weak regulator's excuse to allow radionuclide releases into the environment. The 'As Low As Reasonably Achievable' (ALARA) principle, which has to take into account social and economic issues, is often used to justify the low dose exposure of people and the environment without reasoned and informed debate. There is a general understanding that people are the most sensitive to ionizing radiation and if they are protected, the environment should be also. To separate people from the environment is clearly irrational (eg. the Ranger Inquiry included people in 'environment') and against the global push for sustainability, of which the Precautionary Principle is a key standard adopted by many governments and communities in their ongoing journey in this regard. The onus of proof should be on industry and government to demonstrate that there are no impacts on the environment from ionizing radiation and radionuclides. Until there is a broader consensus (from all), it is perhaps more appropriate to follow the ALATA or 'As Low As Technically Achievable' principle. Given the future potential for expansion of the nuclear industry (eg. uranium mining), it is imperative that the sources and environmental impacts of ionizing radiation are better quantified and understood.

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# **ARPANSA's regulatory role in the protection of the environment from ionising radiation**

## ***Licensing the remediation of abandoned uranium mine workings in Kakadu National Park***

**J.S. Prosser**

Australian Radiation Protection and Nuclear Safety Agency, Sydney, Australia

**Abstract.** ARPANSA was established by the *Australian Radiation Protection and Nuclear Safety Act 1998* to perform a number of specific functions including the licensing of those activities involving radioactive material that are undertaken by Commonwealth entities. This paper describes the role of the regulator in the rehabilitation of an area of Kakadu National Park, which has been licensed as a Prescribed Radiation Facility.

When making a decision on whether to issue a licence, the CEO of ARPANSA is obliged by the *ARPANS Act* to consider, *inter alia*, whether it has been established that the proposed conduct can be carried on without undue risk to the health and safety of people, and to the environment. However, when licensing the remediation of environmental and radiological consequences of mining activities special difficulties may arise in determining the regulatory approach to be adopted with regard to an intervention of this nature. One specific example is the limited internationally accepted objective radiological criteria on which to base regulation of any remediation. In the particular case of an intervention in Kakadu National Park, the multiplicity of interest groups directly or indirectly involved in management of the affairs of this world heritage listed area adds an extra element to the regulatory equation.

### 1. INTRODUCTION

The Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) was established by the Australian Radiation Protection and Nuclear Safety Act 1998 [1] – an Act to regulate activities involving radiation, and for related purposes.

Section 3 of the Act states: – The object of this Act is to protect the health and safety of people, and to protect the environment, from the harmful effects of radiation.

To this end the legislation defines the functions of the CEO, which include:

- promotion of uniformity in radiation protection policy and practices across jurisdictions;
- provision of advice and services;
- undertaking of research; and
- determination of licences to permit Commonwealth entities to deal with radiation sources or to operate radiation facilities or nuclear installations.

The legislation is intended to control the activities of Commonwealth organisations that are exempt from State regulation and were therefore previously not formally regulated. Specific reference is made to the protection of the environment, in addition to protection of health and safety of people. Such reference is however very general.

### 2. LICENSING

In determining whether to grant a source or facility licence, the Regulations describe information that the CEO may request, including details of various plans and arrangements for managing safety, security, waste, emergencies etc. There are certain prescribed matters included that the CEO is obliged to take into account. These include whether the information provided in the application establishes:

- a net benefit;
- that doses are as low as reasonably achievable; and
- that the proposed undertaking can be carried out *without undue risk to the health and safety of people, and to the environment.*

The licensing philosophy espoused by the Regulatory Branch of ARPANSA is that of being non-prescriptive as to the means by which the various plans and arrangements establish net benefit, ALARA and acceptable risk to people and the environment. To this end documented expectations [2] have been compiled using international best practice and Australian Standards and Codes of Practice as guidance. In addition, compliance with certain National Health and Medical Research Council Codes of Practice is specifically required by the legislation.

The Code of Practice for the Near-surface Disposal of Radioactive Waste in Australia [3] is one such code. Its stated purpose is to *'provide a basis for the near-surface disposal of solid radioactive waste in a way which ensures that there is no unacceptable risk or detriment to humans, other biota or the environment...'*

In principle therefore environmental concerns, with respect to radiation, are addressed in a number of existing controls that apply to Commonwealth entities.

International best practice in relation to radiation protection and nuclear safety must also be taken into account in determining whether to grant a licence. This is of course strongly influenced by pronouncements from the International Commission on Radiological Protection. In ICRP 60 [4] it is stated that the Commission believes *'that the standard of environmental control needed to protect man to the degree currently thought desirable will ensure that other species are not put at risk. Occasionally, individual members of non-human species might be harmed, but not to the extent of endangering whole species or creating imbalance between species'*. This position has been restated as recently as 1998 [5] but is currently under review [6].

In a recent report [7], the CEO of ARPANSA stated his view *'that there is not yet an established radiation protection system for non-human species that can be regarded as international best practice ... other than focussing on the protection of humans'*. This statement was made in the context of determining a licence for a research reactor but applies equally to other licence decisions. In anticipation of developments in this evolving field, conditions with regard to the provision of summary information on protection of the environment have been placed on the licence issued to Parks Australia North, as discussed below.

### 3. LICENSING THE REMEDIATION OF ABANDONED URANIUM MINE WORKINGS IN KAKADU NATIONAL PARK

The first involvement of ARPANSA in the issue of radiological matters in the South Alligator River area of Kakadu came after the publication of a report in 2000: *γ Radiation survey of exposed tailings in the area around Rockhole mine* [8]. This internal report of the Office of the Supervising Scientist received significant publicity in the media. The authors measured increased radiation levels during a routine periodic survey of a public thoroughfare and an area adjacent to that used for burial of miscellaneous waste materials from uranium mining activities conducted in the 1960s and 1970s. Elevated radiation levels, up to 6 µGy/h in some places, were measured following disturbance of mine tailings buried alongside the road. Further measurements established that there was no significant hazard to people using the road, from external exposure or from dust inhalation but the authors advised that remediation work be undertaken as soon as practicable.

A subsequent inspection by ARPANSA determined that this and additional similar areas of disturbance due to previous mining activities should come under the regulatory control of ARPANSA. The Director of National Parks, who with the Board of Management jointly administers Kakadu National Park (through Parks Australia North), was therefore advised to make an application for a facility licence. The purpose would be to authorise remediation of all areas affected by uranium mining activities within the South Alligator River area of the National Park.

There may appear to be a certain incongruity in licensing part of a world heritage listed area as a prescribed radiation facility. However the ARPANS Regulations include the capacity to licence a facility where a mixture of controlled materials is stored or managed. By including within the definition of facility a specified geographical area, in legal terms at least the incongruity disappears and management of radioactive mining residues can be (and can be seen to be) appropriately regulated.

The initial licence granted to Parks Australia North is very limited. It authorises immediate interim works to stabilise the area initially identified and remove exposed tailings. In addition it requires the development of a remediation plan for all the affected sites. This plan is to be reviewed and approved by ARPANSA before any further works may be undertaken and should include an assessment of:

- stakeholder aspirations and values;
- environmental and safety issues;
- previous site activities, including remediation; and
- environmental, social and cultural values affecting the area.

Finally, before any further works may be authorised, a summary of assessment of protection of the environment must be provided. In essence this should show how Parks Australia North have assessed the risks to the environment and indicate the rationale for their choice of remediation actions intended to be taken as a result.

### **3.1. Criteria for remediation**

ARPANSA's radiological requirements are essentially those provided in ICRP 60 for an intervention to reduce radiation dose due to previous human activities [4]. In principle therefore remediation would be expected to reduce the dose to any individual, from sources associated with previous mining activities, where this is currently greater than 10 mSv per year. The ICRP recommends, however, against the use of its dose limits for deciding on the need for, or scope of, any intervention. The need for and extent of remedial action should be judged by comparing the benefit of dose reduction with the detriment of the remedial work. Appropriate radiation protection criteria must be applied to the practices undertaken in any remediation.

Some limited guidance is also given in ICRP Publication 77 [9], which states that any decision on treatment of mining residues should be made on a case by case basis, should be aimed at doing more good than harm and should not be based on a preselected dose. More recent ICRP publications (ICRP 81 [5] and ICRP 82 [10]) are similarly non-committal with regard to numerical criteria for avoidable dose: repeating the advice that intervention should be managed on a case by case basis. They each repeat the suggestion that: intervention is not likely to be justifiable for some prolonged exposure situations for annual doses approaching 10 mSv, whereas an existing annual dose rising towards 100 mSv will almost always justify intervention.

However, experience suggests that the dose limit for practices is likely to dominate any discussion of what is acceptable. The public and decision-makers are unwilling or unable to recognise the distinction between a practice and an intervention or to deviate from the established limit for practices of 1 mSv per year. Indeed it appears that this may be the more usual dose criterion that has been applied to the clean up of abandoned contaminated areas [11].

Inevitably, questions were raised by Parks Australia North as to the standard of remediation required by the regulator. The preceding paragraphs indicate that it is not reasonable to set fixed generic numbers for radioactivity and contamination levels for every site. It was agreed that a separate case would need to be made for each individual site. Parks Australia North would need to set out arguments for the degree of remediation works and their justification based on various factors including remoteness, site access, predicted occupation and use, wishes of the traditional owners, political and social concerns etc. In a number of instances social, economic and (non-radiological) environmental considerations may have the major influence in determining the degree of remediation activity to be proposed. For example, the degree of radiological and environmental remediation to be recommended will take into account the degree of disturbance permitted in some areas by traditional beliefs. This may be limited by the traditional owners' preference that no heavy machinery or explosives be used and, in some areas of special significance, the requirement that no rock or earth be transported in or out.

TABLE 1. SOME RECOMMENDED VALUES FOR DOSE LIMITS UNDER DIFFERENT CIRCUMSTANCES

Authority	Dose Limit	Basis	Reference
NSW, QLD & WA	2.5 $\mu$ Sv/h	Maximum dose rate for roads, pathways etc	State legislation
ICRP	10 mSv/y to a member of the public	Dose level above which intervention may be required	ICRP 60
ICRP	100 mSv/y to a member of the public	Dose level above which intervention will be required	ICRP 60
ICRP	1 mSv/y to a member of the public	Limit of additional dose from a practice	ICRP 60
National Occupational Health and Safety Commission	1 mSv/y to a member of the public	Limit of additional dose from a practice	National Standard – Radiation Health Series No. 39 [12]

Using this information, plus values of dose limits for practices from a selection of national and international codes and standards (given in Table 1), Parks Australia North have produced an argument for pragmatic guidelines for radiological clearance and these have been agreed with ARPANSA. These values will assist in determining whether radiological remediation is required and provide criteria to establish completion. The values that they have selected are:

- an action level gamma dose rate of 2.5  $\mu$ Sv/h above background at 1 metre above ground level; and
- a soil contamination level of 35 Bq/g of U238 (or Ra226 where U238 is not in secular equilibrium with its decay products).

In the South Alligator River region of Kakadu, the general dose rate in all areas ranges from 0.1 to about 8  $\mu$ Sv/h. The average background dose rate at abandoned mine sites is assumed to be 5  $\mu$ Sv/h. Hence a guideline dose rate for action of 7.5  $\mu$ Sv/h is proposed, which is to be applied with discretion depending on the other various factors mentioned.

Using these criteria and taking into account the proposed occupancy of the areas to be rehabilitated, the expected dose to any of the most exposed group of people due to mining residues is less than 1 mSv per year. As it becomes available, site specific information will be used to calculate realistic doses to the critical group.

### 3.2. Protection of the environment

Having determined the radiological criteria to aid decision-making with regard to mine site remediation, the main outstanding licensing issues for Parks Australia North relate to special licence conditions regarding the provision of summary information on protection of the environment and on disposal of any wastes arising.

The special licence condition with respect to information on protection of the environment is written very broadly, reflecting the wording of the Act. An outline of ARPANSA's expectations in regard to demonstrating compliance with environmental protection obligations using a risk assessment approach are given by Dooley et al [13] (this conference).

Waste management is probably the most intractable problem and is yet to be satisfactorily resolved. A small amount of waste material has already been generated from the interim cleanup and stabilisation of the original roadside area. Deciding how to manage this contaminated waste, and the possibly much larger amounts arising from further remediation, is one of the more difficult issues associated with the proposed intervention. The material will be largely waste rock, plus some tailings, contaminated concrete and other construction materials. Alternative approaches include:

- burial of material on site, where suitable;
- removal to a dedicated waste repository either within the designated area of the national park facility or elsewhere;
- use of the majority of the waste as a resource to be milled for uranium extraction.

All options need to be considered by all of the stakeholders, particularly the traditional owners, before a final plan can be drawn up. Meanwhile it is anticipated that for those areas where little if any radioactive waste will be generated, approval should be granted for remediation to begin this year.

#### 4. STAKEHOLDERS

In requiring a fully developed plan before authorising any further works, ARPANSA was aware of the potential conflicting requirements of the many stakeholders needing to be satisfied by Parks Australia North's strategy for remediation. The interests of Commonwealth and State governments, traditional owners, various environmental groups and a growing number of visitors must all be considered. An indication of the number of interested parties and the political sensitivity of any decisions taken by the Board of Management is given in Figure 1. The ARPANS legislation follows ICRP 60 in requiring that relevant social and economic factors be taken into account when establishing optimal dose criteria. In this instance, the most significant of these factors is undoubtedly the wishes of the traditional owners of the land, most particularly with regard to undertakings in areas of special cultural significance. In the last few years the importance of informing and empowering local stakeholder groups has become well recognised within the radiation safety community. The topic is an important part of current discussions on the way forward in developing a system of radiological protection and a dose optimisation process using egalitarian ethics [14].

It is unlikely that Parks Australia North is aware of the development in ICRP's current thinking on these matters. Their process has probably been arrived at independently. Whatever the reason, their commendable efforts in fully informing and involving the most relevant stakeholder groups parallels the approach supported by participants at the recent OECD/NEA sponsored meeting at Villigen where several examples of successful outcomes from a more politically focussed strategy were discussed. For example Gatzweiler et al [11], in their evaluation of remediation programmes in the uranium mining areas of the former East Germany, argued strongly that there are no general rules for stakeholder involvement except that the attempts should be adapted to local conditions and total openness and consultation are essential.

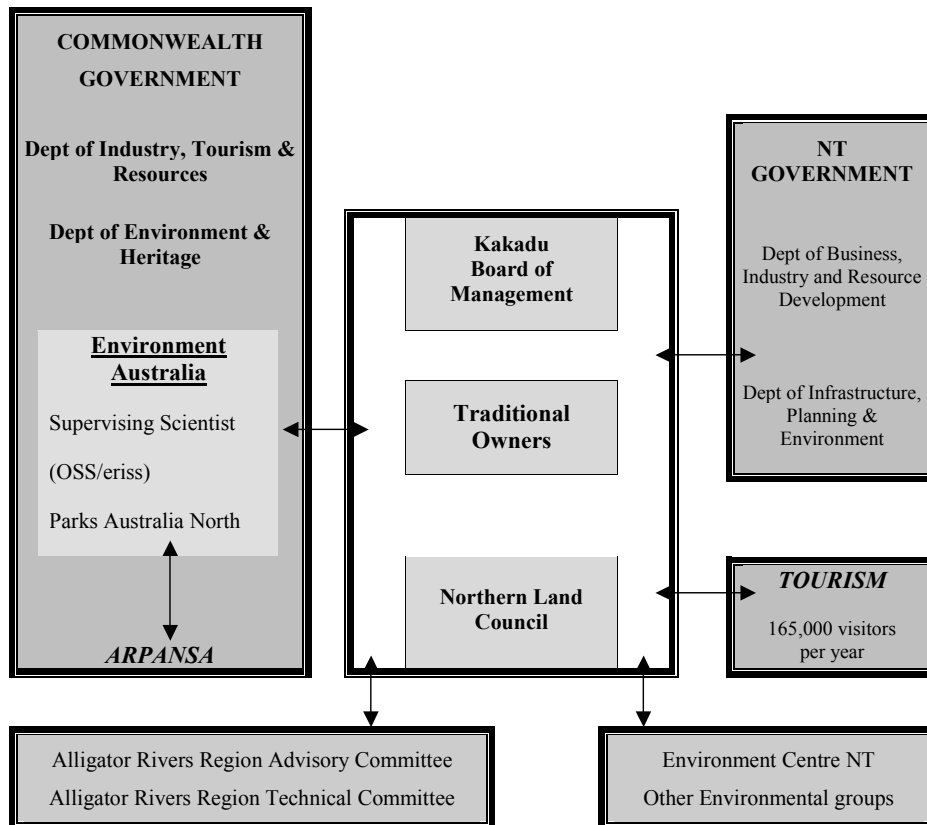


FIG. 1. Stakeholder relationships influencing the management of Kakadu National Park.

## 5. THE FUTURE

An application to amend the existing licence is expected shortly. This will request authorisation to remediate those areas less affected by radiological contamination. Remediation of the remaining areas will not be authorised until a waste disposal plan has been agreed. Long term stewardship, including periodic monitoring is yet to be addressed but is anticipated not to present too many difficulties given the status of the area as a national park managed in close cooperation with the traditional owners.

## ACKNOWLEDGEMENTS

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# Regulatory guidance in England and Wales to protect wildlife from ionizing radiation

I. Zinger-Gize<sup>a</sup>, D. Coplestone<sup>b</sup>, C. Williams<sup>c</sup>

<sup>a</sup> Environment Agency, Warrington, United Kingdom

<sup>b</sup> Environmental Research and Consultancy, University of Liverpool, Birkenhead, United Kingdom

<sup>c</sup> Environment Agency, Bristol, United Kingdom

**Abstract.** The Environment Agency has a legal requirement to assess the impacts of permits for discharges affecting Special Areas of Conservation (SACs) or Special Protection Areas (SPAs) under the Habitats Regulations (1994), including the assessment of radiological risk. As there is no international consensus on the approach to be taken to assess the impact of ionizing radiation on wildlife, the Environment Agency has developed guidance to satisfy its regulatory obligation, based on a published impact assessment methodology. This approach will also feed into the European Commission funded project Framework for ASSESSment of Environmental Impact (FASSET), due to complete in October 2003.

The impact assessment approach focuses on three ecosystems representative of those considered potentially most at risk from the impact of authorized radioactive discharges, namely a coastal grassland (terrestrial ecosystem); estuarine and freshwater ecosystems. Dose calculations have been programmed into spreadsheets to support the methodology. The approach also makes recommendations on the relative biological effectiveness of different types of radiation with respect to wildlife.

Evidence for effects at low dose rates is reviewed and Tables of experimental and field study data on the effects of ionizing radiation are presented with which to compare any predicted doses to wildlife in order to assist in the impact assessment process.

Information reviewed indicates that it is unlikely that there will be any significant effects in:

- terrestrial animal populations at chronic dose rates below 40  $\mu\text{Gy/h}$ ;
- terrestrial plant populations at chronic dose rates below 400  $\mu\text{Gy/h}$ ;
- populations of freshwater and coastal organisms at chronic dose rates below 400  $\mu\text{Gy/h}$ .

The guidance make use of the methodology at various stages of the review process. It is also important to recognise that the assessor must consider the assumptions and constraints in the methodology, especially in the case of site-specific features, such as the presence of rare or endangered species. Finally, the above levels have been adopted within the guidance for the present time, but will be reviewed in the light of FASSET's radiation effects database.

## 1. INTRODUCTION

The Environment Agency of England and Wales has specific responsibilities for assessing the compliance of radioactive discharge and disposal authorizations issued under the Radioactive Substances Act 1993 to the requirements of the EU Birds and Habitats Directives (79/409/EEC & 92/43/EEC) and the UK Conservation Regulations 1994 (the 'Habitats Regulations'). English Nature is responsible for designating, and monitoring the conservation status of, Sites of Special Scientific Interest (SSSIs), as well as other European designated conservation sites; it is also a statutory consultee regarding radioactive discharges and waste disposal authorizations.

Both Agencies worked together to implement the Habitats Directive, as applied to England and Wales. They have produced a series of guidance, with Appendix 8 dedicated to applying the regulations to Radioactive Substances Authorizations [1]. The procedure is laid out in four stages:

- Stage 1: Identifying whether the Habitats Regulations are applicable.
- Stage 2: Identifying whether authorizations to discharge radioactive substances present a potential risk of significant effect on wildlife and habitats.

- Stage 3: Identifying issues to consider when carrying out a more detailed assessment of the potential impact of radioactive discharges for sites identified as presenting a potential risk.
- Stage 4: Determining an application, i.e. issuing an authorization for regulated discharge purposes.

The principal hazard relating to radioactive discharges/emissions, under this guidance, is usually radiotoxic contamination, other effects arising from the discharges (such as chemotoxicity) are rarely of significance except for a few specific nuclides (such as isotopes of uranium).

This paper will concentrate on the methodology used to carry out Stages 2 and 3, which take into account the recent Environment Agency and English Nature review of the effects of radioactivity on wildlife [2]. It is mainly based on the latest regulatory guidance used in England and Wales on applying the Habitats Regulations to radioactive substances authorizations [1, 3].

## 2. IMPLEMENTING THE HABITATS REGULATIONS

### 2.1. Stage 1: Identifying relevant applications

Disposals of radioactive waste occur to land, air or water. The Stage 1 – identification of relevant applications – is a simple screening exercise, designed to filter out applications or activities that by virtue of their nature or location could not conceivably have an effect on the relevant features of a European site of interest. Two primary activities are identified for consideration.

- For nuclear sites (i.e. sites holding an operating licence under the Nuclear Installations Act 1965), proposals for emissions to air within 5 km of a European ‘Natura 2000’ site should be assessed under the Habitats Regulations. The sites covered are Special Areas of Conservation (SAC) and Special Protection Areas (SPA). For discharges to surface freshwater (e.g. rivers or lakes) or ground water, any proposal which is likely to result in an exceedence of World Health Organization recommended levels (total  $\alpha$  = 0.1 Bq/l, total  $\beta$  = 1 Bq/l) for radioactivity in potable water, in or adjacent to the European site, should be assessed. For discharges to the coastal and marine environment, the threshold for triggering an assessment should be any discharge which raises, or is likely to raise, environmental concentrations above background (for naturally occurring radionuclides) or above the historically prevailing concentration (for anthropogenic radionuclides), in line with OSPAR commitments.
- For other sites authorized for the disposal of radioactive waste (i.e. sites such as hospitals or universities, where the handling or use of radioactive substances is not the main activity but minor amounts of radioactive waste are discharged to the environment), proposals for emissions to air within 1 km of a European ‘Natura 2000’ site should be assessed under the Habitats Regulations. For discharges to surface freshwater (e.g. rivers or lakes) or ground water, any proposal which is likely to result in a breach in World Health Organization recommended levels for radioactivity in potable water, in or adjacent to the European site, should be assessed. For discharges to the coastal and marine environment, the threshold for triggering an assessment should be any discharge which raises, or is likely to raise, environmental concentrations above background (for naturally occurring radionuclides) or above the historically prevailing concentration (for anthropogenic radionuclides), in line with OSPAR commitments.

The distance criteria should be applied to the point of discharge/emission if it is some distance from the authorized installation. It should also be noted that some Natura 2000 sites might receive discharges or emissions from several sources, which could act in combination to affect that site. In such cases, assessments of the combined impact should be undertaken, as laid out in the Habitats Regulations [4].

TABLE 1. GUIDELINES RECOMMENDED AS UPPER DOSE LIMITS FOR WILDLIFE [2, 7, 8]

Impacted group of organisms	Guideline Dose Limit ( $\mu\text{Gy/hr}$ ) <sup>a</sup>
Terrestrial animals <sup>b</sup>	40
Marine mammals	40
Terrestrial plants <sup>c</sup>	400
Other freshwater and coastal marine water organisms	400
Deep ocean organisms	1000

<sup>a</sup> Source: IAEA [7], reviewed and assessed for screening purposes [2, 8].

<sup>b</sup> Includes animals which inhabit the marine environment or the freshwater environment, but could have >50% occupancy of the terrestrial environment. For instance, 'freshwater' organisms include amphibians, aquatic mammals and ducks. 'Marine' organisms include seabirds.

<sup>c</sup> Includes bacteria, lichen and fungus.

## 2.2. Stage 2: Screening procedure for identifying potential significant effects

In the past, the Stage 2 screening assessments have relied on the assumption, stated by the International Commission on Radiological Protection [5], that 'if man is adequately protected from ionizing radiation, then so are other species'. This assumption may represent a reasonable rule of thumb. However, it is open to scientific challenge, mainly due to a lack of explicit evidence to support the position, and there has been increasing public and political pressure for the environment to be protected in its own right. It is also inconsistent with situations where the precautionary approach has been adopted to protect the environment from non-radioactive discharges. Accordingly a wildlife-based screening approach has been developed.

The purpose of the Stage 2 Assessment is to screen the RSA 93 Authorizations identified as relevant in Stage 1, and to identify those which could give rise to a significant effect on wildlife, and whose impact will therefore require further detailed assessment (Stage 3). Assessments under Stage 2 adopt a precautionary approach, following the release of the Environment Agency's R&D Publication 128 [2], and the detailed process used by Environment Agency staff is described elsewhere [3]. The methodology devised within the R&D Publication 128 is presented in detail in a separate article in this volume [6].

The initial test of significance is based on dose threshold guidance criteria for biota are laid out in Table 1 [2, 7, 8].

The discharge screening levels are designed to ensure that all doses received by wildlife remain within the guideline environmental dose rate levels. However, given the very broad nature of the groupings of organisms identified in Table 1, the UK methodology [2] recommends that it is appropriate to adopt a precautionary threshold set at 5% of the guideline environmental dose rate levels in order to determine whether further detailed assessments (i.e. Stage 3) are required.

For ease of reference, discharge screening levels (TBq/yr) have been calculated, for comparison with discharge levels at current authorized limits for RSA93 Authorizations which impact on European Natura 2000 sites, to see if the further detailed radiological assessment is required for these sites. The discharge screening level approach has been developed for a representative range of organisms and radionuclides for radioactive discharges to freshwater (rivers), atmosphere and to the coastal marine environment. Liquid radioactive discharges to the deep ocean or freshwater lake and solid radioactive waste disposals to land have not been included in this approach as these scenarios should automatically be considered for Stage 3 assessment.

The screening levels have been calculated by using cautious assumptions about the nature of the environments into which the discharges are made. Cautious estimates were made where data required for the calculations (e.g. biota concentration factor data) were unavailable. The method used to calculate the wildlife screening levels [3] takes account of generic factors such as the radiobiological effectiveness (RBE) of different radionuclides.

Where it has been determined from Stage 1 that the Stage 2 process should be carried out, the assessor will need to identify the RSA Authorizations (if more than one) which could impact on a European Natura 2000 site. The assessor will then need to add together the authorized discharge limits (TBq/y) from each relevant RSA93 Authorization, for each radionuclide discharged to atmosphere (terrestrial), river (freshwater) and sea (coastal marine) and compare these with the appropriate screening level. For example:

$$X_{\text{freshwater organisms}} = (\text{Discharge limit}_{\text{radionuclide1}} / \text{Screening level}_{\text{radionuclide1}}) + (\text{Discharge limit}_{\text{radionuclide 2}} / \text{Screening level}_{\text{radionuclide 2}}) + \dots + (\text{Discharge limit}_{\text{radionuclide n}} / \text{Screening level}_{\text{radionuclide n}}) / \text{Mean summer flow rate}$$

Note that the flow rate of the water determines the extent of rapid dilution. This is site specific and must be entered by the user. As a precautionary approach the mean summer flow rate is to be used (expressed m<sup>3</sup>/s, subject to an upper flow rate of 100 m<sup>3</sup>/s).

If  $X_{\text{freshwater organisms}} > 0.05$  then a further detailed Stage 3 radiological assessment is indicated to determine the impact of freshwater (river) discharges on the European Natura 2000 site.

Appropriate screening levels (TBq/yr) are shown in Table 2. These have been computed by pooling information from various sources [3], as shown in this schematic representation, Figure 1. As data gaps were envisaged, Table 3 identifies analogues for those radionuclides for which adequate data to calculate specific discharge thresholds are unavailable.

### 2.3. Stage 3: Appropriate assessment for activities which may have a significant effect

An appropriate assessment is required for any plan or project that is likely to have a significant effect on a European site, alone or in combination with other plans or projects, i.e. if it exceeds the threshold as laid out in Section 2.1 above. The Habitats Regulations do not specify how the assessment should be undertaken.

Existing assessment methods and standards focus primarily on measures for human radiological protection (see previously). Dose optimization philosophy has been based on the principle that human exposure should be kept as low as reasonably possible (ALARA). This is not necessarily the most effective means of achieving environmental protection. The Stage 2 screening procedure therefore has been modified to take account of recent guidance on protection of wildlife, resulting in appropriate assessment under the Habitats Regulations for a more detailed consideration of the effects on interest features which includes estimates of impact to wildlife.

TABLE 2. DISCHARGE SCREENING LEVELS FOR COMPARISON TO GUIDELINE DOSE LIMITS [1]

Radionuclide	Discharge screening level (TBq/yr)		
	Release to air <sup>a</sup>	Release to coastal waters <sup>b</sup>	Release to freshwater (river) <sup>c</sup>
Tritium (not OBT)	9E+03	2E+07	1E+05
Carbon-14	7E+02	8E+01	5E-01
Sulphur-35	1E+04	See Table 3	See Table 3
Strontium-90	5E+00	3E+00	2E-02
Technetium-99	–	3E+01	2E-01
Iodine-129	3E-01	3E+01	2E-01
Caesium-137	4E-01	1E+01	8E-02
Polonium-210	–	5E+01	2E-04
Radium-226	4E-04	–	–
Uranium-238	1E-03	2E-02	1E-04
Plutonium-239	2E-03	3E-01	1E-03

<sup>a</sup> Impacts on terrestrial organisms.

<sup>b</sup> Impacts on marine organisms.

<sup>c</sup> Impacts on freshwater organisms.

TABLE 3. RECOMMENDED ANALOGUES FOR RADIONUCLIDES WHERE NO SPECIFIC DISCHARGE SCREENING LEVEL IS SET

Radionuclide <sup>a</sup>	Analogue radionuclide		
	Release to air	Release to coastal waters	Release to freshwater <sup>d</sup>
Tritium – as organically bound	Carbon-14	Carbon-14	Carbon-14
Phosphorus-32/33	Caesium-137	Caesium-137	Caesium-137
Sulphur-35	Data available	Carbon-14	Carbon-14
Argon-41 <sup>b</sup>	Caesium-137	N/A	N/A
Cobalt-60 <sup>c</sup>	Caesium-137	Strontium-90	Caesium-137
Krypton-85 <sup>b</sup>	Caesium-137	N/A	N/A
Technetium-99m	Caesium-137	Caesium-137	Caesium-137
Ruthenium-106	Caesium-137	Caesium-137	Caesium-137
Iodine-125	Iodine-129	Iodine-129	Iodine-129
Iodine-131	Caesium-137	Caesium-137	Caesium-137
Uranium-234	Uranium-238	Uranium-238	Uranium-238
Uranium-235	Uranium-238	Uranium-238	Uranium-238
Other alpha-emitting nuclides	Radium-226	Uranium-238	Uranium-238
Other beta/gamma-emitting nuclides	Caesium-137	Strontium-90	Strontium-90

<sup>a</sup> The analogue radionuclides represent cautious choices based on the best available data and should be regarded as indicative only. Alternative analogues or approaches may be used but there must be a clear audit trail through the assessment.

<sup>b</sup> The analogue may be quite restrictive for argon-41 and krypton-85. Work is underway to provide data for these radionuclides. These radionuclides are not discharged to the aquatic environment and generally do not deposit to land. Hence exposure is via external irradiation only.

<sup>c</sup> Cobalt-60 has higher concentration factors than caesium-137 for the marine environment. Therefore the more restrictive strontium-90 analogue has been selected.

<sup>d</sup> Rivers only.

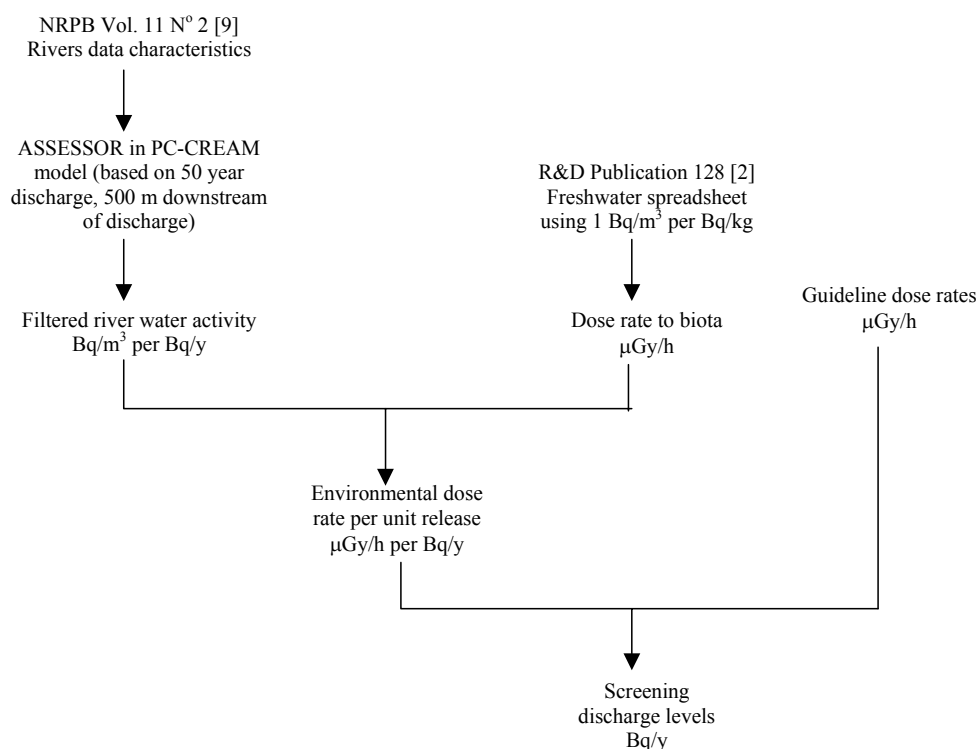


FIG. 1. Assessment Methodology to derive freshwater screening concentrations (rivers only).

The European FASSET (Framework for ASSESSment of Environmental impactT) programme is developing a framework for environmental assessments of radiation impacts with emphasis on biota and ecosystems. Until FASSET is completed, the Guidance Dose Limits presented in Table 1 should be applied in England and Wales to the most highly exposed members of the wildlife populations. Strictly, however, these values relate only to the exposure from the ionizing radiation and take no account of the different impact, either high (e.g.  $\alpha$ ) or low (photons and  $\beta$ -particles) LET (linear energy transfer, or ionization intensity) radiation. It is expected that equal absorbed dose rates of high LET radiation ( $\alpha$ -particles) would produce greater damage. There is no accepted procedure for combining the exposures of plants and animals from both low and high LET radiation (as can be done through the application of weighting factors for humans) to obtain a single measure of the biologically effective dose rate. The Environment Agency R&D Publication 128 adopts an interim approach to assessing the potential impacts from high and low LET radiation [2].

These generic approaches and limits can be used as an indication of the likelihood of an adverse effect but may need to be reconsidered in a site-specific context, as there is considerable variation in radiosensitivity between species and life stages. Where detailed assessments indicate that doses incurred by biota may approach the Guidance Dose Limits, the method adopted to consider high and low LET exposure should be justified.

### *2.3.1. Background exposure of qualifying features to radiation*

The appropriate assessment will also need to determine the contribution of the proposed plan or project to the radiation dose received by the qualifying feature compared with the natural background levels. Dose rates to plants and animals from natural background sources can be found [8], and the sources of natural radiation exposure include:

- Cosmic radiation. Since the components making up the cosmic radiation are penetrating in nature, the resulting exposure of terrestrial organisms living on or close to the surface of the earth will be similar to that determined for humans. For aquatic organisms, the absorption of the cosmic radiation flux through the overlying water can significantly reduce the dose rate to organisms from this source in mid-water and benthic habitats.
- Naturally occurring radionuclides in the surrounding environment. Exposure from this source depends on the local geology. In terrestrial environments for example radiation can build up in burrows. In marine and freshwater environments, the sediment is a more significant source of external natural exposure to benthic organisms than those radionuclides present in the water column.
- Naturally occurring radionuclides accumulated in the body via inhalation or ingestion with food and water. For plants, exposure is likely to reflect the local geology. For animals, exposure will depend on the degree of decay equilibrium and the feeding and behaviour of the species. It is not therefore valid to extrapolate data for humans to other organisms even though they may occupy the same environment. Two particular situations, which may be expected to result in enhanced natural exposure, are the ingestion of lichens by deer, and the occupation of underground burrows by mammals.

## 3. CONCLUDING REMARK

The above approach [1] is an interim means of assessing both Stage 2 (likely significant effect) and Stage 3 (appropriate assessment) until FASSET's recommendations become available in October 2003. It represents a pragmatic approach, which enables the Environment Agency, and English Nature, to fulfil its regulatory responsibilities, using the best available science to provide assurance that the environment is protected.

## ACKNOWLEDGEMENTS

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## **Regulatory assessment of risk to the environment: *Radiation***

**B.L. Dooley<sup>a</sup>, P.J.Colgan<sup>a</sup>, P.A. Burns<sup>b</sup>**

<sup>a</sup> Regulatory Branch, Australian Radiation Protection and Nuclear Safety Agency, Sydney, Australia

<sup>b</sup> Environmental and Radiation Health Branch, Australian Radiation and Nuclear Safety Agency, Melbourne, Australia

**Abstract.** 'Health, Safety and Environment' is a familiar catchphrase in today's government departments, universities and industry. This catchphrase is most often associated with terms such as 'risk assessment', 'risk management' and 'hazard identification'. Such grouping demonstrates the underlying assumption that 'risk(s)' to the environment can be dealt with, and are being dealt with, using the same processes as those used for evaluating health and safety risks. This equally applies when the hazard is radiation.

Management of risk to the environment is often carried out within a framework of an Environmental Impact Assessment (EIA) incorporating Risk Assessment (RA) processes. Environmental Management Systems (EMS) and Risk Management Systems (RMS) also provide frameworks which incorporate RA processes. These systems are often intergrated with existing quality and safety systems. The steps in these frameworks have been explained and comparisions have been drawn of how an EIA process and Environmental Management System compare with Environmental Risk Management processes. These comparisions demonstrate the common elements of each framework.

The Australian Standard for Risk Management (AS/NZS 4360:1999) describes the risk management process in terms of establishing a risk context, identification, analysis, evaluation and treatment of risks. The application of risk management procedures as described in the Australian Standard for Risk Management have been discussed in relation to how they might apply to a simple case scenario of a historical landfill containing radioactive waste.

### 1. RISK TO THE ENVIRONMENT AND THE ARPANS LEGISLATION

The object of the *Australian Radiation Protection and Nuclear Safety Act 1998* (ARPANS Act) [1] is to 'protect the health and safety of people, and to protect the environment, from the harmful effects of radiation.' In addition when the CEO of ARPANSA considers whether or not to issue a facility licence or a source licence to an applicant, he must take into account 'international best practice in relation to radiation protection and nuclear safety.' He also is required to take into account matters set out in the Regulations. One of these matters 'is whether the information establishes that the proposed conduct can be carried out without undue risk to the health and safety of people and the environment.' Whilst the Act sets out the matters that the CEO must have regard to, he is also able to take into account other matters that assist him to meet the object of the Act. It is not clear as yet how far this may go in relation to the environment.

In terms of radiation protection and nuclear safety a concensus has not yet been reached on international best practice with regard to protection of the environment. However, environmental risk assessment is currently carried out in Australia and world wide within several well established frameworks aimed at managing environmental risk. Often this occurs alongside management of risk to people. Following is a description of three of those frameworks and a hypothetical example of how a risk management process could be applied to a historical landfill containing radioactive waste.

Remediation of historic landfill sites is common practice in Australia, as it is world wide. Brisbane City Council [2] estimates on its website that it is currently 'remediating up to 150 old landfill sites.' Many government departments and environmental consultancies in this country have sections and departments dedicated entirely to management of landfill remediation.



In the field of health physics, risk management processes define acceptable levels of identified lethal and sublethal effects to people in the step of 'establishing the context.' Health physics assessments are then used in the 'analysis' stage to evaluate the level of risk to people. By inclusion of environmental considerations in 'establishing the context' and logically environmental analyses methods in the 'analysis' stage the risk management process may also be extended to include risk to the environment.

## 2. ENVIRONMENTAL CONSIDERATIONS; HOW ARE THEY DIFFERENT TO HUMAN CONSIDERATIONS?

When considering risk to people, risk assessors are considering the lethal and sublethal effects on human health. Over the last few decades the effects to the environment to be considered in environmental risk management have been documented in forums like the First United Nations Conference on the Environment (1972), the Second United Nations Conference on the Environment, 'the RIO Convention' (1992) and Earth Summit II (1997). Policy and working documents have been produced by government from international to local levels. Broadly speaking these documents have included a distillation of the principles of Ecologically Sustainable Development (ESD) as presented in *Agenda 21* [3], produced at the RIO convention. In Australia in 1992 the *National Strategy for ESD* [4] defined ESD as 'using, conserving and enhancing the community's resources so that ecological processes, on which life depends, are maintained, and the total quality of life, now and in the future, can be increased.' This strategy identified five key principles of ESD. ESD Principles identified by different government bodies vary in number and wording but appear to cover the same broad issues.

Principles of ESD, as identified by the EPCB Act [5], include the following:

- Intergration of both long term and short term economic, environmental, social and equitable considerations.
- **Precautionary principle.** Consideration of threats of serious or irreversible environmental damage. Lack of full scientific certainty should not be used as a reason for postponing measures to prevent environmental degradation.
- **Intergenerational equity.** The present generation should ensure that the health, diversity and productivity of the environment is maintained or enhanced for the benefit of future generations.
- **Biological diversity.** The conservation of biological diversity and ecological integrity should be a fundamental consideration in decision making.
- **Environmental economic valuation.** Improved valuation, pricing and incentive mechanisms should be promoted. The true costs of environmental impacts should be factored into the market.

This list is a distillation of a more comprehensive list from *Agenda 21* [3].

## 3. TAKING ENVIRONMENTAL CONSIDERATIONS INTO ACCOUNT

Although it is 'commonly assumed by the public and some individuals involved in site remediation that protection of human health will also result in protection of non-human species' [6], there are many examples of environmental objectives where this is not shown to be the case. In relation to protecting significant species, Suter [6] goes on to discuss the reasons many organisms may have a greater sensitivity to given contaminants than humans. These included greater exposure because pathways are different, inherent sensitivity to a particular contaminant for a given species and secondary effects such as loss of primary production. In measuring the effects on human health, consumption models are often used. For example, in the case of contaminated water, it may be calculated that 2 litres of water per day, with a particular level of a given contaminant would give a statistically defined risk in terms of death or sublethal effects. The information these models are based on (information related to toxicology and epidemiology) is most often not available for significant species and, because of the large number of species and diverse routes of exposure which may be encountered, it is unlikely that it will be available in most circumstances. For this reason, analyses of effects on significant species and biological diversity depend on approaches where real responses from real receptors are detected or not.

Environmental criteria, and therefore endpoints, related to the principles of ESD are not inclusive of protection of significant species and biological diversity. Examples of other criteria include protection of a non-physical component of the environment such as the soil for use by future generations, avoidance of acid rain, prevention of release of new chemicals which 'may' cause harm, and prevention of unjustified economic cost of future remediation. According to Harding [7], maintaining intergenerational equity, in the case of a landfill, may mean that 'the present generation manages production of waste in a way that there is no cost incurred to future generations, in terms of loss of environment quality and/or costs of remediating degraded environmental resources.' Analyses of the impact of waste production could then be in terms of waste minimisation and long term disposal strategies. So it is important to identify criteria and objectives which relate to ESD principles for a given practice before deciding on methods of measuring risk.

#### 4. SOME FRAMEWORKS USED TO ASSESS RISK TO THE ENVIRONMENT

The framework provided in the *Australian Standard for Risk Management* [8] is recommended as a generic tool that can be used across a variety of industries and economic sectors to identify opportunities and mitigate losses. An Environmental Impact Statement (EIS) is, according to Thomas [9], a tool to 'help us make decisions about whether we can live with the consequences of proceeding with an action.' An EIS is the highest level of environmental impact assessment reporting which can be required in Australia for new proposals. An Environmental Management System (EMS) as described by *AS/NZ ISO 14001/ 1996, Environmental management systems – Specifications with guidance for use* [10] is a tool used to control the impact of activities, products and services on the environment. These frameworks are tools used to provide information about the consequences of actions that can be used to determine whether the actions are appropriate.

The requirements of *ISO 14001* [10] have been compared by National Association of Testing Authorities (2001) [11] to the ISO 9001-2000 quality system standard [12] and the Australian Safety Standard 4801:2000 [13] demonstrating that compliance with this standard would comfortably fit in with the frameworks commonly used for safety and quality.

##### 4.1. The Australian Standard for Risk Management [8]

The Australian Standard for Risk Management, AS/NZS 4360: 1999 [8] prescribes six elements in the risk management process. These elements are linked as an iterative process by 'monitoring and review' and 'communicate and consult' steps as shown in the flow chart in Figure 1.

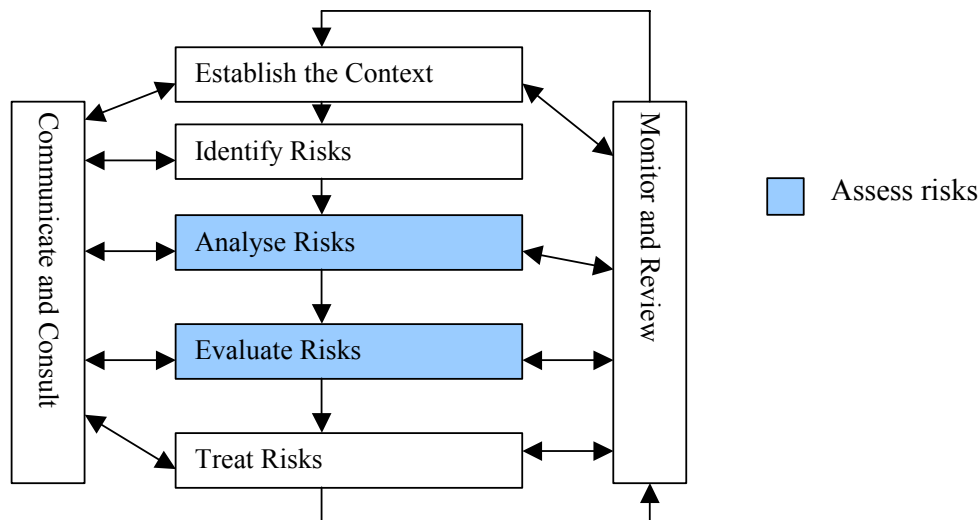


FIG. 1. Risk Management Overview (from AS/NZS 4360:1999 [8]).

The following descriptions of each step are adapted from the AS/NZS 4360:1999 [8]:

- **Establish the context.** In this step risk managers should look at the strategic, organisational and risk management context in which the process will take place. It is important to determine who the audience is. At this stage it is appropriate to decide criteria against which risk will be evaluated. In the case of environmental risk management these criteria could be precipitated from the principles of ESD. A risk management committee should comprise representation of all stakeholders and relevant technical experts.
- **Identify risks.** It may be more appropriate to identify hazards and associated risks. The standard defines hazard as 'a source of potential harm or a situation with a potential to cause loss'. Whilst risks are defined as 'the chance of something happening that will have an impact on objectives.' (Risk is measured in the 'analyse risk' step in terms of consequence and likelihood.) This step could be described as 'scoping'. It is where the assessors delineate what, why and how hazards may arise. Methods for scoping include such things as lists, brainstorming, surveys and flashcard associations (as used in HAZOP).
- **Analyse risks.** In this step a quantitative or qualitative analysis of the frequency and severity of the risks is carried out. This is the step which might involve tools such as pathway analysis or pharmacokinetic modelling. Level of risk should take into account the consequence and likelihood of risks.
- **Evaluate risks.** In this step a comparison of estimated levels of risks (made in 'analyse risks') is made against the pre-established criteria (determined in 'establish the context').
- **Treat risks.** Low priority risks may be accepted and monitored. For significant risks a specific management plan should be developed. Considerations should be given to development of a management plan including level of risk, financial and socio/political considerations.
- **Monitor and review.** Monitoring results for all risks are interpreted and the management plan is reviewed as considered appropriate.
- **Communicate and Consult.** This is carried out with all stakeholders at all stages of the process as is considered appropriate.

Note: The terms 'risk management' and 'risk assessment' are often used interchangeably across disciplines and borders. In this paper 'risk assessment' refers to the 'analyse risk' and 'evaluate risk' steps of 'risk management'.

#### 4.2. The Australian/New Zealand Standard for Environmental Management Systems [10]

The *Australian/New Zealand Standard Environmental management systems – Specifications with guidance for use* [10] has been reproduced directly from ISO 14001(Int):1995. ISO 14001 is widely used internationally.

Continual improvement is made through changes arising from management reviews as a result of feedback from all stages of the EMS process:

- **Identify aspects and impacts.** An 'aspect' is an element of an organisation's activities, products or services that can interact with the environment. An 'impact' is an adverse or beneficial change to the environment.
- **Identify legal and other requirements.** These might include requirements of various legislation and/ or agreements. They would also include government planning documents.
- **Determine significance of environmental impacts.** The organisation must have a procedure in place to determine which of its aspects can have a significant impact on the environment. This would be carried out against criteria determined by the risk management team. The principles of ESD may be used to precipitate these criteria.

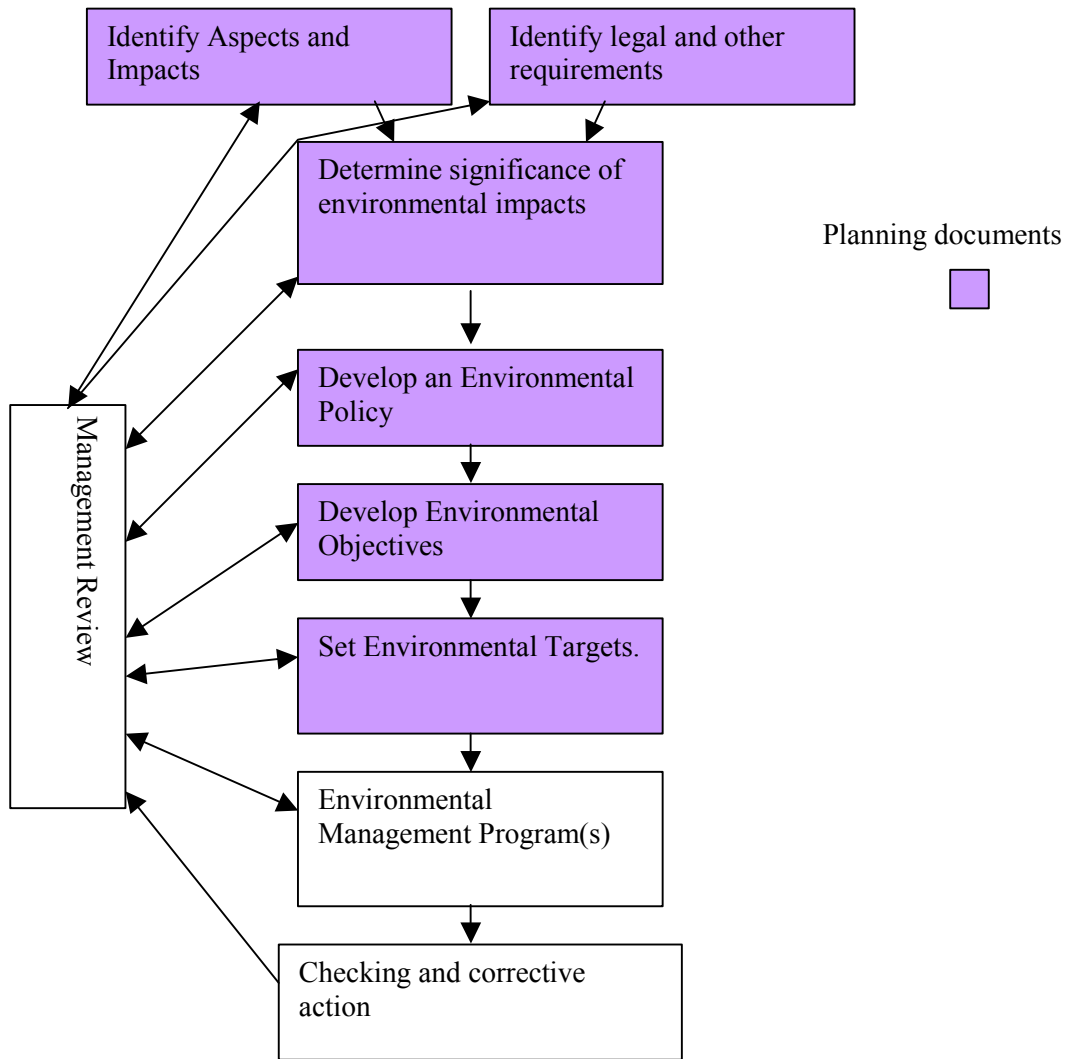


FIG. 2. Maintaining an environmental management system overview.

- **Develop an environmental policy.** This should be appropriate to the nature, scale and environmental impacts of its activities; include commitments to continual improvement, prevention of pollution; and compliance with relevant environmental legislation, regulations and with other requirements to which the organisation subscribes. It should provide the framework for settling and reviewing environmental objectives and targets and be documented, implemented and maintained and communicated to all employees.
- **Develop environmental objectives.** These must consider the legal and other requirements, technological options and its financial, operational and business requirements, and the views of interested parties. Environmental objectives must be documented.
- **Set environmental targets.** The targets should achieve the objectives and be consistent with the environmental policy.
- **Environmental management program(s).** These programs should implement activities which achieve the environmental objectives and targets.
- **Checking and corrective action.** The environmental monitoring regime should demonstrate that the objectives are being achieved and targets are being met. If not, corrective action should be implemented.

### 4.3. Environmental impact assessment

Environmental impact assessment is a tool which is commonly used to decide whether or not a project is viable with regard to its effect on the environment. Steps involved in the EIA process as described by Wood [15] are shown in Figure 3. A brief description of these steps follows:

- **Relating the concept to broad environmental policy and criteria (eg SEA).** Strategic Environmental Assessment (SEA) is the process of taking the environment into account earlier in the planning process. This should include looking at the cumulative effects of development and broad policies such as the greenhouse policy and environmental planning documents. Stakeholder involvement should be considered at this time.
- **Incorporating environmental considerations into the action's design and planning.** In this stage it is important to define key environmental objectives which may be distilled from the ESD and other environmental principles.
- **Developing a proposed course of action.** In this step a proposal is put forward including actions and timeframes.
- **Determining the need for an EIA (screening).** In Australia the *Australian and New Zealand Environment and Conservation Council (ANZECC) Guidelines and Criteria for Determining the Need for and Level of Environmental Impact in Australia* [14] provide criteria (including checklists) to determine the need for EIS. These criteria include:
  - (1) 'The character of the receiving environment.
  - (2) The potential of the proposal.
  - (3) Resilience of the environment to cope with change.
  - (4) Confidence of prediction of impacts.
  - (5) Presence of planning or policy framework or other procedures which provide mechanism for managing potential environmental impacts.
  - (6) Other statutory decision making processes which may provide a forum to address the relevant issues of concern.
  - (7) Degree of public interest.'
- **Determining the coverage of an EIA (scoping).** This is where important environmental factors to be taken into account are decided. The form of an EIS and other EIA will be prescribed by relevant government organisations.
- **Describing the action and the environment.** This should include a full description of the activity including its objectives, the existing environment including air, water, soil and biota.
- **Predicting the impacts (magnitude and significance).** This should include an indication and analysis of the likely interactions between the proposed activity and the environment.
- **Evaluating the impacts.** Justification of the proposed activity should be given taking environmental considerations and likely/ plausible impacts into account. An assessment should be made of measures to be implemented to protect the environment and their likely effectiveness. Viable alternatives to the proposal should be described and considered with respect to the proposal put forward.
- **Reviewing the EIS and preparing the assessment report.** The EIS is reviewed against a variety of objectives depending on the government department(s). These may include meeting environmental social, political and economic objectives. The and depth and adequacy of technical detail would be assessed at this time. The report produced should be based on information taken from the EIS, public comment, comment from government departments and additional information supplied by the proponent.
- **Deciding about proceeding with the proposal perhaps with conditions.** The decision and additional conditions would be guided by the assessment report.
- **Monitoring impacts form the action.** Environmental monitoring should confirm predictions of impacts or trigger further decision making in the management of the proposal. This would ideally be carried out in an iterative EMS framework.

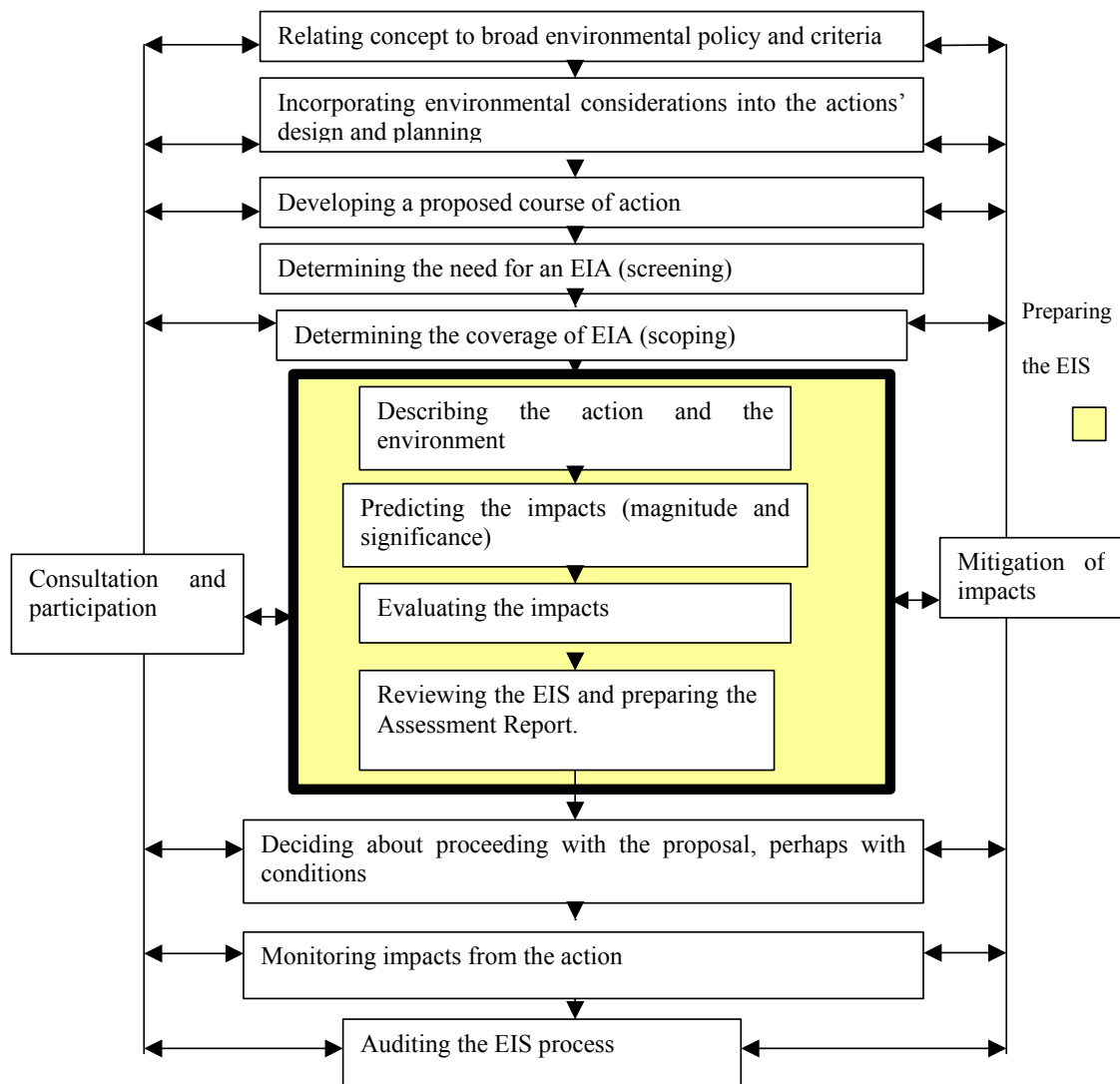


FIG. 3. Environmental Impact Assessment Process (adapted from Wood [15]).

- **Auditing the EIA process.** The auditing process should identify whether or not predicted impacts have occurred, whether methods used to make these predictions were reliable, whether recommendations were followed and whether safeguards were effective.
- **Consultation and participation.** This should be done at all stages of the process and involve all stakeholders. It may incorporate public information sessions, surveys, meetings and any other effective means of involving all stakeholders.
- **Mitigation of impacts.** This is an iterative step which should occur at any stage. Where an undesirable impact is identified steps should be taken to mitigate the impact.

## 5. COMPARISON OF THREE FRAMEWORKS

Table 1 shows that the same or similar broad steps are included in all three frameworks. Although the flow charts show steps in these frameworks to be in different order there are many common elements. One common element in each process is the iterative nature of steps corresponding to the 'communicate and consult' and 'monitor and review' processes.

It is recommended that the EIS process is audited, however, in practice this step is often not carried out. Thomas [9] concludes that for this reason the EIS process is often not as iterative as it could be.

TABLE 1. COMPARISON OF RISK MANAGEMENT, ENVIRONMENTAL MANAGEMENT AND ENVIRONMENTAL IMPACT ASSESSMENT

	AS/NZS 4360:1999	ISO 14001	EIS
	Establish the context	Develop an environmental policy Develop environmental objectives Set environmental targets.	Relating the concept to broad environmental policy and criteria Incorporating environmental considerations into the actions design and planning Develop a proposed course of action Determine the need for an EIS screening
	Identify risks	Identify aspects and impacts Identify legal and other requirements	Describing the action and the environment.
	Analyse risks	Checking and corrective action?	Predicting the impacts (magnitude and significance).
	Evaluate risks	Determine significance of environmental impacts	Evaluating the impacts. Reviewing the EIS and preparing the Assessment Report.
	Treat risks	Environmental management program(s)	Mitigation of impacts Deciding about proceeding with proposal perhaps with conditions.
Iterative	Communicate and consult	Management review	Consultation and Participation
Iterative	Monitor and review	Checking and corrective action	Monitoring impacts from the action Auditing EIS processes

### 5.1. Application of a risk management process to a historical landfill containing radioactive waste

Following is a description of how the risk management process could be applied to a historical landfill containing radioactive waste. For the sake of simplicity only two criteria will be identified and discussed in terms of subsequent steps of the project. For a comprehensive risk management process a number of criteria would usually be identified.

In 'establish the context' the information to be considered includes:

- Physical site information including physiographic setting, climatic and hydrology information.
- The inventory of material disposed of at the site. Chemical/ radiological form, toxicity and transport parameters. Any other relevant information such as the origin of the waste.
- Location of the site. What is the site used for? What could it be used for in the future? A description of surrounding land use and possible future surrounding land use should be included.
- Identification of stakeholders. Who is legally and/or otherwise responsible for the site? Who is the regulator? Who are the surrounding landholders? Are there any local community/ environmental groups?
- Identification of all applicable legislation, policy documents and other agreements.
- Criteria against which risk will be evaluated. The criteria could be precipitated from ESD principles, but should be quantitative. For example, considering the ESD principle of 'intergenerational equity.' The following criterion might be identified:
  - Criterion 1 -'preservation of groundwater quality to the level of use for which it is currently viable.'
- When considering the ESD principle of 'conservation of biological diversity', the criterion might be:
  - Criterion 2 -'maintenance of the current level of species diversity with regard to flora.'

TABLE 2. COMPARISON OF RISK MANAGEMENT STEPS WHICH MAY FOLLOW ESTABLISHING THE CONTEXT FOR CRITERIA 1 AND 2

Risk Management Step	Criterion 1	Criterion 2
Establishing the context	ESD principle – ‘intergenerational equity’. Related criterion ‘preservation of groundwater quality to the level of use for which it is currently viable.’ Note: Assume groundwater is currently potable in the aquifer of concern and hydrogeologically connected aquifers.	ESD principle – ‘conservation of biological diversity’. Related criterion ‘maintenance of the current level of species diversity with regard to flora.’
Identify risks Hazard – movement of radionuclides from containment.	Degradation of ground water.	Loss of plant species.
Analysis of risks The level of these risks should be given a risk ranking which relies on frequency and severity of the risk. Note: good baseline data is important in confirming predictions.	Contaminant transport modelling Physical analysis of groundwater samples.	Literature surveys of the toxicology or epidemiology individual or similar species. Comparative surveys of biological diversity both on site and in surrounding areas.
Evaluate risks Comparisons of estimated levels of risks (made in ‘analyse risks’) is made against the pre-established criteria (determined in ‘establishing the context’).	Does the predicted or confirmed level of contaminant in the groundwater fall below drinking water standards. Note: It is important to remember that other criteria such as protection of a significant species may require a more restrictive assessment of water quality.	If a loss of species diversity is predicted or measured then criterion one cannot be satisfied. This is an unacceptable outcome which needs treating.
Treat Risks This step must be undertaken unless criteria are considered to be satisfied.	Implement plan of management. Treatment options may include stabilising the waste, isolating the waste or removal of the waste.	Treatment options for loss of species diversity may be removal of the waste or isolation of the waste from species concerned.
Monitor and review	A monitoring regime is established to confirm results of previous analyses, predicted contaminant transport and find any unexpected anomalies.	A monitoring regime is established to confirm results of previous analyses, predicted species diversity and find any unexpected anomalies.
Communicate and consult	Throughout the process, if discrepancies are found they should be communicated and consulted at an appropriate level and further action taken as deemed necessary. Any decision for action or no action should be justified and documented.	

Importantly this process is iterative. The 'monitor and review' and 'communicate and consult' processes should continue until the hazard is removed.

## 6. CONCLUSIONS

ARPANS legislation requires assessment of environmental risk management with regard to radiation. There is no agreed international best practice in the fields of radiation protection and nuclear safety with regard to protection of the environment. However, this is not the case in the general field of environmental protection where the principles of ecologically sustainable development are widely used to precipitate environmental objectives and criteria aimed at protecting the environment in industries and government. It has been proposed that the principles of ESD are equally relevant to radiation.



Well established frameworks for management of environmental risks used in the field of environmental protection have been discussed and compared. This comparison showed that the same broad steps are common to the frameworks discussed. A simplified description of how the risk management framework could be applied to a historical landfill containing radioactive waste has been put forward. The authors of this paper consider that using the existing frameworks for environmental management to protect the environment from the harmful effects of radiation would be the most sensible available option and would avoid duplication of effort for regulators and proponents.

### ACKNOWLEDGEMENTS

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### **3. METHODS AND MODELS FOR EVALUATING RADIATION AS A STRESSOR TO THE ENVIRONMENT**

# Evaluating the effects of ionising radiation upon the environment

**R.J. Pentreath**

Environmental Systems Science Centre, The University of Reading, Reading, United Kingdom

**Abstract.** As a result of several recent initiatives, there is now a reasonably well-established international appreciation of the need to evaluate the effects of radiation on the natural environment in a more explicit and comprehensive manner [1–8]. This need derives not only from what are seen as the inadequacies that result from simply assuring the protection of human beings, but also from a need to satisfy the broader ethical, social, and various forms of legal requirements for the demonstration of adequate environmental protection. In this respect, therefore, because a very wide and disparate range of such requirements already exists, and are likely to continue to arise, it is important to try and maintain some sort of clarity about the different objectives that one might be trying to achieve in any environmental protection evaluation exercise, and how best to achieve them.

The presence – or even the risk of the presence – of radionuclides in the environment as a result of human activity is a very emotive subject, and is likely to remain so. It is therefore all the more important for the ‘protection’ of the environment to be based upon a set of international protocols, and standardised procedures and data bases – as has been the case for the protection of man – that are free from political interference or pressure, either in relation to national bodies or to specific pressure groups. In this regard, suggestions have already been made for the establishment of a ‘system of protection’ that would parallel, and indeed could be combined with, the ‘system’ that has been developed to protect human beings under different circumstances from different sources of radiation [9–11].

This suggestion has been supported by the IUR [12], and the concept is being actively researched for regional application in Europe and the Arctic [13] as well as at a national level [14], and has been discussed by an ICRP Task Group [8]. This paper therefore attempts to explore some of the means by which the elements of such a system could be applied, because in order to evaluate environmental protection it is first necessary to make assessments, and to make them numerically, and in their proper context.

## 1. INCREASING SOCIETAL DEMANDS FOR ENVIRONMENTAL PROTECTION AND THEIR IMPLICATIONS FOR RADIATION CONTROL

The system of human radiation protection initially developed in response to very real and practical circumstances, as both the scientific and medical communities appreciated the harmful effects, and the potential benefits, of exposure to radiation. These circumstances were those arising from a working environment, or in relation to medical care. They were therefore not concerned primarily with exposures of the public through environmental pathways of radionuclide transfer. Such circumstances, and approaches for dealing with them, arose later. But the focus has remained on protection of human beings, and the notion arose that, as humans are part of the environment anyway, delivering the level of protection afforded to them would suffice, in some way, for the protection of other living components of it.

At the same time, the need to protect other biotic – and abiotic – components of the environment has developed in ways that deal more directly with the many threats to them. Thus, in contrast to the case of exposure to radioactive materials, concerns about pollution from other chemicals has usually centred upon what fauna and flora are most likely to be exposed to them, and thus affected by them, and therefore about any resultant ecological consequences. In this respect it has not gone unnoticed that, with regard to the deliberate release of radionuclides into the environment, human beings probably count amongst those fauna that are least likely to be exposed and, if exposed, are likely to be exposed at the lowest dose rates. This has been shown to be the case in modelling exercises [15], and environmentally in those few circumstances where actual measurements have been made [16–18].

Concern for the biotic components of the environment has also evolved in other ways, with a greater emphasis on the direct protection of species and habitats from all or any threats, not only from pollution. Indeed, public attitudes to environmental matters, together with a greater scientific understanding of wildlife and the environment generally, have also continued to evolve, and this

evolution has ultimately been reflected in political responses to all of these concerns, some of which have been translated into new legal requirements.

Recent developments in this evolving ethical background to environmental protection, and its reflection in international law, have been reviewed by the IAEA [4] and need not be repeated here. But the result is that the general subject of environmental protection now covers many topics, and has many facets to it. Managing the interfaces between them is often considered to be largely a matter of identifying what level of ‘protection’ is required under different circumstances, but it also involves a deeper understanding of more basic issues, including the various ethical and moral considerations that underlie the origin and practical consequences of different forms of agreements and legislation. In this respect, therefore, it is sometimes useful to differentiate amongst different forms of environmental management, such as ‘*environmental exploitation*’, ‘*pollution control*’ and that which may generally be referred to as ‘*nature conservation*’ [19], as follows.

### 1.1. Environmental exploitation

Exploitation of the environment, as in such practices as fisheries, forestry, and agriculture, takes for granted the fact that the environment will be ‘damaged’ in that individual animals or plants will die. Its relevance to environmental protection, however, is that the objective is usually to ensure that the practice can be carried out in a sustainable way, and although it is essentially concerned with effects on the environment at the ‘population’ level, it may also be concerned about the genetic ‘integrity’ or ‘stability’ of those populations.

### 1.2. Pollution control

Pollution control is usually concerned with protecting the environment generally from specific pollutants. The requirements – to take some European examples – are often couched in terms of having to take steps or measures in order to prevent *pollution of the environment* (that is to say, something that is *harmful* to the *quality of the environment* [20]) or, more explicitly, by referring to pollution as being the causing of “..*harm to man or any other living organism...*” where *harm* means “..*harm to the health of living organisms or other interference with the ecological systems of which they form a part*” [21]. Elsewhere, as for example in Canada, industrial activities may be constrained to ensure that they do not present ‘*unreasonable risks to the environment*’ [22]. Pollution control can be taken to include control over sources of chemicals from a specific practice, from a specific location, or from a specific area – such as contaminated land. Control is usually exercised by way of requiring specific and auditable actions to be undertaken, and by the setting of numerical values in relation to emissions and one or more components of the environment that are not to be exceeded (environmental quality standards).

### 1.3. Nature conservation

In contrast, the objectives for nature conservation are usually to protect specific species, habitats, or areas from threats (including pollution) generally, and are thus framed in other forms of legislation. This “*nature conservation*” legislation is often necessarily less precise, but has essentially stemmed from the following three, broad, requirements:

- the *conservation* needs of particular species or areas, where the term ‘conservation’ usually implies active management of a situation to achieve a particular objective and includes the term *preservation*, which usually implies the need to maintain the *status quo* absolutely, and is therefore usually applied to inanimate components of the environment;
- the *maintenance of biological diversity* (‘biodiversity’) which is usually construed to include biodiversity *within* species (i.e. the morphological and physiological variations to be found within a particular species); biodiversity *amongst* species (i.e. the overall number and variety of species); and the biodiversity of habitats (i.e. the number and variety of species present *in a* particular habitat and *amongst* different habitats); and
- the protection of particular types of *habitats*.

Both conservation and the maintenance of biodiversity take note of the need to protect the abiotic as well as the biotic components of the environment, but the concept behind habitat protection recognises the fact that *habitats* (both abiotic and biotic components) need to be protected from direct and indirect pressures, even though their specific faunal and floral assemblages may continually vary and be primarily affected by events outside the habitat. Similarly, biological diversity is not a static entity, but the aim is to ensure that it is allowed to develop without avoidable and undue human interference.

An example of the implications of all of the above is again provided by some European Directives. Two of them, in relation to particular species and habitats, collectively require that steps be taken to ensure that designated areas are maintained in, or restored to, “*favourable conservation status*” [23, 24]. This ‘status’ may be differently, and explicitly, defined for each and every site in a numerical way – such as percentage changes in the numbers of certain species, ratios of different species to each other, age structures of populations of species and so on. Similarly, a third Directive requires action to be taken to ensure “*good ecological status*” of aquatic ecosystems [25]. It will probably therefore be necessary to demonstrate in all of these cases that controllable activities would not have a detrimental effect on such factors, as variously defined for specific locations. For the possible effects of radiation, in relation to such ‘wildlife’ or pollution control legislation, it has therefore been suggested that these could usefully be compiled in terms of different broad categories, such as *early mortality*, *reduced reproductive success*, and “*scorable*” *cytogenetic effects* [10].

## 2. DIFFERENT ASSESSMENT AND MANAGEMENT REQUIREMENTS

It is immediately apparent from this brief summary of these different approaches to environmental management that there are clear – and often contradictory – aspects about them. But for the purposes of this paper, it is also important to note that the specific requirements relating to any of them will also differ considerably. And all of these subject areas are continuing to develop at an international level. Thus the need to make evaluations of the impact of radiation on the environment, now or in the future, might arise for reasons that stem from any or all of the above environmental management requirements, but particularly in relation to pollution control and nature conservation. The practical consequence, however, is that this need may now include any of the following *objectives*, each of which would need to be expressed, and deemed ‘acceptable’ or otherwise, in different ways:

- *assurance* of the public or their politicians, at national or international level, of the likely environmental impact of any actual or proposed specific practices, and demonstration of the ability to deal with any consequences if it all goes wrong;
- *compliance* with the spirit or the letter of *trans-national* general *pollution* or *wildlife-protection obligations*;
- *compliance* with *national pollution control licensing requirements* relating to particular industrial practices or to specific sites or areas; or
- *compliance* with the requirements of specific *national wildlife and habitat protection legislation*.

Common to all of them, however, is the process of having to *assess* the situation, to analyse its component parts and then, if necessary, consider the various options for *managing* whatever situations may arise. This is particularly important when attempting to understand the purpose of the environmental evaluation, because each component may need to make use of completely different approaches and interpretations. But what should also be common to both assessment and management is the basic scientific understanding, plus the means of expressing and using the relevant scientific information. This has been the basis of success for the radiological protection of humans, and therefore needs to be carefully considered with respect to protection of the environment generally.

For the purpose of *pollution control*, the above protection *objectives* may, in turn, require the explicit demonstration of:

- the protection of the public;
- the avoidance or minimisation generally of harm to the environment; or
- the ability to deal with the environment that is already harmed.

And, for the purpose of *nature conservation*, the above protection *objectives* may, in turn, require assessments to be made of:

- the likelihood of harm to individuals of particular species;
- potential or actual effects on populations of one or more species, in terms of population integrity and viability (this would also apply to environmental exploitation);
- potential or actual effects on the principal (or majority) components of a specific habitat, or at a specific place; or
- potential or actual effects at ecosystem level, within a local area or more generally, but without specific reference or preference to any particular faunal or floral type.

And there may be other considerations, as where the mere *presence* of radionuclides, “contaminating” an area, may be of concern to certain individuals or sectors of the public for ethical, moral, or social reasons [4].

### 3. GENERAL REQUIREMENTS FOR ENVIRONMENTAL EVALUATIONS

In order to make an evaluation of the effects of radiation on the environment itself, with respect to any particular situation or practice, there are clearly several basic requirements to consider including:

- the radionuclides of interest, their sources, and their rates of introduction into the environment; and
- the distribution and fate of those radionuclides in the environment.

This basic information is also required in order to protect the general public. Many numerical models therefore exist that can be applied to different practices, situations, and ecosystems. Their focus has been on the fate of radionuclides in the environment with respect to their potential to deliver doses to different members of the public in relation to what they do and what they eat. An assessment can then be made of the ‘risk’ to people (usually via the risk of not complying with the legislation that exists to protect them) and thus how best to manage the situation.

But for environmental protection purposes the ‘questions’ are different, and thus other information is necessary including:

- the potential exposure to radiation of the fauna and flora within the area of radionuclide distribution; plus
- the likely consequences for them, in terms of radiation effects.

Of these two, being able to address the former should not be too difficult, the nature of the problem having much in common, again, with the environmental information needed for human radiation protection. The latter, however, is more difficult, and the term ‘consequences’ is far more open-ended than it is for human protection; many other factors therefore need to be considered, not least the original objectives of the assessment.

#### 3.1. Points of reference

All of this environmental information, of course, is only of value in the case of protection of the public because it can then be related to basic reference sets of biological and radiobiological information that exist for human beings. Indeed, all assessments with respect to the biological effects of human exposure to ionising radiation have, for many years, been based on the concept and use of a *Reference Man*. It has therefore been proposed that a similar concept – that of a system using discrete and clearly defined *Reference Fauna and Flora* (RFF) – could also be useful for environmental protection [9–11, 19]. This use of reference fauna and flora is **not** to suggest that such entities are the **only** objects of protection *per se*, (anymore than it is for a thirty year old Caucasian – the basis for Reference Man) but that they could provide the basis for extrapolation, interpolation, and for developing other frameworks and applications, as deemed necessary, through secondary and tertiary levels of development and interpretation – in much the same way as the numerical limitations

currently placed upon discharges from nuclear installations are, ultimately, traceable back to numerical values (dose limits) that are based on models and data that relate to Reference Man.

### 3.1.1. Primary Reference Fauna and Flora

Essentially, therefore, this approach first envisages the establishment of a (preferably international) *Primary* set of Reference Fauna and Flora. The purpose and objective of the *Primary* set would be to obtain as complete a data base as possible, and as complete an understanding as possible, of the basic biology and the doses that could be received by, and the resultant *effects* of radiation on, a limited set of faunal and floral types.

The effects' data would need to be set out initially in terms of, at least, the categories of *early mortality*, *reduced reproductive effects*, and *scorable cytogenetic effects* for each faunal and floral type – possibly relative (in orders of magnitude) to background radiation in the form of *Derived Consideration Levels* [11]. Thus logarithmic increments in dose above background would be related to known radiation effects for those types of fauna and flora. This would allow such information to be interfaced with other data sets in order to assess and analyse different situations, thereby enabling a reasonable degree of flexibility in their use and interpretation for different kinds of environmental protection assessments and control, or even for incorporation into legislative frameworks. It would also allow environmental radiation protection to be addressed in a similar manner to that currently being discussed for human radiation protection.

Criteria that might be considered for the selection of Primary Reference Fauna and Flora would necessarily include the ability to identify any adverse effects (at the level of the individual organism) that could be related to radiation exposure; the amount of radiobiological information that is already available on them, including data on probable radiation effects; and their amenability to future research in order to obtain the necessary missing data on radiation effects [26].

Once selected, such Primary Reference Fauna and Flora would still need to be described. Such description, in taxonomic terms, is necessary both to assign existing data sets to them with different levels of confidence (depending on the extent to which they have been extrapolated or interpolated from other faunal or floral types) and in order to target further research to provide the necessary missing information. In this respect, therefore, it is necessary to define a taxonomic level that is not too narrow, because of such factors as limited geographic range of whatever is selected, and the limited data bases that exist or could be obtained. Equally, however, it is necessary not to be so broad that almost any existing data, or any future experiments on any type of animal or plant, could serve as the reference data set. Thus characterisation at the level of *species* or *Genus* would appear to be too narrow; whereas *Order* or *Class* is probably too broad for the basis of future research work. This leaves *Family* as about the correct level for a wide range of organisms, although this would have to be justified in each case.

### 3.1.2. Secondary Reference Fauna and Flora

It is however unlikely that the sole use of such a limited set of Primary Reference Fauna and Flora would of itself serve to satisfy all assessment needs – any more than the strict application of the thirty year old Caucasian Reference Man data set does in the case of humans. Thus one might imagine that the use of 'secondary' sets would probably be required where, for example, there was a need for:

- a greater *overall range* of faunal and floral types of organisms in the assessment exercise;
- *locally characteristic* types of fauna and flora for particular ecosystems, either in terms of habitats (forests, freshwater lakes), or with respect to particular geographic regions or areas (e.g. the Arctic, or temperate Europe); or
- *very specific faunal or floral types* (e.g. in order to satisfy or comply with specific 'nature conservation' legislation).

The *differences* between the *Primary* set and the *Secondary* sets would therefore be either:

- (a) *biological*, in terms of life history, life span, habitat, diet, physical dimensions, position in food chain, population dynamics, ecological niche and so on; or

- (b) *radiobiological*, in terms of the expected external exposure, the accumulation of radionuclides, and the likely responses to the radiation doses received in terms of observable or expected effects.

Differences in either biological or radiobiological factors between the *Secondary* and the *Primary* Reference Fauna and Flora would affect assessments of exposure to radiation in any particular circumstance, and thus the dose received by them and the potential consequences. The principal difference of relevance, however, is the fact that although it is likely that a reasonably-comprehensive set of '*biological*' information may be obtainable for the *Secondary* set, the '*radiobiological*' information is likely to be far less complete, and may well be virtually impossible to obtain (e.g. a knowledge of radiation dose-effects for marine fauna, large mammals and so on). The '*secondary*' data would therefore usually be both less complete overall, as well as at variance from the '*primary*' data set with regard to particular properties or values. Nevertheless, one would be able to evaluate, numerically, the likely differences (the deviations) between the two, both in terms of exposure and in terms of *Derived Consideration Levels*. (This, again, is no different from what is effectively done for human radiation protection in terms of deriving '*secondary*' data sets for Reference Man in relation to age, size, race, diet, exposure to different chemical forms of radionuclides and so on, together with interpolations with respect to a knowledge of radiation effects.)

Where necessary, one might also require what one might regard as '*Tertiary*' sets that related to *actual* fauna and flora, applicable to certain locations, or sites, under specific circumstances.

### **3.2. Applications**

The manner of application of a Reference Fauna and Flora approach will clearly depend upon the objectives of the evaluation exercise – essentially, with respect to what particular question one was trying to answer. This could be a general one, or relate to specific legal requirements. Thus it might be sufficient to demonstrate that an '*evaluation*', '*appraisal*', '*assessment*' or whatever had been done: for example, in relation to the provision of public or political reassurance, or in response to '*what if*' questions in a public inquiry. Equally, however, it may be necessary to demonstrate how particular situations are to be handled, or how compliance with existing or forthcoming legislation is to be achieved. As already indicated, this latter requirement may relate to '*pollution control*' or '*nature conservation*' legislation, or to both – in addition to protection of the public.

The fundamental issue, therefore, is not simply to determine what levels of radiation are '*safe*' for the environment, because that always leaves open the question of what the consequences might be if such levels were to be exceeded – the answer to which would depend upon many other pieces of information. The basis purpose of *Derived Consideration Levels* is a more basic one: to derive and organise information to help manage any situation, as is the case for the management of human exposure.

The '*success criteria*' could therefore also be different. Success in terms of pollution control would be measured in terms of what was achieved with regard to management of the source, its containment, behaviour and so on. Success in terms of nature conservation would be measured in terms of the properties (health) of the individuals, populations, ecosystems or habitats that were the object of original concern – and for which any particular threat (such as radiation) may only have been one amongst many.

A range of numerical values might therefore be necessary to deal with different situations, or to answer different questions – as, again, is the existing case for human radiation protection. The set of *Derived Consideration Levels* for different fauna and flora could therefore be used to evaluate different situations, or to inform the development of different management strategies. It would also enable the origin of different numerical values used in legislation or decision making to be transparent, and allow for revisions of them as new data, or new interpretations of existing data, arise.



### 3.2.1. *Environmental exploitation*

Environmental exploitation, by its very nature, impacts upon the environment primarily by way of the activity itself – such as fishing, forestry, or agriculture. Nevertheless, there is an increasing awareness of the need to assess other environmental pressures that may act, simultaneously, upon the same population or community, particularly if they have the same effect – such as early mortality or reduced reproductive success. Where environmental *over-exploitation* has already greatly weakened a population or community (as in over-fishing), then the need to assess all other licensable/controllable threats may need to be considered.

### 3.2.2. *Pollution control*

Evaluations in relation to *pollution control*, for example, may include the need to take steps to avoid the creation of any unnecessary waste, to render any such waste as harmless as possible, and to minimise the need to dispose of, or release, any waste into the environment. They may also relate to situations where the environment has already been unacceptably contaminated and requires remediation. Management controls are therefore exercised in relation to the point of release, or to the manner in which a contaminated area is to be cleaned up. For environmental protection, various safeguarding measures or evaluations may be undertaken – often by way of the use of one or more “standards”, as is already the case for radionuclides at certain sites in the USA [27, 28]. Such standards could be set – as they have been – in terms of generalised ‘dose standards’ for organisms, and for which methods for compliance then need to be developed and applied [29, 30]. Or they could be set in terms of concentrations of radionuclides that could give rise to such dose rates. Different numerical values may be relevant to different situations. Other approaches might be favoured for specific practices, circumstances, or locations. Much thought has been given to the development of what have become known as ‘*ecotoxicological*’ type assessments for many chemicals. Such assessments, using models for characterising the distribution and fate of chemicals in the environment, may focus on what is considered to be the most exposed or the most ‘sensitive’ individuals, species, or life stages of fauna or flora in a particular environment or ecosystem [31].

### 3.2.3. *Nature conservation*

Equally, however, the assessment may arise from a requirement to address quite different or explicit issues relating to *nature conservation*; for example by commenting on the likely effects of exposure to ionising radiation on the genetic diversity of a particular species (e.g. salmon in rivers); or on population dynamics (e.g. for a commercial fisheries); or on the ecology of a habitat (a lake or embayment). In these cases many other factors – plus data bases – would probably need to be taken into account in order to ‘manage’ the likely ‘risk’, however expressed. This is particularly the case where the objective is focussed on the need to ‘protect’ the environment in terms of consequences for ‘populations’, ‘ecosystems’ or ‘habitats’. Meeting such requirements may often require information that is outside the scope of ‘radiobiology’ as such. Nevertheless, it is perfectly feasible to assess the consequences of radiation effects for populations – bearing in mind that each population is characterised by such statistical variables as number, birth rate, death rate, sex ratio, age distribution and so on. There have been many ideas on how to model such consequences [32]. Recent work by Woodhead [33] has demonstrated that it is certainly feasible to extrapolate from data on individuals to the level of populations, and to examine how a population might respond to radiation-induced changes in different attributes such as mortality, reproductive success and so on. The management controls for nature conservation, using this information, may also be different. It might therefore be more important to be able to demonstrate that adequate managerial and operational steps have been taken to ensure that the actual or perceived risks posed by radiation to a particular species, area, or habitat have been addressed – by using a range of ‘accredited’ radiobiological information – than to use one or more generalised dose-limit standards.

#### 4. DISCUSSION

As can be seen from all of the above, at a practical level there may well be different reasons for making evaluations of the effects of radiation on the environment, and there will be different situations to assess. How these evaluations are made, and how any particular situation is then managed, is clearly a national responsibility. Concerns may range from a need to assess effects at the level of the individual, the population, the community or the ecosystem. Situations may be real or hypothetical. Approaches will therefore necessarily differ: they may include eco-toxicological type assessments, risk-models, the application of dose limits, and so on; or they may require quite different forms of evaluation. A wide range of information on the effects of radiation on plants and animals already exists in order to achieve some of these objectives; it has been reviewed many times. Much of it relates to our knowledge of the effects of radiation at the level of the individual; some relates to studies on populations. But as virtually all reviewers have pointed out, there are a great many inconsistencies and inadequacies in the information obtained to date. All approaches could therefore probably benefit from some form of 'benchmarking' based on an international 'system of protection'. Given the amount of information already available, and the current national and international work programmes already in place, this should not be difficult to achieve in practice.

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# **Radioactive contamination of aquatic ecosystem within the Chernobyl NPP exclusion zone: 15 years after accident**

**D.I. Gudkov<sup>a</sup>, V.V. Derevets<sup>b</sup>, M.I. Kuzmenko<sup>a</sup>, A.B. Nazarov<sup>b</sup>**

<sup>a</sup> Institute of Hydrobiology of the National Academy of Sciences of Ukraine, Department of Radioecology, Kiev, Ukraine

<sup>b</sup> State Specialised Scientific Enterprise “Ecocentre” of the Ministry of Ukraine on the Emergency and Affairs of Population Protection Against the Consequences of the Chernobyl Catastrophe, Chernobyl, Ukraine

**Abstract.** Aquatic ecosystems and especially lakes are efficient collectors for the wide range of radionuclides, which are deposited in abiotic and biotic components after their intake by aquatic environments. For the time being, ecosystems of water bodies of the Chernobyl NPP exclusion zone continue to suffer from heavy radioactive contamination, dictating, thus, the need to carry out further research works within the framework of comprehensive regional radioecological monitoring. Particular features of radionuclide accumulation by hydrobionts substantially depend on formation of hydrochemical composition of water, which, in its turn, is a complex process, depending on functioning of aquatic ecosystems and natural conditions of a water catchment basin. Hydrochemical regimes in reservoirs are determined by external factors and internal water body processes. The results of radionuclides <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>238</sup>Pu, <sup>239+240</sup>Pu and <sup>241</sup>Am content in hydrobionts tissues of different trophic levels of water objects within the Chernobyl NPP exclusion zone have been analysed. Our studies were conducted: (1) to identify dynamics of radionuclide distribution in components of freshwater biocenose of water bodies within the Chernobyl NPP exclusion zone; (2) to study dynamic profiles of radioactive contamination levels in species of different ecological groups; (3) to assess the major factors, which determine distribution of radionuclides in the freshwater ecosystems; (4) to study the seasonal dynamics of radionuclides content in macrophytes and the role of main aquatic plant associations in processes of radionuclides distribution in biotic component of biohydrocenose; (5) to assess a possibility to use hydrobionts of different trophic levels as biological indicators of radioactive contamination of water objects and (6) to assess the absorbed dose rate for hydrobionts from different water bodies.

## 1. RADIONUCLIDES IN COMPONENTS OF AQUATIC ECOSYSTEMS

The territories of the Chernobyl NPP (ChNPP) exclusion zone are characterised by significant heterogeneity of radionuclide contamination, which is significantly reflected by the radioactive substances contents in aquatic ecosystem components. Primarily this is due to the composition and the dynamics of radionuclide emissions into the environment as a result of accident in 1986, as well as to the subsequent processes of radioactive substances transformation and biogeochemical migration in the soils of catchment basin and bottom sediments of reservoirs. Relatively low contents of radioactive substances are found in the river ecosystems. Due to high water change rate the river bottom sediments have undergone decontamination processes (especially during floods and periods of high water) and over the years that passed since the accident have ceased to play the essential role as a secondary source of water contamination. The main sources of radionuclides in rivers are currently the washout from the catchment basin, the inflow from more contaminated water bodies, as well as the groundwater. On the other hand, the closed reservoirs, and in particular the lakes in the inner exclusion zone, have considerably higher levels of radioactive contamination caused by limited water change and by relatively high concentration of radionuclides deposited in the bottom sediments. Therefore, for the majority of standing reservoirs the level of radionuclide content is determined mainly by the rates of mobile radionuclide forms exchange between bottom sediment and water, as well as by the external wash-out from the catchment basin.

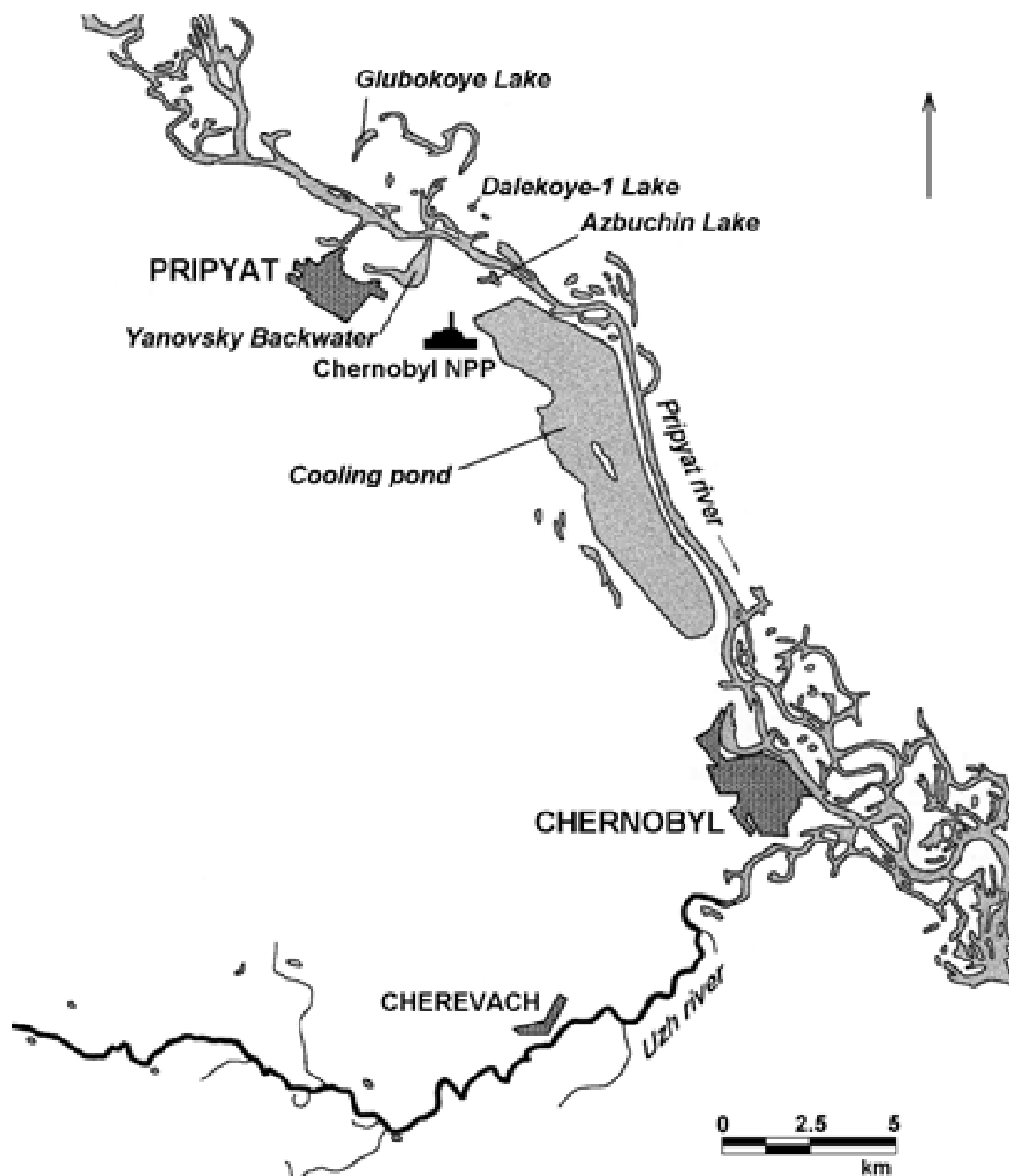


FIG. 1. Map of reservoirs within the Chernobyl NPP exclusion zone.

Our research was carried out during 1998–2001 on Azbuchin Lake, Yanovsky (Pripyatsky) Backwater, the cooling pond of the ChNPP, the lakes of the left-bank flood plain of Pripyat River – Glubokoye Lake and Dalekoye-1 Lake and also on Uzh River and Pripyat River. The sampling station on Uzh River is situated near the river mouth (Cherevach village), on Pripyat River – near the town Chernobyl (Figure 1). The radionuclide content in biological tissues was measured for 28 higher aquatic plant species, 6 species of molluscs and 18 species of fish. The results of the radionuclide content measurements in hydrobionts are expressed in  $\text{Bq kg}^{-1}$  of wet weight at natural humidity. The tendency of the aquatic organisms to accumulate radionuclides, traditionally expressed as the concentration factor (CF), which is determined by calculating the ratio of the specific activity of radionuclides in tissue to the average annual content (for molluscs and fish) or to the average content in the environment water during the vegetation period (for higher aquatic plants). The estimation of the absorbed dose rate for hydrobionts was carried out according to the method [1].

## 1.1. Water and bottom sediment

The highest radionuclide activity in water among the studied objects was found in the Azbuchin Lake. During 1998–2001, the average content of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in lake water reached 120–190 and 18–43  $\text{Bq l}^{-1}$  respectively. The radionuclide contamination density values found in the lake bottom sediments for  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{238+239+240}\text{Pu}$  and  $^{241}\text{Am}$  averaged at 6.70, 11.50, 0.24 and 0.22  $\text{TBq km}^{-2}$  respectively, with the maximum values of 33.30, 14.40, 1.10 and 0.29  $\text{TBq km}^{-2}$ . The  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  content in the water of Glubokoye Lake come to 99–120 and 13–14  $\text{Bq l}^{-1}$  respectively. The average values of contamination density in the bottom sediments by  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{238+239+240}\text{Pu}$  and  $^{241}\text{Am}$  in 1998 were 2.6, 5.6, 0.07 and 0.06  $\text{TBq km}^{-2}$ , with the maximum values being 10.0, 13.7, 0.22 and 0.23  $\text{TBq km}^{-2}$  respectively. In Dalekoye-1 Lake the average content of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the research period reached 82.5 and 11.8  $\text{Bq l}^{-1}$  respectively. The maximum value of radionuclide contamination density in the bottom sediments by  $^{90}\text{Sr}$  in 1999 was 18.9, by  $^{137}\text{Cs}$  – 15.2, by  $^{238+239+240}\text{Pu}$  – 0.6 and by  $^{241}\text{Am}$  – 0.4  $\text{TBq km}^{-2}$ . The average values were, accordingly, 4.0, 3.1, 0.08 and 0.08  $\text{TBq km}^{-2}$ .

The average specific activity values for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in water of Yanovsky Backwater for the period 1998–2001 were 75.2 and 5.6  $\text{Bq l}^{-1}$  respectively. The radionuclide contamination of the bottom sediments of reservoir is extremely heterogeneous, which is obviously caused by the non-uniform character of the nuclear fall-out and by the absence of wind-induced turbulence in deep water. The average content of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{238+239+240}\text{Pu}$  and  $^{241}\text{Am}$  in bottom sediments was, respectively, 16.3, 14.8, 0.4 and 0.3  $\text{TBq km}^{-2}$ . At the same time, within the bounds of silt sediment deposition, some sites with abnormally high density of contamination by  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{238+239+240}\text{Pu}$  (307.1, 251.6 and 5.3  $\text{TBq km}^{-2}$  respectively, which is 20 times higher than the average values in the backwater), were found.

The cooling pond of the ChNPP has undergone the highest radionuclide contamination in comparison with other reservoirs of exclusion zone. In the course of time, after the cessation of radioactive emissions into the atmosphere and due to disintegration of short-lived isotopes,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  have become the main radioactive contaminants of the cooling pond water. During 1998–2001, the specific activity of  $^{90}\text{Sr}$  in the cooling pond water was found to be within the range of 1.7–1.9, with the range being 2.7–3.1  $\text{Bq l}^{-1}$  for  $^{137}\text{Cs}$ . The heterogeneity of the bottom sediment contamination in the cooling pond is currently determined by the nature of the silt accumulation processes. The height of silt layers at the depth of over 11 m (for up to 35 per cent of the bottom area) reaches up to 100 cm., with the density of contamination by  $^{137}\text{Cs}$  at 18.5–133.2  $\text{TBq km}^{-2}$ . The bottom at the depth of 3–11 m consists of primary soils, which are covered, with a 1–6 cm layer of silt, with the contamination density by  $^{137}\text{Cs}$  in the range of 1.5–5.9  $\text{TBq km}^{-2}$  [2].

Uzh River is the main tributary of Pripjat River within the exclusion zone and the runoff of its inflow covers the southern part of a zone with a rather low level of  $^{90}\text{Sr}$  contamination. The contents of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in water of Uzh river during 1998–2001 averaged at 0.11 and 0.31  $\text{Bq l}^{-1}$ . The specific activity of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in water of Pripjat River during 1998–2001 was found in the ranges of 0.22–0.50 and 0.11–0.14  $\text{Bq l}^{-1}$  respectively. The radionuclide content in the bottom sediment of main riverbed sites of Uzh River and Pripjat River is currently only slightly in excess of the pre-accident levels. Considerably higher activity in the bottom sediments is still observed in the backwaters, the old riverbeds and other slow-running sites of the rivers.

## 1.2. Higher aquatic plants

The radionuclide contents in higher aquatic plants (macrophytes) in the studied water bodies were largely determined by the nature of radionuclide contamination of the water objects and nearby territories, as well as by the hydrochemical regime in the reservoirs. The latter affects the forms of radionuclides in the reservoirs, thus affecting the level of their bioavailability to the hydrobionts. The specific activity data for the main radionuclides in macrophyte tissues are shown in Table 1.

TABLE 1. CONTENT OF RADIONUCLIDES IN MACROPHYTES (1998–2001), Bq kg<sup>-1</sup> WET

Water reservoir	<sup>90</sup> Sr			<sup>137</sup> Cs		
	max	min	average	max	min	average
Glubokoye Lake	14060	67	2212	36470	1215	8730
Dalekoye-1 Lake	5100	200	1808	19470	1167	5170
Azbuchin Lake	24210	730	4895	23860	220	2025
Cooling pond	1600	36	218	3125	310	1379
Yanovsky Backwater	2200	110	779	1702	38	814
Uzh River	234	3	10	185	4	42
Pripyat River	357	5	15	164	5	30

The patterns of <sup>90</sup>Sr and <sup>137</sup>Cs accumulation have been shown to be species-specific. Among the species with relatively high <sup>137</sup>Cs content are the helophytes (air-water plants) of genus *Carex*, *Phragmites australis*, *Glyceria maxima*, *Typha angustifolia*, as well as strictly water plant species *Myriophyllum spicatum* and *Stratiotes aloides*. The low values of <sup>137</sup>Cs activity in all reservoirs were found in the representatives of family Nymphaeaceae – *Nuphar lutea* and *Nymphaea candida* as well as *Hydrocharis morsus-ranae*. Relatively high content of <sup>90</sup>Sr was shown by the species of genus *Potamogeton*. Obviously this is related to this plant's tendency to accumulate large quantities of calcium (which is not washed off during standard sampling) on its surface during photosynthesis. At the same time, calcium carbonate that is removed from the plant could contain 7–20 times more radioactive strontium than the plant tissue [3]. Thus, *Potamogeton* species makes a good prospective radioecological monitoring object as a specific accumulator of <sup>90</sup>Sr.

The analysis of the <sup>90</sup>Sr and <sup>137</sup>Cs content at various vegetative stages has revealed seasonal dynamics of radionuclide accumulation by macrophytes. The majority of higher aquatic plant species showed the increased content and CF of <sup>90</sup>Sr and <sup>137</sup>Cs at the peak of vegetation (end of July – August) in comparison with the spring and autumn periods.

The content of radionuclides <sup>238+239+240</sup>Pu and <sup>241</sup>Am in higher aquatic plants of the left-bank flood plain of Pripyat River was found, respectively, in the ranges of 1–66 (11) Bq kg<sup>-1</sup> with CF – 24–4175 and 1–45 (11) Bq kg<sup>-1</sup> with CF – 83–7458. *Typha angustifolia* showed the highest CF, which was 5–7 times higher than the average CF values for other studied plant species. That allows to consider this species as a specific accumulator of transuranic elements in reservoir conditions within the ChNPP exclusion zone.

### 1.3. Freshwater molluscs

Freshwater molluscs are often considered as bioindicators of radionuclide contamination of water objects. These invertebrates accumulate practically all the radionuclides found in water and, due to their high biomass, molluscs play an important part in bioaccumulation processes and radionuclide redistribution in aquatic ecosystems. The <sup>137</sup>Cs and <sup>90</sup>Sr content in molluscs of reservoirs of the ChNPP exclusion zone are shown in Table 2.

The lowest CF for both <sup>90</sup>Sr and <sup>137</sup>Cs were found in *Lymnaea stagnalis* (440 and 137 respectively). Whereas the differences in <sup>90</sup>Sr CF for gastropods could be explained by their shell morphological structure and its specific weight, the distinctions in value of <sup>137</sup>Cs CF are related to the functional ecology and feeding modes of these invertebrates.

TABLE 2. CONTENT OF RADIONUCLIDES IN MOLLUSCS (1998–2001), Bq kg<sup>-1</sup> WET

Water reservoir	<sup>90</sup> Sr			<sup>137</sup> Cs		
	max	min	average	max	min	average
Glubokoye Lake	170300	39770	61830	27150	3156	9067
Dalekoye-1 Lake	62830	17123	32310	2410	847	1523
Azbuchin Lake	87670	34010	51120	4750	2704	3620
Cooling pond	2500	1600	2133	2900	830	1450
Yanovsky Backwater	13350	7720	10350	590	430	510
Uzh River	216	98	177	30	14	25
Pripyat River	101	77	86	61	31	35

The highest CF for both  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were found in bivalve molluscs *Dreissena polymorpha* and *Unio pictorum*, which are the most active filtrators. The highest CF for  $^{90}\text{Sr}$  was noted in *Dreissena polymorpha* – in excess of 1100, while for  $^{137}\text{Cs}$  the highest CF (about 500) was found in the tissues of *Unio pictorum*. Considerably lower CF were determined for the gastropod species *Lymnaea stagnalis*, *Planorbarius corneus* and *Viviparus viviparus*.

The average contents of transuranic elements  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  in mollusc tissues in Glubokoye Lake and Dalekoye-1 Lake were as follows: the lowest value was determined for *Lymnaea stagnalis* – 0.1 and 0.2 Bq kg<sup>-1</sup> respectively in Dalekoye-1 Lake, 2.7 and 6.4 in Glubokoye Lake. The highest content was determined for *Stagnicola palustris* from Glubokoye Lake – 14 and 36 Bq kg<sup>-1</sup> respectively. The highest activity among gastropods was shown in *Planorbarius corneus* – 1 and 2 Bq kg<sup>-1</sup> respectively from Dalekoye-1 Lake; 25 and 53 Bq kg<sup>-1</sup> in Glubokoye Lake. *Dreissena polymorpha* from the cooling pond of the ChNPP showed  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  contents of 3 and 6 Bq kg<sup>-1</sup> respectively.

The contents of  $^{241}\text{Am}$  in *Lymnaea stagnalis* tissue was the lowest – in the range of 4–30 (15) Bq kg<sup>-1</sup> in Dalekoye-1 Lake and 6–51 (27) Bq kg<sup>-1</sup> in Glubokoye Lake. For *Stagnicola palustris* from Glubokoye Lake the value was about about 75 Bq kg<sup>-1</sup>. The highest value was found in *Planorbarius corneus* – 18–29 (24) Bq kg<sup>-1</sup> in Dalekoye-1 Lake and 80–310 (170) Bq kg<sup>-1</sup> in Glubokoye Lake. The content of  $^{241}\text{Am}$  in *Dreissena polymorpha* tissue from the cooling pond of the ChNPP was at the level of 8 Bq kg<sup>-1</sup>.

#### 1.4. Fish

The fish species that are found at the upper levels of the food webs may also constitute a part of human diet and, therefore, are of a particular interest in radioecological research of water ecosystems. The comparative contents of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in fish of the exclusion zone reservoirs are represented in Table 3.

The concentration of transuranic elements  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  was measured in fish of Glubokoye Lake and Dalekoye-1 Lake. The activity of  $^{238}\text{Pu}$  in fish tissue was found in the range of 0.4–0.5 (0.4) Bq kg<sup>-1</sup> with CF – 72–98 (83),  $^{239+240}\text{Pu}$  – 0.7–0.9 (0.8) Bq kg<sup>-1</sup> with CF – 68–87 (75) and  $^{241}\text{Am}$  – 2.2–10.0 (6.2) Bq kg<sup>-1</sup> with CF – 367–1667 (1028).

The contents of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  radionuclides in lake fish of the left-bank flood plain of Pripyat River in all cases considerably exceeded maximum permissible level (MPL), according to the standards accepted in Ukraine for fish production: for  $^{90}\text{Sr}$  on average 146 times higher (MPL – 35 Bq kg<sup>-1</sup>), for  $^{137}\text{Cs}$  – 134 times (MPL – 150 Bq kg<sup>-1</sup>). The highest measured values were 373 and 180 times in excess of MPL. The content of  $^{90}\text{Sr}$  in fish of the cooling pond practically in all caught specimens also exceeded MPL (on average 8 times higher), with the highest registered values being 43 times higher than MPL. The  $^{137}\text{Cs}$  contents in all cases also considerably exceeded MPL – on average 33 times higher, with highest registered values exceeding MPL 84 times.

Despite the fact that the average values of the radionuclide contents in fish of Pripyat River did not exceed MPL, the cases of excess  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  concentrations for the research period have constituted about 15 per cent of the total studied individuals. The highest values of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  contents exceeded MPL by a factor of two. No values in excess of MPL for the research period were found in Uzh River.

TABLE 3. CONTENT OF RADIONUCLIDS IN FISH (1998–2001), Bq kg<sup>-1</sup> WET

Water reservoir	$^{90}\text{Sr}$			$^{137}\text{Cs}$		
	max	min	average	max	min	average
Glubokoye Lake	3300	660	2582	11000	5200	8520
Dalekoye-1 Lake	13060	410	5103	27020	16110	20030
Cooling pond	1518	18	272	13260	1855	4990
Uzh River	7	1	3	75	3	44
Pripyat River	87	9	23	287	32	109



## 2. DOSE RATE FOR HYDROBIONTS

The values of the absorbed dose for hydrobionts from reservoirs of the ChNPP exclusion zone were found to be in the range from  $1.8\text{E}-03$  to  $3.4\text{ Gy}\cdot\text{year}^{-1}$ . The highest value was found for hydrobionts from lakes within embankment territory on the left-bank flood plain of Pripjat River, the lowest – for specimens from the running water objects – Uzh River and Pripjat River (Table 4). The ratio of external and internal doses varied considerably for hydrobionts from different reservoirs and depended on the contents of  $\gamma$ -emitting radionuclides in the littoral zone bottom sediment, as well as in soils close to the river bank. Thus, in Glubokoye Lake, which contains a so-called abnormal contamination strip at the shoreline border, about 95% of absorbed dose results from external exposure and only about 5% – from internal exposure to radionuclides incorporated in tissue (Figure 2). The similar ratio is observed for the exclusion zone rivers – Uzh River and Pripjat River; however, in these objects the ratio is related to the high flow rate as well as to the relatively low radionuclide content in water and, consequently, in hydrobiont tissues.

In Azbuchin Lake and Yanovsky Backwater, at a relatively low external radiation dose, the main contribution to the absorbed dose is made by radionuclides incorporated in hydrobiont tissues. It is linked to the high radionuclide content in water and at the same time to the low contamination level of the bottom sediment within littoral zone and the soils of nearby areas (with sandy soils showing low levels of radionuclide fixation). In this respect, the cooling pond of the ChNPP is in a mid-way position.

TABLE 4. DIAPASONS OF ABSORBED DOSE FOR HYDROBIONTS OF LITTORAL ZONE FROM DIFFERENT WATER OBJECTS WITHIN THE SAMPLING SITES,  $\text{Gy}\cdot\text{year}^{-1}$

Dose		Water object						
		Uzh River	Pripjat River	Cooling pond of the ChNPP	Yanovsky Backwater	Azbuchin Lake	Dalekoye-I Lake	Glubokoye Lake
Internal dose from incorporated radionuclides	$^{90}\text{Sr}$	9.9E-06	1.5E-05	2.2E-04	7.7E-04	4.9E-03	1.8E-03	2.2E-03
	$^{137}\text{Cs}$	1.8E-04	8.6E-05	2.1E-03	8.1E-04	5.1E-02	3.2E-02	6.1E-02
	$^{238}\text{Pu}$	1.0E-04	1.2E-04	5.7E-03	3.2E-03	8.3E-03	6.2E-03	3.6E-02
	$^{239+240}\text{Pu}$	1.8E-04	4.5E-04	2.0E-02	4.2E-02	1.5E-02	8.2E-03	4.9E-02
	$^{241}\text{Am}$	×	×	5.2E-06	5.4E-05	4.2E-05	6.5E-05	1.5E-04
	$^{241}\text{Am}$	×	×	7.9E-05	2.5E-04	3.3E-04	3.7E-04	1.3E-03
	Total	×	×	8.3E-05	1.3E-04	1.3E-04	1.6E-04	3.2E-04
External water dose	$^{90}\text{Sr}$	×	×	1.6E-04	5.9E-04	7.2E-04	7.9E-04	2.3E-03
	$^{137}\text{Cs}$	×	×	2.3E-05	4.5E-05	6.7E-05	9.7E-05	7.2E-04
	$^{241}\text{Am}$	×	×	3.6E-04	6.3E-04	7.5E-04	8.0E-04	2.5E-03
	Total	1.1E-04	1.4E-04	6.0E-03	4.2E-03	1.3E-02	8.5E-03	3.9E-02
	$^{90}\text{Sr}$	3.6E-04	5.4E-04	2.3E-02	4.4E-02	6.8E-02	4.2E-02	1.2E-01
	$^{137}\text{Cs}$	4.9E-08	6.8E-08	5.2E-07	2.1E-05	4.4E-05	2.8E-05	3.2E-05
	Total	9.8E-08	1.5E-07	5.8E-07	2.8E-05	7.0E-05	3.3E-05	4.0E-05
External $\gamma$ -dose	$^{137}\text{Cs}$	2.7E-07	2.9E-07	7.2E-06	1.3E-05	4.5E-05	2.7E-05	3.8E-05
	Total	3.8E-07	4.3E-07	8.3E-06	1.6E-05	1.3E-04	4.0E-05	4.5E-05
Total absorbed dose	$^{90}\text{Sr}$	3.2E-07	3.6E-07	7.7E-06	3.4E-05	8.9E-05	5.5E-05	7.0E-05
	$^{137}\text{Cs}$	4.8E-07	5.8E-07	8.9E-06	4.4E-05	2.0E-04	7.3E-05	9.5E-05
External $\gamma$ -dose	$^{90}\text{Sr}$	1.7E-03	2.3E-03	7.4E-03	3.3E-03	4.8E-03	4.3E-02	1.5–3.3
	$^{137}\text{Cs}$	2.9E-03	3.6E-03	7.9E-03	6.2E-03	1.2E-02	5.0E-02	
Total absorbed dose	$^{90}\text{Sr}$	1.8E-03	2.4E-03	1.3E-02	7.5E-03	1.8E-02	5.2E-02	1.6–3.4
	$^{137}\text{Cs}$	3.3E-03	4.1E-03	3.1E-02	5.0E-02	8.0E-02	9.2E-02	

\* the measurements were not carried out.

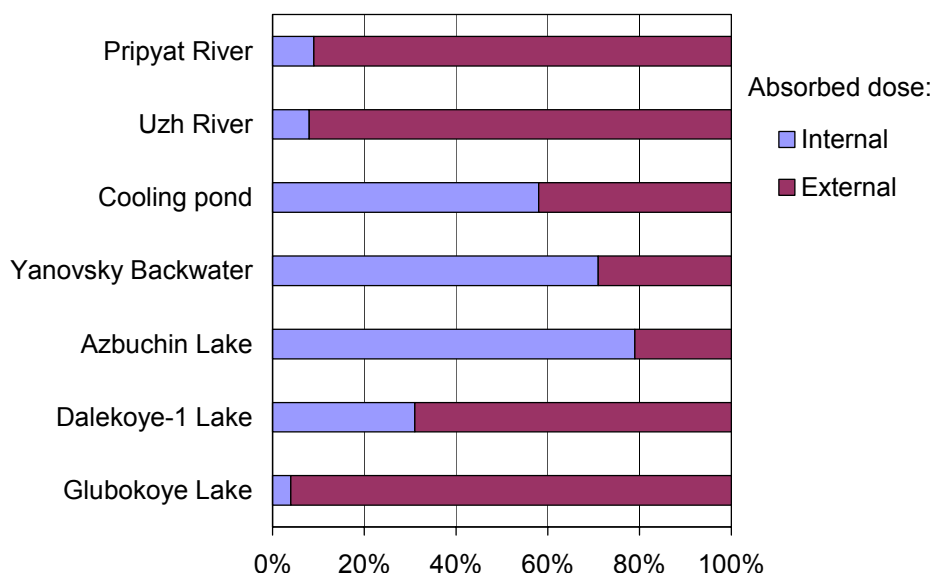


FIG. 2. Ratio of internal and external absorbed dose for hydrobionts from different water objects within the Chernobyl NPP exclusion zone.

According to the classification by G. Polikarpov [4, 5], the studied littoral sites of Uzh River and Pripyat River can be assigned to the radiation safety zone; the sampling stations of Azbuchin Lake, Yanovsky Backwater, the ChNPP cooling pond and Dalekoye-1 Lake – to zones of physiological and ecological disguise, which are also approaching the ecosystem effect zone (Glubokoye Lake), where reduction in aquatic organisms numbers and loss of radiosensitive species can be observed. Populations of benthos organisms from the studied water objects cannot, however, be assigned to the above mentioned zones as, at the known levels of bottom sediment contamination in non-flowing reservoirs within exclusion zone, the absorbed dose for these hydrobionts could be considerably higher.

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# Multi-tiered process in the characterization of a uranium mine waste dump in Lathrop Canyon, Canyonlands National Park, Utah

R.V. Graham<sup>a</sup>, J.E. Burghardt<sup>b</sup>, M. Mesch<sup>c</sup>, R. Doolittle<sup>d</sup>

<sup>a</sup> Radiation and Indoor Environments Unit, United States Environmental Protection Agency, Denver, CO, United States of America

<sup>b</sup> Geologic Resources Division, National Park Service, Denver, CO, United States of America

<sup>c</sup> Division of Oil, Gas and Mining, Abandoned Mine Land Reclamation Program, Utah Department of Natural Resources, Salt Lake City, Utah, United States of America

<sup>d</sup> Division of Oil, Gas and Mining, Abandoned Mine Land Reclamation Program, Utah Department of Natural Resources, Salt Lake City, Utah, United States of America

**Abstract.** The National Park Service (NPS), Utah's Department of Natural Resources Abandoned Mine Reclamation Program (AMRP), and the U.S. Environmental Protection Agency-Region 8 (EPA) entered into a partnership to develop and implement a tiered framework that utilized environmental assessment and management protocols. Implementation of this framework for abandoned mine restoration combined societal and National Park Service program objectives, ecological attributes, indicator biota measurement endpoints, and regional environmental program goals and scientific attributes in the evaluation of environmental risks from abandoned uranium mines' waste rock materials. The NPS and AMRP have worked cooperatively to evaluate and close hazardous openings at these sites throughout Utah, but mine waste rock piles at remote sites have often not been reclaimed. Representative sites on NPS lands were considered for this study. Criteria for site selection included size of mine waste rock piles, proximity to ephemeral desert washes, and evidence of waste rock pile erosion. Twelve abandoned uranium mines in the Lathrop Canyon area of Canyonlands National Park were selected for field analysis. Radiological surveys of the uranium-238 series isotopes were conducted *in situ*. Further waste rock materials and opportunistic water samples were also collected for laboratory analysis. Biological observations and inventories were conducted and environmental risks estimated. Preliminary results potentially indicate that the Lathrop Canyon sites, abandoned in the 1950s, have stabilized over time and currently pose minimal radiological dose impacts on specific ecological receptors. Data comparison to other sensitive environment toxicological endpoints similarly indicate minimal ecological risks and impacts to other potential biological receptors utilizing this sensitive environment. From this initial analysis the NPS, AMRP, and EPA will further evaluate other comparable sites in order to apply this tiered evaluation to conserve, restore, assess and manage environmental objectives on a regional scale.

## 1. INTRODUCTION

The NPS has identified 80 abandoned mine land (AML) sites on its lands in Utah. These sites are, for the most part, vestiges of the uranium boom of the 1940s and 1950s: uranium mines or prospects consisting of adits, shafts, and their associated waste rock piles. They are often in remote settings that now receive minimal human visitation. The mines are typically dry, having no standing water or effluents draining from the mine portals. Spoils material in drainages has long-since been washed down the drainage, leaving a relatively static or stabilized landscape today. There are many sites of this type beyond NPS boundaries throughout the Colorado Plateau Region, a physiographic province centered on the "four corners" area of Utah, Colorado, New Mexico, and Arizona, which has been a major center of production for the nation's radioactive ores since 1900. Mostly these mines follow stratiform deposits in the Chinle (upper Triassic) or Morrison (upper Jurassic) formations: the two principle formations known to host significant uranium deposits in the Colorado Plateau.

The NPS and AMRP have worked cooperatively since 1988 to evaluate and close hazardous openings at these sites throughout Utah, but mine dumps have often not been reclaimed for reasons such as limited funding, minimizing new impacts, and preserving historic fabric. Reclamation standards for

active uranium mines, based on returning the land to a residential use scenario, require cleanup of Radium-226 in the first 15 cm of the soil surface to 185 Bq/g above background (40 CFR §192.12(a)). However, the NPS and AMRP have taken the position that application of this standard to remote, arid, abandoned sites that will never see residential use is unnecessary, cost-prohibitive, and may in fact be counterproductive [1]. These agencies contend that exposures to reclamation crews attempting removal or encapsulation of spoils material could far exceed minimal exposures to occasional visitors passing by the site. They further contend that in attempting to remove or encapsulate spoils piles, may potentially release far more contamination into the environment than if the piles were left alone in their current condition. Oxidized, relatively stable material currently at the surface would be removed, exposing unoxidized, more mobile material. The NPS and AMRP have maintained that these disturbed landscapes have stabilized through time and are currently having little if any effect on the environment and potential biological receptors, although the environmental risks of leaving these dumps unmitigated have admittedly not been fully assessed. For this reason, the NPS, AMRP, and EPA entered into a partnership to develop environmental management goals and implement field sampling protocols for evaluating the environmental risk of abandoned uranium mine waste dumps.

Specifically, the objectives of this partnership were: 1) collect and measure waste rock materials, opportunistic surface water samples, and field-measured external radiation exposures, in an attempt to evaluate the potential for the waste rock piles to leach contaminants into the environment; 2) evaluate potential of contaminants to adversely affect human and ecological receptors; and, 3) select field measurements, monitoring equipment, and monitoring criteria which would best enable NPS and AMRP to screen, evaluate and prioritize other similar sites for future mitigation with minimal time and expense. As part of their Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) Program, the EPA contributed funding and expertise for this study. Representative sites on NPS lands were considered for field analysis, giving highest priority to those sites where there was evidence of contaminants being released to the environment. Criteria for site selection included size of mine spoils piles, proximity to active drainages, and evidence of erosion of the spoils piles. Twelve abandoned uranium adits in the Shinarump member of the Chinle formation within Lathrop Canyon, Canyonlands National Park, were selected.

The extent of lateral workings and waste rock pile volumes for each mine are detailed in Table 1. Since this is a narrow, strata-bound deposit, the mines are all relatively horizontal adits on one stratigraphic level. There are no vertical workings except in localized areas of minor stoping several feet above the normal mine roof. The adits are typically 5 feet wide by 7 feet high with some areas broadening to as wide as 20 feet. Spoil pile volumes vary depending on mine size, the amount of material removed from the site for processing, and the amount of material transported down-gradient by erosive forces through time. Because of their size and distinct drainages leading to the main Lathrop Canyon drainage, field analysis focused on Mine 4, the Mine 5,6,7 Complex, and Mine 12.

## 2. MATERIALS AND METHODS

The original sampling plan was to take multiple cores throughout the spoils piles using 3-inch-diameter hand augers to a depth of 5 feet. Site access and budgetary restrictions precluded the use of mechanized sampling equipment. Once onsite it was quickly determined the coarse fraction in the piles precluded the effective use of hand augers. Since mine dumps are very similar to placer deposits, in that they are essentially horizontally stratified due to the method of emplacement through time, a very basic placer deposit sampling technique was adopted. Shovels were used to dig several vertical or near-vertical trenches through each pile sampled. These trenches were cut as deep as 3 feet into the piles to ensure sampling of relatively fresh, unweathered material. Each trench was cut perpendicularly across different layers in the pile, which represented the various mineralized zones encountered through the life of the mine. Each trench was swept clean of loose debris, then a sample of uniform width and depth was cut along the entire length of the trench. The material collected was reduced using a No. 8 sieve. The fines were homogenized, then approximately 2 kilograms (kg) were bagged for analysis. Equal portions from the fines of the various sample trenches in each spoils pile were also combined to form one 2 kg composite sample. A total of 13 trench samples and 3 composite samples were collected from the main spoils piles at Lathrop Canyon Mine 4, the Mine 5, 6, 7 complex, and at Mine 12. One composite background sample was collected in similar fashion from 3 sample trenches cut in an unmined exposure of the Shinarump member just west of Mine 4.

TABLE 1. LATHROP CANYON MINE WORKINGS AND ASSOCIATED WASTE ROCK VOLUMES

Mine	1	2	3	4	5	6	7	8	9	10	11	12
Lateral Workings (feet)	82	28	75	865	450	230	215	188	20	70	40	235
Waste (cubic yards)	120	–	–	800	470			220	35	165	100	400

External gamma radiation measurements were recorded using both Mt. Sopris® SC-132 and Exploranium® GR-130 hand-held monitoring instruments. External alpha radiation readings from radon decay products (the primary source of alpha radiation in natural settings) were also recorded in the spoils pile sample trenches using an MDA® Model 811 Working Level Meter.

Lathrop Canyon is a semi-arid environment with approximately 25 cm of precipitation per annum. Sufficient rainwater fell during the fieldwork, however, to create adequate drainage for opportunistic sampling both up-gradient and along the ephemeral water course evident through the spoils area. All water samples were collected in 200 milliliter (ml) sample bottles that were pre-acidified with nitric acid. Two background water samples were collected: one from drainage off an overhang immediately above the Shinarump member of the Chinle formation at Mine 6, and the other from drainage that had passed entirely through the Shinarump member up-gradient from the mined area. Three aqueous samples were taken immediately down-gradient from the waste piles where the solid samples were taken, and a final aqueous sample was taken in the main Lathrop Canyon drainage down-gradient from the spoils piles sampled.

Spoils solids and aqueous samples were analyzed for metals and radionuclides by the US Environmental Protection Agency National Air and Radiation Environmental Laboratory (EPA-NAREL). Non-radiological metals were run using ICP-MS (EPA Total Analytical List Method ILM04.0). Appropriate quality assurance / quality controls (QA/QC) of the samples were run as duplicates and spike samples for each analytical sample batch for a total of 5% internal controls.

Ecological receptors that utilized the impacted area and watershed were observed and categorized. Biomonitoring was not included in this screening assessment of these waste rock spoil piles at this time. Ecological sensitive species (those species known to fulfill critical niches within this desert environment), organism foraging and reproductive ranges, and radiologically sensitive receptors [2, 3] were chosen as indicators of the impacts potentially caused by these waste piles. These species selected included grasses, shrubs (*Artemisia spp.*, *Tamarix spp.*, *Pinus spp.*, *Sarcobatus spp.*, *Quercus spp.*), small and large mammals. The most sensitive non-radiological toxicological endpoint for comparison to potential effects upon the biological receptors was considered to be the “No observable adverse effects” level. This level or concentration of a contaminant does not cause toxicological observable effects upon an individual test species exposed to the contaminant during laboratory toxicity testing. Terrestrial toxicity values were based upon both phyto-toxicity and terrestrial organisms response reported in open peer reviewed literature. Aquatic toxicity responses were based upon the US National Recommended Water Quality Criteria [4]. Radiological doses for these biological indicator species was also estimated using the RESRAD-BIOTA® code. The radiological dose estimated to these radiologically sensitive terrestrial species by the model was then compared to the IAEA [5] and NCRP (1994) [6] guidelines.

### 3. RESULTS

The results for water samples collected in the ephemeral stream running throughout the Lathrop Canyon are shown in Tables 2 and 3, below. The background or reference samples taken up-gradient from the spoil piles showed elevated levels of dissolved (0.45µm filter) metals but not statistically significant when compared with water samples collected immediately down-gradient from the piles. Although it appears that the spoil piles are leaching into the stream, a more detailed analysis of uranium-238 and radiological decay products in the U-238 series shows only minor radionuclide contributions from the piles with Ra-226 being the major radiological constituent of concern detected.

TABLE 2. WATER SAMPLE RESULTS COLLECTED IN LATHROP CANYON ( $\mu\text{G/L}$ )

Analyte	GM $\pm$ GSD of Up-gradient Water Samples	95% CI of GM Up-gradient Water Samples	GM $\pm$ GSD of Down-gradient Water Samples	95% CI of GM Down-gradient Water Samples
Arsenic	20.7 $\pm$ 9.0	33.2	20.7 $\pm$ 5.8	25.4
Copper	35 $\pm$ 34.4	82.4	28.6 $\pm$ 23.4	47.4
Manganese	723 $\pm$ 256	1036	908 $\pm$ 11653	13429
Selenium	2.95 $\pm$ 0.4	2.95	19.3 $\pm$ 32.3	45.1
Vanadium	51 $\pm$ 18	75.2	24.4 $\pm$ 39.3	55.8

TABLE 3. RADIONUCLIDE CONCENTRATIONS IN LATHROP CANYON WATER (Bq/L)

Analyte	Sample 1 Reference: Upgradient	Sample 2 Reference: Downgradient	Sample 3 Lathrop Channel	Sample 4 Mines 5, 6, 7 drainage	Sample 5 Mine 4 drainage above Channel	Sample 6 Downgradient Mine 4
Gross Alpha	1.3 $\pm$ 0.7	0.5 $\pm$ 0.4	2.3 $\pm$ 0.9	7.8 $\pm$ 2.0	21 $\pm$ 7.4	9.4 $\pm$ 2.1
Gross Beta	1.8 $\pm$ 0.9	0.5 $\pm$ 0.8	2.5 $\pm$ 1.0	8.3 $\pm$ 1.5	36.4 $\pm$ 7.0	12.4 $\pm$ 1.7
Protactinium-234m	Not Detected	Not Detected	Not Detected	Not Detected	26 $\pm$ 44.4	Not Detected
Radium-226	5.4 $\pm$ 8.5	4.9 $\pm$ 7.4	ND	8.3 $\pm$ 8.1	19.3 $\pm$ 8.5	5.2 $\pm$ 8.5

Distribution coefficient ( $K_d$ ) values for each sampled spoils pile were calculated and shown in Table 4.  $K_d$  values are defined as the ratio of the contaminant concentration associated with the solid to the contaminant concentration in the aqueous solution, assuming equilibrium conditions. These values were determined using the 95% confidence interval of the geometric mean for both soil and water samples. Each spoil pile was also analyzed for soil pH and soil electrical conductivity (EC). Soil pH values ranged from 7.5–8.6. Soil EC values ranged from 1.7–13.7 with reference soil samples reflecting a value of 19 mmhos/cm.

The spoil piles were divided by their physical locations within Lathrop Canyon drainage. The data was log-normally distributed with results for the various mines as shown in Tables 5 and 6, below. The geometric standard deviation (GSD) and the 95% confidence interval (CI) about the geometric mean (GM) were also determined. The 95% confidence interval about the geometric mean defines the maximum concentration of each analyte of interest. The piles show mineral heterogeneity. Composite sampling collected throughout each spoil pile was initially thought to be more representative of the pile's metal and radionuclide concentrations. Laboratory analysis showed that compositing did not underestimate either metal or radionuclide concentrations within the piles.

TABLE 4.  $K_d$  VALUES CALCULATED IN THE LATHROP CANYON DRAINAGE

Analyte	Mine 4	Mines 5, 6, 7	Mine 12 Composite	Reference Site
Arsenic	1.8	9.4	1.1	0.4
Copper	21.0	115	6.0	4.0
Manganese	2.1	2.50	3.0	0.70
Selenium	0.02	0.04	0.03	0.10
Vanadium	1.03	1.5	2.8	0.40

TABLE 5. METALS IN LATHROP MINE WASTE ROCK SPOIL PILES (mg/kg DRY WEIGHT)

Reference	Arsenic 12 ± 2.0	Copper 322 ± 26	Manganese 702 ± 59	Selenium 0.3 ± 0.02	Vanadium 30 ± 2.5
Mine 4 (1)	42.3	601	630	0.4	7.9
Mine 4 (2)	24.8	216	864	1.2	18.4
Mine 4 (3)	46.5	900	701	0.8	8.0
Mine 4 (4)	38	499	605	0.3	5.8
Mine 4 Composite	50.7± 5.7	429± 79	850± 34	0.7± 0.03	8.1 ± 0.8
Mine 4 GM ± GSD (95%CI)	34.8 ± 13.7 (46.8)	544.8± 678.3 (1139.3)	794.5 ± 214.5 (982.5)	0.7 ± 0.4 (1.1)	9.2 ± 4.8 (13.4)
Mines 5–7 (1)	115	2310	962	1.7	8.8
Mines 5–7 (2)	374	7910	842	0.6	25.4
Mines 5–7 (3)	75.6	1430	1120	0.4	12.6
Mines 5–7 (4)	109	3740	1060	2.4	10
Mines 5–7 Composite	124 ± 13.3	3500 ± 982	948 ± 119	2.7 ± 0.8	9.8 ± 1.3
Mines 5–7 GM ± GSD (95%CI)	154 ± 107 (248)	3865 ± 2708 (6238.7)	1043 ± 150 (1174.5)	1.3 ± 1.2 (2.3)	14.1 ± 6.0 (19.3)
Mine 12 (1)	24.5	112	1250	0.3	20.2
Mine 12(2)	18.7	289	1410	1.2	17.3
Mine 12 (3)	9.2	192	1270	0.3	35.6
Mine 12 Composite	19 ± 3.3	130 ± 32	1130 ± 107	1.7 ± 0.6	19.6 ± 2.2
Mine 12 GM ± GSD (95%CI)	18.4 ± 10 (29.6)	206 ± 104 (323.3)	1311 ± 86 (1408.1)	0.7 ± 0.6 (1.4)	25 ± 10 (36)

TABLE 6. RADIONUCLIDES IN LATHROP MINE WASTE ROCK SPOILS PILES (Bq/kg DRY WEIGHT)

Location	Gross Alpha	Gross Beta	Uranium- 238 <sup>a</sup>	Bismuth- 214	Protactinium- 234m	Lead-214	Thorium- 234	Radium- 226
Reference	2490± 444	3352± 296	559± 74	747± 37	396± 111	814± 37	306± 37	977± 74
Mine 4 (1)	10249	7881	1391	1184	1639	1295	1446.7	2157
Mine 4 (2)	2708.4	4144	614	500	766	548	636	596
Mine 4 (3)	3626	3160	574	525	618	48	585	829
Mine 4 (4)	11692	9546	1203	1103	1373	1203	137	1491
Mine 4 Composite	11211± 888	10027 ± 481	1251±111	1158± 67	1373± 189	1225± 74	1247± 74	1702±104
Mine 4GM±GSD (95%CI)	9520± 8517 (16983)	80475± 5343 (12728)	1195± 696 (1806)	1084± 677 (1676)	1358 ± 766 (2035)	1184± 766 (1850)	1154±611 (1691)	1565±1036 (2472)
Mines 5–7 (1)	71040	64010	12203	12099	1184	13283	11581	26048
Mines 5–7 (2)	3700	49950	9113	9176	8880	10064	8325	18500
Mines 5–7 (3)	24420	2442	3530	3441	3589	3737	3349	7511
Mines 5–7 (4)	23162	19684	4340	3922	4699	4292	4440	9250
Mines 5–7 Composite	41070 ± 1702	36408 ± 888	6209±370	6438 ±370	6771 ± 444	7067 ± 407	4551± 259	14208 ± 814
Mines 5–7 GM±GSD (95%CI)	40097 ± 24546 (61612)	36142 ± 23199 (56462)	6712 ± 4292 (10471)	6605 ± 4381 (10434)	6660 ± 4829 (10223)	7237 ± 4829 (11470)	6364± 3959 (943.5)	13901 ± 10397 (23014)
Mine 12 (1)	814	1432	137	126.5	146	137	139	90
Mine 12 (2)	640	1502	129	125	125	139	125	269
Mine 12 (3)	444	1591	63	68.5	100	75	24	141
Mine 12 Composite	429	1162± 222	78 ± 22	74 ± 4	68 ± 74	80 ± 4	87 ± 11	167 ± 15
Mine 12GM±GSD (95%CI)	740 ± 126 (873)	1469± 48 (1524)	133 ± 7 (141)	126 ± 1 (126)	137 ± 15 (152)	137 ± 1 (141)	133 ± 10 (144)	211± 189 (422)



TABLE 7. ECOLOGICAL TOXICITY NO OBSERVABLE ADVERSE EFFECTS LEVEL (NOAEL) SCREENING CRITERIA (MG/KG) AND RADIOLOGICAL DOSE ESTIMATES (mGy/day) FROM WASTE ROCK SPOILS PILES

Analyte of Concern	Terrestrial Criteria	Aquatic Criteria	Radiological Dose
Arsenic	10–14 mg/kg	150 µg/L	Not applicable
Copper	4 mg/kg (sheep) 100 mg/kg (plants)	9 µg/L	Not applicable
Manganese	500 mg/kg	1100 µg/L	Not applicable
Selenium	0.5 –1 mg/kg	5 µg/L	Not applicable
Vanadium	0.5 mg/kg (ruminants) 2 mg/kg (plants)	0.05 – 0.5 mg V/L	Not applicable
Uranium (total)	5 mg/kg	200 µg/L	0.1 – 0.001 mGy/d
Radium 226	N/A	N/A	152 – 1.8 mGy/d (animal) 13 – 0.2 mGy/d (plant)

External gamma and alpha radiation measurements taken in the sample trenches during the fieldwork ranged from background levels (0.1–0.2µSv/hr) to a maximum reading of 4.4 µSv/hr. Alpha radiation levels from radon progeny were recorded in Working Levels (WL) units, where 1WL ≈ 7.4 Bq/m<sup>3</sup> (200 pCi/L). To obtain alpha readings, 5 liter air samples were passed through a filter paper using an air sampling pump (running 2 minutes at 2.5 liters per minute) under a tarp covering each trench, then the filter paper was analyzed onsite with an MDA® Model 811 Instant Working Level Meter. Radon readings within the pile ranged from 0.07–0.14 Bq/m<sup>3</sup>.

Radiological dose criteria used in this study (0.1Gy/d for terrestrial animals and 1Gy/day for terrestrial plants) were modeled using RESRAD-BIOTA®. Toxicity values gathered from literature were compared to site soil and water concentrations (Table 7). Estimated dose rates using soil and water radionuclide site concentrations and “surrogate” terrestrial species indicated dose levels exceeding screening levels. However, modeled comparison of the mines to upgradient reference site showed similar background soil dose rates to terrestrial organisms, 105 mGy/d vs.152 mGy/d. Reference site modeled doses for plants for Ra-226 and U-238+234 showed a dose of 1mGy/d whereas the maximum modeled dose from the spoil piles was 14 mGy/d. A conceptual site model was utilized in order to sample and integrate site specific fate and transport physio-chemical migration routes and biotic exposure pathways into a full site picture of the ecological condition of the site (Figure 1).

#### 4. DISCUSSION AND CONCLUSIONS

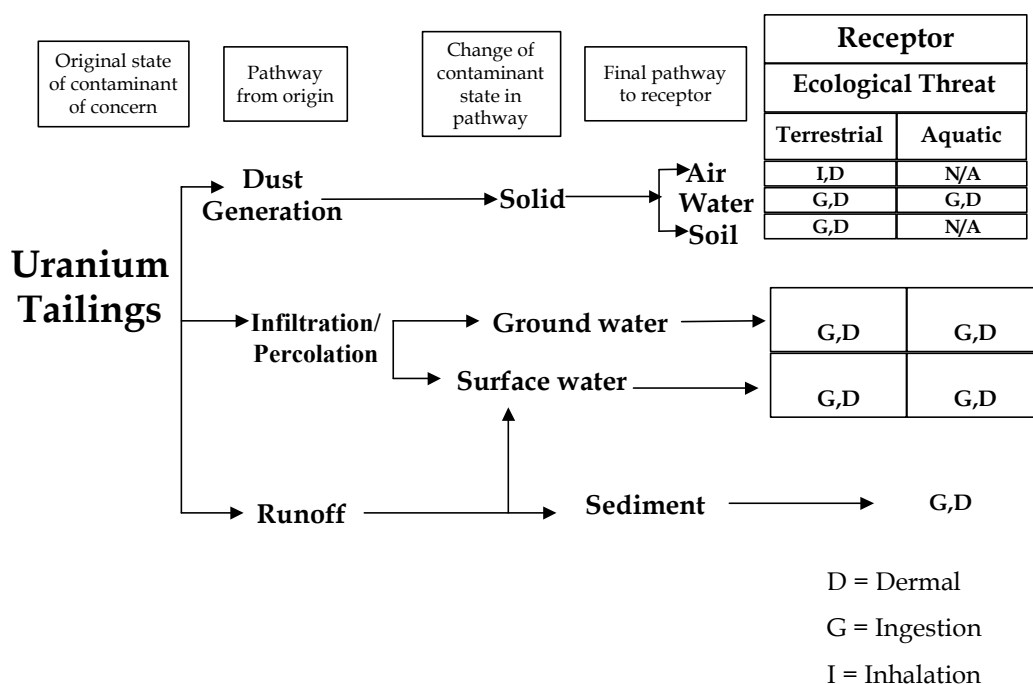
This study looked at various methods that land management agencies might use to determine potential impacts from abandoned mines, waste piles, and subsequent leachates in small, remote sites typical of the Colorado Plateau Region. Abandoned mine mitigation efforts should utilize a multi-disciplinary approach in order to evaluate the waste rock piles’ toxicological impacts upon benchmark or focal

biological receptors. This approach would utilize the physio-chemical properties of these piles, the hydrology and topography of the drainage area, environmental processes such as primary productivity or growth efficiency, biotic conditions, toxicological screening benchmark parameters, and radiological screening models to assemble the indicators into a real-world evaluation of the site.

Field instruments and measurement protocols that measure soil pH and electrical conductivity are beneficial in trying to determine whether spoils material is leaching into ephemeral streams. Soil pH reflects the potential for metal mobility or metal binding. Soil EC reflects salinity content and potential for contaminants to be dissolved and transported into the neighboring streams. Soil K<sub>d</sub> measurements are valid in showing whether various minerals are leaching into the environment, but this measurement requires soil and water analytical analysis for confirmation. A field leachate process was recently described by K. Smith et al., [7] that may provide expedient answers adaptable from hard-rock mining endeavours.

In-situ K<sub>d</sub> values show values lower than reported in literature even though it appears that copper and selenium are leaching into the drainage area and may be at toxic concentrations to aquatic receptors.

## Migration Routes of a Solid (Metal) Contaminant from Origin to Receptor



*FIG. 1. Site Conceptual Framework.*

Whether erosional processes occurred immediately after the waste piles were deposited is unknown, but speculated. Spoil materials are not readily evident in the down-gradient drainages. Leaching maybe retarded due to alkaline to circum-neutral soil pH and reducing environments within the piles which aid in reducing metal solubilities [8]. Another factor that may reduce the leaching of minerals out of the spoils piles may be the elevated manganese concentrations in the background geological formations from which the ore was extracted. In the Lathrop Canyon area “desert varnish,” a micro-thin patina composed of manganese and iron oxides, may aid in covering the surfaces of rocks in the canyon country and inhibit the leaching process.

At abandoned uranium mines, radiation meters that indicate external dose readings from uranium decay products and meters that can determine *in situ* radium concentrations may give a good indication of surface contamination and possibly reduce the number of samples necessary for site characterization. This would assist the land management agency in determining potential human and environmental health concerns. However, these meters would yield no information on leachability of spoil materials or anticipated concentrations of contaminants in down-gradient, nor would they be useful in determining impacts at depth in the spoil piles due to the attenuating effect of overlying material or determining radiological impacts upon the environment.

The primary ecological receptors of concern in the Lathrop Canyon area were based upon benchmark keystone or focal species [3, 9] that are critical to the arid environment, have smaller foraging or home ranges thereby potentially utilizing the spoil piles and drainages throughout their lifespan, and toxicologically and radiologically sensitive to contaminants identified in these spoils piles. Sampled soil and water results show that metal levels were indeed elevated above toxicity relevant levels. Even “Reference Site” soil concentrations were above toxicity levels for terrestrial animal and plants except for arsenic and selenium. Water concentrations showed elevated levels of copper up-and-downgradient from the piles ranging from 30–80 µg/L, over four times recommended chronic toxicity levels for fish. Similarly, vanadium and selenium were elevated downgradient along the drainages but vanadium was also elevated in the upgradient reference area. Arsenic, manganese, and uranium were not shown to be in the collected water samples as levels statistically different than the reference site.

Radiological impacts showed modeled doses exceeding screening criteria of 0.1 Gy/d and 1 Gy/d. As suspected the external exposures from Ra-226 drives the radiological impacts of the area. However it is speculated that the uranium series decay products are not causing an effect at the populational level due to: 1) Concentration Ratios (CR) for naturally occurring U and Ra-226 have been shown in other studies not to be readily bioavailable to terrestrial species [10–12]; 2) Dilution of leachates through storm events; 3) Heavy metal phytotoxic concentrations in spoil soils which discourages foraging habitat; 4) Sandy spoils encourage water runoff instead of water retention; 5) Low organic carbon content of soils; 6) Wide foraging range of small and large mammals with choice of a variety of pools and water holes in the area. The small mammals in the area are not particularly dependent upon standing water in the drainage. They have highly effective metabolic systems that enable them to extract water from the fats stored in seeds and excrete high nitrogenous, high-solid, low-water urine. The larger mammals have large foraging ranges, which minimizes impacts from the spoil materials and ephemeral streams on these receptors. The bighorn sheep and mule deer that frequent the area would be the receptors of major concern due to the societal and ecological value of standing water when it occurs in this ecosystem. After reviewing the water and spoil contaminant concentrations and noting the lack of vegetation on the spoils piles that might be ingested, it appears that the most likely route of toxicity would come from direct ingestion of water; 7) Short reproductive life and larger turnover rate for small mammals; 8) Ephemeral stream prevents aquatic life from exposure at radiologically sensitive life stages; and, 9) Competitive or lack of bio-uptake and sequestering of heavy metals in plant and animal tissues.

This study investigated legacy uranium and vanadium mines that have been abandoned for over 50 years in a semi-arid environment. Through a variety of physical, chemical, hydrological, and biological sampling this paper attempted to identify field expedient attributes that would enable the land manager to fully assess and evaluate ecological affects from these mines. It would appear that soil particulate sizing, pH, EC, and analytical measurements of metal and radionuclide concentrations were instructive whereas Kd derivation was not as important as once thought. Radionuclide surveys were beneficial in determining “hotspots” and potential dose estimates but biomonitoring is the only method which would indicate whether the materials are readily bioavailable to biotic receptors in the area. Soil organic carbon and field leachate testing will be added to further field studies in order to investigate whether the piles are leaching into area drainages. Better ecological attributes must be developed that utilize biotic conditions (trophic structure, population size, habitat suitability), ecological processes (growth efficiency), hydrology/geomorphology (surface flows, water storage, channeling), chemical/physical characteristics (nitrogen, carbon, pH, salinity, organic matter), and natural disturbance regimes (frequency, intensity).

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# Assessment of the impact of radionuclide releases from Canadian nuclear facilities on non-human biota

G.A. Bird<sup>a</sup>, P.A. Thompson<sup>a</sup>, C.R. Macdonald<sup>b</sup>, S.C. Sheppard<sup>c</sup>

<sup>a</sup> Canadian Nuclear Safety Commission, Ottawa, Canada

<sup>b</sup> Northern Environmental Consulting and Analysis, Pinawa, Manitoba, Canada

<sup>c</sup> ECOMatters Inc., Pinawa, Manitoba, Canada

**Abstract.** The radiological impact of radionuclides released from nuclear facilities is being assessed for regulatory purposes using an ecological assessment framework. Hazard quotients are determined by dividing an estimated exposure value (EEV) by an estimated no effect value (ENEV). Values less than one indicate that environmental harm is not likely, whereas in Tier 2 or Tier 3 assessments values greater than one indicate the potential for environmental harm. Radiation exposure values are calculated using annual mean radionuclide concentrations in water, sediment and biota, and either published screening DCFs or dosimetry equations taking into account the geometry and size of the organism. A relative biological effectiveness (RBE) weighting factor of 40 for alpha emitters and 3 for tritium is used in the dose calculations. When radionuclide concentrations are not measured in biota, they are estimated from published geometric mean concentration ratios. Radionuclide concentrations in benthic invertebrates are assumed to be equivalent to those in the sediment. The radiation dose is the sum of the internal and external doses, except for benthic invertebrates where the total radiation dose is assumed to be from internal radiation exposure. The ENEVs for the various taxonomic groups are determined from literature data using an ecotoxicological approach. The ENEVs derived for radiation effects on biota are: 0.2 Gy·a<sup>-1</sup> for fish, 2 Gy·a<sup>-1</sup> for both benthic invertebrates and terrestrial invertebrates, and 1 Gy·a<sup>-1</sup> for algae, macrophytes, mammals and terrestrial plants.

The assessment conducted for uranium mines and mills is presented as a case study outlining the recommended approach. The results suggest that a probabilistic Tier 3 assessment may not be necessary when environmental data are readily available.

## 1. INTRODUCTION

In Canada, the regulation of nuclear facilities under the Nuclear Safety and Control Act requires that their effects on the environment be evaluated. This information is used in the licensing process to determine whether or not mitigation measures are effective. The impact of the release of radionuclides from Canadian nuclear facilities on non-human biota was also assessed by Environment Canada under the Canadian Environmental Protection Act.

Nuclear facilities release complex effluents with both radioactive and non-radioactive substances (organics and metals). Therefore, a consistent methodology needs to be used to assess environmental effects of all the contaminants of concern. For this reason, an ecological risk assessment (ERA) approach is used for radionuclides released from nuclear facilities. The following sections describe the recommended methodology to assess risks, to calculate exposure values (radiation dose) and the recommended radiation effects benchmarks. A case study of the assessment of three operating uranium mines and mills is also briefly described.

## 2. ECOLOGICAL RISK ASSESSMENT METHODOLOGY

The chosen assessment methodology follows the guidelines in Environment Canada (1997) and is generally consistent with methodology developed by the U.S. EPA [1]. The potential for environmental harm is assessed as a risk quotient of the Estimated Exposure Value (EEV) divided by the Estimated No-Effects Value (ENEV), where the value is radiation dose. If this risk quotient (RQ) is less than unity, it is unlikely that the contaminant is harmful. First, a tier 1 assessment using conservative assumptions and data is performed [2]. Conservative RQs <1 indicate very low probability of harmful effects. Conservative RQs >1 require further evaluation evoking a more realistic Tier 2 assessment using realistic EEVs [2]. A probabilistic assessment may be conducted if Tier 2 risk quotients (RQs) are greater than one. Spatial effects were accounted for by calculating RQs for taxa in the first receiving water body, then each subsequent downstream water body (providing data were available) until a RQ <1 is obtained. Consistent ENEVs are used throughout the assessment.

## 2.1 Critical toxicity values and estimated no effect values

Critical toxicity values (CTVs) were identified from which ENEVs are derived using appropriate application (safety) factors to account for uncertainties related to differences between the observed effects endpoints and the success of organisms in the field [2]. An application factor of 1 has been used to estimate the realistic radiation ENEVs. Preferred CTVs are estimates of low toxic effects, such as the LC<sub>25</sub> or EC<sub>25</sub> for more sensitive species [2] for responses applicable to survival of the species and are based on chronic exposures as much as possible. If sensitive species are protected, then other less sensitive taxa will also be protected. Considerable information is available on the effects of acute and high exposure to radiation on organisms. However, there are few studies involving chronic radiation exposure at environmentally relevant dose rates.

The CTVs for radiation effects and derived ENEVs are presented in Table 1. It is recommended that they be used in all assessment tiers. The ENEVs are 0.2 Gy·a<sup>-1</sup> for fish, 1 Gy·a<sup>-1</sup> for mammals, terrestrial plants, macrophytes and algae and 2 Gy·a<sup>-1</sup> for both benthic invertebrates and terrestrial invertebrates. In comparison, NCRP, IAEA and UNSCEAR [3–6] have suggested that doses less than or equal to 0.4 Gy·a<sup>-1</sup> and 3.7 Gy·a<sup>-1</sup> should not result in effects on populations of terrestrial plants and animals and aquatic organisms, respectively.

The ENEV for fish is based on the CTV of 0.6 mGy·d<sup>-1</sup> (0.2 Gy·a<sup>-1</sup>) for reproductive effects in carp in the Chernobyl cooling pond [7]. This value is in the range where both effects and no effects have been observed and should ensure that long-lived (10 to > 20 years), slow-growing fish species that start reproducing at the age of 3–7 years or older are adequately protected.

In the case of mammals the LD<sub>50</sub> of 3 mGy·d<sup>-1</sup> (1 Gy·a<sup>-1</sup>) (corrected by a RBE factor of 3 for tritium) for immature oocytes of the squirrel monkey [8] was chosen as the CTV. Given that this level of effect did not result in sterility, an application factor of 1 yields an ENEV of 1 Gy·a<sup>-1</sup>. In a recent review, Harrison and Knezovich [9] indicated that in mammals adverse effects on fertility are first observed at a critical value of about 0.48 mGy·d<sup>-1</sup> (0.18 Gy·a<sup>-1</sup>). Therefore, an ENEV of 1 Gy·a<sup>-1</sup> is not considered overly conservative.

The NOEL of 2.4 mGy·d<sup>-1</sup> reported by Amiro [10] and Amiro and Sheppard [11] was chosen as the chronic CTV for terrestrial plants. Given the very large number of plant species tested, an application factor of 1 was used to derive an ENEV of 1 Gy·a<sup>-1</sup>. Because of the scarcity of data on chronic radiation effects on aquatic plants, the ENEV for terrestrial plants (conifer) of 1 Gy·a<sup>-1</sup> was adopted for aquatic macrophytes and algae. Conifers are more sensitive to radiation than lichen [5] and lichens are composed of fungus in symbiotic union with an alga. Therefore, the use of the conifer data for the ENEV for aquatic plants is likely conservative.

TABLE 1. RECOMMENDED ENEVS TO ASSESS THE POTENTIAL TOXICITY OF RADIATION TO NON-HUMAN BIOTA

Taxa	CTV (GyAa <sup>B1</sup> )	Endpoint	ENEV (GyAa <sup>B1</sup> )
Fish	0.2	reproduction	0.2
benthic invertebrates	2.0	reproduction	2.0
algae*	1.0	NOEL	1.0
macrophytes*	1.0	NOEL	1.0
amphibians	1.0 Gy	survival	1.0
small mammals	1.0	reproduction	1.0
terrestrial plants	1.0	NOEL	1.0
terrestrial invertebrates**	2.0	reproduction	2.0

\* based on no-observed-effect level for terrestrial plants.

\*\* based on effects to benthic invertebrates.

Of the aquatic invertebrates tested, the polychaete worm ( $1.7 \text{ Gy}\cdot\text{a}^{-1}$ ) appears to be the most sensitive to radiation. Therefore, the value of  $4.6 \text{ mGy}\cdot\text{d}^{-1}$  ( $1.7 \text{ Gy}\cdot\text{a}^{-1}$ ) for effects on reproductive indices was chosen as the CTV. An application factor of 1 was chosen because both the nature and magnitude of effects on reproductive indices were similar at  $4.6 \text{ mGy}\cdot\text{d}^{-1}$  and  $50.4 \text{ mGy}\cdot\text{d}^{-1}$  in the key study of Harrison and Anderson [12]. The ENEV of  $2 \text{ Gy}\cdot\text{a}^{-1}$  from this study is in the range of radiation doses experienced by *Chironomus tentans* larvae that resulted in genetic effects but no population level effects [13]. Therefore, using the weight of the evidence, an ENEV of  $2 \text{ Gy}\cdot\text{a}^{-1}$  should adequately protect freshwater benthic invertebrate species. Because of the paucity of data on the effects of chronic low dose radiation exposure in terrestrial invertebrates, the ENEV for terrestrial invertebrates is based on the ENEV of  $2 \text{ Gy}\cdot\text{a}^{-1}$  for aquatic benthic invertebrates.

## 2.2 Calculation of estimated exposure values

For the conservative Tier 1 assessment, the EEVs are the highest concentration in environmental media (e.g., in water, sediment and biota) for each radionuclide recorded in recent monitoring data. In a realistic Tier 2 assessment, the EEV is based on the mean concentration in a specific receiving water body (e.g., in water, sediment and biota). The species having the highest radionuclide concentrations are assessed. Where measured concentrations in biota are not available, concentrations in biota are estimated by multiplying the mean aqueous concentrations by an appropriate geometric mean (GM) concentration ratio (CR). For the radiation dose to benthic invertebrates (e.g., chironomids), it is assumed that ionization is uniform throughout the sediment and that the radionuclide concentrations in benthic invertebrates are equivalent to those in sediment. Field data support this assumption that small benthic invertebrates have radionuclide concentrations similar to those in sediment [14].

When no monitoring data are available for a daughter radionuclide, the daughter is assumed to be in secular equilibrium with its parent. This assumption was tested using environmental data from the uranium mining area in northern Saskatchewan, Canada. The assumption of secular equilibrium is reasonable between  $^{230}\text{Th}$  and its daughters with values being usually within a factor of 2. Therefore, the assumption of secular equilibrium is applicable between  $^{230}\text{Th}$  and  $^{226}\text{Ra}$  and among  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ . In all cases,  $^{222}\text{Rn}$  was assumed to be 0.3 times the concentration of  $^{226}\text{Ra}$ , because of the lack of data on the retention of  $^{222}\text{Rn}$  in the various media.

The EEV for biota is the total radiation dose to the organism, which is the sum of the internal and external doses. However, in the case of benthic invertebrates the radiation is calculated as the radionuclide sediment concentration multiplied by the internal DCF. An external radiation dose is not included. Radionuclide concentrations in soil and litter invertebrates are derived from soil radionuclide concentrations following the same approach used for benthic invertebrates living in sediment. Internal radiation doses were estimated using tissue radionuclide concentrations and, for screening purposes (Tier 1), the internal dose conversion factors (DCFs) of Amiro [15], corrected for a relative biological effectiveness (RBE) of 40 for alpha emitters and 3 for tritium, a beta-emitter [16]. The external dose was estimated using radionuclide concentrations in the external medium (water and sediment) and the external dose factors of Amiro [15]. Amiro's [15] internal DCFs assume that all the emitted radiations are totally absorbed within the organism, which is a reasonable for larger organisms and alpha emitters. When RQs are greater than 1, dosimetry equations are used to correct for geometry and size for gamma and beta emitters [4, 17].

## 3. CASE STUDY – URANIUM MINES AND MILLS

There are currently five fully operating U mines and mills in northern Saskatchewan and several decommissioned mines in both Saskatchewan (Beaverlodge Lake area) and Ontario (Elliot Lake area). Operating facilities that are the focus of the case study for which both a realistic (deterministic) Tier 2 assessment and a probabilistic assessment were performed are Rabbit Lake, Key Lake, and Cluff Lake sites. Baseline (pre-operational) data for northern Saskatchewan were also assessed to account for background concentrations at these U mine sites.

Realistic RQs calculated for radiation exposure at the U mines and for GM and 90% confidence limits for baseline (pre-operational) conditions are presented in Table 2. The RQs were much  $<1$  for biota at both GM and 90% confidence limits for baseline conditions in northern Saskatchewan. This indicates that the general methodology and ENEVs are reasonable.

TABLE 2. REALISTIC RISK QUOTIENTS (RQS) CALCULATED FOR THE RADIO-TOXICITY OF RADIONUCLIDES RELEASED TO THE ENVIRONMENT FROM URANIUM MINES AND MILLS

	Northern Saskatchewan Baseline <sup>1</sup>			Cluff Lake Area <sup>2</sup>			Realistic		
	GM	90% CL		Sandy Lake 1998	Island Lake 1999				
Fish	0.01 <sup>2</sup>	0.02 <sup>2</sup>		0.04	0.21				
benthic invertebrates	0.012	0.03		0.1	0.45				
Algae	0.01 <sup>2</sup>	0.04 <sup>2</sup>		0.01	0.54				
macrophytes	0.03 <sup>2</sup>	0.04 <sup>2</sup>		0.01	0.13				

	Rabbit Lake Area <sup>2</sup>					Key Lake Area <sup>2</sup>			
	Upper Link Lake	Lower Link Lake	Horseshoe Lake	Collins Bay	Collins Bay Stn S9	McDonald Lake	Little McDonald Lake	Fox Lake	Wheeler River
fish	4.1	0.79	0.82			1.1			
Benthic invertebrates	11.2	2.2	2.8	0.25	0.19	0.18 – 0.3	0.96	0.28	
algae	2.5	0.6	0.42			0.03			0.36
macrophytes	0.53	0.6	0.05						

<sup>1</sup> Calculated from GM or 90% CL baseline radionuclide water concentrations using GM concentration ratios.

<sup>2</sup> Area lakes separated out to show spatial extent of potentially harmful effects on various taxa.

The Cluff Lake mine is located in the Island Creek watershed. Treated effluent from the tailings management area enters Island Lake, which drains into Sandy Lake. At Cluff Lake, RQs are <1 in Island Lake (Table 2), the first lake to receive effluent discharges, which indicates that there is little potential for harmful effects.

The Rabbit Lake mine is the oldest operating U mine and mill facility in Saskatchewan. Milling commenced in 1975. At this mine site, high RQs were calculated for fish, benthic invertebrates and algae in Upper Link Lake and for benthic invertebrates in Lower Link Lake and Horseshoe Lake (Table 2) indicating the potential for harmful effects to these organisms.

The Key Lake mine is located in north-central Saskatchewan. Treated mill effluent is released to Wolf Lake, which discharges to Fox Lake, eventually flowing into Wollaston Lake. Groundwater from dewatering activities at the ore bodies is discharged to Horsefly Lake, which flows into Little McDonald Lake, McDonald Lake, to the Wheeler River. A RQ slightly greater than 1 was calculated for fish (Table 2) in McDonald Lake indicating a potential for harmful effects to fish. All other RQs are less than 1.

### 3.1 Probabilistic assessment of Cluff, Key and Rabbit Lake Projects

The probabilistic assessment for the radiation effects on fish, benthic invertebrates and algae was carried out as described in the realistic assessment, except that probability distribution functions were used for radionuclide concentrations in water, sediment and biota based on monitoring data. Probability distributions for radionuclide concentrations in water, sediment and biota were lognormally distributed with the measure of central tendency, with the geometric mean based on data for the most recent 2–5 years of monitoring data. A full description of the methodology is available in ECOMatters Inc. [18].

The probability of a RQs >1 due to radiation exposure in lakes at these mine sites is given in Table 3. At the Rabbit Lake site, high probabilities (i.e., > 0.5) of exceeding a RQ of 1 exist for fish, benthic invertebrates and algae in Upper Link Lake and to a lesser extent in Lower Link Lake. The probability of exceeding a RQ of 1 is also high for benthic invertebrates in Horseshoe Lake. The probability of exceeding a RQ of 1 is more limited at the other two mine sites. Probabilities >0.50 are observed at the Cluff Lake site for fish in Sandy Lake based on water concentrations. In the case of the Key Lake operation, a high probability of exceeding a RQ of 1 is observed for fish in MacDonald Lake.



TABLE 3. PROBABILITY OF A RISK QUOTIENT GREATER THAN UNITY AS A RESULT OF RADIATION EXPOSURE IN THE CLUFF LAKE, RABBIT LAKE AND KEY LAKE AREAS. A GEOMETRIC MEAN RADIATION DOSE THAT RESULTS IN A RISK QUOTIENT OF ONE WOULD HAVE A PROBABILITY OF 0.5

	Algae	Fish based on water	Fish based on tissue analysis	Benthic Invertebrates
<b>Cluff Lake Area Lakes</b>				
Cluff Lake	0.03	0.40	0.06	0.03
Douglas River	0.03	0.00	–	–
Island Lake	0.40	0.26	0.18	0.26
Lac Phillip	0.17	0.45	0.00	0.19
Sandy Lake	0.04	0.54	0.00	0.00
Snake Lake	0.14	0.14	0.12	0.02
<b>Rabbit Lake Area Lakes</b>				
Collins Bay	0.02	0.96	–	0.17
Collins Bay at Eagle Eagle Point	0.25	0.91	–	0.12
Hidden Bay 1	–	–	–	0.04
Hidden Bay 2	0.26	0.42	–	0.03
Hidden Bay 4	0.01	0.30	–	0.04
Horseshoe Lake	0.38	0.91	0.44	0.71
Lower Link Lake	0.45	0.91	0.42	0.58
Pow Bay	0.02	0.77	–	0.05
Upper Link Lake	>0.7	1.00	0.82	0.97
Wollaston Lake	0.05	0.12	–	0.08
<b>Key Lake Area Lakes</b>				
David Lake	0.11	0.00	0.00	0.00
David Creek	0.07	0.00	–	0.00
Delta Lake	0.06	0.00	0.00	0.00
Fox Lake	–	–	–	0.18
Little MacDonald Lake	0.13	0.00	–	0.44
MacDonald Creek	0.05	0.00	–	–
MacDonald Lake	0.08	0.00	0.51	0.14
Martin Lake	0.04	0.00	0.00	0.00
Outlet Creek	0.05	0.00	–	–
Russel Lake	0.03	0.00	0.00	0.00
Unknown Lake	–	–	–	0.04
Wheeler Lake	0.09	0.01	–	–
Wheeler Creek	0.06	0.00	–	–
Wilson Lake	–	–	0.00	0.00
Yak	0.14	0.01	–	–

It is noteworthy that the probability of a risk quotient greater than unity is much higher for fish when the radionuclide concentrations in fish are based on the use of water concentrations and CRs rather than measured concentrations in fish. An exception is the probability for fish in MacDonald Lake (Key Lake site), where the probability based on the observed concentrations in fish is higher than that estimated from concentrations in water. However, the majority of cases with measured tissue concentrations have RQs lower than those based on water concentrations. This suggests that the use of CRs may result in overly conservative risk estimates.

The results of the probabilistic assessment (Table 3) confirm the results of the realistic assessment (Table 2). Both the realistic and probabilistic assessments demonstrate that there is little likelihood of harmful effects from radiation exposure at the Cluff Lake and Key Lake mine sites. At the Rabbit Lake mine site harmful effects of radiation are likely in Horseshoe Lake and in the Link lakes.

The results of this case study also suggest that a realistic Tier 2 assessment adequately describes environmental risks and that a probabilistic assessment does not improve the bases for decision-making. Probabilistic assessments are likely only of significant value when environmental data are not readily available, for example for fore-casting the potential environmental effects of proposed projects.

#### 4. SUMMARY AND CONCLUSIONS

For the assessment of the effects on non-human biota of discharges of hazardous substances and radioactive nuclear substances to the environment from nuclear facilities, an ecological risk assessment approach is recommended. An ERA approach allows both radioactive and non-radioactive substances to be assessed using consistent methodology and relative risks to be compared in a transparent manner because assessment benchmarks (i.e., ENEVs) for both radiation and hazardous chemicals are derived using an ecotoxicological method. The work conducted to date, as illustrated by the uranium mines and mills case study, indicates that the ERA approach, as developed by CNSC staff for radiation exposure, can appropriately take into account background concentrations of naturally occurring radionuclides and the spatial extent of effects. Furthermore, the results of the probabilistic assessment (Tier 3) are in agreement with those for the deterministic realistic assessment (Tier 2). This suggests that a probabilistic assessment is not necessary as long as realistic values are used in the assessment. In the case of facilities for which risk characterization stops at a realistic Tier 2, valid, scientifically sound conclusions on the likelihood and spatial extent of effects can be made. This is supported by the work of Richardson [19] indicating that the deterministic approach is no less realistic than a probabilistic approach. A probabilistic approach, however, is needed when data are scarce or for predicting future environmental performance of a facility.

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# A method of impact assessment for ionising radiation on wildlife

S. Jones<sup>a</sup>, D. Coppleson<sup>b</sup>, I. Zinger-Gize<sup>c</sup>

<sup>a</sup> Westlakes Research Institute, Cumbria, United Kingdom

<sup>b</sup> Environmental Research and Consultancy, University of Liverpool, Birkenhead, United Kingdom

<sup>c</sup> Environment Agency, Warrington, United Kingdom

**Abstract.** A method for undertaking an impact assessment of ionising radiation on wildlife has been developed for the UK Environment Agency, who has a legal requirement to assess the impacts of consents and authorisations for discharges under the Habitats Regulations (1994). A simple approach has been adopted based on the latest thinking in the field, in which the calculation of doses to wildlife are determined by their size, internal incorporation of radionuclides and external exposure in the environment. The doses are calculated either by using literature derived values or empirical measurements of radionuclide concentrations at the site of interest. The data required to enable dose calculations are:

- Concentrations of each radionuclide in the soil/sediment, water or air (from empirical or modelled approaches);
- Concentration factors for each radionuclide in each organism to be assessed relative to soil, water or air (based on literature values or actual measurements at the site of interest);
- Organism dimensions (as an ellipsoid);
- The proportion of time the organism spends in different 'compartments' of the ecosystem.

The impact assessment approach then focuses on three ecosystems representative of those considered potentially most at risk from the impact of authorised radioactive discharges, namely a coastal grassland (terrestrial ecosystem); estuarine and freshwater ecosystems. A range of target organisms have been chosen to be representative of species found in each ecosystem. Several radionuclides were initially selected for the impact assessment:

- Estuarine and freshwater ecosystems:  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{239+240}\text{Pu}$ ,  $^{238}\text{U}$ ,  $^{129}\text{I}$ ,  $^{210}\text{Po}$
- Terrestrial ecosystem:  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{35}\text{S}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{239+240}\text{Pu}$ ,  $^{238}\text{U}$ ,  $^{129}\text{I}$ ,  $^{226}\text{Ra}$ .

Subsequently, the method has been extended to additional radionuclides, including all the major natural series radionuclides. Dose calculations have been programmed into user-friendly Excel spreadsheets using Visual Basic for Applications. The user may vary the default values used in the assessment for all major parameters. In order to demonstrate the flexibility of the method, results for an example scenario based on published data for natural series radionuclide concentrations in wildlife of the Magela and Cooper Creek systems in the Northern territory of Australia are presented.

## 1. INTRODUCTION

The 1990 Recommendations of the ICRP [1] include the statement that “radiological control of the environment to the standard necessary to protect humans will ensure that other species are not put at risk”. Over the last six years, a broad international consensus has developed to the effect that this statement, whilst it may be true in many circumstances, lacks detailed scientific justification. In addition, the overwhelming focus on human protection appears out of step with the principles of the Rio Declaration [2], which place environmental protection *per se* as a cornerstone of sustainable development. Many have urged the development of a more detailed and transparent framework for the protection of the environment from ionising radiation (e.g. [3, 4]), and the European Commission has funded the project “Framework for Assessment of Environmental Impact” (FASSET), due to complete in October 2003, to develop such a framework.

Within the UK, the Environment Agency has a statutory duty to ensure the protection of the environment, including the setting of authorisations for radioactive discharges and the disposal of radioactive wastes. English Nature is responsible for designating, and monitoring the conservation status of, Sites of Special Scientific Interest (SSSIs), as well as other European designated conservation sites; it is also a statutory consultee regarding radioactive discharges and waste disposal authorisations. During 2001, these two agencies initiated the development of an interim methodology for assessing the impact of ionising radiation on the environment, in order to assist them fulfil their statutory responsibilities under the Habitats Regulations, pending ongoing international developments through FASSET, ICRP, IAEA and UNSCEAR. This culminated in a detailed report issued in June 2001 as Environment Agency R&D Report 128 [5], of which this paper provides a very brief summary.

As a result of that publication, the two organisations have revised their guidance to incorporate the methodology. The new guidance is presented in detail in a separate article in this volume (Regulatory guidance in England and Wales to protect wildlife from ionising radiation, I. Zinger-Gize, D. Copplestone, and C. Williams).

## 2. RADIATION EFFECTS AND RELATIVE BIOLOGICAL EFFECTIVENESS

There is a very extensive literature on the effects of ionising radiation on individual organisms from a wide variety of different taxa; a smaller part of the literature refers to effects at higher levels of organisation such as ecosystems, communities or populations [6]. Interpretation of the literature in the context of environmental protection is complicated by the variety of endpoints studied and uncertainty as to which of these endpoints would be of major relevance for environmental protection; and also by the preponderance in the published literature of studies which involve relatively high doses of gamma and/or beta radiation delivered over relatively short periods of time. For the purpose of environmental protection the effects of chronic radiation dose rates are of most interest, and it is necessary to take account of radiation types with high linear energy transfer (in particular, alpha particle radiation) as well as low linear energy transfer types such as beta and gamma.

Presently, the best considered advice on chronic radiation dose rates unlikely to harm ecosystems is that advanced by UNSCEAR [6], based in part on earlier reviews [7–9] together with consideration of more recent literature. Thus chronic radiation dose rates of 40 and 400  $\mu\text{Gy h}^{-1}$  are considered unlikely to harm terrestrial and aquatic ecosystems respectively. The interim methodology has adopted these dose rates as reference levels, applying factors of caution to allow for uncertainty in the assessment (see below).

It is well established that radiation with high linear energy transfer is more biologically damaging, for the same absorbed dose, than radiation with low linear energy transfer. The quantity *relative biological effectiveness* (RBE), which expresses this difference numerically, depends on radiation type, dose rate, and the endpoint under consideration. Generally, RBE values for high LET radiation are greatest for low dose rates and stochastic effects (such as carcinogenesis) and lowest for high dose rates and non-stochastic effects (such as impairment of fertility). In human protection, *radiation weighting factors* are defined as a broad interpretation of the RBE data, focussing on the effects which are most important in determining health detriment at low dose rates, and are used in converting absorbed doses into effective doses for comparison with dose limits. The interim methodology defines ‘default’ radiation weighting factors for alpha and low energy beta radiation, which are used in the calculation of doses to biota.

In human radiological protection, a radiation weighting factor of 20 is used for alpha radiation, based largely on RBE data relevant to carcinogenesis as an endpoint of concern [1]. In relation to protection of biota, several authors have argued that lower factors (between 5 and 10) would be appropriate, on the basis that the endpoints of ecological concern are more likely to be non-stochastic effects such as impairment of fertility [6, 10, 11]. Others have argued for continued use of the factor of 20 since this would allow also for potentially important stochastic effects such as genetic damage [12, 13], whilst a value of 40 has been proposed for use in Canada [14]. The interim methodology uses 20 as an adequately cautious ‘default’ value, although this and other factors can easily be modified by the assessor (see below).

In human radiological protection, beta radiation of all energies is considered to have a radiation weighting factor of 1; however a relationship between RBE and linear energy transfer suggests values greater than 1 for beta particles with energies of a few keV or lower [1]. There is also experimental evidence that beta radiation from tritium (which has an average beta energy of 6 keV) has a relative biological effectiveness of between 1 and 3 for a range of endpoints [15]. Therefore, the interim methodology uses 3 as a cautious default value for all beta particles and electrons with an average energy of less than 10 keV, and 1 for all other beta and gamma radiations.

### 3. DOSIMETRIC METHODS

The dosimetric quantity used for biota in the methodology is absorbed dose, adjusted by the above radiation weighting factors for alpha particles and low energy beta particles or electrons. In calculating absorbed doses from internally incorporated radionuclides it is necessary to calculate the fraction of decay energy which is absorbed within the organism (the absorbed fraction); likewise, in calculating absorbed doses from radionuclides in the medium surrounding the organism it is necessary to allow for self shielding by the organism. The method used is based on point specific absorbed fractions [16, 17], which have already been applied to ecosystem dosimetry [7, 18, 19], and provide a good balance between physical realism and computational practicality. The key assumptions in the dosimetric calculations are:

- organisms are represented by ellipsoids of appropriate dimensions;
- radionuclides are uniformly distributed within the organism;
- radionuclides are uniformly distributed within surrounding media, which are infinite or semi-infinite in extent;
- density differences between the organism and surrounding media, or within the organism itself, are ignored;
- absorbed dose is calculated as an average throughout the volume of the organism.

Absorbed fractions are calculated numerically using a method which repeatedly samples source-receptor pairs within the volume of the organism [5, 20, 21].

### 4. REFERENCE ECOSYSTEMS AND ORGANISMS

Definition of reference ecosystems and reference organisms [3, 22] is a necessary simplifying step in the evaluation of doses to biota, and appears to be broadly accepted as the basis for development of a system for radiological protection of the environment. The interim methodology has defined reference ecosystems for a coastal environment; a freshwater river; and a terrestrial grassland. These ecosystems are populated by reference organisms as follows:

Freshwater	Coastal	Terrestrial	
Bacteria	Bacteria	Bacteria	Carnivorous mammal
Macrophyte	Macrophyte	Lichen	Rodent
Phytoplankton	Phytoplankton	Tree	Bird
Zooplankton	Zooplankton	Shrub	Bird egg
Benthic mollusc	Benthic mollusc	Herb	Reptile
Small benthic crustacean	Small benthic crustacean	Seed	
Large benthic crustacean	Large benthic crustacean	Fungus	
Pelagic fish	Pelagic fish	Caterpillar	
Benthic fish	Benthic fish	Ant	
Amphibian	Fish egg	Bee	
Duck	Seabird	Woodlouse	
Aquatic Mammal	Seal	Earthworm	
	Whale	Herbivorous mammal	

For each of these reference organisms, the following parameters are defined:

Factor	Relevance
Ellipsoid dimensions (3 axes)	Determines absorbed fraction for internally incorporated radionuclides.
Concentration ratio (radionuclide dependent)	Determines equilibrium radionuclide concentration in organism for unit radionuclide concentration in surrounding medium.
Occupancy factors	Determines fraction of time spent by organism burrowed into soil/sediment, at soil/air or sediment/water interface, in air or in water column, etc – for the purpose of external dose calculation.
Dose per unit concentration – internal (radionuclide dependent)	Absorbed dose for unit concentration of radionuclide incorporated within the organism. Calculated separately for low energy beta, other betas and photons, and alpha to allow variation of radiation weighting factors.
Dose per unit concentration – external (radionuclide dependent)	Absorbed dose for unit concentration of radionuclide in the medium surrounding the organism. Calculated separately for low energy beta, other betas and photons, and alpha.

These parameters, together with the separately specified radiation weighting factors for low energy beta and alpha radiation, permit the calculation of weighted absorbed doses to each reference organism based on predicted or measured radionuclide concentrations in water, soil or air [5]. For the aquatic environment, concentration ratios are specified relative to water; for the terrestrial environment, concentration ratios are specified relative to soil for radionuclides which have half-lives long enough to accumulate in soil. For shorter lived radionuclides and radionuclides such as  $^3\text{H}$  and  $^{14}\text{C}$ , for which an isotopic dilution approach is appropriate, concentration ratios for the terrestrial environment are specified relative to air.

The principal limitation of the equilibrium concentration ratio approach is the difficulty of adequately describing the scenario of ongoing atmospheric deposition into a terrestrial ecosystem; in order to do this a dynamic modelling approach is required, but at present it is not possible to construct an adequate dynamic model for all the required reference organisms. However, where measured concentrations of radionuclides in organisms are available, these can be used directly in place of values derived using concentration ratios.

## 5. SPREADSHEET ASSESSMENT PROGRAM

The necessary calculations within the developed methodology have been coded into a program utilising Microsoft® Excel and Visual Basic for Applications. For each of the three ecosystems, this program allows the user to:

- Calculate weighted absorbed doses to reference organisms based on predicted or measured radionuclide concentrations in soil, air or water;
- use supplied default concentration ratio values and occupancy factors, or available site specific data, in the assessment;
- validate calculated radionuclide concentrations in organisms against measured values and/or use measured values in the calculation in place of calculated values;
- alter the radiation weighting factors from the supplied default values;
- present dose rate results for each reference organism graphically;
- carry out sensitivity analyses to explore the effect of changes in assumptions on the calculation results;
- save calculation results and input parameters/assumptions into a separate Excel workbook.

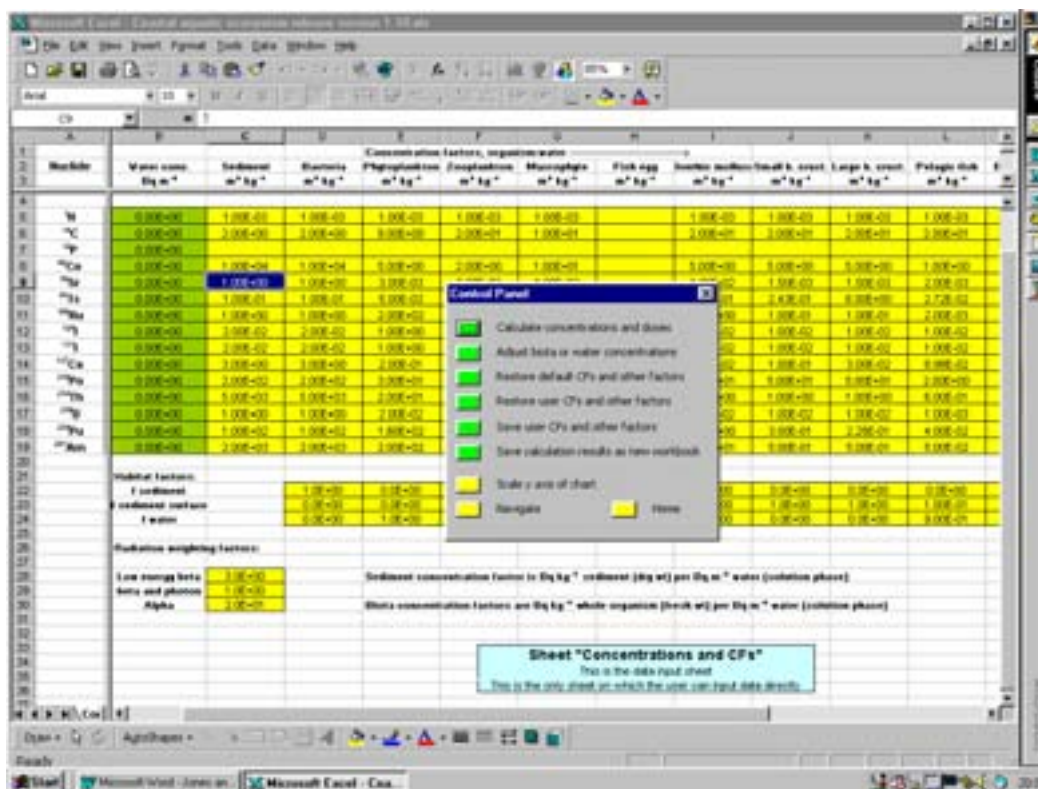


FIG. 1. Data input sheet for the assessment spreadsheet program for the coastal/marine environment, showing the 'control panel' menu.

The spreadsheet program is protected to prevent changes by the user to default parameter values and the underlying Visual Basic code; all operations are executed through user-friendly menus (Figure 1), which ensure that calculations are carried out in a correct and consistent sequence. All assumptions and limitations are clearly stated within the spreadsheet, and the program should only be used in conjunction with the associated report [5].

The initial release of the three spreadsheet programmes provided dosimetric and concentration ratios for nine radionuclides (estuarine/freshwater ecosystems:  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{239+240}\text{Pu}$ ,  $^{238}\text{U}$ ,  $^{129}\text{I}$ ,  $^{210}\text{Po}$ ; terrestrial ecosystem:  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{35}\text{S}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{239+240}\text{Pu}$ ,  $^{238}\text{U}$ ,  $^{129}\text{I}$ ,  $^{226}\text{Ra}$ ); the next release, due out in May 2002 *via* the Environment Agency, will extend this coverage to 16 radionuclides (by adding  $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ ,  $^{131}\text{I}$ ,  $^{125}\text{I}$ ,  $^{234}\text{Th}$ ,  $^{234\text{m}}\text{Pa}$ ,  $^{241}\text{Am}$ ,  $^{32}\text{P}$ ).

## 6. INTERPRETATION OF CALCULATION RESULTS

As explained above, based on the conclusions of UNSCEAR and IAEA, dose rates of 40 and 400  $\mu\text{Gy}^{-1}$  are used as 'reference levels', which are unlikely to cause harm to terrestrial and aquatic ecosystems respectively. However it is recognised that the calculation of doses to organisms involves a degree of uncertainty, especially if default parameters, rather than site specific parameters, are being used in the calculations.

Therefore, if dose rates calculated using this methodology exceed 5% of the above reference values (i.e. 2 and 20  $\mu\text{Gy h}^{-1}$ ), further consideration of the situation is required, which may include one or more of the following actions or investigations:

- consideration of the likely radiosensitivity of the organisms receiving the highest calculated doses, and the main sources of uncertainty in the calculation for those organisms;
- additional measurements to determine site specific parameters and/or concentrations of key radionuclides in biota;



- for an existing contaminated ecosystem, initiation of appropriate ecological or biomarker studies;
- for a prospective assessment, precautionary action to limit or reduce radionuclide emissions.

The detailed report [5] also provides summary tables of experimental and field study data on the effects of ionising radiation to wildlife, which may also assist in the impact assessment process.

## 7. EXAMPLE OF APPLICATION – FRESHWATER ECOSYSTEMS IN THE NORTHERN TERRITORIES, AUSTRALIA

To illustrate the flexibility of the methodology, we have applied it to the assessment of radiation doses to freshwater organisms in the Magela Creek system, close to the Ranger uranium mine, using published data on concentrations of uranium series radionuclides in water and in a range of organisms which form part of the aboriginal diet [23, 24]. For this assessment we used the latest version of the freshwater ecosystem spreadsheet, further extended by the addition of dosimetric factors for  $^{40}\text{K}$ ,  $^{222}\text{Rn}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{230}\text{Th}$  to provide complete coverage of the key natural radionuclides. This version is being used in ongoing development work as part of the EU FASSET project, to assess the range of exposures to biota arising from natural sources.

Concentrations of filtered water were taken from the Georgetown billabong site, which shows the highest reported concentrations in the creek system (Table 1). Concentration ratios were taken for the Magela creek system where cited [23, 24], and from other referenced sources [25–28] where no site specific data were available (Table 2).

Dose rates to organisms were assessed using both provisional ‘default’ Concentration Factor (CF) values for generic assessments, and the site-specific data for the Georgetown billabong derived as explained above. The calculated dose rates include the weighting factors of 3 for low energy beta particles, and 20 for alpha particles. The results of the dose assessment are shown in Table 3.

Based on our criteria for use of the method, dose rates to phytoplankton and benthic molluscs exceed  $20 \mu\text{Gy h}^{-1}$ , and further investigations including the use of additional site specific data would be required. Site specific data for freshwater mussels appear to be available [24] but values are not cited in the published paper, so we have not been able to include those data in these example calculations. Calculated dose rates to other organisms for which site specific data are available are lower than, or similar to, the dose rates calculated with provisional default CF values, with the exception of dose rates to benthic bacteria. Here, the apparent accumulation factors from water to sediment at the site appear high; this may reflect a contribution to activity concentration in sediments from a purely mineralogical component.

Concentrations of natural series radionuclides in the Georgetown billabong are clearly elevated well above the normal natural levels [8]. However, they do not necessarily reflect anthropogenic enhancement due to uranium mining activities; rather, they are likely to reflect localised natural enhancement due to the uranium mineralisation in the area [23]. The results of the example dose calculations may therefore be considered indicative of enhanced radiation exposures to freshwater organisms resulting from the variability of natural radionuclide concentrations in the environment.

TABLE 1. RADIONUCLIDE CONCENTRATIONS IN FILTERED WATER FROM THE GEORGETOWN BILLABONG, USED FOR THE EXAMPLE ASSESSMENT

Radionuclide	Concentration ( $\text{Bq m}^{-3}$ )
$^{210}\text{Po}$	10
$^{226}\text{Ra}$	9
$^{228}\text{Th}$	3
$^{230}\text{Th}$	5
$^{232}\text{Th}$	2
$^{234}\text{U}/^{238}\text{U}$	8

TABLE 2. DERIVATION OF CONCENTRATION FACTORS FOR THE EXAMPLE ASSESSMENT

Reference organism	Interpretation	Reference for CF data
Bacteria	Sediment	[23]
Phytoplankton	Phytoplankton	[25, 28]
Macrophyte	Waterlily (rhizome)	[23]
Benthic mollusc	Mussel	[26–28]
Small crustacean	Freshwater shrimp	[24]
Large crustacean	None assessed	
Amphibian	Turtle	[24]
Pelagic fish	Fish ARR group 2	[24]
Benthic fish`	Fish ARR group 1	[24]
Aquatic mammal	None assessed	
Waterbird	Magpie goose	[24]

TABLE 3. RESULTS OF THE EXAMPLE DOSE ASSESSMENT

Organism	Default parameters		Site specific parameters	
	Dose rate $\mu\text{Gy h}^{-1}$	Principal nuclide(s)	Dose rate $\mu\text{Gy h}^{-1}$	Principal nuclide(s)
Bacteria	14	$^{226}\text{Ra}$	99	$^{226}\text{Ra}$
Phytoplankton	24	$^{210}\text{Po}$		
Macrophyte	10	$^{210}\text{Po}$	6.5	$^{226}\text{Ra}$
Benthic mollusc	51	$^{210}\text{Po}$		
Small crustacean	7	$^{210}\text{Po}$	2.1	$^{226}\text{Ra}, ^{210}\text{Po}$
Amphibian	1.3	$^{210}\text{Po}, ^{226}\text{Ra}$	1.6	$^{226}\text{Ra}, ^{210}\text{Po}$
Pelagic fish	1.2	$^{226}\text{Ra}$	0.75	$^{226}\text{Ra}$
Benthic fish	10	$^{210}\text{Po}$	5.1	$^{226}\text{Ra}$
Waterbird	0.34	$^{210}\text{Po}, ^{226}\text{Ra}$	0.39	$^{210}\text{Po}, ^{226}\text{Ra}$

Note: calculated dose rates include weighting factors of 3 for low energy beta particles, and 20 for alpha particles.

## 8. CONCLUSIONS

This interim methodology has shown that it is practicable to put together an approach for the assessment of radiological impacts on the environment, along the lines being pursued by FASSET and other initiatives.

The use of the method on a variety of assessments, including the example described in this paper, has established that it provides a logical approach to assembling the data required for an assessment, and for progressing from a generic assessment, using default parameters, to a more realistic assessment using site specific data. Once the required data are assembled the spreadsheet program makes the actual calculation of doses extremely easy.

The principal challenges to be overcome in further development appear to be:

- achieving transparency in the derivation of ‘reference’ levels of dose or dose rate to biota, including clear linkage to endpoints and the recognition of differences in sensitivity between different taxa;
- derivation of concentration ratios or transfer factors for a sufficiently wide range of reference organisms and radionuclides;
- adequately describing the dynamics of radionuclide transfers within ecosystems, particularly for the case of ongoing atmospheric deposition into terrestrial ecosystems.

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# **An ecosystem approach to assess radiation effects on the environment used for nuclear waste disposal facilities\***

**U. Kautsky<sup>a</sup>, L. Kumblad<sup>b</sup>**

<sup>a</sup> Swedish Nuclear Fuel and Waste Management Company, Stockholm, Sweden

<sup>b</sup> Department of Systems Ecology, Stockholm University, Stockholm, Sweden

**Abstract.** Traditionally safety assessments of repositories with long-lived nuclear waste have used a stylised biosphere and usually only addressed pathways to humans. However, new regulations, e.g. in Sweden, require that also effects on the environment should be evaluated. This means that important pathways not leading to exposure to humans needs to be considered because they may affect other biota than humans. Moreover, the predictions over long time periods required, often 1000–100000 years, can shift an initially high accumulating, but abiotic environment to a highly exposed biotic environment due to land-rise, climate changes etc.

In the last three safety assessments of radioactive waste facilities, the Swedish nuclear fuel and waste management company (SKB) has used an ecosystem approach to address these issues. The ecosystem approach, which earlier has been used for other substances, such as PCBs and PAHs, includes site-specific dispersal modelling, strictly based on mass-balance. Methods from systems ecology have been used, which have the advantage to constrict the mass-flow within the ecosystem as well as the flows over the system borders. The models are based on natural processes such as total photosynthesis, decomposition and material transfer with carbon (i.e. food) and extrinsic drivers are insolation, nutrient supply and water transport. Since the models are founded on fundamental processes, they can be scaled to future ecosystems describing successions e.g. from estuaries, lakes, mires to farmland. Our conclusion is that these methods probably are the only way to address radiation effects on the environment as well as humans. Moreover, the models are independent on whether the hazard substance is a radio-nuclide or another pollutant, thus it enables to overbridge and maybe unify ecotoxicological and radio-ecological issues.

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\* Only an abstract is given as the full paper was not available.

# Consideration of biota dose assessment methodology in preparation of environmental impact statements

T. Harris<sup>a</sup>, E. Pentecost<sup>b</sup>

<sup>a</sup> United States Nuclear Regulatory Commission, Washington, DC, United States of America

<sup>b</sup> Argonne National Laboratories, Argonne, IL, United States of America

**Abstract.** The U.S. Nuclear Regulatory Commission (NRC) has been working with the U.S. Department of Energy (DOE) and U.S. Environmental Protection Agency on developing methods, models, and guidance within a graded approach for evaluating radiation doses to biota. This effort has been headed by DOE and is predominately a screening tool to demonstrate compliance with dose to biota standards at existing DOE facilities. The graded approach does, however, have the capability of incorporating site specific pathway and biota data to estimate doses to biota. Two questions under consideration by the NRC are: is a biota dose assessment methodology needed in the existing National Environmental Policy Act (NEPA) framework and how should a biota dose assessment methodology be incorporated into the preparation of an Environmental Impact Statement (EIS). The paper discusses the protection of biota in context of the existing framework of NEPA and the Endangered Species Act, and how protection of biota is typically addressed in EISs prepared by the NRC. The paper also discusses whether the proposed biota dose methodology needs to be considered in preparation of NEPA documents, given the protection afforded under these Acts. The paper also identifies circumstances where an NRC NEPA document would use the dose methodology for impact assessment to biota.

## 1. INTRODUCTION

In recent years, reliance on the International Commission on Radiological Protection (ICRP) principle *that if man is adequately protected, then other living things are also likely to be sufficiently protected* [1, 2] has been questioned. Issues regarding the development of a separate dose standard and methodology for the protection of biota from ionizing radiation are the subjects of significant international debate. The International Atomic Energy Agency (IAEA) has been working to develop a framework that could be used by member countries to establish biota protection standards. This framework maps ethical and cultural views to the protection of biota from ionizing radiation. Cultural views may be anthropocentric (human focused), ecocentric (ecosystem focused), or biocentric (individual species focused). There are also numerous international efforts to develop methodologies to conduct dose assessments for biota (e.g., Framework for Assessment of Environmental Impact (FASSET), Environmental Protection from Ionizing Contaminants in the Arctic (EPIC)).

Within the United States, the U.S. Department of Energy (DOE) currently has a dose limit for protection of aquatic organisms (DOE Order 5400.5) and has proposed dose limits for terrestrial biota (10 CFR 834, Subpart F). These standards are based on the recommendations for non-human biota in IAEA Technical Report Series No. 332 and National Council on Radiation Protection (NCRP) Report No. 109 [3–5]. DOE has developed a methodology [6] for DOE facilities to demonstrate compliance with these standards. DOE is also developing a computer code (i.e., RESRAD-BIOTA) [7] to support its efforts.

Currently, the U.S. Nuclear Regulatory Commission (NRC) does not have a separate dose standard for biota. Rather, NRC has relied on the principle that by protecting man, the environment is protected. The broader question of whether a biota dose standard should be developed within the U.S. is not the focus of this paper. This is a very complex and potentially controversial question that includes consideration of potentially diverse cultural views in determining how to protect biota (i.e., protection of ecosystems, populations, individuals).

This paper discusses protection of biota from ionizing radiation in the context of preparing Environmental Impact Statements (EISs) in accordance with the National Environmental Policy Act of 1969, as amended (NEPA). The assessment of ecological impacts in EISs could be enhanced by considering new information and methodologies on biota impacts from radiation that are being

developed both internationally and within the United States. Specifically, this paper focuses on two questions currently under consideration by the NRC regarding dose impacts to biota and the preparation of EISs. First, is a biota dose assessment methodology needed in the existing NEPA framework? Second, how could a biota dose assessment methodology be incorporated into the preparation of EISs?

## 2. BACKGROUND

This section provides the context for two U.S. environmental laws discussed in this paper. It also discusses guidance documents related to biota protection developed by the U.S. Environmental Protection Agency (EPA) and DOE.

### 2.1. National Environmental Policy Act

NEPA established a national policy that encourages a productive and harmonious relationship between man and his environment. It promotes efforts to prevent or eliminate damage to the environment and biosphere and stimulate the health and welfare of man. The goals of NEPA are to fulfill the responsibility of each generation as trustee of the environment for succeeding generations, and to assure for safe, healthful, productive, and aesthetically and culturally pleasing surroundings [8].

To accomplish these goals, NEPA directed Federal agencies to utilize a systematic, interdisciplinary approach integrating natural, social, and environmental sciences in decision-making for projects that may impact man's environment. That is, the Federal government of the U.S. would specifically consider impacts to the environment in its decision-making. To document the decision-making, Federal agencies, such as the NRC, prepare EISs for major Federal actions that have the potential to significantly affect the quality of the human environment. EISs address the environmental impact of proposed actions and alternatives to the proposed actions. Additionally, EISs describe the relationship between short-term uses of the environment and the maintenance and enhancement of long-term productivity. NEPA encourages consultation with other related Federal agencies in the preparation of EISs.

### 2.2. Endangered Species Act

Numerous other environmental laws have been promulgated within the U.S. following the enactment of NEPA. One of the most significant, relative to protection of biota is the Endangered Species Act of 1973 (ESA). ESA was developed because various species of fish, wildlife, and plants in the U.S. had become extinct or were in danger of, or threatened with, extinction, as a result of economic growth and development untempered by adequate concern and conservation. These species were determined to be of aesthetic, ecological, educational, historic, recreational, or scientific value [8].

ESA provides a means of conserving the ecosystems upon which threatened and endangered species depend (i.e., critical habitats) and establishes a program for the conservation of threatened and endangered species through the U.S. Department of the Interior (DOI). Within DOI, the Fish and Wildlife Service (FWS) determines which plant and animal species are threatened or endangered and publishes a list of these species. FWS also has responsibility for defining geographic areas as critical habitat, pursuant to the ESA, that are necessary for continued survival of an endangered species. The FWS and state agencies also maintain a repository of information on occurrence data and ongoing surveys and research on threatened and endangered species. This information database is made available to potential proponents of new projects for which a federal permit or license is required. This list is revised periodically to include new species that may be threatened or endangered, and to remove species that are no longer threatened or endangered. Under ESA, it is unlawful to take (i.e., kill or harm), import, export, deliver, receive, transport, or sell any listed species. On a national scale, DOI has developed and implemented plans for conservation, survival, and recovery of threatened and endangered species [8].

Consistent with NEPA, ESA also sets forth a requirement for inter-agency cooperation. Under ESA, federal agencies are required to consult with the FWS on any proposed agency action that might involve threatened or endangered species or critical habitat to determine whether the proposed action is, or is not, likely to jeopardize listed species or critical habitat. If threatened or endangered species

may be present, the FWS may require a Biological Assessment (BA) to identify species that are likely to be affected by a proposed action. The BA presents impacts of likely takings; steps to minimize and mitigate such impacts; and alternatives to the proposed action. If a permit is issued to allow incidental takes, then monitoring and reporting would be required in addition to possible mitigation measures [8]. This consultation with the FWS, as it relates to NRC's consideration of biota in EISs, is discussed in the Section 3.1.

Other U.S. laws, which are not discussed in detail in this paper but are relative to biota protection, include Fish and Wildlife Coordination Act; Migratory Bird Treaty Act; Marine Protection, Research, and Sanctuaries Act; Coastal Zone Management Act; Federal Water Pollution Control Act (Clean Water Act); Bald and Golden Eagle Protection Act; and Marine Mammal Protection Act.

### **2.3. Ecological risk assessment**

In addition to the laws discussed above, guidance on evaluation of risks to the environment has been developed by the EPA. Specifically, EPA published "Guidelines for Ecological Risk Assessment" in 1998 [9]. Ecological risk assessments have been used to evaluate estimates of impacts to biota at contaminated sites both for regional and site-specific contaminated areas. Such assessments are often conducted in support of site remedial investigations and feasibility studies necessary for deciding between cleanup alternatives at Superfund sites as required under the Comprehensive Environmental Response Compensation and Liability Act (CERCLA) [10]. The guidelines involve three steps: (1) problem formulation; (2) analyses; and (3) risk characterization. The problem formulation includes integrating available information and establishing assessment endpoints and conceptual models (i.e., how a contaminant moves through the environment to be accessed by receptors). The analyses include an exposure analysis and an ecological response analysis. These analyses are used to develop the risk characterization. The guidelines discuss the use of tiered and iterative approaches in the analyses, starting with screening. The screening process allows users to eliminate obvious non-problem sites from sites where further consideration is warranted [11].

EPA's guidelines are general in nature and could be applied in the evaluation of radiation as a stressor to the environment, but not without some modifications and provision of additional guidance. DOE, through its Biota Assessment Committee, has developed methods, models, and guidance in a Graded Approach (i.e., a multi-tiered process) for evaluating radiation doses to biota [6]. In partnership with EPA and NRC, DOE is developing a computer code (i.e. RESRAD-BIOTA) that uses the Graded Approach as a starting point but incorporates added capabilities to allow for both screening and realistic evaluations of doses to biota [7]. The biota dose criteria used by DOE are 1 mGy/d (0.1 rad/d) for terrestrial animals, and 10 mGy/d (1 rad/d) for aquatic animals and terrestrial plants. The Graded Approach begins with general screening in which users compare radionuclide concentrations in environmental media (e.g., soil, water, sediments) obtained during periodic environmental sampling and monitoring with a set of media and radionuclide specific Biota Concentration Guides (BCGs) for the different media. The BCGs represent the limiting concentrations in environmental media that would not cause the biota dose limits to be exceeded. Site-specific screening and analysis phases, if needed, are also provided. The methodology uses simplified models, conservative generic organism dosimetry, and conservative generic effects estimates. The methodology generally provides protection at the population level, which is generally accepted as the appropriate endpoint [12]. The methodology also allows for the use of site specific parameters to further refine the biota dose estimates. The potential use of the EPA and DOE methodologies in NRC's consideration of biota protection is discussed in Section 3.2.

## **3. DISCUSSION**

Considering the legal framework presented in the previous section, this section discusses how biota protection is typically addressed in EISs prepared by the NRC. It also discusses how EPA ecological risk assessment guidelines and the DOE biota dose methodology could be considered and incorporated into EIS documents. Finally, a discussion of circumstances where an NRC NEPA document would benefit from explicitly considering doses to biota is provided. The conclusion summarizes these discussions and includes actions that may cause NRC to revise its current approach.



### 3.1. Current NRC approach

Although NRC regulates commercial uses of nuclear material, EISs prepared by NRC typically consider both radiological and non-radiological impacts from construction and operation of nuclear facilities over a wide range of resource areas. These resource areas include: air quality; surface water quality; soil and groundwater quality; noise; ecology; cultural, historic and archeological resources; and socioeconomics. Protection of biota is considered in all but the last two resource areas. Human health is also considered in air quality, surface water quality, and groundwater quality.

Impacts to biota focus on “important” species, which are defined as:

- (1) either threatened or endangered species (e.g., Federally listed, proposed for listing, State listed);
- (2) commercially or recreationally valuable species;
- (3) species essential to maintenance of rare or commercially or recreationally valuable species;
- (4) species critical to the structure and function of the ecosystem; or
- (5) species that are biological indicators to potential impacts.

Typical construction impacts to biota would include loss of habitat, obstacles that limit movement of species, noise, erosion, and wetland destruction. Typical impacting factors from normal operations include effluents or discharges to air from stacks and vents, and surface water from pipe outfalls. The concentration of these effluents is limited to protect humans (e.g., general public). These effluents may have thermal effects such as cooling water discharges at nuclear power plants. There may also be chemical effects from the releases of biocides, pesticides, scale or corrosion inhibitors, chlorination, or process chemicals. In addition, there may also be physical effects from facility operations such as loss of habitat; bird injury and mortality from collisions with structures (e.g., natural draft cooling towers, transmission line conductors, buildings); road kill; loss or alteration of unique habitat; or alteration of migratory pathways.

As discussed above, ESA requires that NRC consult with FWS to determine if threatened or endangered species are present and could be impacted from proposed actions. If FWS determines that threatened or endangered species will not be impacted, then no further analyses are performed. The consultation may result in surveys of project areas and the surroundings to better assess potential impacts. If the potential for impacts to threatened or endangered species, or critical habitat is identified, a more detailed BA may be required. The BA would evaluate various stressors (e.g., physical, chemical, biological, radiological) to the habitat and threatened or endangered species. These assessments can be qualitative in nature. The FWS would review the BA and issue a biological opinion. This opinion could state if threatened or endangered species are in jeopardy, in which case additional mitigation and monitoring may be required. If the potential impact is significant enough, the proposed action may not be allowed. The opinion could also state that the potential impact is insignificant, and no further consideration of threatened or endangered species is required.

Ecological impacts from direct exposure of biota to radioactive material or exposure from food ingested that contains radionuclides are currently not explicitly evaluated. Instead, the impacts explicitly evaluated focus on destruction or modification of habitat and protection of threatened and endangered species. The NRC has developed guidance to aid in the preparation of EISs, including assessing the significance of ecological impacts [13, 14].

In addressing protection of biota and the environment from ionizing radiation, the NRC relies heavily on the ICRP principle that “if man is adequately protected, then other living things are also likely to be sufficiently protected.” The standard review plan (SRP) for environmental reviews for nuclear power plants [14] states that the dose to biota should be determined when pathways are identified that could result in higher doses to biota than humans. Because there is not a specific regulatory dose criterion for biota, the SRP suggests using the public dose limit (i.e., 1 mSv [100 mrem]). The SRP notes that it is generally agreed that the limits established for humans are also conservative for other species. Emphasis is placed on maintenance of biota population stability rather than on the fate of individual species, except for threatened and endangered species. The SRP further states that no pathways that would cause significant or harmful exposure to biota have been identified at nuclear power plants.

### 3.2. Incorporating biota dose methodology in EISs

As discussed above, NRC's current biota protection system relies on the evaluation of habitat impacts and on the protection of humans limiting doses to biota. Potential situations where this system could be enhanced are presented in the next section. This section discusses how existing biota protection guidance could be incorporated into evaluations to support EISs, and possible mechanisms for implementing this approach. Impacts to ecological resources from construction activities are not discussed, because licensed radioactive material would not be present during the construction phase and biota would not be radiologically impacted during construction. Therefore, the following focuses on nuclear facility operation and decommissioning phases.

Incorporating EPA's guidelines for ecological risk assessment [9] to protection of biota from ionizing radiation would require some modifications to the guidelines to incorporate radiation as a stressor. Although the guidelines provide a structure for assessing the impacts and risk to biotic systems, the models and effects criteria (i.e., exposure and ecological response analyses) for biota dose assessments are not as well developed as those for chemicals [4, 10–12]. Because radiation as a stressor has unique characteristics, additional considerations would be required in the development of conceptual models (i.e., problem formation) and analysis plans. For example, a significant fraction of the impact (i.e., dose) from radiation can come from external exposure. This is different than for chemicals, in that most of the impact comes from internal exposure and only when the contaminant is biologically available for uptake. The magnitude of external exposure is a function of the penetrating power of the radiation (i.e., radiation weighting factor). Exposure from radioactive progeny would need to be considered. Moreover, the progeny can have different environmental fate characteristics than parent radionuclides [10].

Once appropriate exposure models have been developed to incorporate radioactivity as a stressor, the effects of that exposure would need to be estimated. The appropriate level for assessing the effects of radiation on ecological receptors is generally accepted as the population level [3, 10, 12]. However, the radiosensitivity (i.e., effects from radiation) can vary significantly between phylogenetically similar species and at different stages of life, and the science is far from complete in this area. To date, exposure limits of 1 mGy/d (0.1 rad/d) for terrestrial animals, and 10 mGy/d (1 rad/d) for aquatic animals and terrestrial plants have been proposed as safe exposure levels, which would provide adequate protection [3–6]. The estimation of risk in the EPA guidelines uses a tiered approach. If a screening calculation indicates acceptable risk (e.g., hazard index less than one), then a detailed quantification of risk is not performed. A detailed risk assessment would be difficult to perform, because more sophisticated dosimetric models are not available and the uncertainty associated with the effects data is large [4, 10, 11].

Incorporating DOE's methodology for protection of biota from ionizing radiation [6] into NRC's EIS process would be relatively easy. Available computer codes could be used to model the transport in the environment for air and liquid effluents. The resulting media concentrations (e.g., soil, sediment, water) estimated over time could be easily compared with the screening BCGs to evaluate impacts to biota populations. Existing environmental monitoring could be used to validate predictive models and to further demonstrate that operations were not impacting biota.

Both the EPA guidelines and DOE methodology are not directly applicable to the protection of individuals. Therefore, if threatened or endangered species were present, then the use of species specific effects data and endpoints (i.e., exposure limits) sufficient to protect individuals would be required. The exposure limits discussed above were based on consideration of reproductive effects rather than other effects that might be important to protection of individuals (e.g., early mortality, morbidity). These endpoints are based on a proportion of the lowest observed adverse affect level (LOAEL). However, in order to be protective of individuals rather than populations, effects endpoints would need to be based on the no observed adverse effect level (NOAEL) [11]. It is also possible that, because of the degree of conservatism used in developing the screening BCGs for populations, a site-specific BCG for the protection of threatened and endangered species could be higher than the screening BCGs. (i.e., the screening BCGs may be protective of threatened and endangered species). It would also be critical to determine that a threatened or endangered species is a likely receptor [11]. In addition, the application of either the DOE or EPA approach to evaluate radiation effects on threatened or endangered species would likely need to be coordinated with the FWS.

A mechanism exists for NRC to evaluate potential impacts to biota from exposure to radioactive materials. Potential radioactive contaminants and potential pathways are identified at two initial stages of a permit or license review. The environmental report submitted by an applicant or licensee to support a request (e.g., construction authorization, combined nuclear reactor license application, operation license, decommissioning) typically must identify radiological issues and potential exposures of onsite workers and the nearest residents beyond the facility boundary. At the time of the NRC acceptance review of an application for the above actions, staff members identify potential environmental issues and deficiencies requiring additional data to support an EIS analysis. Radioactive contamination and potential pathways impacting biota and human populations would be reviewed in the context of operational effluents, accident scenarios or a cleanup analysis, such as at a licensed nuclear fuel fabrication facility seeking approval for decommissioning.

### **3.3. Possible situations when biota protection could be explicitly considered**

One of the first situations regarding protection of biota in addition to humans was sea disposal of radioactive waste in the 1980's [3]. (Note: sea disposal is now prohibited) This was certainly a situation where biota could be exposed to a greater degree than humans were. Since that time, other generic situations have been identified where doses to biota may be a concern. During the preparation of the Graded Approach for evaluating radiation doses to biota [6] DOE held a series of workshops. Participants agreed with the IAEA and NCRP that human doses limits would generally protect biota, except when (1) human access is restricted without restricting access by biota, (2) unique exposure pathways exist, (3) rare or endangered species are present, or (4) other stresses are significant [4]. It should be noted that these situations do not necessarily indicate that biota are being harmed, only that further evaluation may be warranted to demonstrate that they are being protected.

Several situations occur during both operation and decommissioning of nuclear facilities where human access is limited. For example, large land buffers exist around some NRC licensed facilities that are precluded from use by members of the public. In these situations, human access is restricted, but access by wildlife species is not (e.g., animals that are not limited by fences such as deer, rodents, birds, and migratory waterfowl). Other situations would involve restricted use decommissioning, where residual contamination, higher than that allowed for unrestricted human access, remains at a site and institutional controls limit human uses and access. Potential radiation impacts to biota have also been identified as a stakeholder concern in DOE's long-term stewardship program [15].

Other situations that involve unique exposure pathways may result from liquid discharges produced by facility operation. For example, radionuclides that accumulate in sediments would typically not be of significance to humans primarily because of the limited exposure time in a recreational scenario. In contrast, benthic fish species and aquatic invertebrates spend significant residence time near or within the sediment layers. Estuary cleanup to limit biota and human impacts from heavy metal contamination have posed a challenge for both EPA and DOE [16–18]. Heavy metals, including radionuclides, have received considerable attention in ecological risk because they are known to bioaccumulate and may biomagnify within the food webs of aquatic and terrestrial ecosystems [19, 20]. Accumulation of radionuclides in sediments could exist where multiple nuclear facilities discharge liquid effluents into the same watershed.

Moreover, for operating and decommissioning facilities, pathways for human exposure are often not significant enough to be considered in radiological assessments [19]. Although these pathways may not be significant for humans, they may be more significant for biota. For example, subsurface contamination that is not very mobile in the environment (e.g., thorium) might not pose a significant impact to humans; however, biota such as burrowing animals may experience direct exposure to subsurface contamination at levels greater than for humans.

## **4. CONCLUSION**

One of the ways NRC documents protection of biota and the environment is through the preparation of EISs. NRC's current biota protection system emphasizes threatened and endangered species, and relies on protecting critical habitat and on the principle that by protecting man, the environment is protected. NRC is closely following work and discussions regarding the validity of this principle in the international community. The broader question, of whether a biota dose standard should be developed

within the U.S., is not addressed in this paper. The focus here is on protection of biota from ionizing radiation in the context of preparing EISs, such as more explicitly including radiation impacts to biota. NRC has started to consider the possible effects a change in the ICRP recommendations would have on preparing EISs. Possible answers to the questions posed in the introduction are discussed below.

First, *is a biota dose assessment methodology needed in the existing NEPA framework?* For protection of threatened and endangered species, loss of critical habitat has the most significant impact on these species. ESA focuses on loss of critical habitat and actions leading to the direct mortality of listed species. NEPA is predominately a decision-making tool for consideration of environmental impacts among various alternatives. Impacts to resource areas do not always need to be precisely quantified, and NEPA does not require the decision maker to choose the least environmentally harmful option. NEPA analyses should provide a basis of comparison among alternatives. Although protection of humans from ionizing radiation does provide some level of protection to biota that inhabit the same areas, NEPA analyses could benefit from documenting biota impacts for alternatives, when, for example, there are limitations to human access. Incorporating the results of an initial screening level ecological risk assessment into the NEPA analyses could be important in certain situations discussed above.

Second, *is how could a biota dose assessment methodology be incorporated into the preparation of EISs?* EISs currently consider ecological impacts that encompass biota impacts. However, the measure of impact is largely based on habitat impacts. If reliance on the ICRP principle were no longer appropriate, then it may be reasonable to adopt a tiered approach of assessing radiological impacts to biota. Additional work would be required with EPA and FWS to ensure a nationally consistent approach to protection of threatened and endangered species that considers both habitat impacts and impacts from radiation. Furthermore, the science to support more detailed biota impacts analyses is not mature at this time and additional research is needed. The contribution of on-going studies such as the FASSET and EPIC programs could significantly enhance this area.

For situations that warrant detailed analyses, using a combination of the EPA guidelines and DOE Graded Approach seems to have several advantages. First, the EPA guidelines would allow the assessment of all stressors (e.g., physical, chemical, biological, radiological). The methods selected would need to take into account unique factors such as bioaccumulation; ecodosimetry radiation weighting factors; and spatial and temporal variability [4, 12, 21]. Second, the DOE Graded Approach would allow for the use of various end points as appropriate to protect either populations or individuals, and the models have been specifically developed to consider radiation as a stressor. Lastly, the EPA approach allows the consideration of risk (i.e., likelihood of adverse effects). Consideration of risk and uncertainty can provide for more realistic and valuable decision-making process.

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# SYMBIOSE: A modeling and simulation platform for environmental chemical risk assessment

M.-A. Gonze<sup>a</sup>, L. Garcia-Sanchez<sup>b</sup>, C. Mourlon<sup>b</sup>, P. Boyer<sup>b</sup>, C. Tamponnet<sup>b</sup>

<sup>a</sup> Present address: C.E. Cadarache, Saint-Paul-lez-Durance, France

<sup>b</sup> Division of Environmental Protection, Institute of Radioprotection and Nuclear Safety, Saint-Paul-lez-Durance, France

**Abstract.** Environmental systems are considered among the most complex ones because they involve a large number of diverse components, interactions, scale issues, spatial heterogeneity and significant sources of uncertainty. An Environmental Chemical Risk Assessment (ECRA) require therefore the integration of a wide range of data and modeling approaches, while accounting for sources and propagation of uncertainties in the system. Further, the level of detail to be achieved in an assessment depends mainly on environmental management objectives and the difficulty of adequately describing exposure, toxicity and other properties of the chemicals with site-specific data. This can range from simplistic conservative analyses to more realistic spatio-temporal modeling approaches. As a consequence, there is a pressing need for *integrated, flexible* (and *user-friendly*) tools that could adapt to this shifting and expanding assessment context. The SYMBIOSE project aims at developing such a modeling and simulation platform, for assessing the fate, transport and effects of chemicals – radionuclides and heavy metals, mainly – on humans and biota, in a multi-media environment. The various aspects of an environmental chemical risk assessment process, and existing relationships between them, are first revisited in a comprehensive way with emphasis on valuable modeling techniques. The modeling approach that will be implemented in the platform is then described through keystone aspects such as *conceptual, mathematical* and *spatial modeling* aspects. Finally, some key ideas about the object-oriented software architecture that is foreseen are presented.

## 1. MAJOR COMPONENTS OF ECRA

As the SYMBIOSE modeling platform is designed as a tool to support the process of environmental chemical risk assessment, it is important to first revisit the various aspects (or *components*) of the exercise, and relationships (or *interactions*) between them. The interaction matrix formalism, first introduced by Hudson [1] for analyzing rock engineering systems, is a convenient way to summarize a somewhat consensual vision of an environmental management problem [2–10], see Figure 1. This kind of conceptual framework gives the assessors a high-level view of the various interacting components, and can help to ensure that the assessment is properly comprehensive, addressing the whole end-to-end problem. We recognize at least five major components, to be more or less thoroughly investigated in SYMBIOSE according to mainly the objectives and the level of complexity of the assessment that is performed. Namely: (1) pollutant sources dynamics, (2) biosphere system dynamics, (3) impact and risk endpoints, (4) sources of uncertainty, and (5) responses. They are now discussed, with emphasis on valuable modeling techniques that might be implemented in the SYMBIOSE framework.

### 1.1. Pollutant sources

The first component of a chemical risk assessment framework is the pollutant sources that exert a pressure on the environment, i.e. an emission of chemicals that are potentially a risk for biological populations (humans and biota). These sources may be natural or anthropogenic (e.g. chemical industries, waste disposals, nuclear power plants), in a normal or accidental mode, point-scale or diffuse in space. Modeling the chemical sources dynamics aims at assessing the physico-chemical forms which are released, quantifying the magnitude, kinetics and space distribution (for a diffuse contamination) of the source releases, and evaluating their probability density functions for a range of scenarios. We won't further discuss modeling approaches dedicated to the evaluation of sources dynamics; this is another subject.

PROBABILISTIC CHEMICAL RISK ASSESSMENT				RISK MANAGEMENT
<b>1</b> <b>CHEMICAL SOURCES</b> Anthropogenic/natural chronic & acute sources of pollutants (e.g. NPP, waste disposal)	<b>1,2</b> Pollutant emissions			
	<b>2</b> <b>BIOSPHERE SYSTEM DYNAMICS</b> Pollutant dynamics Ecological dynamics Fluid & dispersed matter dynamics	<b>2,3</b> Exposure inducing an individual-level impact on biotic components Extrapolating organism-level to higher-level endpoints		
	<b>3,2</b> Impact on biota may cause changes in the state (e.g. through food-web perturba.)	<b>3</b> <b>ENDPOINTS ( Impact &amp; Risk )</b> Human health Ecosystems health, biodiversity, etc. Economic Financial		<b>3,5</b> Risk-based decision making
<b>4,1</b> Uncertainties on sources dynamics	<b>4,2</b> Uncertainties in the structure & functioning of the biosphere Uncertainties in initial & boundary conditions	<b>4,3</b> Uncertainties in exposure-response relationships Uncertainties in toxicity-extrapolation methods	<b>4</b> <b>SOURCES of UNCERTAINTY</b> Inherent spatial & temporal variability Scientific ignorance (e.g. parameter & model errors)	
<b>5,1</b> Reducing emissions	<b>5,2</b> Cleaning contaminated lands & reinforcing protection of biota Remediation actions may affect ecological resources (e.g. habitat loss)		<b>5,4</b> Implementing actions can induce hardly predictable changes in anthropogenic behaviour	<b>5</b> <b>RESPONSES</b> Appropriate actions to optimize the quality of the environment (cost- benefice analysis)

FIG. 1. A high-level view of an environmental chemical risk assessment and management problem, clarifying major components of the system (diagonal boxes) and interactions among them (off-diagonal boxes).

## 1.2. Biosphere system dynamics

Biosphere can be assumed to consist of a set of natural, semi-natural or artificial interacting sub-systems (e.g. atmosphere, aquatic and terrestrial semi-natural ecosystems, sewer or irrigation networks, groundwater, urban systems), the biotic/abiotic components\* of which are evolving within a given space and time domain. Modeling the state and dynamics of this system aims mainly at assessing potential exposure of spatially-distributed biological populations to the chemicals, and the characteristic transfer functions of the multiple potential pathways from sources to critical receptors. Such an exposure assessment is typically addressed through the development and use of chemicals transport and fate, ecological and fluid dynamics models (cf. Figure 2), each of which requires some relevant initial and boundary conditions to be implemented with.

\* *Components* refer here to real-world entities of the biosphere that potentially contain pollutants, without any consideration to their geographical extent. There is sometimes a confusion with the *compartment* concept, although compartments are truly-speaking spatial entities of the domain in which a homogeneity and instantaneous mixing assumption is made. The component concept is a much more general concept. See paragraph 3 for further discussion.

DETERMINISTIC EXPOSURE ASSESSMENT			DETERMINISTIC IMPACT & RISK ASSESSMENT	
<b>2.1 CHEMICAL FATE &amp; TRANSPORT MODELING</b> Mass balance equations approach			<b>2.1,3.1</b> Exposure inducing an individual-level impact on biotic components	
<b>2.2,2.1</b> Supporting the modeling of pollutant dynamics for biotic components Selecting keystone biotic components	<b>2.2 ECOLOGICAL DYNAMICS MODELING</b> (Meta)population Food-web Ecosystem Landscape			<b>2.2,3.2</b> Assessing impact at higher levels <i>via</i> an explicit resolution of ecological interactions
<b>2.3,2.1</b> Supporting the modeling of pollutant dynamics for fluid and dispersed matter components		<b>2.3 FLUIDS &amp; SUSPENDED MATTER DYNAMICS MODELING</b> Hydrodynamics Watershed/urban hydrology & erosion Meteorology		
	<b>3.1,2.2</b> Disturbing biotic component dynamics		<b>3.1 ORGANISM-LEVEL ENDPOINTS</b> Dose Mortality, fecundity, growth, morbidity, physiolo. responses	<b>3.1,3.2</b> Inter-endpoint & inter- species toxicity- extrapolation methods (e.g. safety factors, species-sensitivity distributions)
				<b>3.2 HIGHER-LEVEL ENDPOINTS</b> Population endpoints Biodiversity Trophic structure Spatial structure

FIG. 2. A view of a deterministic chemical exposure and impact assessment problem, clarifying important sub-components of the system (diagonal boxes) and interactions among them (off-diagonal boxes).

### 1.2.1. Pollutant dynamics

A major aspect of the biosphere dynamics component is the study of pollutant dynamics, once released into the biosphere system, which aims at predicting the fate and transport of chemicals in a multi-media environment. This is an inevitable step, where models are really valuable tools, especially when combined with a treatment of *in-situ* monitoring data. Many different modeling approaches can be adopted, depending on the level of detail required. They range from empirical point-scale equilibrium models, sometimes suitable for a rough and conservative analysis, to semi-mechanistic, dynamic and spatially-distributed models (e.g. spatial biogeochemical models), dedicated to a more realistic analysis and a more thorough understanding of the system. Even more realistic predictions require in some cases the coupling with nutrient dynamics models, as long as nutrients “compete with or drive” pollutants in some biogeochemical processes (e.g. Cs/K and Sr/Ca competition for soil-to-plant transfer, [11, 12]). A good compromise can usually be built upon the resolution of mass balance equations, in which time and space evolution of pollutant stocks in the biosphere components are mathematically related to the various physical, chemical and biological fluxes participating to either pollutant transformation or pollutant exchange among components [13, 14].

As one of the major objectives of numerical ecology is to address the quantification of mass (e.g. nutrients, carbon and toxicants), energy and information flows in most environments (e.g. food-web, ecosystem and landscape models), ecosystem and landscape modeling approaches can play a major role in helping define hypotheses about the structure and functioning of the biosphere system, in terms of chemical exposure pathways (interaction 2.2,2.1 in Figure 2), and nutrients pathways when required. This is particularly important, as environmental risk assessments are in essence multi-faceted problems.



### 1.2.2. Ecological dynamics

Population dynamics modeling provides valuable methods to predict the spatial and temporal behavior of biological populations (e.g. plant population density, biomass, growth rate, migratory behavior of mobile animal populations). This is more or less explicitly required for fate and transport calculations, as it may significantly influence the pollutant mass transfer between biota and the contaminated biotope (e.g. wet and dry deposition fluxes of contaminated aerosols onto a plant cover strongly depends upon plant biomass or leaf area index). Many different approaches have been developed in quantitative ecology, some of them being potentially useful in a chemical risk assessment context, such as population, meta-population, food-web, ecosystem and spatially-structured landscape modeling approaches (see [9, 15, 16] for a review). Mobile populations, such as individual mammals or birds, move in the landscape while retaining their identities as unique individuals. In modeling endangered species, for example, it is important to keep track of individuals over space and time, interactions among individuals and interactions of individuals with the landscape. This is accomplished by agent-based models (see [17] for a review). But they are usually too complex and time-consuming for them to be used as practical assessment tools. Scalar-abundance population models that represent a single-species population abundance with a scalar dimension without consideration of age or stage structure, or life-history population models that allow exploration of vital rates as a function of stage or age (e.g. matrix models, [18]) can provide sufficient details, especially when performing screening assessments in data- or knowledge-poor situations. Multiple-species predator-prey and food-web modeling may provide information about the keystone species in a community. This kind of ecological selection criteria is useful when selecting representative receptors and relevant endpoints.

### 1.2.3. Fluid and suspended solid matter dynamics

Physical modeling of fluid dynamics and *in-situ* suspended matter dynamics (e.g. aerosols, hydrosols, phyto-plankton, colloids) in the various geophysical media encountered in the biosphere (e.g. atmospheric boundary layer, surface water systems, vadose zone) play also an important role, as these mobile components stand as potential vectors for contaminant migration in space. In an environmental assessment context, physical modules typically aim at evaluating turbulent advection and dispersion properties of fluid media. Quite realistic and prognostic approaches can be adopted by solving the fully coupled mass (here, mass of fluid and suspended matter), momentum and energy balance equations (e.g. Navier-Stokes equations or approximations). Nevertheless, simplified approaches such as those that combine real on-time data acquisition and mass balance principle (usually under stationary and/or homogeneous assumptions) are more tractable and appropriate for screening-level analysis (diagnostic approaches).

## 1.3. Impact and risk assessment

Many risk assessments done in support of environmental regulatory programs rely on simplistic conservative approaches, where comparison is made of some exposure estimate (e.g. Predicted-Environmental-Concentration) for each chemical of interest with a corresponding toxicity threshold for individual-organism endpoints such as mortality, fecundity, or growth (e.g. No/Lowest-Observed-Effect-Level). As the complete dose-response curve for the chemical, the species or the endpoint of interest at the desired level of organization is usually unknown, arbitrary uncertainty factors or other simple toxicity-extrapolation methods (3.1,3.2 interaction in Figure 2) are often applied to translate available laboratory test results onto individual organisms into the endpoint of interest (e.g. from acute to chronic exposures, from one species to others, extrapolation to population and communities). Based on conservative assumptions about exposure and toxicity, deterministic hazard quotients (e.g.  $\text{Max(PEC)/Min(NOEL or LOEL)}$ ) cannot be used to reliably rank chemicals, identify priority receptors, or identify problem areas because the degree of conservatism is inconsistent among the elements ranked. Such worst-case analysis using individual-level endpoints prove to be effective and practical tools for screening out chemicals, receptors or site areas that are clearly not a problem, however (when hazard quotients are considerably less than 1). For all other cases, application of population and higher-level models would be warranted [4, 9, 19], as these may provide a valuable method to predict the responses of population and communities to perturbations in individual biotic components, with higher-level endpoints such as population abundance, biomass, population growth rate or productivity, population stage or age structure, trophic structure and species richness (3.1,2.2 and 2.2,3.2 interactions in Figure 2).

#### 1.4. Uncertainty calculations

Uncertainty plays a major role in risk assessment. Risk is the probability that a specified harmful effect will occur, or, in the case of a graded effect, the relationship between the magnitude of the effect and its probability of occurrence. The importance of uncertainty in risk assessment derives from its central role in risk-based decision-making; the decision maker needs to understand the uncertainties associated with the scientific information on which the responses will be based. Uncertainties have three basic sources: (1) the inherent spatial and temporal variability, or randomness, of the system at levels of interest in risk assessment (stochasticity), (2) imperfect or incomplete knowledge of some aspects of the system that is potentially knowable (ignorance, that is parameter and model errors), (3) human error including incorrect measurements, misidentifications, data recording or entry errors and computational errors (errors) [4]. The diversity and complexity of biosphere systems ensures that the ignorance uncertainty component in environmental risk assessments will be very high. Model errors include inappropriate selection or aggregation of variables, incorrect functional forms and incorrect initial or boundary conditions.

Much of the technical literature on uncertainty pertains to identifying, quantifying and computing uncertainty propagation to the final endpoints, with uncertainty increasing with the hierarchical level of an assessment endpoint. Stochasticity and parameter-induced errors are often quantified in relatively straightforward ways using well-known Monte-Carlo techniques because of their generality and ease of application. Fuzzy set theory is a serious and powerful candidate when knowledge about parameter estimates and distribution is poor, or computing time is a limiting factor [20]. However, the major source of uncertainty can be model error. A possible approach to deal with involves the use and comparison of alternative models, this implementation process being greatly facilitated when working with a flexible and integrated modeling platform (see paragraph 3).

#### 1.5. Responses

This component, which is the key step in risk management process, aims at determining appropriate responses, in order to direct the final impact or risk in the desired direction (a reduction in environmental harm to a given level). These actions can influence the source dynamics or the state of the biosphere, thus introducing a new feedback in the ECRA system. Examples of responses might be: (1) requiring industry to reduce pollution emissions, (2) implementing programs to clean contaminated land or reinforcing protection of humans and biota (e.g. countermeasures). As the impact can only be changed indirectly at the end of the cycle of interactions among components 1 to 4, a decision-maker must therefore use a model to predict what effect these actions will have on the impact. Once communicated to the public, responses might induce indirect effects such as profound changes in human behavior (e.g. panic behavior, migration outside the contaminated region, changes in dietary habits). Such hardly predictable events may add sources of uncertainties in the system (5,4 interaction).

## 2. THE NEED FOR INTEGRATED AND FLEXIBLE MODELING PLATFORMS

As demonstrated above, human health and ecological risk assessments require the integration of a wide range of environmental data and modeling approaches related to the fate, transport and effects of chemicals. Researchers recognize the need for *integrated* and comprehensive approaches to modeling that can access many of the risk assessment components together [5, 21–24]. Such integrated approaches may provide rapid, scientifically based evaluation of potential risks and of remedial alternatives. It is a major challenge to build a simulation system that can successfully capture the chemical, ecological and physical aspects of the biosphere dynamics.

The level of detail required for a given risk assessment depends on environmental management objectives, the complexity of the site or the behavior of the chemical contaminants, and the difficulty of adequately describing exposure, toxicity and other properties of the chemicals (ignorance). It is recognized that a human health or ecological risk assessment must be conducted following a tiered, or graded, approach [2–4, 6, 7, 9]. An initial screening-level risk assessment is first conducted that uses available data, conservative assumptions about exposure and toxicity, simplified approaches for fluid and suspended solid matter dynamics, and quantitative uncertainty analysis. This can provide valuable information. From there, chemicals, habitats and species of potential concern are identified and

decisions are made about additional data and knowledge gathering. In the next tiers, more realistic spatio-temporal modeling approaches are used to better describe the chemical fate, transport and effects, and the role of physical and ecological modules is likely to become more important here. Therefore, it has also been recognized that there was a need for *flexible* and *user-friendly* tools that could adapt to this expanding and shifting assessment context, and offer a cost-effective way to support alternative environmental simulations. This is another modeling and computational challenge.

### 3. THE SYMBIOSE MODELING APPROACH

Examples of model applications to assess ecological risks of toxic chemicals are common in the literature. However, little guidance exists on the step-by-step process of developing integrated spatially-distributed models in a risk assessment context, though the use of a rigorous methodology reinforces transparency, coherence and defensibility of the model which is built up. Methodologies recommended for ecological risk assessments are basically the same as those practiced for human health risk assessments (see [8] for example). They all involve at least three keystone developments: *conceptual*, *mathematical* and *spatial modeling*, which are in practice carried out iteratively. We propose here to briefly examine the conceptual, mathematical and spatial approach that will be implemented in SYMBIOSE for describing the biosphere dynamics, and its interactions with the source and impact assessment components. This approach is currently being applied for investigating the fate and transport of radiocaesium, radiostrontium and technetium in agricultural and forest soil-plant systems, at increasing levels of detail [25]. The way environmental and demographical stochasticity can be implemented in the modeling approach will not be analyzed in this paper.

#### 3.1. Conceptual modeling

The conceptual modeling step aims first at analyzing the pollutant dynamics in the biosphere system in terms of mass-containing material entities (*material components*) and potential flows of contaminants among them (*mass transfer processes*). At this stage of the development, no spatial consideration is brought up, just as if all real-world entities were located at the same geographical place and could exchange pollutant mass with each other. A *material component* represents any collection of abiotic/biotic individuals (or both) that share, according to expert judgment, some important physical, chemical or biological characteristics that govern pollutant dynamics. Here are some examples of components: water as a (continuous) medium made of infinitesimal fluid parcels, a (discrete) collection of sub-micrometer inorganic aerosols, a plant/animal population or community, a life-stage or age-stage cohort, a given type of human food-stuff. In some way, a component is a representative individual with some statistics accounting for variability among individuals. A *mass transfer process* represents any combination of physical, chemical and biological mechanisms prone to drive: (1) in-space migration due to the movement of mobile material components (e.g. advection/dispersion of pollutants in fluids or attached to suspended matters, migratory behavior of wild animals, food-stuffs circulation), (2) mass transfer between material components (e.g. wind erosion/deposition of contaminated airborne materials, soil-to-plant transfer, food-web processes) or (3) physicochemical transformation of the pollutant in a component (e.g. chemical speciation in aqueous media, radioactive filiations). In addition to the mass transfer description, conceptual models must also help to identify some extrinsic factors or events that may influence the pollutant dynamics in the system, through their influence upon material components or processes (e.g. nuclear accident, countermeasures, forest fires, extreme weather events, water temperature or pH). Therefore, the conceptual scheme must include additional key features of the system (referred here as *information-containing components*) and their influence upon the pollutant behavior (referred here as *information flows*).

The above-mentioned concepts are also valuable concepts to model ecological and fluid dynamics. They can be, and have been, applied in exactly the same way to analyze interactions governing the behavior of ecological and fluid components (e.g. plant growth, predator-prey or competition relationships, food-web processes, fluid and suspended matter movements).

Environmental systems are considered among the most complex because they are characterized by a large number of diverse components, (sometimes nonlinear) interactions, scale multiplicity, and spatial heterogeneity. Hierarchy theory, as well as empirical evidence, suggests that complexity often takes the form of modularity in structure and functionality (see [26] for further discussion). Therefore, a

hierarchical modeling approach is strongly recommended for analyzing and understanding such complex biosphere systems. In SYMBIOSE, a biosphere system will be, as far as possible, modeled as a nest of increasingly detailed components, processes and interactions, ranging from ecosystem levels to organism, or even organ levels (e.g. physiologically-based pharmaco-kinetics approaches). This kind of hierarchical approach that provides an increasingly complex description of the system contributes to the flexibility required for conducting any risk assessment. An interesting way to develop, represent and communicate a conceptual model (to other assessors, risk managers and stakeholders) is using the interaction matrix formalism, whereby we built up as “Russian dolls” nested matrices, from the very top level (see Figure 1) to much more detailed levels.

### 3.2. Mathematical modeling

This modeling step aims at quantifying the behavior of the various components introduced above, by providing mathematical parameterizations for processes and interactions, and specifying initial and boundary conditions when required. These developments must be carried out for pollutant, ecological and fluid dynamics. A quite difficult task is to ensure the coherence between the levels of complexity in the conceptual and mathematical descriptions. It is useless to develop a fine conceptual approach, when ignorance of mathematical relationships or site-specific data values prevails. However, we believe that building the mathematical model upon approximations to the dynamic mass, energy and momentum budget equations, offers a quite general and defensible approach. Moreover, the ability to swap one parameterization for another in these equations gives some flexibility, and reinforces the ability to cover a wide range of environmental risk assessment contexts.

#### 3.2.1. Pollutant dynamics

At any time  $t$  and location  $\mathbf{x}$  in the space domain, let us introduce for any *material component*  $i$  its concentration in chemical  $M$ , that is  $[M]_i(\mathbf{x},t)$  [mole· $U_i^{-1}$ ], where  $U_i$  refers to the unit which best characterizes this component (e.g. surface, volume, fresh or dry (bio)mass). The “degree of presence” of component  $i$  at given  $\mathbf{x}$ ,  $t$  can be quantified by a characteristic density  $\chi_i(\mathbf{x},t)$  [ $U_i \cdot U_x^{-1}$ ]. This state variable expresses the number of units of  $i$  per unit of space, typically length, surface or volume respectively for 1D, 2D or 3D spatial problem. Examples of densities are: air density, moisture content for soil solution, water level in a 1D river network, leaf area index or surface biomass for plants. Therefore,  $[M]_i \chi_i(\mathbf{x},t)$  [mole· $U_x^{-1}$ ] is the total stock of  $M$  in  $i$ . Pollutant dynamics is governed by the following equations (in mole· $U_x^{-1} \cdot s^{-1}$ ) written for all material components  $i$  and chemicals  $M$  introduced into the conceptual model:

$$\frac{\partial}{\partial t} \chi_i [M]_i = \nabla \cdot \mathbf{FM}_i + SM_i + RM_i - \sum_{j \neq i} \left( TM_{i,j} - TM_{j,i} \right) \quad (1)$$

Where the right-hand side terms represent the various contributions of the *mass transfer processes* to the time evolution of stocks.  $\nabla \cdot \mathbf{FM}_i$  is the divergence of density migration fluxes of  $M$ .  $SM_i$  is the chemical source term<sup>\*\*</sup>.  $RM_i$  represents all physico-chemical transformations affecting  $M$  in  $i$ , and  $TM_{i,j}$  (resp.  $TM_{j,i}$ ) express mass transfers of  $M$  from  $i$  to  $j$  (resp.  $j$  to  $i$ ).

Usual parameterizations for advection/dispersion fluxes in fluid components (e.g. in atmosphere, surface water systems, ground waters, sewer networks) are as follows:

$$\mathbf{FM}_i = -\chi_i \mathbf{u}_i [M]_i + \chi_i D_i \nabla [M]_i \quad (2a)$$

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<sup>\*\*</sup> This source term is induced by *internal* sources located within the time and space domain under study. *External* sources, i.e. past emissions or outside located sources, exert a pressure which can be mathematically treated either as initial or boundary conditions to these equations.

Where  $\mathbf{u}_i$  [ $\text{m}\cdot\text{s}^{-1}$ ] is the mean velocity vector of  $i$  and  $D_i$  [ $\text{m}^2\cdot\text{s}^{-1}$ ] is the dispersion coefficient (resp. a tensor of dispersion coefficients) of  $M$  in fluid  $i$  for an isotropic (resp. anisotropic) medium. Parameterizations are very similar for abiotic or biological transported matters in fluids (e.g. aerosols, hydrosols, zoo and phytoplankton). For discrete mobile components, like animal populations or foodstuffs, we can assume:

$$\mathbf{FM}_i = \mathbf{F}_i [M]_i \quad (2b)$$

Where  $\mathbf{F}_i$  stands for the density migration flux of component  $i$  (see paragraph 3.2.2). In a screening assessment context, transfer processes are usually parameterized assuming first order kinetics, or even “equilibrium” through the use of concentration ratios. *Information flows* as defined previously are accounted for through some functional dependence upon extrinsic factor or event characteristics.

To be completely closed, the system of equations (Equation 1) further requires: (i) estimating parameter values, (ii) giving initial conditions for concentrations, (iii) defining boundary conditions for mobile components, and (iv) estimating density functions and migratory quantities (e.g. velocities/dispersion coefficients for fluids, fluxes  $\mathbf{F}_i$  for others). Characteristic density functions, and migratory behaviors, are basically assessed by ecological and fluid dynamics studies: namely, interactions 2.2,2.1 and 2.3,2.1 in Figure 2.

### 3.2.2. Ecological and fluid dynamics

The spatio-temporal dynamics of any *material component* density  $\chi_i(\mathbf{x},t)$  obeys the general mass balance equation (in  $\text{U}_i\cdot\text{U}_x^{-1}\cdot\text{s}^{-1}$ ):

$$\frac{\partial}{\partial t} \chi_i = \nabla \cdot \mathbf{F}_i + S_i + \left[ \frac{\partial \chi_i}{\partial t} \right]^{(*)} + \left[ \frac{\partial \chi_i}{\partial t} \right]^{(**)} \quad (3)$$

Where  $\nabla \cdot \mathbf{F}_i$  is the divergence of density migration fluxes of  $i$  (e.g. infiltration rate affecting soil water content in a soil layer, suspended matter discharge in a river system affecting suspended loads, migration of wild animals affecting local densities).  $S_i$  is a potential source term of  $i$  (e.g. aerosols injected into the atmosphere through a chimney).  $[\ ]^{(*)}$  term stands for the local appearance or disappearance of individuals in a population apart from migration (e.g. precipitation/evapotranspiration rates, deposition/erosion rates of suspended matter, fecundity/mortality rates of wild animal populations, sowing/harvesting of agricultural plants).  $[\ ]^{(**)}$  term is essentially the average biological growth/senescence rate of individual characteristics (e.g. growth of individual plant or animal weight, size, etc.).

For fluids, we usually write  $\mathbf{F}_i = \chi_i \mathbf{u}_i$ , and for suspended matters,  $\mathbf{F}_i = \chi_i \mathbf{u}_i + D_i \nabla \chi_i$ . In general,  $\mathbf{u}_i$  and  $D_i$  will be estimated in SYMBIOSE after some treatments of: (i) *in-situ* data or (ii) results issuing from external fluid dynamics dedicated codes. For other mobile components like animal populations, no generic formulations exist: migratory behavior may be assessed using simple time budgets or more elaborated migratory rules (e.g. agent-based approaches). For single species populations,  $[\ ]^{(*)}$  and/or  $[\ ]^{(**)}$  terms can be parameterized through the introduction of a more or less complex density-dependent growth rate  $r(\chi_i)$  [ $\text{s}^{-1}$ ] (e.g. scalar abundance models and matrix models). When multiple species are strongly interacting in the system, such interactions as feeding, predator-prey or other competition relationships are accounted for through the introduction into the mathematical parameterizations of some functional dependence upon other  $\chi_j$  component densities (e.g. food-chain and food-web models).

Once parameterizations are developed for the right-hand side terms of Equation 3, this set of deterministic differential equations still requires: (i) estimation of associated parameters, (ii) initial conditions for densities and (iii) boundary conditions for mobile components.

### 3.2.3. Impact assessment

Ecotoxicological approaches typically aim at quantifying the relationship between fecundity/mortality rates, or other representative individual-level endpoints, and the chemical concentration. Introducing such mathematical or statistical relationships in Equation 3 for the sensitive biotic components enables

us to theoretically compute the overall deterministic impact assessment at population or community levels by solving the coupled systems Equation 1 and Equation 3. Actually, a hierarchical description of the biosphere through nested components results in nested sub-systems of equations, which characteristic time and space scales can be very different. This requires numerical splitting techniques for getting sufficient accuracy and efficiency. Typically, in data and/or knowledge poor situations where Equation 3 for ecological dynamics can be very uncertain, extrapolation to higher levels of biological organizations should be by-passed through the use of safety factors (see paragraph 1.3).

### 3.3. Spatio-temporal modeling

Spatially explicit (geographic) approaches, which account for small-to-large scale spatial heterogeneity in nature, are especially important if one's modeling goals include developing relatively realistic description of the biosphere and predictions of the impacts, at least because the distribution of pollutants at a contaminated site is heterogeneous. In aquatic systems, such spatially-distributed models are most relevant for large-scale systems (e.g. lakes, wetlands and estuaries), where spatial homogeneity cannot be reasonably assumed. In terrestrial ecosystems, substantial heterogeneity tends also to occur at much smaller scales, especially when the landscape has been fragmented by human activities.

An approach to spatial landscape modeling which has proven successful [24, 26–29] will be implemented in SYMBIOSE. The spatial domain is broken down into a set of sub-regions (i.e. spatially referenced sub-areas), corresponding each to a typical component of the system: a typical ecosystem, medium, habitat or any other grouping of sub-components (e.g. a vineyard landscape including dispersed villages). Thus, each sub-region can be associated with the conceptual mathematical module dedicated to the locally present component. A sub-region is then discretized into cells (meshes): volumes, surfaces, segments, surfaces or points, depending on the spatial dimensionality of the mathematical module (e.g. 3D rectangular grid, 2D Triangular Irregular Network, 1D graph and network, unlinked cell sets). Cells are then linked by fluxes of mass (pollutants, nutrients, mobile components) and information, plus energy and momentum when required. Typically two cells are linked if they share a boundary, but more general linkages are possible (e.g. an irrigation process transferring mass from a river point cell to some distant agricultural plot cell). The use of GIS maps, remote sensing data, and methods for extrapolating or aggregating data from one scale of observation to other scales, greatly support the development of such spatial explicit approaches.

From there, two basic assumptions can be made for each of the media of interest. First, a *compartmental assumption* can be made whereby we hypothesize that concentrations, densities, and other properties of components (and associated processes) are homogeneous within a cell; mass, energy and information being instantaneously mixed. This is particularly appropriate for screening assessment analysis. For media dominated by fluids, such as atmosphere and surface water systems, predictions are usually more realistic and accurate when medium is treated locally as a *continuum* in which the above-mentioned quantities vary continuously in space. This is particularly recommended when advection/dispersion processes dominate dynamics.

The coupling of the multiple scales (and assumptions) requires methods for extrapolating or aggregating data. The implementation of the concept of space in SYMBIOSE must be general enough to allow resolution of a wide range of specific space-time representations, while the details of linking and transferring data between them, should be invisible to the modelers. Multiple spatial resolution can be processed by stacking grids representing the same spatial sub-region at different resolutions. The platform should maintain a catalog of generic grids, which could be customized (configured) with GIS maps, and generic methods to map data and/or results between geographical grids.

## 4. AN OBJECT-ORIENTED SOFTWARE ARCHITECTURE

A suitable approach to support integrated graded assessments is the use of an object-oriented integration framework [24, 27, 30–32]. An object-oriented approach to the biosphere modeling and simulation typically relies on the creation of *objects* that represent real-world entities (mass- and information-containing components).

In contrast to procedural programming techniques where code and data are kept separate, an object-oriented approach employs objects that receive and send messages, contain specific attributes that describe the object intrinsic state (e.g. pollutant concentration, characteristic density function) and contain programming code describing “*what kind of*” behaviors this object may be concerned with (e.g. chemical, ecological and physical interactions introduced in the conceptual model). In most object-oriented approaches, objects also implement programming code describing “*how*” to calculate these interactions in space and time (spatio-temporal mathematical model). An alternative approach is to split the “*what*” from the “*how*”, whereby the “*how*” is addressed by separate *models* or *applications* that explicitly compute one or several interactions associated with this object (e.g. a *plant* object whose characteristic attribute leaf *area index* ( $x,t$ ) is updated by a separate *plant growth* module). By adopting such an *object-model* interaction approach (see [30, 31] for further discussion), time and effort required to develop alternative simulations by swapping one model for another or adding new models to an existing simulation are substantially reduced, while the structure of the biosphere and the type of interactions remain unchanged. It remains that, if the new model requires additional parameters or generates output that differ from the original model, the entity objects need to be edited to add new attributes or modify existing attributes to accommodate those changes.

In complex simulations, *external legacy models* or *applications* should participate in a simulation through a “wrapping” procedure [27, 30–32]. An important feature is that the “wrapped” models and applications run in their native languages, and do not require translation to the common standard system language (C++ for SYMBIOSE). We typically plan to connect external ecological or fluid dynamics models for specifically addressing Eq. 3, while internal models would focus on pollutant dynamics given by Eq. 1. Such a technique also enables us to feed in a particular attribute or behavior of an object through real time monitoring data or GIS applications. This ability to link external dedicated models gives the platform the ability to scale very well to increasingly complex problems as it is recommended in ECRA.

Once objects are developed, they may be stored into a library of re-usable objects and placed into a hierarchy in which one kind of object can inherit the behavior of another (e.g. a “deer population” object which inherits attributes and behavior from a “mobile wild animal population” object). A programmer can simply create a new object that inherits many of its features from existing objects (parent objects). Each time a new application will be developed with SYMBIOSE, existing objects will become more mature and new objects will be added to the library, giving rise to a wide variety of natural and artificial elements of the biosphere available for future applications.

This SYMBIOSE infrastructure should make it feasible to build, manipulate and simulate complex biosphere dynamics in which multiple objects interact with internal/external models or applications, through multiple mass- and information-flow processes.

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# Defining the spatial area for assessing doses to non-human biota

R.C. Morris

TREC, Inc., Rigby, ID, United States of America

**Abstract.** In order to apply any biota dose assessment methodology, assessors need a way to decide the spatial scale over which it should be applied. Any useful scaling procedure should take into account the level of biological organization at which impacts are to be assessed (usually the population). Both grain and extent of the selected scale should be considered, and the procedure must achieve an optimum balance between cost of sampling or analysis, and assuring protection of biota. That is, the procedure must define the smallest possible total area to be evaluated, divided into the maximum sized assessment units compatible with assuring protection of populations. Ecological processes occur at multiple scales which vary widely between contaminated sites. Therefore, the procedure must also allow for flexibility to apply site specific knowledge. One such procedure was developed by the U.S. Department of Energy's Biota Dose Assessment Committee to support our proposed dose assessment methodology. The procedure includes the following steps: 1) determine whether this procedure is necessary by using screening models, 2) determine and map the boundaries of areas contaminated with the same radionuclides in similar quantities, 3) determine and map the boundaries of discrete habitat types, and 4) overlay the maps and identify the intersections. Each area of discrete habitat that lies within a discrete contaminated area can be appropriately defined as an assessment unit. Our approach satisfies all the requirements for a useful scaling procedure by using site knowledge of, and professional judgement about, environmental contamination and ecological habitats to develop maps of the largest patch sizes over which biota dose assessment parameters can be averaged.

In any assessment of radiation dose to biota, it is necessary to average over some assessment area. One cannot generally sample every square millimeter of a site or every organism that lives there. In virtually every case, one must collect representative samples and argue that they represent all the soil, or all the organisms, in some defined spatial area. Usually, one collects a number of samples and assumes their average, their maximum, their upper 95% confidence limit, or some other metric, represents the entire area over which they were collected. Deciding on that area is not necessarily straightforward, but there are some general guiding principles.

My objectives in this paper are to discuss some of the general principles one must think about to establish an assessment area, and to describe the procedure the U.S. Department of Energy's Biota Dose Assessment Committee recommends to define an assessment area for the methodology we developed.

## 1. GENERAL PRINCIPLES GOVERNING SELECTION OF SPATIAL AREA

The most basic information to keep in mind when selecting an assessment area is the lowest level of biological organization at which impacts are to be assessed. If you choose to assess dose to an individual, while the size of your assessment area will vary depending on the size of the organism, it will almost certainly be different than the size of the area you would choose for a population of individuals. Some of the choice here is out of our hands. There is a general societal understanding that the lowest level of organization in which we're interested, is the population. This is reflected in most of our environmental laws. For example, we routinely authorize the legal killing of individuals, through licensed hunting, with the partial justification that populations are not harmed and may even be benefited. The single exception of which I'm aware is the body of laws protecting Threatened or Endangered species, such as the U.S. Endangered Species Act. These laws force us to be concerned about individuals because, almost by definition, every individual of an endangered species is important to the survival of the species.

Society's opinion on an appropriate level of biological organization at which to assess environmental impacts may be changing. In areas surrounding Yellowstone National Park, in recent years, it has not been uncommon for people to throw themselves in front of bison hunter's rifles in order to prevent their being killed. Dangerous conflicts have occurred between anglers and animal rights activists who are concerned, primarily, about the suffering of individual fish. If our society continues to move in the direction of valuing individual non-humans more than populations on non-humans, we may be required to reevaluate the level of biological organization at which to assess impacts.

Given a decision that plant and animal populations are the biological level of interest, one next must consider the *susceptibility* of the population to the contaminant [2]. The organisms of interest must come into contact with the contamination in such a way that they are susceptible to harm. For large facilities, such as the U.S. Department of Energy's Idaho National Engineering and Environmental Laboratory (INEEL), the entire 2470 km<sup>2</sup> reservation would be too large an assessment area. Most of the biota on the reservation would not ever come into contact with the contamination, and organisms which don't come into contact with contaminants, don't receive dose. The inclusion of measurements from non-contaminated areas in the calculation of mean concentrations would result in low doses not representative of the actual impacts to the affected biota.

If there is contamination and harm to a population, it is important to consider the *ecological relevance* of the harm [2]. The organisms of interest must be harmed in such a way that there is a measurable ecological impact. At the opposite end of the scale from the entire reservation, the individual operable unit, waste trench, or contamination source would, in most cases, be too small to be ecologically meaningful. For example, although the small mammals living in a 100 m<sup>2</sup> waste trench may be greatly affected by trench contaminants, their loss will likely have little impact on the population of small mammals in the region, or on broader scale ecosystem function. Using a realistic average density of 0.02 small mammals per square meter on the INEEL, that 100 m<sup>2</sup> trench would support two small mammals. The INEEL as a whole supports over 46 million small mammals, half of which are females, that give birth, conservatively, to 12 pups each year. If populations remain stable from year to year, which they do within broad variability, these births must be matched by deaths equaling 276,610,730 per year. Given this many deaths from "natural causes," the two killed in the waste trench are lost in the noise, and their loss would have little ecological relevance.

In general, any scale selection procedure must achieve an optimum balance between cost of sampling or analysis, and assuring protection of biota. That is, the procedure must define the smallest possible total area to be evaluated, divided into the maximum sized assessment units compatible with assuring protection of populations. Ecological processes occur at multiple scales which vary widely between contaminated sites. Therefore, the procedure must also allow for flexibility to apply site specific knowledge. Nevertheless, there are some general recommendations which can be made.

## 2. THE U.S. DEPARTMENT OF ENERGY PROCEDURE

The U.S. Department of Energy (DOE), through its Biota Dose Assessment Committee (BDAC) has published a technical standard for assessing doses to non-human biota [1]. This standard includes a recommended procedure for selecting the spatial area over which the methodology should be applied. That procedure is described here.

The DOE's Graded Approach was developed primarily for application at DOE National Laboratories and other DOE facilities which are generally intensively monitored and well studied. Thus, one of the primary assumptions of the approach is that both substrate contamination and biota population distributions are adequately determined. The procedure for determining an adequate assessment area depends heavily on this assumption.

An appropriately scaled area of application is defined by the intersection of areas contaminated with the same radionuclides in similar quantities (contaminated areas) and areas of similar ecological characteristics (habitat types). Determining this intersection is most easily done using area maps, and Geographic Information Systems (GIS) will prove an invaluable tool. The following steps can be applied:

### 2.1. Determine whether this analysis is necessary

One of the first phases in the Graded Approach is the "General Screening" phase. In this phase, users apply the default screening models with the input contaminant concentration set at the highest concentration found in your area of interest (e.g., the entire reservation). Because the default screening models are necessarily conservative (i.e., calculate the lowest credible substrate contaminant concentration guideline), this sets up a worst case scenario: the highest measured concentration compared to the lowest calculated concentration guideline. Under these conditions, if the screen is passed, there is no need to proceed any further.

TABLE 1. RADIONUCLIDE CONCENTRATIONS (kBq·m<sup>-2</sup>) IN HYPOTHETICAL CONTAMINATED AREAS SHOWING DIFFERENCES IN RADIONUCLIDES AND/OR CONCENTRATIONS WHICH MIGHT LEAD TO DISTINGUISHING THESE AS DIFFERENT CONTAMINATED AREAS FOR THE PURPOSE OF DETERMINING AN ASSESSMENT AREA

Radionuclide	Contaminated Area 1	Contaminated Area 2	Contaminated Area 3
<sup>137</sup> Cs	5.5	55	55
<sup>90</sup> Sr	4.0	40	40
<sup>60</sup> Co	6.0	60	–
<sup>241</sup> Am	–	–	20
<sup>238</sup> Pu	–	–	30

## 2.2. Determine and map the boundary of the contaminated areas

A contaminated area can be defined by any criterion appropriate to the site's geography, geology, ecology, and history. One possible boundary might be the background isopleth of a contamination plume, but there are other possibilities, particularly if the types and quantities of contaminants differ from location to location (Table 1). Figure 1A shows a portion of the Idaho National Engineering and Environmental Laboratory (INEEL) divided into contaminated areas based on concentration isopleths for <sup>129</sup>I [3]. The site environmental monitoring organization, in consultation with the stakeholders, should determine the most meaningful and justifiable boundaries for their site.

## 2.3. Determine and map the boundary of discrete habitat types

Like contaminated areas, habitat types may be determined by a number of appropriate criteria such as vegetation, geology, geography, wildlife distribution, history, etc. Figure 1B shows a portion of the INEEL divided into discrete habitat types based on the three dominant vegetation types [4]. In general, within a habitat type, one assumes ecological structure and function are sufficiently homogeneous to be represented by a single parameter, and the species of concern are distributed throughout the habitat type. Between habitat types one assumes that structure and function are dissimilar.

Ecologists familiar with the site, in consultation with the stakeholders, should use best professional judgement and all available data to justify these habitat boundaries. Because different habitat parameters vary at different scales, it is very difficult to set boundaries without knowing a great deal about the parameters and species of interest. In the absence of sufficient knowledge, and in the interest of providing the best possible protection in a timely manner, it will be necessary to recognize the limits of knowledge, and negotiate a workable solution.

## 2.4. Overlay the maps and identify the intersections

Each area of discrete habitat that lies within a discrete contaminated area can be appropriately defined as an assessment area (Figure 1C). This intersection of contaminated areas and habitat types may occur in four ways:

- (1) A single contaminated area may intersect a single habitat patch (Figure 2A). In this case, the contaminated area bounds the assessment area. An example of this kind of intersection might be a small pond with uniformly contaminated sediment.
- (2) A single contaminated area might also intersect multiple habitat patches (Figure 2B). This might be the case at any site which releases airborne contaminants from a stack. In this case, there will be multiple assessment areas bounded by habitat type and contaminated area.
- (3) Multiple contaminated areas of the same type may intersect a single discrete habitat patch (Figure 2C). In this case, the contaminated areas bound the assessment area and it is acceptable to integrate or average over multiple contaminated areas within a single habitat type. This type of intersection might occur in large areas of uniform habitat in which there are multiple disposal pits or pads.

- (4) Finally, there may be multiple habitat patches of the same type which intersect one or more areas with the same contamination profile. In this case, multiple assessment areas are bounded by habitat type and contaminated area. However, because it is reasonable to argue that patches of the same habitat type have similar species assemblages and similar structure and function, we recommend intersections of a given habitat type with contaminated areas having the same contaminants in roughly the same concentrations be considered one assessment area, even though they are separated in space.

In all these examples, it is important that contamination levels or parameters only be averaged over the intersection of the contaminated area and the habitat type of interest and not the areas between the intersection. If the areas outside the intersection were included, the averages would not likely be representative of the habitat type and/or contaminant levels of interest. The contaminated areas outside this intersection will be included in a different intersection of habitat type and contaminated area.

This guidance is not meant to be prescriptive. Each step of the recommended process involves a significant element of professional judgement and requires appropriate justification and documentation. In particular, the environmental monitoring organization at the site will be required to determine, justify, and document appropriate boundaries for areas with similar contamination profiles. Similarly, ecologists familiar with the site will be required to determine, justify, and document appropriate boundaries of similar habitat types.

### 3. CONCLUSION

Regardless of how one chooses to do it, it is always necessary to select some spatial area over which to average environmental measurements. It's often the "hidden assumption" in environmental monitoring programs. The U.S. Department of Energy Biota Dose Assessment Committee believes the procedure we've defined will result in rational, defensible areas of assessment that will be useful in demonstrating compliance with proposed dose limits.

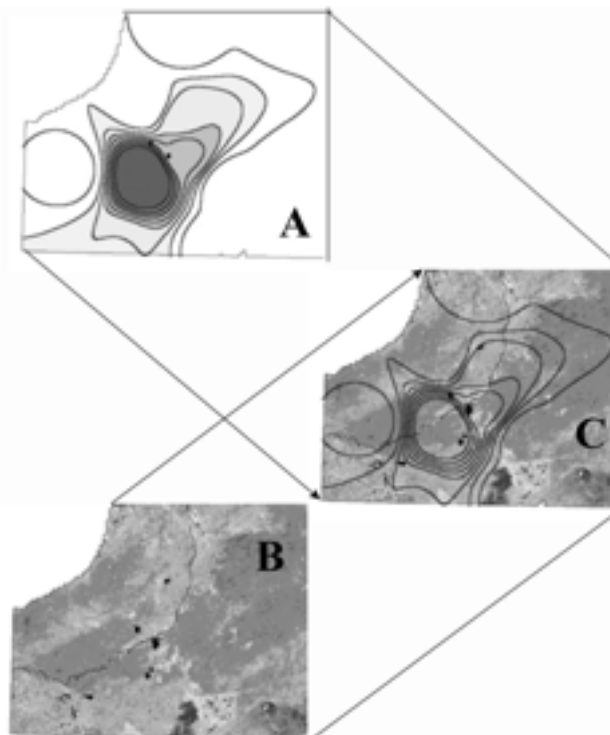


FIG. 1. Maps of the southwest corner of the Idaho National Engineering and Environmental Laboratory. A) Contaminated areas defined by isopleths of <sup>129</sup>I concentration [3]. B) Habitat types defined by the three dominant plant species [4]. C) An overlay of B and C showing potential assessment areas. All area of the same vegetation type bounded by an inner and an outer isopleth can be defined as a distinct assessment area.

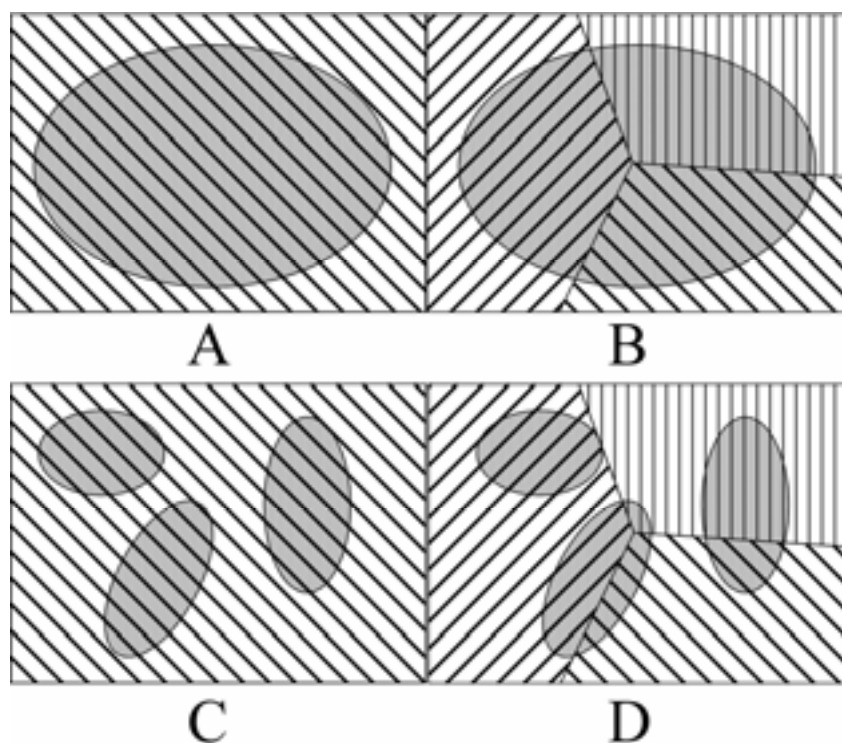


FIG. 2. Schematic representations of different ways contaminated areas may intersect with habitat types. The shaded ovals represent discrete contaminated areas and the cross hatched areas represent different habitat types.

### ACKNOWLEDGEMENTS

The author would like to acknowledge the contribution of David Kocher, Robin VanHorn, and other members of the Biota Dose Assessment Committee who contributed substantially to the ideas presented here. This work was supported, in part, by the U.S. Department of Energy Idaho Operations Office and, in part, by TREC, Inc.

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# **The RESRAD-BIOTA code for application in biota dose evaluation: *Providing screening and organism-specific assessment capabilities for use within an environmental protection framework***

**C. Yu<sup>a</sup>, D. LePoire<sup>a</sup>, J. Arnish<sup>a</sup>, J.-J. Cheng<sup>a</sup>, I. Hlohowskij<sup>a</sup>, S. Kamboj<sup>a</sup>, T. Klett<sup>a</sup>,  
S. Domotor<sup>b</sup>, K. Higley<sup>c</sup>, R. Graham<sup>d</sup>, P. Newkirk<sup>d</sup>, T. Harris<sup>e</sup>**

<sup>a</sup> Argonne National Laboratory, Argonne, IL, United States of America

<sup>b</sup> United States Department of Energy, Washington, DC, United States of America

<sup>c</sup> Oregon State University, Corvallis, OR, United States of America

<sup>d</sup> United States Environmental Protection Agency, Denver, CO, United States of America

<sup>e</sup> United States Nuclear Regulatory Commission, Washington, DC, United States of America

**Abstract.** The RESRAD-BIOTA code was developed through a partnership between offices of the U.S. Department of Energy, the U.S. Environmental Protection Agency, and the U.S. Nuclear Regulatory Commission. RESRAD-BIOTA is being designed to provide a full spectrum of analysis capabilities, from practical conservative screening methods using Biota Concentration Guides – to more realistic organism-specific dose assessment. A beta version of the RESRAD-BIOTA code is currently available for unofficial use and testing. Continued coordination and partnerships with U.S. agencies and international organizations is providing opportunities for the inclusion of additional evaluation approaches and capabilities, such as: (1) development of BCGs for additional radionuclides; (2) additional flexibility for specifying and expanding organism options; (3) improvements to environmental transfer factor parameter datasets; (4) inclusion of additional “reference organism geometries” (e.g., dose conversion factors for ellipsoids of appropriate size and shielding properties for different sized organisms, appropriate for specific ecosystem types); and (5) sensitivity and uncertainty analysis capability for calculated dose estimates.

## 1. INTRODUCTION

The U.S. Department of Energy (DOE) has developed standardized screening and analysis methods[1] provided within a graded approach for evaluating radiation doses to biota. These methods were made available through a DOE Technical Standard document and a series of electronic spreadsheets termed the RAD-BCG Calculator. The methodology provides limiting (dose/risk-acceptable) concentrations of radionuclides, termed Biota Concentration Guides (BCGs) for use in screening water, sediment, and soil media to determine if user-specified dose limits for biota are exceeded. As the graded approach methodology received increasing interest from other U.S. agencies and from international organizations, it became apparent that a more robust and sophisticated software platform was needed to support the refinements and additional capabilities desired by this broader community of users. The RESRAD (RESidual RADioactivity) software platform was used to convert the RAD-BCG Calculator as the basis for a new PC code, “RESRAD-BIOTA”. The code was developed through a partnership among offices of the DOE, the U.S. Environmental Protection Agency (EPA), and the U.S. Nuclear Regulatory Commission (NRC).

## 2. RESRAD FAMILY OF CODES

The RESRAD family of codes[2], developed at Argonne National Laboratory, is a suite of dose/risk assessment tools designed to evaluate radiological and chemical contamination in the environment. The RESRAD family of codes, shown in Figure 1, includes (1) RESRAD, which evaluates doses and related risks to human health and the environment resulting from exposure to radiologically contaminated soils, (2) RESRAD-CHEM, which assesses chemical risk from soil contamination, (3) RESRAD-ECORISK, which estimates the risk from chemical contaminants to ecological receptors, (4) RESRAD-BUILD, which evaluates potential health impacts in buildings contaminated with radioactive materials, (5) RESRAD-RECYCLE, which estimates radiation doses to various receptors resulting from the recycle and/or reuse of radioactively contaminated materials and equipment, (6)

RESRAD-BASELINE, which performs baseline risk assessments following EPA’s human health risk assessment guidelines, (7) RESRAD-OFFSITE, which extends the RESRAD (onsite) model to evaluate dose/risk to receptors located at off-site locations, and (8) RESRAD-BIOTA, which uses a graded approach in assessing biota dose for radionuclides. All of the RESRAD codes are easy to install, have user-friendly interfaces, and provide on-line help messages. They can be used for different applications and are maintained and updated regularly; comprehensive documents are available to support their operation and application.

### 3. DEVELOPMENT OF RESRAD-BIOTA

The development of the RESRAD-BIOTA code was initiated by DOE in June 2000. The Microsoft Excel-based RAD-BCG Calculator continues to meet DOE’s needs but is approaching the limits of its software platform capabilities. As such, the RESRAD platform was selected as the basis for this “next generation” biota dose evaluation tool because: it has a highly-regarded pedigree, supported by extensive quality assurance (QA) and validation studies; it is widely recognized and implemented within DOE, other U.S. Federal and State agencies, and the U.S. nuclear industry; and it provides the software architecture and degree of sophistication needed for expanding upon biota methods and capabilities.

The RAD-BCG Calculator was successfully converted into a beta version of RESRAD-BIOTA in 2001. The beta code was made available for unofficial use and testing, and to encourage feedback from potential users regarding additional modifications and capabilities that should be incorporated into future versions of the code. The beta version was reviewed within DOE’s Biota Dose Assessment Committee.

In May 2001, an interagency partnership was initiated, whereby EPA and NRC provided funding and staff commitment to collaborate with DOE to further develop the RESRAD-BIOTA code. This interagency partnership is being coordinated through the informal interagency ECORAD working group. In December 2001, an improved beta version of RESRAD-BIOTA was developed. Some features of this version of RESRAD-BIOTA are described in the following sections.

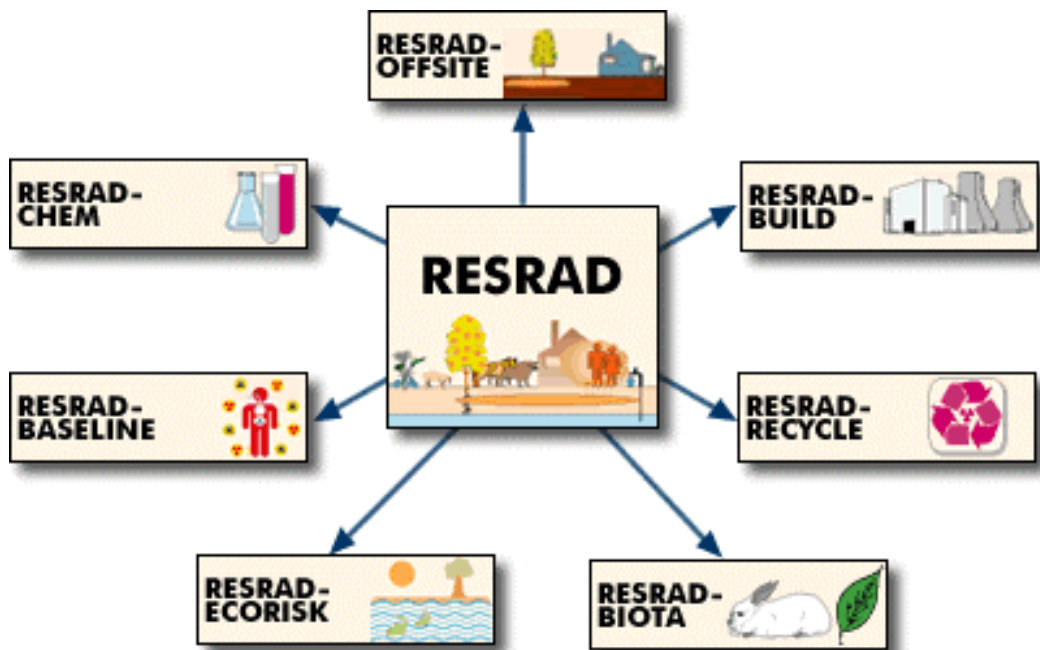


FIG. 1. RESRAD family of codes.



#### 4. FEATURES OF RESRAD-BIOTA

RESRAD-BIOTA is freely distributed at <http://www.ead.anl.gov/resrad> and at <http://homer.ornl.gov/oeпа/public/bdac/>. It is easy to install and user-friendly. The application's main window is shown in Figure 2.

There are three levels of biota dose evaluation available, ranging from Level 1 where conservative assumptions are made through provision of a general screening process but few inputs are required, to Level 3 where fewer assumptions are made a priori but more site- or receptor-specific input data are required. Analysis can start at the general screening level and proceed to level 3 if the assumptions in level 1 are too conservative.

There is also an option for which type of ecological system, terrestrial or aquatic, to assess. However, the layout of the form of an aquatic assessment is almost identical to that of a terrestrial assessment (shown in Figure 2). The only differences in the forms are the default organisms considered and the ability to specify a distribution coefficient if only one of the two media concentrations is known.

After a contaminant (radionuclide) is added to the *Contaminants List Box*, its characteristics are shown in the corresponding boxes. Once selected, the characteristics of the contaminant can be modified. The dose limits of each organism also can be modified. The values for all of the ratios and limits are updated immediately upon data entry.

The organism icons next to the characteristics of each contaminant reflect the organism that is responsible for the limiting dose. For example, a raccoon icon indicates that this terrestrial animal is responsible for the limiting dose. A *results window* with the concentrations, Biota Concentration Guide (BCG) values, and ratios for each individual contaminant, as well as the summed ratios, can be accessed by clicking on the *BCG Results* button. The dose associated with each organism is accessed by clicking on the *Dose Results* button. Examples of the *BCG Results* and *Dose Results* windows are shown in Figures 3. The results can also be shown in graphics (bar charts). Figure 4 is a bar chart of calculated doses.



FIG. 2. RESRAD-BIOTA main window.

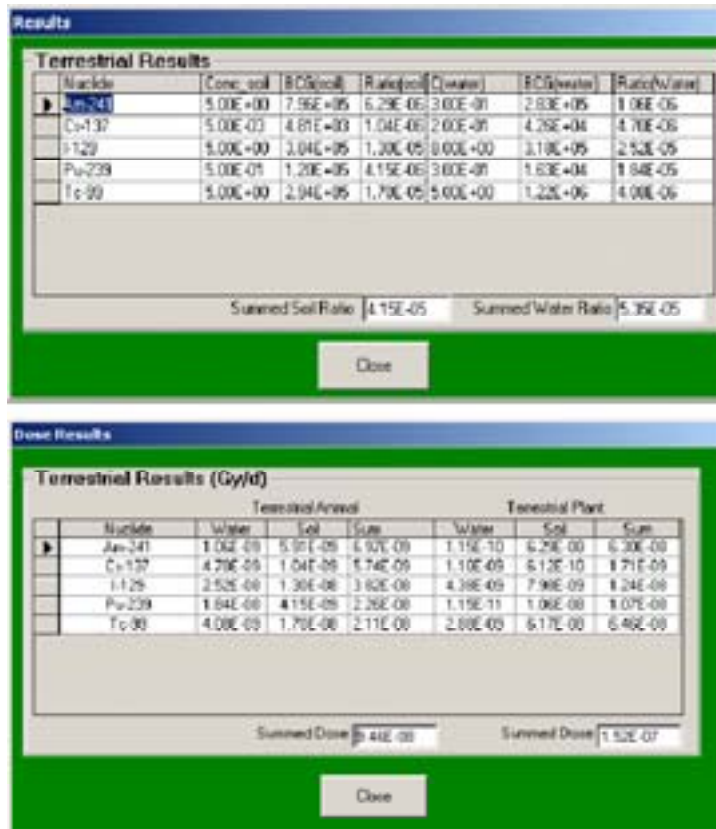


FIG. 3. Text result.

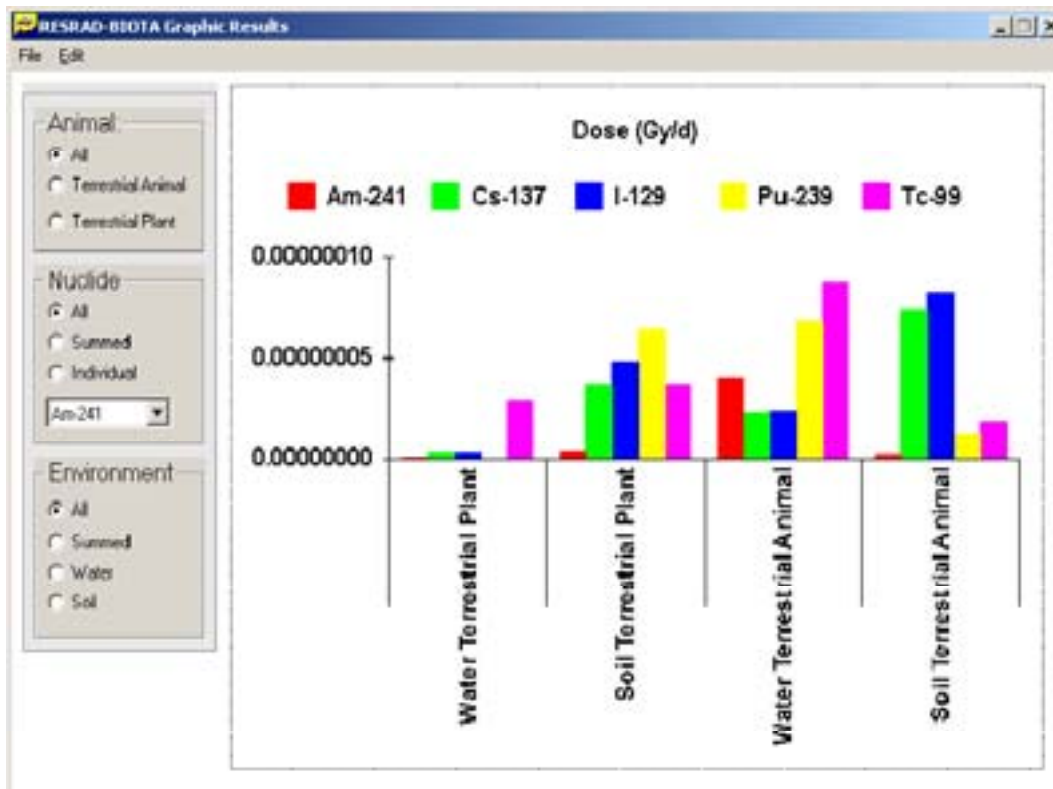


FIG. 4. Graphic results.

The layout of the Level 2 form is shown in Figure 5. Data entry is more extensive in Level 2 than in Level 1. The additional data requirements and flexibility whereby users can customize or make modifications to a Level 2 assessment include the following: a check box that allows users to deselect the inclusion of progeny radionuclides (if applicable) in the internal dose conversion factors, the ability to modify the default radiation weighting factor for alpha radiation, the ability to enter a site- or receptor-specific B-value for each contaminant, specification of an area factor for each organism, and a dose conversion factor for each radionuclide selected.

Parameters used to calculate the dose conversion factors can also be modified including the internal and external dose conversion factor input data. The dose conversion factors will be calculated and displayed on the bottom of the form in the frame titled *Calculated Dose Conversion Factors*. Figure 6 provides an example of the *Dose Conversion Factor Input Window*.

As in Level 1, the Ratio for All nuclides summed text box(es) will turn red if the ratio exceeds 1. The user can then move to the final level (Level 3) and perform an assessment at the greatest level of detail currently supported by RESRAD-BIOTA, consistent with the DOE graded approach methodology.

The layout of Level 3 is shown in Figure 7. The key feature of Level 3 is the *Allometric Parameters window* (upper right in figure), which allows specification of parameters specific to the Riparian or Terrestrial Animal that contribute to the estimation of internal dose, including weight, ratio of active to basal metabolic rate, fraction of energy ingested that is assimilated and oxidized, caloric value of food, fraction of soil in diet, and airborne dust loading. Organism- and Nuclide-specific parameters that can be manipulated are fraction of intake retained (f<sub>I</sub>), the constant (a) and exponent (b) of the biological decay constant, and an adjustment for inhalation relative to ingestion (PT/IT). The organism's default food source Biv values can also be modified. After all of the data have been specified the summed ratios will be updated accordingly.



FIG. 5. Level 2 window.

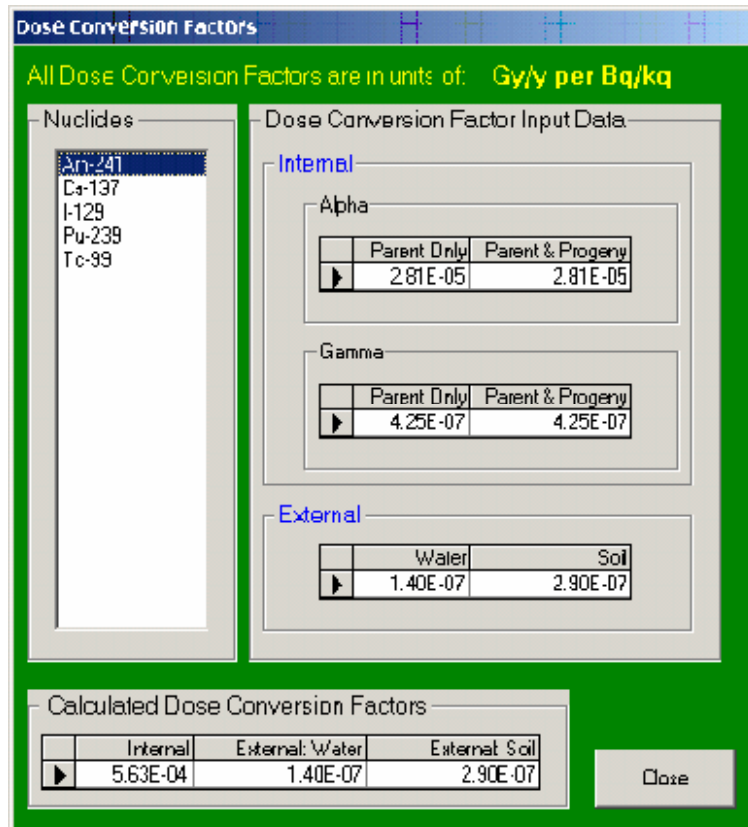


FIG. 6. Dose conversion factor input window.

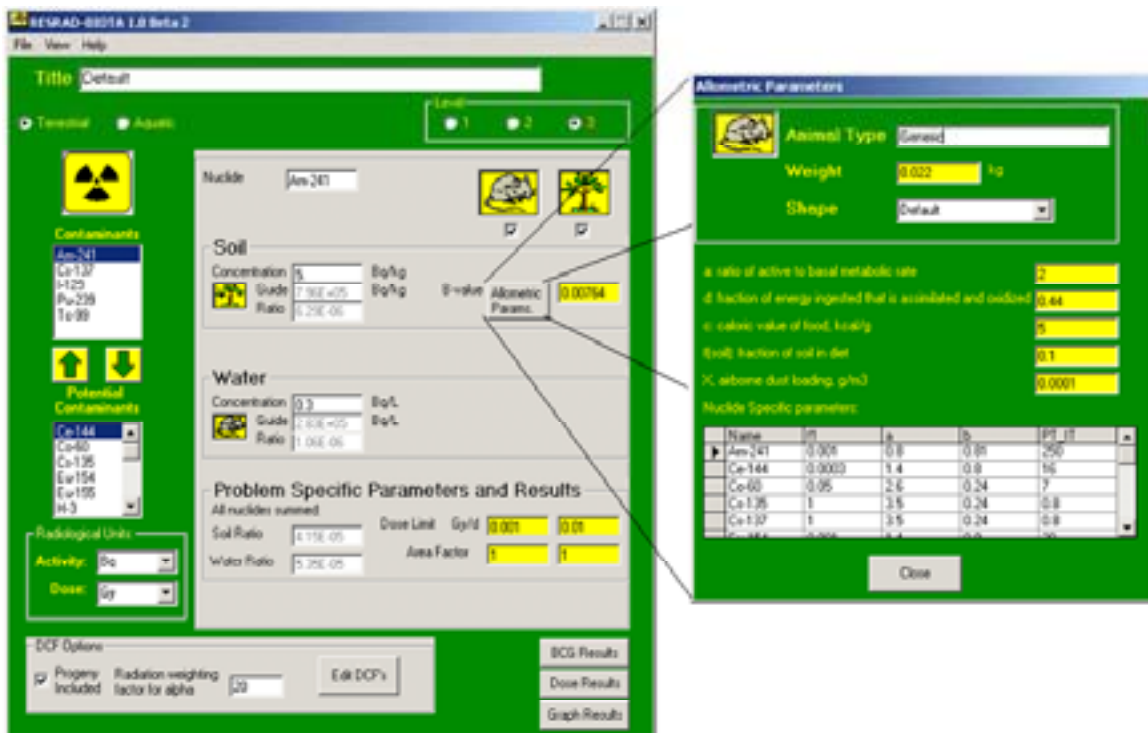


FIG. 7. Level 3 window.

## 5. FUTURE PLAN

Co-ordination and partnerships with U.S. agencies through the interagency ECORAD workgroup will be continued; coordination and partnerships with international organizations is desired. At a recent meeting of the ECORAD work group held at Argonne National Laboratory, a consensus-based work scope to meet the anticipated analysis needs of each agency was agreed upon by the participating agencies. The work scope provides for the inclusion of additional evaluation approaches and capabilities, such as: (1) development of BCGs for additional radionuclides; (2) additional flexibility for specifying, expanding, and sharing organism specification through an organism-editor; (3) improvements to environmental transfer factor parameter datasets; (4) inclusion of additional "reference organism geometries" (e.g., dose conversion factors for ellipsoids of appropriate size and shielding properties for different sized organisms, appropriate for specific ecosystem types); and (5) sensitivity and uncertainty analysis capability for calculated dose estimates. It is also planned that an improved ability be developed that allows user to transfer radionuclide environmental media concentrations generated from other modeling codes for subsequent biota dose evaluation in RESRAD-BIOTA. The availability of the next beta version of the RESRAD-BIOTA code, incorporating many of the features outlined above, is tentatively planned for Spring 2003.

## 6. CONCLUSION

The RESRAD-BIOTA code provides a cost-effective and flexible tool for conducting biota dose evaluations that could be applied within an international framework for protection of the environment. Once finalized, it will support a variety of environmental assessment needs, including: (1) demonstrating compliance of routine facility and site operations with available dose limits for biota; (2) conducting ecological screening assessments of radiological impact at contaminated sites; (3) estimating doses to biota in an Environmental Impact Statement when coupled with predictive dispersion codes that model a facility's effluents prior to construction; and (4) predicting future doses to biota when coupled with pathway codes as part of assessing the decommissioning of facilities.

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# **RadCon: A radiological consequence assessment model for environmental protection**

**J. Crawford, R.U. Domel**

Australian Nuclear Science and Technology Organisation, Menai, NSW, Australia

**Abstract.** This paper highlights the development of a radiological consequences model (RadCon) for use in the Australian and South East Asian Region, the acquisition of available data for use with the model, and sensitivity analysis ability to target future research. RadCon is a simple easy to use model that assesses the radiological dose consequences to humans resulting from the short-term release of radionuclides to the environment and in the process provides the concentrations of the chosen radionuclides in the selected plants and animals. This way, environmental protection can be addressed. Internal and external exposures from the passing cloud and from radionuclides deposited on the ground can be assessed in humans, and given the required data also in animals. RadCon does not provide atmospheric dispersion and ground deposition estimates and requires input data from meteorological models or measured data. RadCon was tested in an international model inter-comparison program (BIOMASS) sponsored by the International Atomic Energy Agency and performed well. The model continued to evolve over the period of the exercise and the final estimates generated by RadCon were comparable to the test data. From this BIOMASS experience, a module has been added to RadCon to allow the viewing of concentrations of radionuclides in plant and animal products together with the generation of a 95% confidence interval for the estimates.

## 1. INTRODUCTION

Over time, a number of models have been developed to simulate the transfer of radionuclides through air, water and terrestrial ecosystems. However, the major studies have focussed mainly on the temperate and cold regions of the world and almost exclusively in the Northern hemisphere. Only limited information is currently available for tropical and subtropical regions.

A model, RadCon, has been designed and used by ANSTO for application in the Australian and South East Asian region. RadCon was implemented with the intended application of calculating the dose received by individuals following an accidental release of radionuclides to the environment. As a result of participation in a model inter-comparison exercise a module has been added to generate the radionuclide concentration in plants and animals from atmospheric dispersion or ground concentration, together with a 95% confidence interval for the estimates. This has permitted RadCon to be considered for use in an environmental protection capacity, although additional data would need to be sourced and added and some additions made to the output screen.

Aside from the calculation of concentrations, RadCon implements parameter sensitivity analysis to assist in the identification of the most relevant parameters in dose calculation. The results of this analysis can be used in directing the acquisition of additional site specific data for the region under investigation.

Other features of RadCon include, separation of the data from the code, incorporation of a graphical user interface at the design stage (opposed to pre and post processing of data) and portability.

The following sections describe the RadCon implementation as well as the evaluation carried out by participating in the BIOMASS program [1].

## 2. THE RADCON MODEL

### 2.1. Model overview

While the main motivation behind the implementation of RadCon was to assist in the identification of the most relevant parameters in dose assessment estimation, it also acts as a tool in estimating the concentrations in foods, i.e. crops and animal products, consumed by individuals as well as the dose to the individual. One, but not the only, potential application of RadCon, is the estimation of

concentrations in foodstuff and dose to individuals at some distance from the source. For example, the impact on Australia from a release in the South East Asian region.

Atmospheric dispersion has been decoupled from the dose calculation and a separate atmospheric transport code is required for use with RadCon. This can be any code which is suitable for the conditions under study and generates time varying air concentration and ground deposition over a two dimensional region of simulation. For example, for a study carried out by ANSTO [2], the air and ground concentrations over the simulation period was provided by the Australian Bureau of Meteorology. Alternatively, measured ground or soil concentrations can be used.

For the calculation of contamination from crops, uptake by the roots from the contaminated soil is considered as well as direct deposition onto the foliage. In estimating uptake from the soil the effective soil contamination is adjusted for radioactive decay, leaching and fixation of the radionuclides to the soil, thus reducing the availability for root uptake. For the deposition onto foliage, two crop types are considered, those that are consumed totally (e.g. leafy vegetables) and those that are used only partially (e.g. cereals, potatoes and fruit). The concentration in the crops from direct deposition is determined by the amount of activity deposited followed by an adjustment for activity loss due to weathering, radioactive decay and dilution due to growth. For crops that are partly consumed, translocation from the deposited material to the edible part is modelled. Contamination of grass and the associated loss processes are also modelled.

The mathematical models have been adapted from ECOSYS-87 [3] and CLRP [4]. The models together with the assumptions have been documented in an ANSTO report [5].

## 2.2. RadCon evaluation

In order to evaluate RadCon's predictions against field data, the RadCon developers participated in a model inter-comparison exercise sponsored by the IAEA. A number of developers and/or users of radiological consequences models participated in the exercise, which involved the estimation of  $^{137}\text{Cs}$  in plants, animals and humans and dose to humans in the Bryansk Region in Russia following the Chernobyl accident.

The exercise [1] can be seen in two parts:

- A model test, i.e. comparison of model estimates against measured data (test data). Measured data is available for ground concentration in the region in the period following the accident, as is air concentration, concentration in crops and animals. In addition, relevant information on agricultural practices, soil types, lifestyle and diets is also available.
- A model comparison in which the total doses to the local population from  $^{137}\text{Cs}$  in the test environment are predicted by different models and compared with each other and with estimates made by the authors of the scenario.

While the results from a model depend on a number of factors, and are strongly influenced by the suitability of the chosen parameter values for the site under consideration, the exercise was used by the RadCon developers to evaluate if the most important processes have been included in RadCon and implemented appropriately. The model and implementation evolved over the period of the exercise and the final estimates generated by RadCon were compatible to those reported by other participants and the test data provided.

The exercise highlighted the need for the chosen parameter values to reflect the site conditions. Further, a number of countermeasures had been implemented at the test site, and as such these had to be considered in estimating concentrations and the final dose. Application of countermeasures was not an intended function of RadCon. However, we were able to simulate some of the countermeasures by adjusting some of RadCon's parameters to reflect the effect of the countermeasures.



### **2.3. Data and parameter input into the model**

In parallel with the development of the RadCon model, a data acquisition and evaluation exercise was undertaken. The aim was to obtain required parameter values for use with RadCon, to enable the estimation of radiological consequences in the Australian and South East Asian Region [6].

Literature and library searches initiated the data acquisition process, with the available data being analysed and evaluated and then stored in tables in a spreadsheet application. The bulk of the Tropical and Subtropical Data were acquired from the draft IAEA, 1997 [7]. The data in this document were presented as individual experimental data points for each radionuclide and required further summation and statistical analysis. This study also highlighted that there are still large gaps in the knowledge relating to the transfer of radionuclides in tropical and subtropical environments. To fill the gaps and be able to run the RadCon model, temperate data were used as the default parameters.

### **2.4. Parameter sensitivity analysis in RadCon**

One of the aims in the RadCon implementation was to assist in the identification of those parameters which contribute most to the final dose, such that any future research could be directed towards the most important parameters. To assist with this, two parameter sensitivity analysis techniques have been implemented in RadCon:

- One-at-a-time Sensitivity Measures: Ranking of the importance of the parameters can be obtained quickly by using this technique. The ranking is obtained by calculating the dose with each parameter set to a base value, e.g. the mean, followed by calculation of the dose by perturbing each parameter by a user selected percentage (e.g. 1%) while leaving all other parameters constant. The change in dose gives the rank of the corresponding parameter.
- Extended one-at-a-time: This technique [8] examines the change in output as each parameter is individually increased from its mean by a factor of its standard deviation. This sensitivity measure takes into account the parameter's variability and the associated influence on model output.

## **3. THE USE OF RADCON FOR ENVIRONMENTAL PROTECTION**

### **3.1. General**

Given ground and airborne radionuclide concentrations, RadCon is able to calculate plant and animal concentrations, provided the specific input parameters for the nominated plant or animal are known. In fact, the majority of calculations and parameter inputs in RadCon deal with aspects of plants and animals, such as root uptake from the soil, deposition on the plant and translocation within the plant.

What needs to be determined internationally and databanked, is the dose conversion factors to be used for plants and animals, and then of course the limits to be applied.

### **3.2. Specifying parameter values for plants and animals**

The RadCon model uses a number of formulae to calculate radionuclide uptake and translocation in plants [5]. Many of these formulae and parameter values have been obtained from Müller and Pröhl [3], where the parameter values relate to the northern hemisphere – the equivalent data for the tropics still needs to be measured and/or sourced. One formula covers the uptake of the radionuclide by the roots of plants. The parameter values required include a number of soil related aspects such as soil density, migration of the isotope below the root zone, fraction of the isotope that is mobile in the soil, rate of fixation, and a soil reduction factor which depends on agricultural practices (eg ploughing).

Deposition onto plants from airborne radionuclides is modelled followed by foliar uptake. Grass, where it is continually cropped, is modelled separately. The deposition formulae calculate total deposition from both wet and dry conditions. Parameters required include an interception fraction, deposition velocity, leaf area index at the time of deposition, retention coefficient, translocation factors (fraction of the activity deposited on the foliage that is transferred to the edible part of the plant – for plants that are only partially consumed, such as fruit), time-integrated activity concentration in air and



amount of rainfall during the deposition event. Grass requires additional parameters for the yield of grass at the time of deposition.

The RadCon model evaluation through the BIOMASS project required outputs for concentrations in plants and animals. The results generated, using RadCon, for concentration in potatoes, hay, wild berries and beef are given in Figure 1.

In all the estimates generated by RadCon a sharp decline is seen in year 2 due to the effects of countermeasures being approximated by a step function. A module to model countermeasures has not yet been implemented in RadCon. Other differences in the results are due to RadCon not implementing seasonal variations and resuspension, in the case of the wild berries. However, results generated using annual averages did reflect the test data well.

The results of the BIOMASS program are scheduled to be reported in an IAEA-TECDOC. A full description of the scenario under consideration as well as the results and analyses generated by the participants will be detailed in the report.

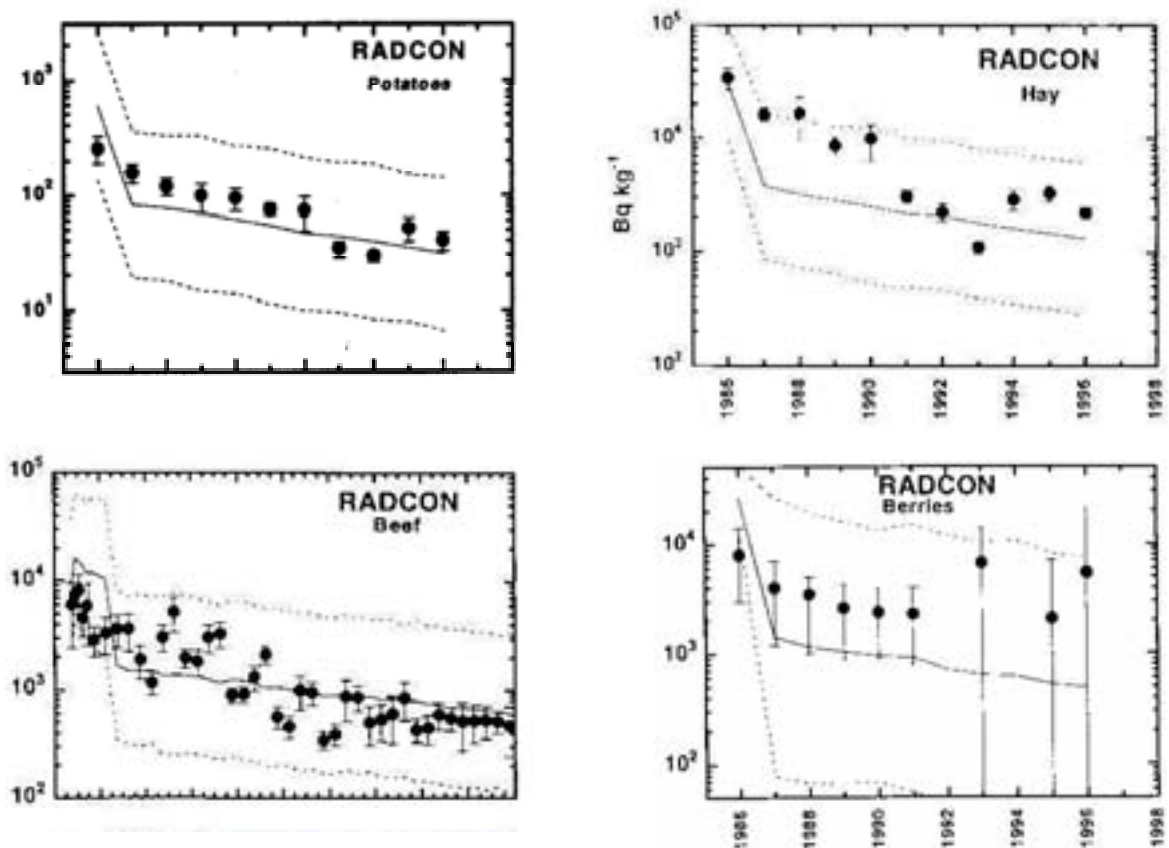


FIG. 1.  $^{137}\text{Cs}$  concentration in potatoes, hay, beef and berries in  $\text{Bq kg}^{-1}$  (dry weight in hay, fresh otherwise) against time from the accident in years (1986–1998). Measured data is represented by the filled circles. RadCon's prediction are given by the solid line and the two dotted lines give the 95% confidence interval estimated by RadCon.

## 4. RADCON IMPLEMENTATION

RadCon was designed and implemented with an initial focus on its potential use in the Australian and the South East Asian region. Given the large variability in this region, emphasis was placed on the ability of the computer code to handle the diversity of parameters such as diets of individuals, soil types and the food crops.

The heart of the code is the implementation of the mathematical models, i.e. the computational model. Interfacing to the same computational model are two user interfaces:

- A graphical user interface allowing the setting of scenario characteristics leading to the choice of parameter values and the calculation of dose over a two dimensional region. Parameter sensitivity analysis can also be carried out through the graphical user interface.
- A text base interface, allowing the calculation of dose to individuals from given values for air and ground concentration. Aside from the calculation of the dose to individuals, this interface also outputs the concentrations in food crops and animal products together with 95% confidence intervals.

To achieve portability across computer platforms, RadCon has been implemented in the Java [9] programming language.

### 4.1. Specifying parameter values

As a primary function, RadCon was designed and programmed to carry out calculations over a two dimensional region with the input values of the parameters at any location reflecting the underlying characteristics at that location and the characteristics of the group under study, e.g. soil type, lifestyle of individuals in the region, diets of people and animal, etc. Instead of requesting the user to specify parameter values over the region, a system was designed and implemented to enable the user to select from a pre-defined set of alternatives for each of the characteristics. For example, the soil types have been grouped into clay, sand, loam etc.; lifestyle of individuals has been grouped into rural, urban and suburban, etc.

The subset of characteristics on which a model parameter depends were identified and the actual values are stored, against the different characteristics, in the data files used by RadCon. For example, the soil to plant transfer factor depends on the plant type, the soil type and the radionuclide of interest, thus a separate value for each combination of these characteristics is required in the data files. While the data files can be edited using most system editors, a program has been written, RadConEd [10], which presents a graphical user interface for the editing of data files, and allows some checking of the input data as well as keeping logs of changes undertaken.

The required information in the data files includes transfer rates – both from soil to plants and plants to animals, animal diet, dose conversion rates, *etc.* A detailed description of the information in these files is given in [11]. The parameter values used for Australian tropical/subtropical regions is presented in [12].

In order to carry out a calculation, the required set of alternatives have to be established, e.g. for the ingestion pathway alternatives have to be chosen for:

- The soil type; Sand, Loam, Clay or Coral.
- The race of the target group; Aboriginal, Asian, Caucasian, Indian or Islander.
- The diet of the target group; race default diet or any of the other predefined diets. Separate diets are generated by listing the quantities of each of the food items consumed by each of the age groups. These diets are set up using the RadCon data editor, and each diet is identified by a unique name which is then user selectable at calculation time.
- The diets of each of the animals are also pre-defined in a manner similar to the human diets. Again, each has a unique name assigned to it, which are the alternatives from which the user selects from at calculation time.

- The age of the target group; Adult, Adult\_male, Adult\_female, Child or Infant.
- The lifestyle; Rural, Residential or Urban.

The alternatives specified above can be modified relatively easily during customisation of RadCon for a particular application.

#### 4.2. RadCon, graphical user interface

A graphical user interface is used for the calculation of dose to individuals and for the generation of relative dose sensitivity to each of the model parameters. To use RadCon in this mode, in addition to the tables of the required parameter values, time varying air and ground concentration over the region of calculation, and an image of the region (e.g. a map, for visualisation purposes) are required. The graphical user interface offers two main screens – the input and output screens. These are described in [13]. An example of the output screen is presented in Figure 2. The user can further select a grid location to view and optionally save to a text file the actual dose at that location together with the contribution from each pathway and radionuclide.

In RadCon, parameter sensitivity analysis is initiated from the calculation screen (Figure 2), following which parameter sensitivity calculations are carried out over the region and presented to the user. The user can examine the parameter ranking at any point; the ranking of each parameter against each radionuclide is given. These ranks can be sorted based on parameter or radionuclide, and can be saved to a file for future processing using the export option.

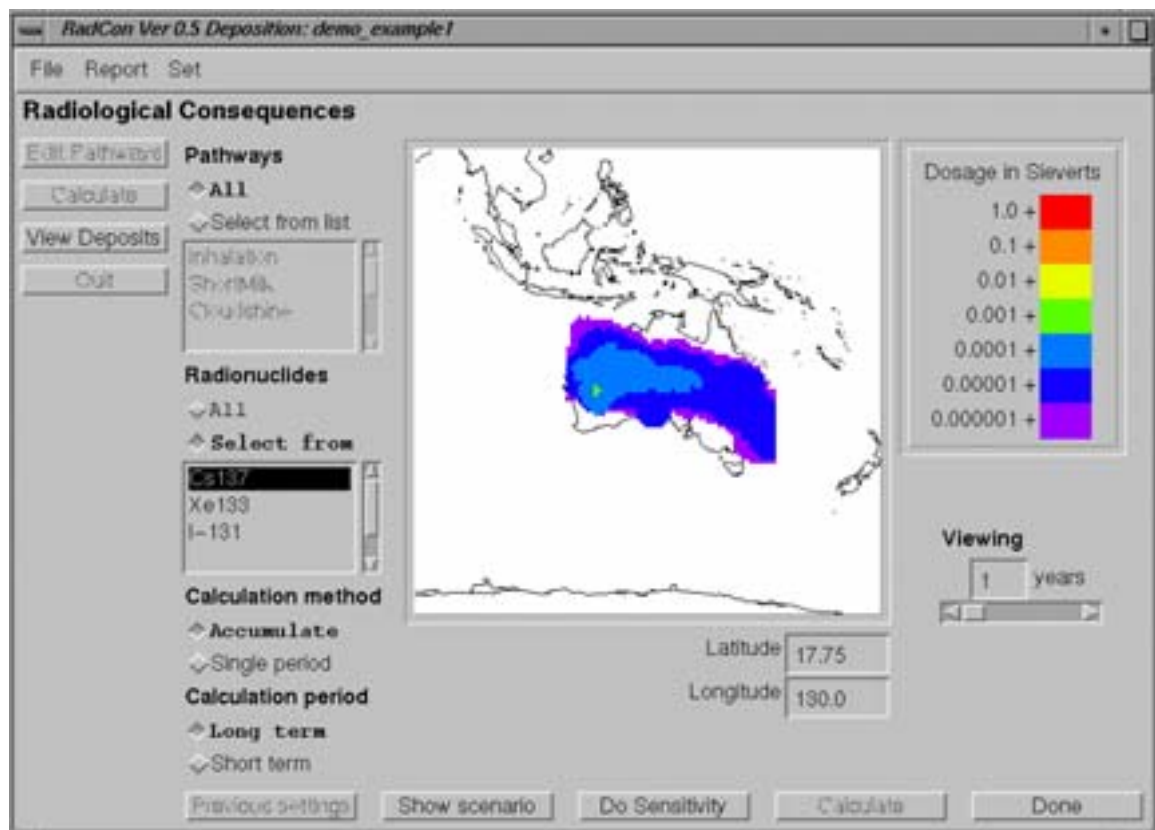


FIG. 2. RadCon's calculation and output screen.

### 4.3. RadCon, text based interface

A module providing a text based interface has been implemented to generate the concentrations in food crops and animal products. At the time of writing this paper the concentrations in crops and animal products can be accessed only through the text based interface.

Both the graphical user interface and the text based interface use the same computational module and input data files, thus, the same options can be selected from either interface. The main difference between the graphical user interface and the text based interface, is that when using the graphical interface, calculations are carried out over a two dimensional region, while for the text based interface the calculations are carried out for a single location for which the output is the radionuclide concentration in each plant or animal chosen, together with a 95% confidence interval of the estimates, see Figure 1. The 95% confidence intervals are estimated by the user specifying a distribution of each input parameter. A Monte Carlo sampling technique is then applied to propagate parameter uncertainty to the calculated estimate.

## 5. CONCLUSION

The RadCon model has been implemented providing a graphical user interface to set up required characteristics over a two dimensional region and carrying out the calculations. All the required data are provided in data files, thus enabling easy adaptation to a new site. Parameter sensitivity analysis can be carried out to identify the most important parameters for a given scenario. A second, text based interface allows the viewing of concentrations in plant and animal products, also allowing the calculations of 95% confidence intervals. By using the Java programming language for implementation, portability across computer platforms is possible.

RadCon's mathematical models and assumptions have been evaluated. Some refinements to the implementation can be carried out and additional exposure pathways can be included, e.g. re-suspension and aspects of the aquatic pathway.

The model inter-comparison exercise has once again highlighted the importance of using parameter values that reflect the underlying characteristics of the site under study.

At this time, the concentrations in plant and animal products are not able to be displayed in Figure 2. A possible extension to RadCon could be the addition of an option specifying which results to display on that screen e.g. concentration in a specified crop, or animal, or dose. For the model to be used in the environmental protection mode, more data needs to be sourced and added to the data tables, and environmental radiation protection limits to be set for plants, animals or ecosystems to allow the outputs of RadCon to be meaningful. In addition, currently RadCon calculates annual average concentrations, i.e. no seasonality is considered.

The transfer parameter data for tropical and subtropical conditions is still very limited, with the most abundant being that available for grains and vegetables. Transfer data for tropical fruit is dependent on only a very limited number of data points and for natural environments almost no transfer data is available. For the studies undertaken, transfer parameter data is taken from temperate studies, for deposition on and translocation in food plants and pastures, and for soil characteristics to name a few. This leaves a huge gap for future research.

## ACKNOWLEDGEMENTS

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# Evaluation and verification of foodweb uptake modeling at the Idaho National Engineering Laboratory

R. VanHorn

Idaho National Engineering and Environmental Laboratory, Idaho Falls, ID, United States of America

**Abstract.** The Idaho National Engineering and Engineering Laboratory (INEEL), a United States Department of Energy facility, is located on 2,300 square kilometers of cool desert ecosystem characterized by shrub-steppe vegetative communities. This large complex superfund site has nine “waste area groups” and multiple contaminants including radionuclides. The INEEL is systematically evaluating risk to non-human receptors located at the facility. Initial assessments used foodweb modeling to calculate the potential exposure of non-human receptors to contaminants in various media. These foodweb models used literature values, which are primarily based on agricultural studies, for transport of contaminants from soil to non-human receptors. Exposure modeling accuracy is dependent on the quality of input parameter values and the validity of the model’s structure (i.e., the degree to which it represents the actual relationships among parameters at the site). Site-specific field measurements of tissue residue levels (concentrations) are the most accurate exposure assessments. In 1997, 1999 and 2000, data was collected both on and off the facility to support the development of site-specific data at the INEEL. These data were compared to the literature values and modeling used in initial risk assessment screening. Results of this study, a comparison and discussion of problems and lessons learned (i.e. non-detects, potential differences in uptake in areas of greater contamination and selection of sites) are presented.

## 1. INTRODUCTION

The Idaho National Engineering and Engineering Laboratory (INEEL), is a United States Department of Energy (DOE) facility, located on 2,300 km<sup>2</sup> on the eastern Snake River Plain in southeastern Idaho of the United States of America. The INEEL facility was established in 1949 for nuclear research and related activities. Today, research, training, and production activities related to defense and non-defense programs continue to be conducted at the INEEL. The facility activities are limited to a small percentage of the total area and grazing by domestic cattle and sheep has been prohibited in the central area of the INEEL (approximately 1,385 km<sup>2</sup>) since 1957. The DOE established the INEEL as a National Environmental Research Park (NERP) in the 1970s. This is one of seven NERPs in the United States and is one of two that contain sagebrush-steppe ecosystems.

The INEEL was designated a Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) site on November 21, 1989. The CERCLA activities at the INEEL include systematically evaluating risk to non-human receptors at multiple sites identified at the facility. Ecological risk assessments primarily used foodweb modeling to calculate the potential exposure of non-human receptors to contaminants in various media. These foodweb models used literature values, which are primarily based on agricultural studies, for transport of contaminants from soil to non-human receptors. Obtaining accuracy in the risk assessment exposure modeling is highly dependent on parameter input quality and the validity of the model’s structure (or how well the model reflects the actual relationships among parameters at the site) [1].

Sampling of co-located biota and soil was performed at the INEEL during 1997, 1999 and 2000. The first sampling was conducted during the summer of 1997 to evaluate the potential for biotic transport or plume dispersion compared to background and to validate the foodweb modeling. The on-site sampling activity was located northeast of the Idaho Nuclear Technology and Engineering Center (INTEC) fence line that houses facilities for reprocessing spent fuel from government defense and research reactors. Facilities at INTEC include spent fuel storage and reprocessing areas, a waste solidification facility and related waste bins, remote analytical laboratories, and a coal-fired steam generating plant. The off-site was located approximately 20 km southwest of the INEEL boundary.

In 1999, a limited number of onion samples were collected at both the on-site and off-site locations to evaluate the potential for uptake of ordnance contamination and to support the Native American scenario. In 2000, soil and biotic sampling was performed outside the fence at the Boiling Water Reactor Experiment (BORAX)-I Reactor Burial Site. This sampling was performed to support

characterization and to evaluate the biotic movement of contamination in the environment. The BORAX-I was a light-water reactor constructed in 1953. This facility was deliberately destroyed in a final excursion in 1954. An engineered barrier was placed over the site in 1996 and all soil with concentrations of radionuclides above human health risk levels was removed. However, low levels of radionuclides remain in the soil at this site.

This paper evaluates the site-specific tissue radionuclide concentrations and compares them to the values used in foodweb modeling performed for the ecological risk assessments at the site. Results of this comparison and its implications to risk assessment modeling and long-term monitoring will be discussed.

## 2. DISCUSSION

A reduced foodweb model for the INEEL is shown in Figure 1. This foodweb model was developed to provide a visual interpretation of the movement of contaminants through the trophic levels in the ecosystem. Transfer factors (TFs) are used in exposure modeling to estimate tissue concentrations for the dietary ingestion pathways for birds and mammals when actual tissue concentrations are not available. A TF is a unitless ratio calculated by dividing the tissue concentration by the soil concentration.

For example, using the soil concentration, a soil-to-deer mouse cesium (Cs) TF is used to estimate the Cs concentration in the deer mouse that would be available to an avian carnivore such as the red-tailed hawk ingesting the mouse. A plant TF (sometimes called a plant uptake factor [PUF]) is likewise used to estimate an analyte concentration in vegetation browsed by an herbivore.

Transfer factors can be dependent on many factors including: the nature and extent of contaminant, the contaminant or radionuclide species, the animal or plant species, the soil/chemical environment, a number of soil- and organism-related variables, and numerous other possible conditions. Appropriate and applicable studies on biotransfer in native species are not generally available. Although, site-specific field measurements of tissue residue levels (concentrations) are considered the most accurate exposure assessments, TFs from comprehensive literature sources [2–6] were evaluated for use at the INEEL. One of the primary criteria for selection of TFs from the literature for use in the INEEL risk assessments was their applicability to semi-arid, non-agricultural scenarios. The values selected for use at the INEEL were all taken from the IAEA [2]. The IAEA [2] provides transfer coefficients for radionuclides primarily in beef, sheep and goat meat. The TFs for feed to meat on a wet weight basis include:  $8.0E-01$  for Cs,  $8.0E-2$  for strontium (Sr), and  $6.2E-02$  for uranium (U). The TFs for soil to plant on a dry weight basis include:  $5.3E-01$  for Cs,  $2.5E+00$  for strontium (Sr), and  $1.4E-02$  for uranium (U).

## 3. FIELD SAMPLING

During 1997, sagebrush (*Artemisia tridentata* ssp. *wyomingensis*), crested wheatgrass (*Agropyron cristatum*), deer mice (*Peromyscus maniculatus*), mountain cottontails (*Sylvilagus nuttallii*), darkling beetles (*Eleodes* spp.), and grasshoppers primarily from the family Acrididae were collected at 5 locations on-site and 5 locations off-site.

The on-site samples were collected in the area of INTEC plume and are referred to as the ecological study areas (ESA) # 1 through #5. The off-site samples were collected in a reference area located approximately 20 kilometers south of the INEEL on public lands and are referred to as the reference study areas (RSA) # 1 through #5.

During 2000, sagebrush, deer mice, cottontails and Ord's kangaroo rats (*Dipodomys ordii*) were collected in the area surrounding the Borax I reactor. These samples are referred to as Borax #1 through # 5. In this same year, additional sampling at limited locations was done for evaluation of wild onions (*Allium textile*).

Where possible the biotic samples were co-located with soil samples. Soils were collected from 0 to 15 cm, 15 cm to 60 cm or bedrock, and/or 0 to 60 cm or bedrock. Sagebrush and crested wheatgrass samples were obtained by collecting new growth leafy material from several plants in the general area near the soil sample location and represented composited materials. None of the samples were washed and the mammals were processed whole body including fur, legs, and ears.

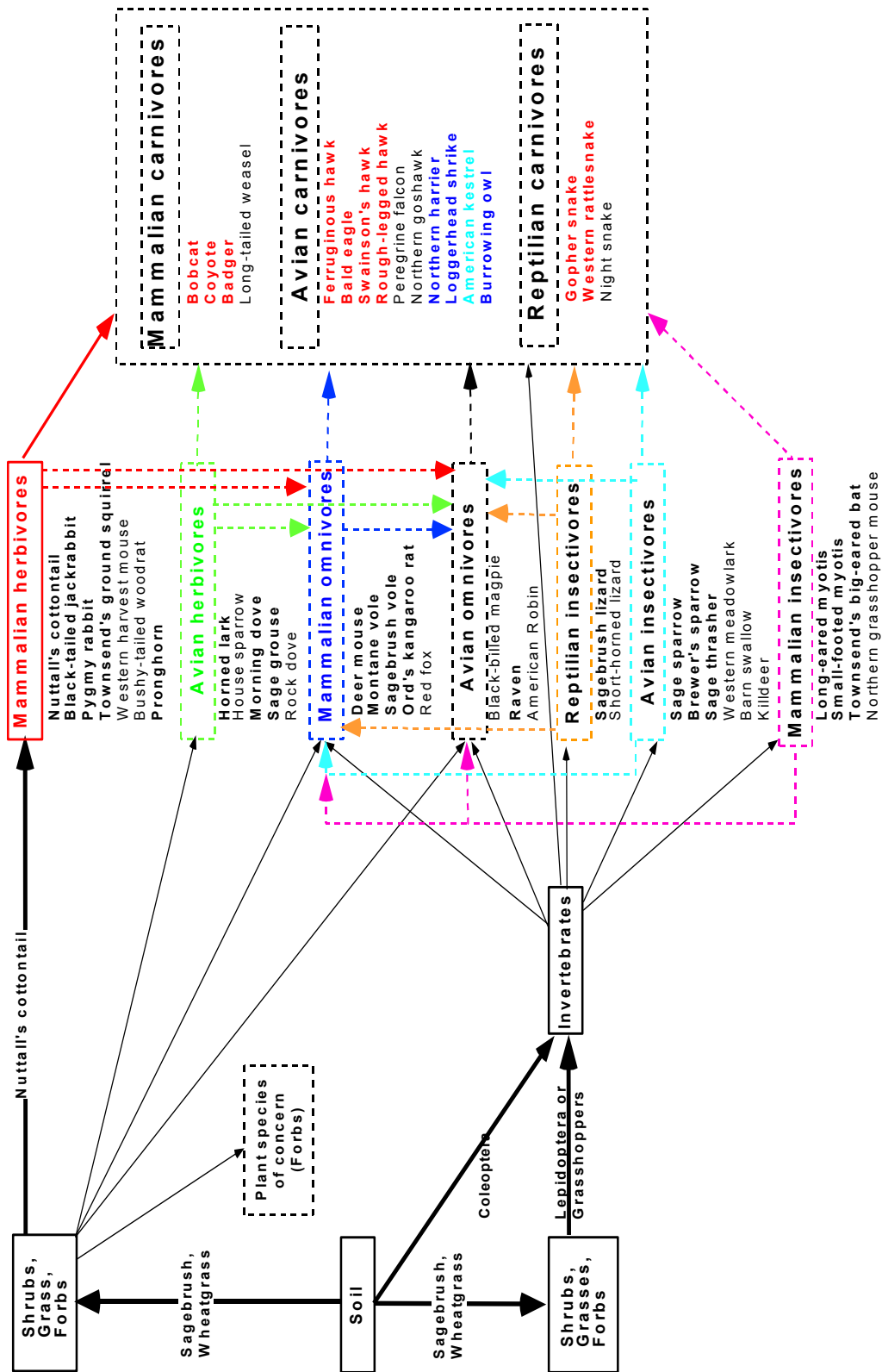


FIG. 1. Section of the INEEL food web selected for characterization [7].



## 4. EVALUATION

Although a complete set of each representative species was collected in the field, due to the low level of radionuclides in the samples, many of the data were designated as non-detects and could not be used in this evaluation. Soil to tissue ratios could be calculated for selected biota for eight out of the fifteen sampling locations for  $^{137}\text{Cs}$ , six out of fifteen for  $^{90}\text{Sr}$ , eight out of fifteen for  $^{235}\text{U}$ , and nine out of fifteen for  $^{238}\text{U}$ .

Transfer factors are generally reported in dry weight to dry weight ratios for plants and wet weight for animal tissues. The sampling results are presented in both dry and wet weight. During the evaluation wet weight and dry weight adjustments were made based on percent solids data specific for the INEEL.

### 4.1. Soil to tissue transfer factors

The site-specific TFs were computed for 6 organisms: beetle, grasshopper, mouse, rabbit, sagebrush and wheatgrass relative to each soil for a range of contaminants. The only contaminants for which ratios are shown are those for having concentrations for both the numerator and the denominator.

The first step was to compute the ratio of measured concentrations of biota to soil individually for each of the five ESA sites and also for each of the five RSA sites locations. Duplicates were averaged prior to computing ratios. The summary of the average soil to tissue concentration ratios by soil depth and the comparison to literature values are presented in Table 1. As shown in Table 1, the amount of data available varied and some values are only represented by one sample.

### 4.2. Foodweb modeling

To model tissue concentrations the approach presented in U.S. Environmental Protection Agency (EPA) was used [8]. This approach is presented for omnivores since they consume both plant and animal material. EPA [8] notes that invertebrates or other animals may not accumulate contaminants at the same rate or to the same level and that the contribution from each type of diet must be summed to obtain total dietary exposure. For risk assessment purposes, the equation used to estimate uptake from ingestion of animal tissue, plant tissue, soil, or sediments, and water was simplified from EPA [8] by assuming that the entire fraction of each media ingested was contaminated. The equation is as follows:

$$C_{OM} = (C_H * TF_{a/m} * FA) + (C_P * TF_{plant} * FP) + (C_{s/sed} * TF_{s/sed} * FS)$$

where:

- $C_{OM}$  is the concentration in omnivore (pCi/g);
- $C_H$  is the concentration herbaceous bird or mammal (pCi/g);
- $TF_{b/m}$  is the transfer factor for bird or mammal (unitless);
- $FA$  is the fraction of diet consisting of animal food items (unitless);
- $C_P$  is the concentration in plants (pCi/g);
- $FP$  is the fraction of diet consisting of plants (unitless);
- $TF_{plant}$  is the transfer factor for soil/sediment to plants (unitless) from Table 1;
- $C_{s/sed}$  is the concentration in soil or bed sediment (pCi/g);
- $TF_{mammal}$  is the transfer factor for soil- or bed sediment-to-omnivore (unitless); and
- $FS$  is the fraction of diet consisting of soil/sediment (unitless).

The ingestion of water is not included for this evaluation. Also, plants and animals were not broken into separate types for modeling due to the uncertainty; thus, there was only one plant component.

TABLE 1. SUMMARY OF SAMPLED SOIL TO TISSUE RATIOS (WET WEIGHT BASIS) COMPARED TO REPORTED LITERATURE TRANSFER FACTORS (TFs) RADIONUCLIDE

Radionuclide/Organism	Average soil concentration to tissue ratio (unitless) <sup>a</sup>			Calculated ratio/reported TF (unitless)		
	0–60 cm	0–15 cm	0.5–60 cm	0–60 cm	0–15 cm	15–60 cm
<sup>137</sup> Cs-Deer mouse	2.0E-01	1.5E-01	1.9E-01	0.25	0.18	0.23
Number of samples	3	3	3			
<sup>137</sup> Cs-Rabbit	1.4E-02	1.1E-01	1.2E-01	0.02	0.14	0.14
Number of samples	1	4	2			
<sup>137</sup> Cs-Kangaroo rat	nc	6.3E-01	3.5E-01	nc	0.78	0.44
Number of samples	0	1	1			
<sup>137</sup> Cs-Sagebrush	nc	4.7E-01	2.6E-01	nc	0.88	0.49
Number of samples	0	1	1			
<sup>90</sup> Sr-Deer mouse	9.0E-01	1.4E-01	2.9E-01	11.20	1.75	3.56
Number of samples	2	2	2			
<sup>90</sup> Sr-Rabbit	6.9E-01	4.3E-01	4.3E-01	8.59	5.33	5.31
Number of samples	2	4	2			
<sup>90</sup> Sr-Kangaroo rat	nc	4.1E-01	1.1E+00	nc	5.07	13.78
Number of samples	0	3	4			
<sup>90</sup> Sr-Grasshopper	6.0E-01	2.0E-01	4.2E-01	7.51	2.49	5.29
Number of samples	1	1	1			
<sup>90</sup> Sr-Onion	1.2E-01	nc	Nc	0.05	nc	nc
Number of samples	2	0	0			
<sup>90</sup> Sr-Sagebrush	nc	1.8E+00	4.4E+00	nc	0.71	1.75
Number of samples	0	2	1			
<sup>235</sup> U-Deer mouse	9.0E-03	1.1E-02	1.0E-02	0.15	0.17	0.17
Number of samples	1	2	2			
<sup>235</sup> U-Rabbit	1.2E-02	1.2E-02	1.3E-02	0.19	0.20	0.22
Number of samples	4	4	4			
<sup>235</sup> U-Kangaroo rat	nc	4.9E-02	5.7E-02	nc	0.79	0.92
Number of samples	0	1	1			
<sup>235</sup> U-Beetles	2.7E-02	3.4E-02	2.9E-02	0.44	0.55	0.46
Number of samples	3	3	3			
<sup>235</sup> U-Onions	4.6E-03	4.8E-03	4.8E-03	0.33	0.34	0.34
Number of samples	5	5	5			
<sup>235</sup> U-Sagebrush	nc	1.5E-01	9.5E-02	nc	10.33	6.81
Number of samples	0	2	2			
<sup>238</sup> U-Deer mouse	7.6E-03	8.0E-03	8.2E-03	0.12	0.13	0.13
Number of samples	1	6	6			
<sup>238</sup> U-Rabbit	8.0E-03	4.8E-03	5.1E-03	0.13	0.08	0.08
Number of samples	4	8	8			
<sup>238</sup> U-Kangaroo rat	nc	6.5E-03	6.9E-03	nc	0.10	0.11
Number of samples	0	5	5			
<sup>238</sup> U-Beetles	2.6E-02	2.5E-02	2.7E-02	0.42	0.40	0.43
Number of samples	3	3	3			
<sup>238</sup> U-Onions	2.0E-02	1.8E-02	2.1E-02	1.44	1.30	1.46
Number of samples	5	5	5			
<sup>238</sup> U-Sagebrush	nc	1.6E-02	1.7E-02	nc	1.12	1.22
Number of samples	0	3	3			

<sup>a</sup> nc indicates that a value was not calculated.

## 5. RESULTS

Much of the data collected was not usable in this assessment due to the large number of non-detections. Due to this lack of data no summary statistics were generated and no distribution testing was performed. For this reason, the results are presented for information and have only been qualitatively evaluated. Consequently, it is difficult to arrive at conclusive results. However, the data that are available can be used to generally compare literature TFs used to those measured, to evaluate model accuracy, and finally provide direction for further sampling.

Three evaluations were performed. First the sampled soil was compared to tissue ratios to literature value TFs (by soil depth). Second the tissue concentrations were compared to the modeled tissue concentrations (by soil depth). Last, the soil concentrations were compared to the tissue concentrations (for 0–15 cm soil only).

## 5.1. Ratio comparisons

The first evaluation is presented in Table 1. This table presents the average of the sample ratio across the sampling locations by soil depth. The calculated sample ratio is divided by the reported literature TF for comparison. If the calculated ratio is less than the reported literature value (ratio is less than 1.0), the literature value should produce higher estimates of concentration in the tissue and are considered conservative in the risk assessment modeling. However, if the value is too much greater it over-estimates risk and possibly results in an unnecessary cleanup. The lowest value of 0.02 in Table 1 is for the rabbit at 0 through 60 cm. In this situation the use of the literature derived TF may possibly over-estimate the risk fifty times.

For  $^{137}\text{Cs}$ , deer mice (3 samples), rabbits (1–4 samples), kangaroo rats (1 sample), and sagebrush (1 sample) could be evaluated. In all cases, the calculated sample ratio divided by the TF yielded values below 1.0, indicating that the calculated sample ratio are less than the literature value and therefore conservative.

The comparison of sampled soil to tissue ratios to literature value TFs for  $^{90}\text{Sr}$  was calculated for deer mice (2), rabbits (2–4), kangaroo rats (3–4), grasshoppers (1), onions (0–2) and sagebrush (1–2). Calculated ratios for soil to small mammals were higher than literature value TFs. The calculated ratios range from 1.7 to 13.8 times greater than the TF. In this instance, use of the literature value may result in a large under-estimation of risk. All but 1 of 3 calculated ratios for soil to plants were lower than literature value TFs used in the risk assessment. The calculated ratios for soil to plant range from 0.05 to 1.7 times the TF. This indicates that although the risk to small mammals may be underestimated the risk to plants would be more adequately estimated. The INEEL selected 2.5 as the TF for  $^{90}\text{Sr}$  for soil to plant tissue. If as is commonly done, a lower number was selected from the literature, it would be possible to under-estimate this tissue concentration.

The comparison of sampled soil to tissue ratios to literature value TFs for  $^{235}\text{U}$  was calculated for deer mice (1–2), rabbits (4), kangaroo rats (1), beetles (3), onions (5) and sagebrush (0–2). The calculated ratios for soil to small mammals were lower than literature value TFs used in the risk assessment (may result in over-estimation of risk) ranging from 0.15 to 0.79 times less. The calculated ratios for soil to onions were lower than literature value TFs used in the risk assessment (0.34) while the calculated ratios for soil to sagebrush were higher than literature TFs (6.8 to 10.3).

The comparison of sampled soil to tissue ratios to literature value TFs for  $^{238}\text{U}$  was calculated for deer mice (1–6), rabbits (4–8), kangaroo rats (0–5), beetles (3), onions (5) and sagebrush (0–3) were evaluated. The calculated ratios for soil to small mammals were lower than literature value TFs used in the risk assessment (may result in over-estimation of risk) and ranged from 0.08 to 0.43 times less. The calculated ratios for soil to plants ranged from 1.1 to 1.4 times greater than the TF.

## 5.2. Tissue concentration comparisons

Comparisons of measured tissue concentrations to modeled tissue concentrations (by soil depth) for all radionuclides are presented in Table 2. For  $^{137}\text{Cs}$ , the results are consistent with the previous evaluation with most sampled tissue concentrations less than that modeled. In this case 6 out of the 21 calculated were higher (1.2 to 2.2), 1 was greatly less than the value (0.04 to 0.9) (rabbit), and 9 out of 21 were in good agreement (0.7 to 1.3).

Based on the previous evaluation, use of the literature value for feed to meat transfer for  $^{90}\text{Sr}$  could greatly under-estimate tissue concentrations while the soil to plant appeared to be in fairly good agreement. Despite this, the modeled tissue concentrations are in fairly good agreement with the measured values. Only 3 out of the 26 calculated were higher (1.3 to 1.8), 1 was greatly less than the value (0.04 to 0.9) (rabbit), and 5 out of 26 were in good agreement (0.7 to 1.3).

Based on the previous evaluation, the literature values for feed to meat transfer for  $^{235}\text{U}$  could over-estimate and the soil to plant could either over- or under-estimate the tissue concentrations. The result indicate that 37 out of the 47 calculated were higher (1.1 to 12.0 for sagebrush) and 13 out of 47 were in good agreement (0.7 to 1.2)

Based on the previous evaluation, the literature value for feed to meat transfer for  $^{238}\text{U}$  could over-estimate and the soil to plant could under-estimate the tissue concentrations. The results indicate that 42 out of the 73 calculated were higher (1.1 to 7.6 for onions) and that 17 out of 73 were in good agreement (0.7 to 1.3).

TABLE 2. COMPARISON MODELED TISSUE CONCENTRATIONS TO SAMPLED TISSUE CONCENTRATIONS

	Concentration (pCi/g)	Sample	Modeled tissue concentrations by soil depth (unitless)			Sampled tissue concentration/modeled tissue concentration (unitless)		
			0-60 cm	0-15 cm	15-30 cm	0-60 cm	0-15 cm	15-30 cm
<b><sup>137</sup>Cs</b>								
ESA-#1	3.90E-01	Deermouse (ww basis)	4.46E-01	5.95E-01	3.44E-01	0.9	0.7	1.1
	4.10E-02	Rabbit (ww basis)	7.90E-01	1.05E+00	6.09E-01	0.1	0.04	0.1
ESA-#2	8.00E-02	Deermouse (ww basis)	1.10E-01	3.42E-01	6.35E-02	0.7	0.2	1.3
ESA-#3	4.00E-02	Deermouse (ww basis)	1.83E-02	2.31E-02	4.23E-02	2.2	1.7	0.9
RSA - #1	1.20E-01	Deermouse (ww basis)		3.49E-02			3.4	
RSA - #2	3.20E-02	Rabbit (ww basis)		3.87E-02			0.8	
Borax-#1	2.70E-02	Rabbit (ww basis)		2.67E-01	3.81E-02		0.1	0.7
Borax-#3	6.00E-02	Rabbit (ww basis)		1.08E-01			0.6	
Borax-#4	4.50E-01	Kangaroo rat (ww basis)		2.14E-01	3.81E-01		2.1	1.2
	3.63E-01	Sagebrush (dw basis)		4.14E-01	7.37E-01		0.9	0.5
<b><sup>90</sup>Sr</b>								
ESA-#1	7.21E-01	Deermouse (ww basis)	6.96E-01	2.09E+00	9.88E-01	1.0	0.3	0.7
	1.63E-01	Rabbit (ww basis)	1.29E+00	3.88E+00	1.83E+00	0.1	0.04	0.1
	5.80E-01	Grasshopper (ww basis)	1.29E+00	3.88E+00	1.83E+00	0.5	0.1	0.3
ESA-#2	2.40E-01	Onions (ww basis)	6.46E-01			0.4		
	2.80E-01	Onions (ww basis)	6.46E-01			0.4		
	4.97E-01	Deermouse (ww basis)	3.43E-01			1.5		
	3.45E-01	Rabbit (ww basis)	6.34E-01			0.5		
Borax-#1	1.58E-01	Kangaroo rat (ww basis)		5.99E-01			0.3	
	5.41E-01	Rabbit (ww basis)		5.99E-01			0.9	
	1.61E-01	Sagebrush (dw basis)						
Borax-#2	2.34E-01	Kangaroo rat (ww basis)		1.24E+00			0.2	
	2.47E-01	Rabbit (ww basis)		1.24E+00			0.2	
	3.47E-01	Sagebrush (dw basis)		2.53E+00			0.1	
Borax-#4	3.60E-02	Deermouse (ww basis)						
	9.21E-01	Kangaroo rat (ww basis)		1.52E+00	1.11E+00		0.6	0.8
	6.10E-01	Rabbit (ww basis)		1.52E+00	1.11E+00		0.4	0.5
	3.98E+00	Sagebrush (dw basis)		3.10E+00	2.27E+00		1.3	1.8
Borax-#5	3.59E-02	Kangaroo rat (ww basis)		3.03E-01			0.1	
	1.93E-01	Rabbit (ww basis)		3.03E-01			0.6	
<b><sup>235</sup>U</b>								
ESA-#2	3.90E-03	Onions (ww basis)	6.45E-03	6.71E-03	5.26E-03	0.6	0.6	0.7
	5.00E-03	Onions (ww basis)	6.45E-03	6.71E-03	5.26E-03	0.8	0.7	0.9
	1.94E-02	Rabbit (ww basis)	7.38E-03	7.67E-03	6.02E-03	2.6	2.5	3.2
ESA-#3	6.82E-03	Rabbit (ww basis)	8.40E-03	6.05E-03	6.43E-03	0.8	1.1	1.1
RSA-#2	1.90E-02	Beetle (ww basis)	3.70E-03	2.19E-03	3.05E-03	5.1	8.7	6.2
RSA-#4	1.00E-02	Onions (ww basis)	5.11E-03	4.84E-03	4.94E-03	2.0	2.1	2.0
	4.00E-02	Onions (ww basis)	5.11E-03	4.84E-03	4.94E-03	7.8	8.3	8.1
	6.05E-03	Deermouse (ww basis)	3.49E-03	3.31E-03	3.38E-03	1.7	1.8	1.8
	6.40E-03	Rabbit (ww basis)	5.84E-03	5.53E-03	5.65E-03	1.1	1.2	1.1
	1.80E-02	Beetle (ww basis)	3.49E-03	3.31E-03	3.38E-03	5.2	5.4	5.3
RSA-#5	1.32E-02	Onions (ww basis)	4.86E-03	5.31E-03	5.07E-03	2.7	2.5	2.6
	4.65E-03	Rabbit (ww basis)	5.58E-03	5.55E-03	6.07E-03	0.8	0.8	0.8
	1.80E-02	Beetle (ww basis)	3.33E-03	3.32E-03	3.63E-03	5.4	5.4	5.0
Borax-#2	1.48E-03	Deermouse (ww basis)		6.65E-04	5.93E-04		2.2	2.5
Borax-#4	5.96E-03	Kangaroo rat (ww basis)		7.15E-04	1.06E-03		8.3	5.6
	1.61E-02	Sagebrush (dw basis)		1.85E-03	1.60E-03		8.7	10.1
Borax-#5	1.21E-02	Sagebrush (dw basis)		1.01E-03	3.42E-03		12.0	3.5
<b><sup>238</sup>U</b>								
ESA - #2	4.60E-03	Onions (ww basis)	6.12E-03	6.11E-03	5.59E-03	0.8	0.8	0.8
	3.90E-03	Onions (ww basis)	6.12E-03	6.11E-03	5.59E-03	0.6	0.6	0.7
	1.57E-02	Rabbit (ww basis)	6.99E-03	6.98E-03	6.38E-03	2.2	2.2	2.5
ESA - #3	4.40E-03	Rabbit (ww basis)	8.56E-03	6.62E-03	6.90E-03	0.5	0.7	0.6
RSA - #2	1.60E-02	Beetle (ww basis)	3.31E-03	3.31E-03	2.98E-03	4.8	4.8	5.4
RSA - #4	1.02E-02	Onions (ww basis)	4.98E-03	5.61E-03	4.85E-03	2.0	1.8	2.1
	3.70E-02	Onions (ww basis)	4.98E-03	5.61E-03	4.85E-03	7.4	6.6	7.6
	4.96E-03	Deermouse (ww basis)	3.41E-03	3.84E-03	3.31E-03	1.5	1.3	1.5
	3.10E-03	Rabbit (ww basis)	5.70E-03	6.42E-03	5.54E-03	0.5	0.5	0.6
	1.50E-02	Beetle (ww basis)	3.41E-03	3.84E-03	3.31E-03	4.4	3.9	4.5

Blanks indicate that no value could be computed.

TABLE 2. (CONTINUED)

	Concentration (pCi/g)	Sample	Modeled tissue concentrations by soil depth (unitless)			Sampled tissue concentration/modeled tissue concentration (unitless)		
			0–60 cm	0–15 cm	15–30 cm	0–60 cm	0–15 cm	15–30 cm
RSA – #5	1.24E-02	Onions (ww basis)	5.28E-03	5.68E-03	5.66E-03	2.3	2.2	2.2
	2.21E-03	Rabbit (ww basis)	6.04E-03	6.49E-03	6.46E-03	0.4	0.3	0.3
	2.10E-02	Beetle (ww basis)	3.61E-03	3.88E-03	3.87E-03	5.8	5.4	5.4
Borax-#1	6.02E-03	Deermouse (ww basis)		4.97E-03	4.48E-03		1.2	1.3
	1.34E-02	Kangaroo rat (ww basis)		8.32E-03	7.50E-03		1.6	1.8
	1.34E-03	Rabbit (ww basis)		8.32E-03	7.50E-03		0.2	0.2
	1.77E-02	Sagebrush (dw basis)		1.46E-02	1.31E-02		1.2	1.3
Borax-#2	1.06E-02	Deermouse (ww basis)		4.74E-03	5.88E-03		2.2	1.8
	3.42E-03	Kangaroo rat (ww basis)		7.92E-03	9.84E-03		0.4	0.3
	1.12E-03	Rabbit (ww basis)		7.92E-03	9.84E-03		0.1	0.1
Borax-#3	7.88E-03	Deermouse (ww basis)		5.07E-03	4.71E-03		1.6	1.7
	5.28E-03	Kangaroo rat (ww basis)		8.48E-03	7.87E-03		0.6	0.7
	1.65E-02	Sagebrush (dw basis)		1.48E-02	1.38E-02		1.1	1.2
Borax-#4	7.34E-03	Deermouse (ww basis)		5.36E-03	4.93E-03		1.4	1.5
	5.99E-03	Kangaroo rat (ww basis)		8.96E-03	8.24E-03		0.7	0.7
	2.16E-03	Rabbit (ww basis)		8.96E-03	8.24E-03		0.2	0.3
	1.61E-02	Sagebrush (dw basis)		1.57E-02	1.44E-02		1.0	1.1
Borax-#5	7.91E-03	Deermouse (ww basis)		4.97E-03	4.18E-03		1.6	1.9
	3.37E-03	Kangaroo rat (ww basis)		8.32E-03	8.08E-03		0.4	0.4
	9.98E-04	Rabbit (ww basis)		1.46E-02	1.41E-02		0.1	0.1

Blanks indicate that no value could be computed.

### 5.3. Soil to tissue concentration comparison

The comparisons of soil concentration to tissue concentration can indicate whether the uptake of these radionuclides into the tissue is dependent on the concentration in the source. For risk assessment purposes, the use of an uptake factor that is dependent on the soil concentration can again result in either an over- or under-estimation of the risk. If the uptake factor declines as the concentration increase, then the risk may be over-estimated. If the uptake factor increases as the concentration increases, then the risk may be under-estimated. Inspecting graphical output provide some indication of the trends but is not a rigorous test due to the limited range of soil concentrations sampled.

If uptake is independent of soil concentration then as the soil concentration increases the tissue concentration has a corresponding increase and this can be reflected graphically. Figures 2 through 5 present the soil concentrations for the 0 through 15 cm depth, as compared to the tissue concentrations (not adjusted for dry or wet weight) with associated trend lines. When the ratio of uptake from soil to tissue is not dependent on the soil concentration then the line should steadily increase with a slope of 1 across the x-axis. When the trend line is horizontal across the x-axis then the ratio of uptake is dependent on soil concentration and is decreasing. When the trend line is vertical then the ratio of uptake is again dependent of the soil concentration and is increasing.

The limited data makes this a more difficult evaluation. However, graphically, it appears that the concentration in deer mice for all four radionuclides is not correlated with soil concentration. Figures 2 through 5 appear to indicate that deer mice tissue concentration increase relative to an increase to soil concentration and that the TF would remain the same at least throughout the range of concentration sampled. This is an important consideration in the selection of a monitoring species. This may also be true of kangaroo rat for  $^{90}\text{Sr}$  but not for  $^{238}\text{U}$ . Again, beetles, sagebrush and onions have limited data and apparent trends seen graphically should be suspect.

## 6. CONCLUSIONS

Primarily this evaluation has re-emphasized the usefulness of site-specific biotic data in the risk assessment process. The use of literature derived TFs to assess exposure may either under-estimate or over-estimate the risk to non-human receptors. This significant uncertainty in the risk assessment process can be greatly reduced by obtaining site-specific information. It is also important to be aware of and evaluate uptake factor dependency on species and/or soil concentration. Risk managers need to balance the cost of sampling against the cost of an unnecessary remediation (both in fiscal and environmental terms) or the cost of not identifying a potential risk.

The INEEL is currently evaluating long-term monitoring based on the results of the ecological risk assessment (DOE-ID 2001). The results of this study will be used to direct additional sampling performed as part of this activity.

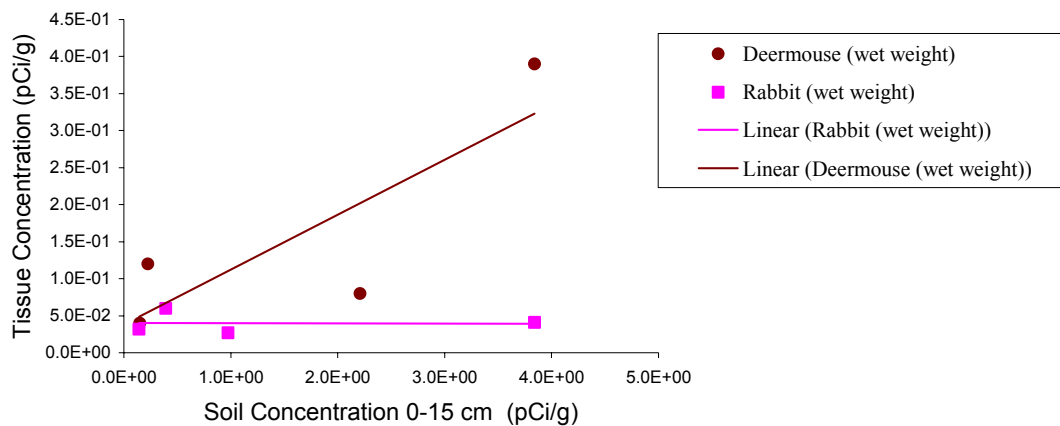


FIG. 2. Comparison of C-137 concentrations in soil to tissue.

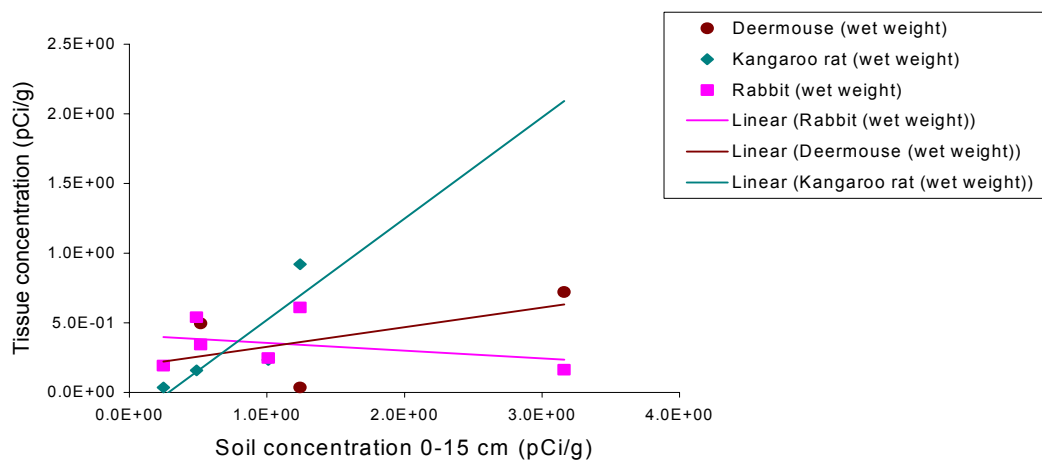


FIG. 3. Comparison of Sr-90 concentrations in soil to tissue.

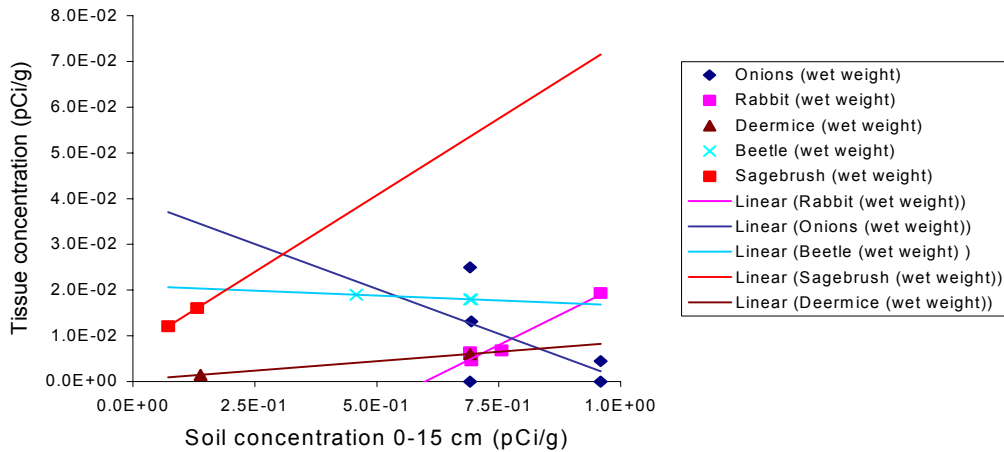


FIG. 4. Comparison of U-235 concentrations in soil to tissue.

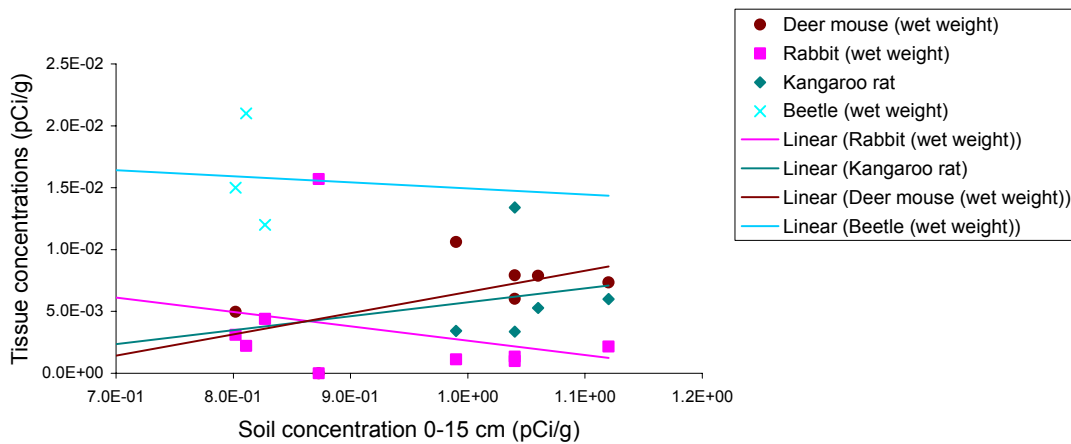


FIG. 5. Comparison of U-238 concentrations in soil to tissue.

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# Application of RAD-BCG calculator to Hanford's 300 area shoreline characterization dataset

**E.J. Antonio, T.M. Poston, B.L. Tiller, G.W. Patton**

Pacific Northwest National Laboratory, Richland, WA, United States of America

**Abstract.** In 2001, a multi-agency study was conducted to characterize potential environmental effects from radiological and chemical contaminants on the near-shore environment of the Columbia River at the 300 Area of the U.S. Department of Energy's Hanford Site. Historically, the 300 Area was the location of nuclear fuel fabrication and was the main location for research and development activities from the 1940s until the late 1980s. During past waste handling practices uranium, copper, and other heavy metals were routed to liquid waste streams and ponds near the Columbia River shoreline. The Washington State Department of Health and the Pacific Northwest National Laboratory's Surface Environmental Surveillance Project sampled various environmental components including river water, riverbank spring water, sediment, fishes, crustaceans, bivalve mollusks, aquatic insects, riparian vegetation, small mammals, and terrestrial invertebrates for analyses of radiological and chemical constituents. The radiological analysis results for water and sediment were used as initial input into the RAD-BCG Calculator. The RAD-BCG Calculator, a computer program that uses an Excel® spreadsheet and Visual Basic® software, showed that maximum radionuclide concentrations measured in water and sediment were lower than the initial screening criteria for concentrations to produce dose rates at existing or proposed limits. Radionuclide concentrations measured in biota samples were used to calculate site-specific bioaccumulation coefficients ( $B_{iv}$ ) to test the utility of the RAD-BCG-Calculator's site-specific screening phase. To further evaluate site-specific effects, the default Relative Biological Effect (RBE) for internal alpha particle emissions was reduced by half and the program's kinetic/allometric calculation approach was initiated. The subsequent calculations showed the initial RAD-BCG Calculator results to be conservative, which is appropriate for screening purposes.

## 1. INTRODUCTION

The 300 Area of the Hanford Site (Figure 1) is located just north of the city of Richland, Washington. This area borders the Columbia River and covers 1.5 km<sup>2</sup>. From the 1940s, most of the research and development for the U.S. Department of Energy's Hanford Site was conducted in the 300 Area. In addition, the 300 Area was used to produce nuclear fuel elements for the Hanford reactors. Metallic uranium was extruded into pipe-like cylinders and encapsulated with aluminum or zirconium cladding to produce nuclear fuel rods. This process resulted in substantial amounts of uranium and heavy metals, such as copper, in the 300 Area liquid waste streams. Initially, liquid waste from the research facility and fuel production was routed to waste ponds in the northern part of the 300 Area that were located near the Columbia River shoreline. Later in the fuel production period, the liquid waste was sent to process trenches in the northern part of the 300 Area. At the present time, all liquid waste from the 300 Area is treated at the 300 Area Treated Effluent Disposal Facility and released to the Columbia River under the requirements of a National Pollutant Discharge Elimination System permit.

The study was conducted in late August to October 2001 to coincide with expected low river stage. Low river stage facilitates locating riverbank springs and collecting riverbank spring water samples along the Columbia River shoreline. A number of contaminants are present in groundwater at the 300 Area [1] and the near-shore environment can be exposed through riverbank springs and groundwater upwelling. Therefore, the sampling locations selected for this study were centered near historic riverbank spring discharges and the contaminants of concern were primarily known groundwater contaminants (i.e., radionuclides).

## 2. METHODS

This section describes methods used to sample water, sediment, and various biotic components of the ecosystem. It also briefly describes the screening and radiological dose calculations performed.

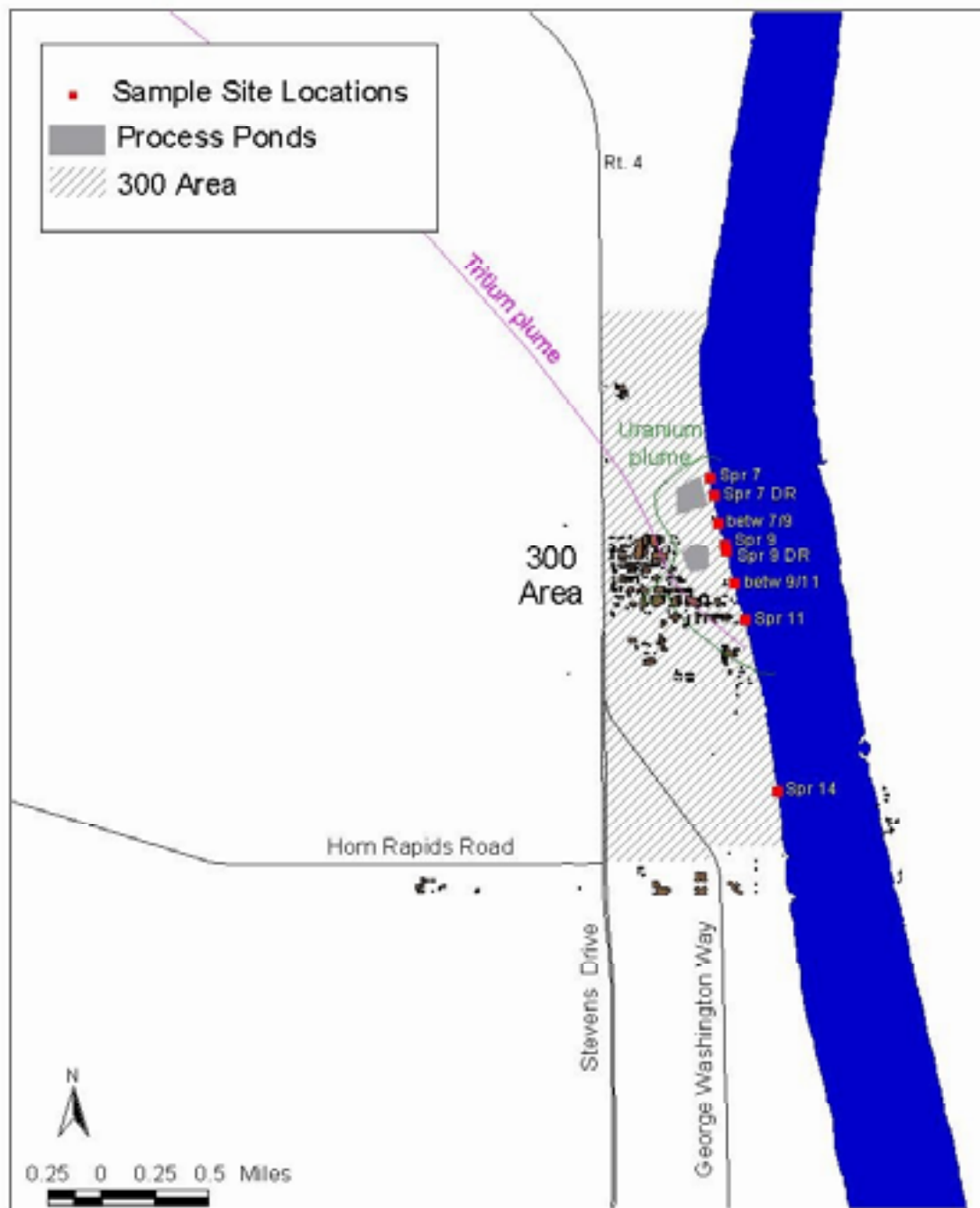


FIG. 1. Sampling locations along the 300 area shoreline.

### 3. WATER AND SEDIMENT SAMPLING

Near-shore river water samples were collected from near the river bottom by using a peristaltic pump and Tygon® tubing with the sample inlet positioned <6 cm above the river bottom. The samples were collected at the major riverbank spring locations (locations #7, #9, #11, and #14 on Figure 1); at downstream locations approximately 60 to 130 m below locations #7, #9, and #11 (e.g., designated “7DR”); and at locations halfway between locations #7 and #9 and halfway between locations #9 and #11 (e.g., designated “betw 9/11”). At each location, four unfiltered river water samples were collected with samples taken at the immediate shoreline (0.25 m depth), and offshore where the river depth was 0.5 m, 1 m, and 1.5 m.

Riverbank spring water samples were collected at locations #7, #9, and Vernita Bridge (considered the background location) using either a hand pump or a peristaltic pump. All samples were unfiltered water, except for samples for metals analysis where both unfiltered and filtered samples were collected (0.45 µm Geotech high volume filter). No riverbank springs were observed at locations #11 and #14.

Sediment samples at the riverbank spring locations and at the background site were collected at each of the major riverbank spring locations and the background site using nylon ladles.

#### 3.1. Biota samples

Riparian vegetation samples (new growth only) of the perennial plant white sweet clover (*Melilotus alba*) and leaves and stems from mulberry trees (*Morus alba*) were cut with stainless steel scissors; samples were placed in glass jars for metals analyses or plastic bags for radiological analyses.

Prickly sculpin (*Cottis asper*) and crayfish (*Pacifasticus leniusculus*) were collected along the near-shore (less than 0.5 m deep) and within 10 m of the spring sites. Sculpin were collected with the use of a Smith-Root Type IV backpack electrofisher and crayfish were netted by hand. Samples were placed in cleaned glass containers, labeled, and stored in ice-filled coolers until the samples were processed. The hepatopancreas was removed from each crayfish, weighed, and split for individual analyses of metals. The metals analysis included uranium.

Asiatic clams (*Corbicula* sp.) were collected concurrently with water samples at all four spring sites, at two down-river locations below locations #7 and #9, and at the reference site above Vernita bridge (i.e., 0.0 [in the riverbank spring], 0.25, 0.5, 1.0, 1.5 m water depths at each site).

Macrophytic vegetation (submerged aquatic vegetation) samples were collected by hand and generally consisted of milfoil (*Myriophyllum spicatum*). Samples obtained for radiological analyses required large (>600 g) quantities of the media and may have included elodea (*Elodea* sp.) and potamogeton (*Potamogeton* sp.).

Adult mayflies (*Ephemeroptera*) and darkling beetles (*Eleoides* sp.) samples were hand picked at each location within 50 m of each spring site. Adult mayfly samples were rinsed in deionized water because they were obtained along the water's edge and were covered in dirt particles. All samples were placed directly into the individual sample containers and labeled and stored for shipment to the analytical labs.

House mice (*Mus musculus*) were chosen to represent the small mammal species because they are highly dependent on the riparian habitat for open water and succulent foods. Animals were collected with the use of pre-cleaned Sherman live traps baited with peanut butter. Whole body weight, length, sex, age, reproductive status, and target organ weights of each individual specimen were measured and recorded.

##### 3.1.1. Contaminant analysis

When sufficient sample mass was available, radiochemical analyses were performed. For small biota samples, inductively coupled plasma mass spectrometry (ICP Mass Spec.) was used to obtain uranium concentrations.

### **3.2. Evaluation using general screening phase**

The U.S. Department of Energy, through its Biota Dose Assessment Committee (BDAC) has developed screening and analysis methods within a graded approach to biota dose evaluation for screening radionuclide concentrations in water, sediment, or soil against existing or currently proposed biota radiological standards [2]. The standard for aquatic animals and is 10 mGy/d [3]. Proposed standards for terrestrial plants is 10 mGy/d, and, the proposed standard for terrestrial animals is 1 mGy/d [2]. Media sampled for this analysis are Columbia River water, riverbank spring water, and sediment collected at riverbank spring locations. In this discussion, this method will be referred to as the graded approach method.

Maximum radionuclide concentrations reported for river water, riverbank spring water and sediment were used for graded approach general screening phase assessments using the RAD-BCG Calculator. The initial screen was based on those samples analyzed by radiochemical techniques. If data was not available for sediment, the sediment concentrations were derived with generic distribution coefficients by the program. Likewise, if a radionuclide was not identified in water, but was identified in sediment, generic distribution coefficients were used to generate data. Maximum measured and derived concentrations in water and sediment were compared to biota concentration guides (BCGs) with the RAD-BCG Calculator. The BCG is a radionuclide concentration in either water or sediment that would result in a modeled dose rate of 10 mGy/d in aquatic organisms or terrestrial plants, or 1 mGy/d for terrestrial or riparian animals. A fraction is generated by dividing the water or sediment concentration by the BCG value for each radionuclide. By summing the fractions for each radionuclide from a site, a sum of fractions value is calculated. A sum of fractions exceeding 1.0 indicates the potential for the dose rate to exceed the existing or proposed dose rate limits of 10 mGy/d or 0.1 mGy/d. The graded approach method also allows in subsequent analyses more definitive assessments based on species- and site-specific considerations.

### **3.3. Calculations using site-specific screening phase**

Site-specific screening and assessment calculations were performed to test the utility of the various phases of the RAD-BCG Calculator. The radionuclide/media combination producing the largest contribution to the sum of fractions is identified and available data were used to calculate site-specific parameters for use in the RAD-BCG Calculator. The ICP Mass Spec. data for uranium in aquatic biota were converted to radioactivity concentrations assuming a natural distribution of uranium isotopes. Site-specific bioaccumulation coefficients were calculated and entered into the RAD-BCG Calculator, which generated site-specific BCGs for uranium isotopes.

### **3.4. Evaluation using site-specific analyses phase**

To test the utility of the site-specific assessment phase, the alpha Relative Biological Effect (RBE) factor for aquatic animals was reduced by half. New site-specific BCGs for alpha emitting radionuclides were calculated by the RAD-BCG Calculator. To further test the site-specific assessment phase, the kinetic/allometric approach built into the RAD-BCG Calculator was initiated. This was accomplished with a 'click' of the computer's mouse in the upper left hand portion of the "Riparian Animal" input page in the RAD-BCG Calculator. This 'click' of the mouse turn off the use of 'lumped parameters' and then employs a basic first order kinetic box model approach. Parameters such as food and sediment intake rates, maximum life span, inhalation and sediment inhalation rates and water consumption rate are estimated based on riparian animal body mass. The program's default riparian animal is an 8.8 kg raccoon.

## **4. RESULTS AND DISCUSSION**

### **4.1. Water and sediments**

The screening assessment was based on maximum radionuclide concentrations measured in either water or sediment (Table 1).

TABLE 1. MAXIMUM RADIONUCLIDE CONCENTRATIONS MEASURED IN WATER AND SEDIMENT

Radionuclide	Water Minimum Detection Limit	Water (Bq m <sup>-3</sup> )	Sediment Minimum Detection Limit	Sediment (Bq kg <sup>-1</sup> )
H-3	370	3.1E+05		
Co-60	222		1.1	4.8E-01
Sr-90	22.2	7.5E+00	1.9	9.6E-01
Tc-99	37	5.6E+02	37	
I-129	37	1.5E-01	37	
Cs-137	370		1.1	8.5E+00
Th-232	2.22	3.2E+00	0.75	5.6E+01
U-234	2.22	2.0E+03	0.75	1.0E+02
U-235	2.22	8.3E+01	0.75	3.8E+00
U-238	2.22	1.8E+03	0.75	9.1E+01

Although measured concentrations of cobalt-60 and strontium-90 in sediments were below contractual detection limits, the errors associated with those measurements were less than the reported value. So, for completeness, they are included in the analyses.

#### 4.2. Biota

Maximum radionuclide concentrations measured in biota samples were used for subsequent site-specific screening and site-specific analyses (Table 2).

TABLE 2. MAXIMUM RADIONUCLIDE CONCENTRATIONS MEASURED IN SELECTED BIOTA SAMPLES

Biota Media	Sr-90 Bq kg <sup>-1</sup>	Tc-99 Bq kg <sup>-1</sup>	Cs-137 Bq kg <sup>-1</sup>	Uranium µg g <sup>-1</sup>
Detection Limits	1.5	5.6	1.5	0.01
Riparian Community				
Sweet Clover	6.7	12.2	10.0	0.12
Mulberry Leaves	6.3	242.0	1.1	0.12
Small Mammal		0.4	1.5	0.02
Aquatic Community				
Milfoil	3.3	11.5	4.4	9.29
Clam				6.77
Crayfish	10.0	4.4	3.0	7.81
Sculpin	0.7	1.5	-0.4	0.06

#### 4.3. General screening phase evaluation

The total sum of fractions based on maximum water and sediment concentrations was 0.50 (Table 3). The relative dose contribution from the water pathway was roughly a factor of 200 greater than the sediment pathway.

This total sum of fraction is the result of the RAD-BCG Calculator screening and indicates that regardless of where the water and sediment samples were collected for this study, the maximum measured radionuclide concentrations for this characterization effort were insufficient to exceed the concentrations necessary to produce dose rates exceeding current or proposed limits. Uranium was the major contributor to radiological dose for both water and sediment pathways. The results did not exceed the screening value and the site passed this initial screen.

TABLE 3. INITIAL 300 AREA SHORELINE STUDY SCREENING ASSESSMENT BASED ON THE RAD-BCG CALCULATOR SUMMATION OF PARTIAL FRACTIONS

Nuclide	Water Limit (Bq m <sup>-3</sup> )	Water Partial Fraction	Sediment Limit (Bq kg <sup>-1</sup> )	Sediment Partial Fraction	Combined Sum of Fractions
H-3 <sup>a</sup>	1E+10	3.2E-05	1E+07	2.2E-08	3.2E-05
Co-60 <sup>b</sup>	1E+05	3.5E-06	5E+04	8.9E-06	1.2E-05
Sr-90	1E+04	7.3E-04	2E+04	4.5E-05	7.8E-04
Tc-99 <sup>a</sup>	2E+07	2.3E-05	2E+06	1.8E-06	2.3E-05
I-129 <sup>a</sup>	1E+06	1.0E-07	1E+06	1.4E-09	1.1E-07
Cs-137 <sup>b</sup>	2E+03	1.1E-02	1E+05	7.4E-05	1.1E-02
Th-232	1E+04	2.9E-04	5E+04	1.2E-03	1.4E-03
U-234	7E+03	2.6E-01	2E+05	5.1E-04	2.7E-01
U-235	8E+03	1.0E-02	1E+05	2.7E-05	1.0E-02
U-238	8E+03	2.1E-01	9E+04	9.9E-04	2.1E-01
Total		5.1E-01		2.8E-03	5.0E-01

<sup>a</sup> Denotes nuclide only identified in water sample, sediment value generated by program default distribution coefficient.

<sup>b</sup> Denotes nuclide only identified in sediment, water value generated by program default distribution coefficient.

## 5. EVALUATION RESULTS FROM SITE-SPECIFIC SCREEN PHRASE

Although, the maximum measured water and sediment data collected passed the initial screen, site-specific screening and assessment calculations were performed to test the utility of the various phases of the RAD-BCG Calculator. The ICP mass spectral results for uranium in aquatic biota were converted to radioactivity concentrations assuming a natural distribution of uranium isotopes. Site-specific bioaccumulation coefficients for uranium isotopes in water were calculated. These coefficients were entered into the RAD-BCG Calculator and revised site-specific BCGs for uranium isotopes were calculated (Table 4).

The total sum of fractions (1.6E-01) for the site-specific screen was 68% lower than the initial screen results indicating a lower dose rate to biota when site-specific parameters were employed. The biggest contribution to the total sum of fraction was still uranium isotopes in water, but the limiting organism changed to a riparian animal when site-specific bioaccumulation coefficients for aquatic animals were employed. The results of this site-specific screen indicate the dose rate to biota were below current or proposed radiological dose limits.

TABLE 4. THE 300 AREA SHORELINE STUDY SITE-SPECIFIC SCREENING ASSESSMENT BASED ON THE RAD-BCG CALCULATOR SUMMATION OF PARTIAL FRACTIONS USING SITE-SPECIFIC AQUATIC ANIMAL B<sub>FS</sub> FOR URANIUM IN WATER

Nuclide	Water Limit (Bq m <sup>-3</sup> )	Water Partial Fraction	Sediment Limit (Bq kg <sup>-1</sup> )	Sediment Partial Fraction	Combined Sum of Fractions
H-3 <sup>a</sup>	1E+10	3.2E-05	1E+07	2.2E-08	3.2E-05
Co-60 <sup>b</sup>	1E+05	3.4E-06	5E+04	8.9E-06	1.2E-05
Sr-90	1E+04	7.3E-04	2E+04	4.5E-05	7.7E-04
Tc-99 <sup>a</sup>	2E+07	2.3E-05	2E+06	1.8E-06	2.5E-05
I-129 <sup>a</sup>	1E+06	1.1E-07	1E+06	1.4E-09	1.1E-07
Cs-137 <sup>b</sup>	2E+03	1.1E-02	1E+05	7.3E-05	1.1E-02
Th-232	1E+04	2.8E-04	5E+04	1.2E-03	1.5E-03
U-234	3E+04	7.9E-02	2E+05	5.1E-04	2.7E-01
U-235	3E+04	3.0E-03	1E+05	2.8E-05	8.0E-02
U-238	3E+04	6.4E-02	9E+04	9.9E-04	6.5E-02
Total		1.6E-01		2.8E-03	1.6E-01

<sup>a</sup> Denotes nuclide only identified in water sample, sediment value generated by program default distribution coefficient.

<sup>b</sup> Denotes nuclide only identified in sediment, water value generated by program default distribution coefficient.

## 6. EVALUATION OF RESULTS FROM THE SITE-SPECIFIC ANALYSIS PHASE

To test the utility of the site-specific assessment phase, the alpha relative biological effect (RBE) factor for aquatic animals was changed from its default factor of 20. Kocher and Trabalka [5] suggested radiation weighting factors should be substantially less than 20, perhaps in the range of 5 to 10. The upper bound of their suggested range was chosen for this test and entered into the RAD-BCG Calculator. New site-specific BCGs were calculated by the program (Table 5).

The resultant BCGs, for alpha emitting radionuclides, calculated for this third analysis were higher than those calculated for the site-specific screen, in the second analysis, resulting in a lower total sum of fractions. By reducing the RBE by half, reduced the sum of fraction to 55% of the site-specific screen value and to approximately 17% of the initial screen sum of fractions value. The limiting organism listed for this site-specific analysis was a riparian animal.

As a next step, one could switch from using default or site-specific bioaccumulation coefficients to using the kinetic/allometric approach built into the RAD-BCG Calculator. This is done in the program on the Riparian Animal page. When this step is taken, new site-specific BCGs are generated and the water and sediment concentrations are ratioed to them and a new sum of fractions is calculated, using the default riparian animal kinetic and allometric parameters. The results are shown in Table 6.

TABLE 5. THE 300 AREA SHORELINE STUDY SITE-SPECIFIC ASSESSMENT BASED ON THE RAD-BCG CALCULATOR SUMMATION OF PARTIAL FRACTIONS, WITH RBE SET TO 10

Nuclide	Water Limit (Bq m <sup>-3</sup> )	Water Partial Fraction	Sediment Limit (Bq kg <sup>-1</sup> )	Sediment Partial Fraction	Combined Sum of Fractions
H-3 <sup>a</sup>	1E+10	3.2E-05	1E+07	2.2E-08	3.2E-05
Co-60 <sup>b</sup>	1E+05	3.4E-06	5E+04	8.9E-06	1.2E-05
Sr-90	1E+04	7.3E-04	2E+04	4.5E-05	7.7E-04
Tc-99 <sup>a</sup>	2E+07	2.3E-05	2E+06	1.8E-06	2.3E-05
I-129 <sup>a</sup>	1E+06	1.1E-07	1E+06	1.4E-09	1.1E-07
Cs-137 <sup>b</sup>	2E+03	1.1E-02	1E+05	7.3E-05	1.1E-02
Th-232	2E+04	1.4E-04	1E+05	5.9E-04	7.4E-04
U-234	5E+04	4.0E-02	4E+05	2.6E-04	4.0E-02
U-235	5E+04	1.5E-03	2E+05	1.9E-05	1.6E-03
U-238	5E+04	3.3E-02	1E+05	7.9E-04	3.4E-02
Total		8.6E-02		1.8E-03	8.8E-02

<sup>a</sup> Denotes nuclide only identified in water sample, sediment value generated by program default distribution coefficient.

<sup>b</sup> Denotes nuclide only identified in sediment, water value generated by program default distribution coefficient.

TABLE 6. 300 AREA SHORELINE STUDY SITE-SPECIFIC ASSESSMENT BASED ON THE RAD-BCG CALCULATOR, USING THE KINETIC/ALLOMETRIC APPROACH, SUMMATION OF PARTIAL FRACTIONS

Nuclide	Water Limit (Bq m <sup>-3</sup> )	Water Partial Fraction	Sediment Limit (Bq kg <sup>-1</sup> )	Sediment Partial Fraction	Combined Sum of Fractions
H-3 <sup>a</sup>	8E+08	3.7E-04	1E+07	2.2E-08	3.7E-04
Co-60 <sup>b</sup>	1E+05	3.4E-06	5E+04	8.8E-06	1.2E-05
Sr-90	1E+04	5.9E-04	3E+04	3.1E-05	6.2E-04
Tc-99 <sup>a</sup>	2E+07	3.1E-05	2E+06	1.8E-06	3.3E-05
I-129 <sup>a</sup>	3E+06	4.8E-08	1E+06	1.4E-09	4.9E-08
Cs-137 <sup>b</sup>	2E+03	7.2E-03	1E+05	6.3E-05	7.3E-03
Th-232	2E+04	1.4E-04	7E+04	8.0E-04	9.5E-04
U-234	3E+07	7.8E-05	5E+05	1.9E-04	2.7E-04
U-235	3E+07	3.2E-06	2E+05	1.6E-05	1.9E-05
U-238	2E+07	7.7E-03	1E+05	7.3E-04	8.0E-04
Total		8.5E-03		1.8E-03	1.0E-02

<sup>a</sup> Denotes nuclide only identified in water sample, sediment value generated by program default distribution coefficient.

<sup>b</sup> Denotes nuclide only identified in sediment, water value generated by program default distribution coefficient.

## 7. CONCLUSIONS

The total sum of fractions in the general screening phase of this analyses indicates that radiological doses to biota residing along the 300 Area shoreline were below applicable or proposed regulatory limits. Results of screening calculations of radionuclide concentrations in water and sediment were conservative when compared to subsequent assessment results.

The RAD-BCG Calculator and the graded approach provide a useful means for determining compliance with current applicable or proposed dose limits. Although the initial screen, using maximum water and sediment data indicated that no further analyses was necessary, subsequent analyses showed a reduction in the total sum of fraction at each progressive step indicating a reduction in dose rates to organisms being evaluated as more site-specific parameters are employed.

The RAD-BCG Calculator employs conservative initial screening levels and is simple to use, allowing for the use of site-specific parameters in screening process.

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# Radiation doses to frogs inhabiting a wetland ecosystem in an area of Sweden contaminated with $^{137}\text{Cs}$

K. Stark, R. Avila, P. Wallberg

The Swedish Radiation Protection Authority, Stockholm, Sweden

**Abstract.** Internal and external radiation doses to frogs living in a wetland ecosystem contaminated with  $^{137}\text{Cs}$  were estimated. The external doses were calculated from measured concentrations of  $^{137}\text{Cs}$  in soil and in water taking into account changes in the habitat during the frogs' life cycle. The internal dose was estimated from measured concentrations of  $^{137}\text{Cs}$  in living frogs (*Rana arvalis*) using a whole-body counter. The average inventory of  $^{137}\text{Cs}$  in the soil was approximately 1000 kBq/m<sup>2</sup> of which 86–99 % was found in the top 12 cm. The concentrations of  $^{137}\text{Cs}$  in frogs varied between 560 and 3450 Bq/kg ww. The estimated external dose rate was between 21 and 160 mGy/y, while the internal dose of beta and gamma was only between 1 and 6.2 mGy/y. The estimated total dose rate to frogs from  $^{137}\text{Cs}$  was below the expected safe level for terrestrial populations but close to the critical dose rate for amphibians suggested in the literature. Therefore, the radiation risk to frogs from radiocaesium in the study area may be one more stressor for an endangered group of animals in this ecosystem.

## 1. INTRODUCTION

In Sweden the highest deposition of radioactive caesium from the Chernobyl fallout occurred in the middle-east part of the country. During the 1990's anomalies with accumulations of  $^{137}\text{Cs}$  have been observed in wetlands by airborne gamma spectrometry measurements with  $^{137}\text{Cs}$ -concentrations up to 10 times higher than in surrounding areas [1]. Wetlands constitute transition zones between terrestrial and aquatic systems, and serves as nurseries and feeding areas for both aquatic and terrestrial species. Wetlands are often situated downstream in watershed and in depressions that receive large surface runoff or where infiltrated precipitation comes to the surface. Also, wetlands alter the hydrology of streams and rivers by impeding water flow and enhancing sediment deposition and thereby function as a filter to coastal waters by retaining nutrients as well as contaminants [2, 3]. Thus, the combination of the high capacity of organic material to adsorb radioactive caesium and the large amount of water seeping through the wetlands may result in high accumulation of radioactive caesium.

When considering radiation doses to animals in wetland anomalies several factors indicate that frogs, due to their physiology and behaviour, are one of the most radiosensitive organisms living in wetland ecosystems. Frogs are poikilothermic animals, i.e., their body temperature is the same as the immediate surroundings [4]. At lower temperature the metabolism is slower and as a consequence the biological half-life for  $^{137}\text{Cs}$  in frogs becomes longer. For every 10°C decrease in temperature the biological half-life is increased by a factor of 2 [5, 6]. Furthermore, frogs live most of their life in a limited area (approximately 200 m<sup>2</sup>; [7]). Frogs hibernate during 6 months in the winter period by digging themselves into the soil or at the bottom of a shallow pond or stream. They can make longer excursions during the summer but often return to the same pond or watershed every year to spawn and lay their eggs. Thus, a frog could spend its whole life, i.e. about 10 years, in the same wetland area. In addition, in comparison with for instance mammals, frogs have a thin and permeable skin that makes them sensitive to changes in the environment. Also, frogs have an external embryonic development and lay their eggs directly in the water lacking shells or amnion that makes them potentially sensitive to exposure from radioactivity.

The content of radiocaesium was measured in adult male green treefrogs (*Hyla cinerea*) in a study conducted in a contaminated swampy floodplain near Savannah River, USA [8]. In this study the measured average concentration of radiocaesium in dried frogs was 7500 Bq/kg dw (1 SD=240).

The aim of this study was to estimate the external and internal radiation doses from  $^{137}\text{Cs}$  that frogs receive living in a contaminated wetland. The study area was a wetland situated in Utnora where the  $^{137}\text{Cs}$  deposition following the Chernobyl accident was above 100 kBq/m<sup>2</sup>. In 2000 the  $^{137}\text{Cs}$ -concentrations in the wetland were up to 10 times higher than in surrounding areas [1]. Two different methodologies were used with different dose conversion factors for estimation of radiation doses.

## 2. MATERIAL AND METHODS

### 2.1. Description of the study area

The study was carried out in a wetland area in Utnora, 10 km north of the town Gävle, in the middle-east part of Sweden (Figure 1). The wetland area (about 0.036 km<sup>2</sup>) is situated in a depression mainly surrounded by coniferous forest and consists of an alder carr that is generally flooded in the spring by the stream of Verkmyra that runs from the lake of Hille, through the alder carr and ends in a nearby estuary in the Baltic Sea (2.5 km downstream). A tall reed belt of common reed dominates the estuary.



FIG. 1. Map over Sweden showing the study site in Utnora, north of the town Gävle [1].

### 2.2. Collection of samples

Twelve soil cores with a diameter of 49.9 mm and a length of 14–36 cm were collected on the 4–5<sup>th</sup> of May 2001 and the 4<sup>th</sup> of October 2001. Eight samples were taken in the alder carr and four samples were taken at the beginning of the reed belt. The soil samples were sliced into 3 cm pieces. Two water samples (5 l) were taken in the flooded alder carr on the 5<sup>th</sup> of May 2001. Five male frogs (*Rana arvalis*) were captured on the 10<sup>th</sup> of May 2001. Four frogs were captured within a 10 × 20 m large area within the alder carr and one was captured 500 m further upstream. The frogs were placed in a bucket with water from the wetland. Leaves and insect larvae were added so that the frogs would have access to food and shelter. The bucket was held at a low temperature throughout the study. After the whole-body measurement the frogs were returned and released in the same wetland as they were found.

### 2.3. Measurement of <sup>137</sup>Cs activity concentrations

The concentrations of <sup>137</sup>Cs in soil samples were measured with an HPGe-detector with a detection level of approximately 10 Bq/kg and a relative statistical measurement uncertainty (1 SD) of between 1 and 35% with a median of 1.47%. To calculate the distribution of the soil samples the programme @RISK [9] was used.

The water samples were filtered to remove soil particles, zooplankton and insect nymphs. To determine the radiochemical yield in each sample approximately 10 Bq of <sup>134</sup>Cs (solution: 12 830 Bq/l 07-02-2001) was added. Each water sample was then filtered through impregnated Cu(Fe(CN)<sub>6</sub>)-filters. The filters were placed in a plastic jar (65 ml) during the measurement. The relative statistical measurement uncertainty (1 SD) was ≤ 20%.

The <sup>137</sup>Cs concentrations in the frogs were measured (one frog at the time) in a whole-body counter for an hour in the dark. The frog was placed in a plastic jar with a few ml of tap water to prevent the frog from dehydrating. Background radiation was accounted for during the measurements by measuring the plastic jar alone. The relative statistical measurement uncertainty (1 SD) was ≤ 10%.

## 2.4. Concentration factors

Concentration factors (CF) between the concentration of  $^{137}\text{Cs}$  in a frog and soil and in a frog and water were calculated. The maximum and minimum values measured in the organisms and the ambient mediums were used to calculate an interval for the CF. The concentration of  $^{137}\text{Cs}$  in the top 12 cm of the soil was used. The formula used was:

$$\text{CF} = \frac{\text{concentration of Cs-137 in organism (Bq/kg ww)}}{\text{concentration of Cs-137 in soil alt. water (Bq/kg dw. alt. Bq/l)}}$$

where: ww = wet weight, dw = dry weight.

## 2.5. Estimation of radiation doses

The external doses to the frogs were conservatively estimated by multiplying the measured activity concentrations of  $^{137}\text{Cs}$  in soil by the dose conversion factors (DCF) reported in Amiro [10]. The average concentration of  $^{137}\text{Cs}$  in the top 12 cm in the alder carr and the reed was used in the calculations. It was assumed that the organisms are submerged 0.1 m below the surface of a semi-infinite, uniformly contaminated body of soil. The doses from photons were calculated at the body surface and from electrons at 70  $\mu\text{m}$  into the skin. This approach will overestimate the doses to sensitive internal organs.

More realistic calculations of the external doses were also made using the methodology described by the Environment Agency of England and Wales (EA) [11]. In this case the organism is represented as an ellipsoid (100  $\times$  20  $\times$  20 mm) and the absorbed external dose (from beta and gamma radiation) is calculated as an average throughout the volume of the organism (weight 20 g). The organism is assumed to spend 50% of its time on the soil surface and 50% buried in the soil.

The internal doses, expressed as an average throughout the whole body of the organism, were calculated by multiplying the measured values of the activity concentrations of  $^{137}\text{Cs}$  in frogs by the DCFs reported by EA [11]. The organism is represented as an ellipsoid of size 100  $\times$  20  $\times$  20 mm and weight 20 g.

## 3. RESULTS

### 3.1. Activity concentrations of $^{137}\text{Cs}$ in soil and water

The average inventory of  $^{137}\text{Cs}$  in the alder carr and at the beginning of the reed belt was 1110 kBq/m<sup>2</sup> (1 SD = 510) and 1000 kBq/m<sup>2</sup> (1 SD = 290), respectively. Between 86 and 99 % of the inventory was found in the top 12 cm of the soil profile (Figure 2). The concentrations of  $^{137}\text{Cs}$  in the water covering the soil were 0.18 and 0.22 Bq/l and the concentrations in the filtered zooplankton, insect larvae and soil particles were 1273 and 8000 Bq/kg dw, respectively. This corresponds to total concentrations of 0.23 and 0.94 Bq/l of unfiltered water.

The activity concentrations of  $^{137}\text{Cs}$  in the top 12 cm of the soil profile followed a lognormal distribution (Figure 3). The values oscillated between 12.2 (5<sup>th</sup> percentile) and 65.7 (95<sup>th</sup> percentile) with a geometric mean of 25.6 kBq/kg dw.

### 3.2. Activity concentrations in frogs

The concentration of  $^{137}\text{Cs}$  in *Rana arvalis* measured alive in the whole-body counter varied between 560–3450 Bq/kg ww (Figure 4). The highest  $^{137}\text{Cs}$  concentrations were measured in the smallest frogs.

CFs defined as the ratio between the activity concentration in frogs (Bq/kg ww) to that in soil (Bq/kg dw) or water (Bq/l) were calculated. The relatively higher concentration of  $^{137}\text{Cs}$  in soil in comparison with the concentration in water yielded a large difference in CFs. The obtained values varied between 0.006 and 6.9 for soil and between 2545 and 19144 for water.

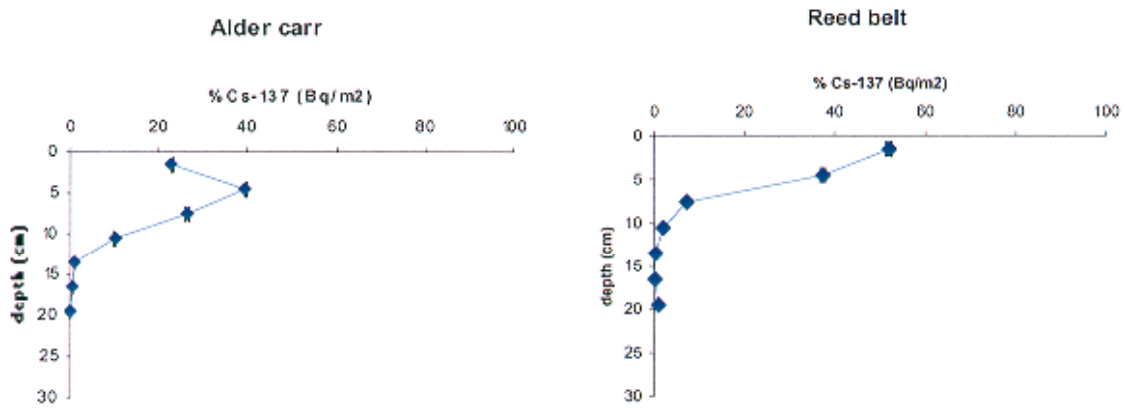


FIG. 2. Examples of soil profiles taken in the alder carr and in the reed belt on the 4<sup>th</sup> of October and 4<sup>th</sup> of May 2001, respectively. The amount of <sup>137</sup>Cs (% of total Bq/m<sup>2</sup>) decrease with increasing depth (cm).

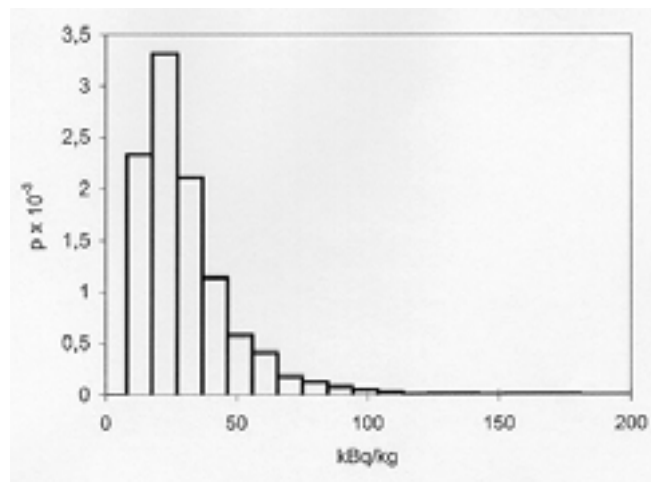


FIG. 3. Distribution of <sup>137</sup>Cs activity concentrations in the top 12 cm of the soil observed in the study area.

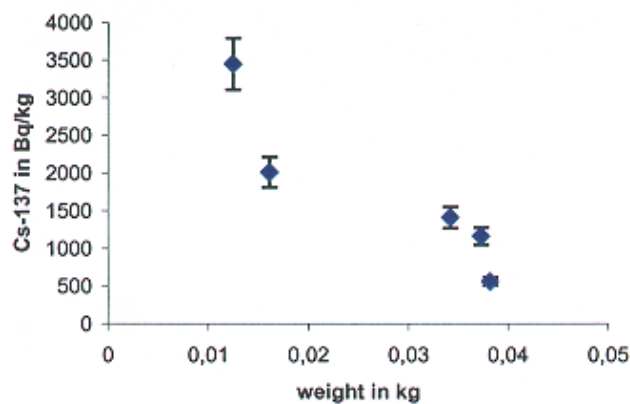


FIG. 4. The concentration of <sup>137</sup>Cs with the relative statistical measurement uncertainty (1 SD) in 5 frogs captured on the 10<sup>th</sup> of May 2001 in relation to their body weight in kg (ww).

TABLE 1. ESTIMATES OF THE EXTERNAL DOSES TO FROGS (MGY/Y) USING CONSERVATIVE [10] AND MORE REALISTIC [11] DOSE CONVERSION FACTORS

Statistical measure	Amiro [10]	EA [11]
Mean	123	54
50 <sup>th</sup> percentile	103	45
5 <sup>th</sup> percentile	49	21.3
95 <sup>th</sup> percentile	160	96

### 3.3. Estimates of external doses

The estimated values of the external dose rates varied between 21 and 160 mGy/y (Table 1). The results show that the use of the DCF reported by Amiro [10] lead to external dose rates that are by a factor of 2.3 higher than the values obtained using the methodology proposed by the EA [11]. The estimated values of the internal dose rate was between 1 and 6.2 mGy/y, which is well below the interval of variation of the external dose rates. The conservatively estimated values of the total doses to frogs are below the expected safe levels of exposure (350 mGy/y for terrestrial populations) suggested in the 1996 UNSCEAR report on Effects of Radiation on the Environment [12]. The expected safe levels of exposure were derived from dose-response relationships for deterministic effects, but it is assumed in the report that as long as the dose was kept below the safe levels of exposure based on reproductive effects, stochastic effects should not be significant at a population level. However, the report does not address the question of whether stochastic effects could cause harm to individual organisms. Environmental critical dose rate values were proposed by the Atomic Energy Control Board, AECB [13], corresponding to values that would assure little probability of underestimating the risk of effects. The value proposed for amphibians was 100 mGy/y, which is close to the 95<sup>th</sup> percentile of the realistically estimated doses to frogs in the studied area (Table 1).

## 4. DISCUSSION

The wetland area in Utnora is at present the area where the highest concentrations of <sup>137</sup>Cs have been measured in Sweden. Airborne gamma spectrometry measurements have indicated that there exist several similar areas where accumulation of <sup>137</sup>Cs has occurred in Sweden.

### 4.1. Activity concentrations in frog

The results from this study suggest that frogs accumulate radioactive caesium to a high extent (Figure 4). The caesium atom resembles the potassium atom and is therefore accumulated in muscle tissue, kidneys and cartilage where there are active metabolising cells [14]. All frogs were captured and measured just after or during the spawning season that occur in early springtime when the frogs emerge from their hibernation shelter. The frogs have by then not been eating any food for approximately 6 months (200 days) and do not start to eat until the end of the spawning season. During the spawning a lot of energy is required and the frogs might loose some muscle mass and by excreting eggs and sperms their body concentration of caesium could decrease. Thus, if the frogs were measured later in the summer time or in the beginning of the fall the concentration of <sup>137</sup>Cs could be higher than found in this study.

The highest <sup>137</sup>Cs concentrations were found in the smallest frogs (Figure 4). This could be due to that these frogs were not yet sexually mature and might have started to eat after the hibernation period. Young frogs are also in a period of very active growth during which they eat more in relation to their body weight than adult frogs. The green tree frogs that were measured in a study conducted in a contaminated area by the Savannah River [8] had about the same weight and therefore the variation of the <sup>137</sup>Cs content in the frogs (7500 Bq/kg dw, 1 SD=240) was suggested to be due to the variation of the <sup>137</sup>Cs content in the food.

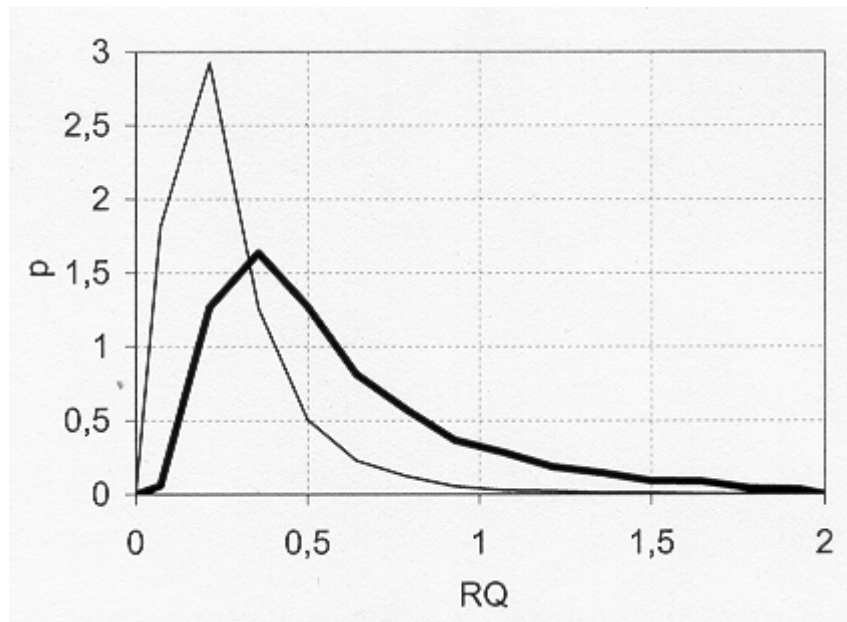


FIG.5. Calculated values of the Risk Quotient corresponding to conservative (—) and realistic (—) estimations of the doses to frogs in the studied area and assuming that the reference dose follows a uniform distribution with values between 100 and 350 mGy/y

#### 4.2. Dose estimations in comparison with reference doses

Currently there is no international agreement on which values should be used as reference dose when estimating the risk to biota from ionising radiation. Lets assume that the reference dose for amphibians could be any value between 100 and 350 mGy/y. By dividing the estimated doses to the frog by the reference dose the risk quotients (RQ) were obtained (Figure 5). If the RQ is <1 it is assumed that there is no risk for the reference dose to be exceeded. For both the realistic and conservative dose estimations the mean RQ was well below 1. Furthermore, for the realistic dose estimations the probability of the RQ being below 1 was more than 98.7%. It should be noted that 14.4 % of the conservative estimations gave RQ values above 1 indicating that realistic estimations are needed at this level of contamination. This illustrates the well-known fact of an increasing need for accuracy in the estimations as doses approach the dose limit (reference dose).

#### 4.3. Different approaches in wildlife dosimetry

In this study two different sets of DCFs were used. DCFs in Amiro [10] are conservative and therefore yield relatively high doses. The DCFs and the methodology described by the EA [11] are more realistic and calculate the average dose to an ellipsoid, which also reduces the possible overestimation. When estimating the internal dose the methodology uses CF for biota in addition to the DCFs. A CF can be used if the concentration of radionuclides in the organism is unknown. However, there is a lack of data both concerning CFs for different radionuclides as well as for different species. Also, CFs generally show a large variation. For instance, in this study the calculated CF varied by a factor of 1000, depending on the variation in soil concentrations. Therefore, the CFs for biota can give an underestimate or an overestimate of the concentration in the organism.

In both estimations conducted in this study a probabilistic approach [15] was used and intervals of possible doses were obtained. Within this interval the probability for an organism to receive a certain dose was calculated. The probabilistic approach corresponds well to the true variation in nature and with specific DCFs a realistic estimation of the dose can be obtained.

## 5. CONCLUSIONS

The results from this study suggest that *Rana arvalis* accumulate radiocaesium to a high extent, but that nevertheless the highest contribution to the total dose in a contaminated wetland will be the external irradiation from radiocaesium in soil. The estimated total dose rate to frogs from  $^{137}\text{Cs}$  was below the expected safe level for terrestrial populations but close to the critical dose rate for amphibians suggested in the literature. Therefore, the radiation risk to frogs from radiocaesium in the study area may be one more stressor for an endangered group of animals in this ecosystem.

Realistic radiation dose estimations to biota are dependent on three major factors, specific dose conversion factors, concentration factors and knowledge of the ecology and physiology of the organism. Dose conversion factors that are calculated for different geometries representing certain animals or group of animals, are more realistic. Within the EC programme Framework for Assessment of Environmental Impact (FASSET) [16] work is presently ongoing to develop a set of models for reference organisms.

One crucial factor for the total dose to an organism is the time spent in a contaminated area. *Rana arvalis* has a small home range and can thus spend its whole lifetime in the studied area. A further improvement in the dose calculations could be achieved by defining better the irradiation geometry at different stages of the life-cycle of the frog and by taking into account radiocaesium vertical distribution in the soil profile.

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# Modelling of consequences for marine environment from radioactive contamination

M. Iosjpe, J. Brown, P. Strand

Norwegian Radiation Protection Authority, Østerås, Norway

**Abstract.** A system for the radiological protection of the environment, with special attention to the derivation of dose limits for flora and fauna, will require the use of modelling tools. This paper promotes one approach for modelling the consequences for the marine environment from radioactive contamination. The method is based on the modified approach for box modelling ("ARCTICMAR 2" model), which includes dispersion of radionuclides during time (non-instantaneous mixing in oceanic space). This approach was created in order to provide a better and more realistic/physical approach compared to traditional box modelling. The model can provide information concerning the dispersion of radionuclides in water, sediment and biota phases of the marine environment. The assessment of internal and external doses to the marine organisms is based on assumptions about uniform distributions of radionuclides in organisms as well as surrounding water and sediment boxes. The dose assessment uses dose conversion factors that account for the absorbed fraction for  $\gamma$ -emitting radionuclides. The potential of the present modelling approach is illustrated by calculations of doses for selected sets of scenarios and "reference" organisms.

## 1. INTRODUCTION

The present doctrine of the International Commission on Radiological Protection (ICRP) concerning protection of flora and fauna under dose limits regarding to man [1, 2] has limitations, which are discussed in recent publications [3, 4]. A system of radiological protection of the environment with special attention to the derivation of dose limits for non-human components, as considered by Pentreath [5], includes modelling as an essential component.

The dose assessment modelling methodology for flora and fauna has many common elements to that adopted in the radiological dose assessment for man from radioactive contamination. Both approaches have to cover whole processes such as dispersion of radionuclides in oceanic space, transfer of radioactivity between sea water and sediments, uptake of radionuclides by biota and, finally, dose calculations. The other common feature of the modelling for the marine environment is a requirement to follow dispersion of radionuclides for some scenarios from sources of contamination for large distances (thousands of kilometres) and long time-scales (up to centuries or millenniums).

Box models can be easily used for describing of all abovementioned processes and time and space scales. Therefore, box modelling has been recommended by the European Commission for dose assessment for man [6] and selected for many investigations (for instance, [7–9]).

The marine box modelling includes the processes of advection of radioactivity between sea water compartments, sedimentation, diffusivity of radioactivity through the pore water, resuspension, mixing due to bioturbation and a burial process of activity in deep sediment. Radioactive decay is included in all compartments (a schematic structure of the processes involved in marine box modelling is shown in Figure 1). The contamination of biota is further calculated from the radionuclide concentrations in filtered seawater in the different water regions on the basis of concentration factors [10].

Contrary to evaluation of doses to man, calculation of doses to biota in the marine environment has to consider a more comprehensive group of of marine organisms than that used in human dose assessment (i.e. fish, crustacea, molluscs and seaweed). This reflects the fact that not all marine organisms form components of the human diet and/or are involved in radionuclide transfer pathways to man. Nonetheless, it is clear that not all marine organisms can be studied, moreover data (primarily transfer, dose-effects) will not be available for the vast majority of species, and therefore some simplifications are required. A possible approach involves the selection of a set of reference organisms [5] that can be used as representatives of the environment in its entirety. These reference organisms could be presented along with specific life history data and uptake/transfer parameters conducive to the calculation of dose.

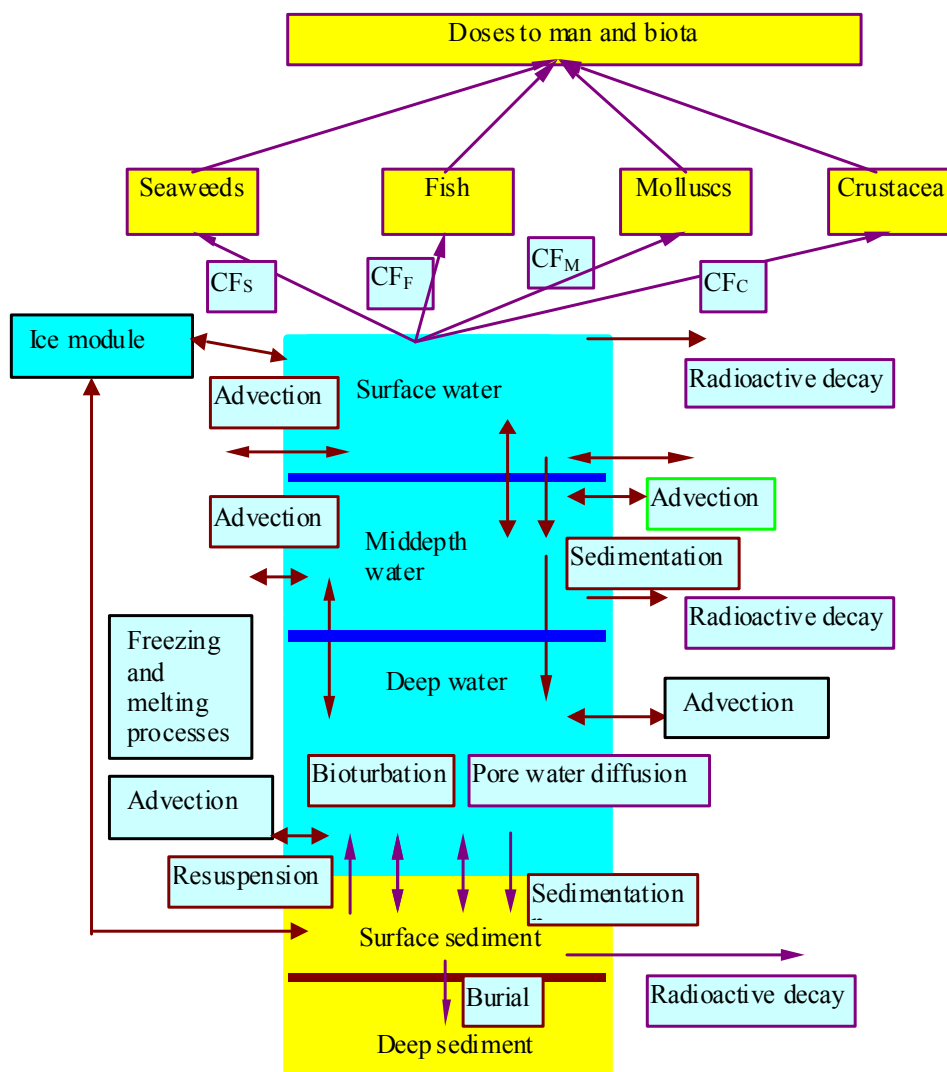


FIG. 1. A schematic structure of the processes involved in modelling.

“The ARCTICMAR 2” model, which will be used in this paper, is based on the modified approach for box modelling [11]. Modelling includes dispersion of radionuclides during time (non-instantaneous mixing in oceanic space). This approach was created in order to provide a better and more realistic/physical description of the radionuclide dispersion comparing to traditional box modelling.

The model can provide information concerning the dispersion of radionuclides in water, sediment and biota phases of marine environment using site-specific data. It should be noted that the present model can be used to simulate the dispersion of different contaminants simultaneously (for instance, radionuclides and heavy metals).

The assessment of internal and external doses to the marine organisms is based on the assumption that radionuclides are distributed uniformly in organisms and the surrounding environment [12].

The abovementioned set of reference organisms includes species with life times that can vary widely. Therefore, the radiation dosimetry models for reference organisms are designed to estimate the actual or potential absorbed dose-rates, as oppose to (cumulative) doses, to the organisms, from internal and external sources of  $\alpha$   $\beta$  and  $\gamma$ -radiation. Various dosimetry model methodologies [13–15] are available for use for this purpose.

## 2. MODEL DESCRIPTION

### 2.1. Dispersion module

The present model is an improved version of the compartmental model [7]. The equations of transfer of radionuclides between the boxes are of the form:

$$\begin{aligned} \frac{dA_i}{dt} &= \sum_{j=1}^n k_{ji} A_j - \sum_{j=1}^n k_{ij} A_i \gamma(t \geq T_j) - k_i A_i + Q_i, t \geq T_i \\ A_i &= 0, \dots, t < T_i \end{aligned} \quad (1)$$

where  $k_{ii}=0$  for all  $i$ ,  $A_i$  and  $A_j$  are activities at time  $t$  in boxes  $i$  and  $j$ ;  $k_{ij}$  and  $k_{ji}$  are rates of transfer between boxes  $i$  and  $j$ ;  $k_i$  is an effective rate of transfer of activity from box  $i$  taking into account loss of material from the compartment without transfer to another, for example radioactive decay;  $Q_i$  is a source of input into box  $i$ ;  $n$  is the number of boxes in the system;  $T_i$  is the time of availability for box  $i$  (the first times when box  $i$  is open for dispersion of radionuclides) and  $\gamma$  is an unit function:

$$\gamma(t \geq T_i) = \begin{cases} 1, & t \geq T_i \\ 0, & t < T_i \end{cases}$$

The times of availability  $T_i$

$$T_i = \min_{\mu_m(v_0, v_i) \in M_i} \sum_{j,k} w_{jk}$$

are calculated as a minimized sum of the weights for all paths  $\mu_m(v_0, \dots, v_i)$  from the initial box ( $v_0$ ) with discharge of radionuclides to the box  $i$  on the oriented graph  $G=(V, E)$  with a set  $V$  of nodes  $v_j$  correspondent to boxes and a set  $E$  of arcs  $e_{jk}$  correspondent to the transfer possibility between the boxes  $j$  and  $k$  (graph elements as well as available paths are illustrated by Figure 2). Every arc  $e_{jk}$  has a weight  $w_{jk}$  which is defined as the time required before the transfer of radionuclides from box  $j$  to box  $k$  can begin (without any way through other boxes). Weight,  $w_{jk}$ , is considered as a discrete function  $F$  of the water fluxes  $f_{jk}, f_{kj}$  between boxes  $j$  and  $k$ , geographical information  $g_{jk}$  and expert evaluation  $X_{jk}$ .  $M_i$  is a set of feasible paths from the initial box ( $v_0$ ) to the box  $i$  ( $v_i$ ).

Furthermore, it is assumed that, at any given time, the activity in the water column is partitioned between the water phase and the suspended sediment material. The fraction of the activity ( $F_w$ ) in the water column, which is in solution, is given by:

$$F_w = \frac{1}{1 + K_d SSL}$$

where  $K_d$  is the sediment concentration factor and  $SSL$  the suspended sediment load.

Activity on suspended sediments is lost to the underlying boxes when particles settle out. The fractional transfer from a water column (box  $i$ ) to the sediments (box  $j$ ) due to sedimentation is given by:

$$k_{ij} \equiv \frac{K_d SR_i}{d_i (1 + K_d SSL_i)}$$

where  $d_i$  is the mean water depth and  $SR$  the mass sedimentation rate.

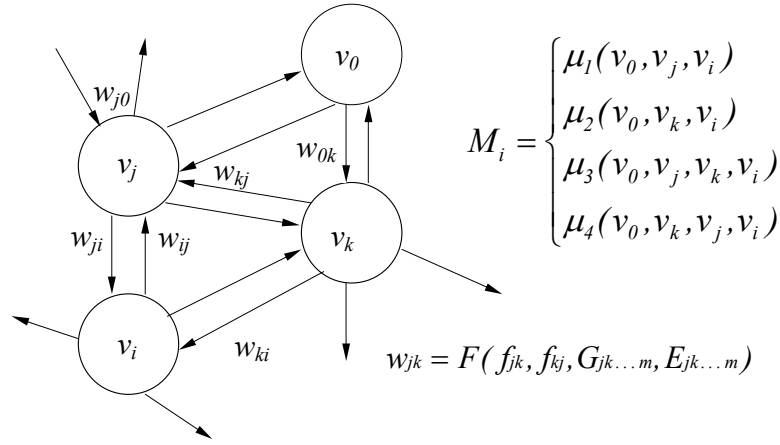


FIG. 2. Graph elements.

The model includes the processes of diffusivity of radioactivity through the pore water, resuspension, mixing due to bioturbation and a burial process of activity in deep sediment on the basis of the Fick law [7, 16]. Radioactive decay is included in all compartments.

The contamination of marine organisms is calculated from the radionuclide concentrations in filtered sea water in the different water regions. Doses to man are calculated on the basis of data for the catch of seafood and assumptions about human diet.

It is easy to see that “traditional” box modelling is a particular case of the present approach when all times of ability in the equation (1) are zero:  $\{T_i\} = 0, i = 1, \dots, n$ . Furthermore, it is significant to note that the uncertainty of calculations does not increase in comparison to traditional modelling, because graph  $G=(V, E)$  is constructed from the same information which is used for traditional box modelling. This means that the developed approach does not incorporate new, additional parameters, but utilises initial information as far as possible.

## 2.2. Dose module

Dose calculations to man will not be considered in detail in this paper, but it is necessary to note that doses to man are calculated on the basis of data for the catch of contaminated seafood and assumptions about human diet. The contamination of fish, crustaceans and molluscs is calculated from the radionuclide concentrations in filtered sea water in the different water regions using concentration factors.

Internal absorbed dose calculations to biota are based on an equilibrium absorbed dose constant model that accounts for the total energy emitted by individual components of electromagnetic and/or particulate radiations after the decay of a specific radionuclide [17]. Assuming that a radionuclide is uniformly distributed throughout the body of an organism, the absorbed dose rate to the biota for the internal radiation can be given by:

$$\dot{D}(t) = f \sum_j A_j(t) \sum_i k_i E_i^{(j)} y_i^{(j)} \phi_i^{(j)}, \quad (2)$$

where  $\dot{D}(t)$  is a dose rate ( $\text{Gy y}^{-1}$ );  $A_j$  is activity concentration for radionuclide  $j$ ;  $E_i^{(j)}$  is energy emitted by component  $i$  of  $\alpha, \beta, \gamma$  radiation for radionuclide  $j$ ;  $y_i^{(j)}$  is a fractional abundance or yield of radiation particles/photons of energy  $E_i^{(j)}$ ;  $\phi_i^{(j)}$  is an absorbed fraction for energy  $E_i^{(j)}$ ;  $k_j$  is a biological factor (relative biological effectiveness) for component  $i$  of  $(\alpha, \beta, \gamma)$  radiation;  $f = 5.04 \cdot 10^{-6}$  is a factor to account for conversions of MeV to Joules ( $1 \text{ MeV} = 1.602 \times 10^{-13} \text{ J}$ ) and seconds to years ( $1 \text{ year} = 3.145 \times 10^7 \text{ seconds}$ ).

All radionuclide data have been derived from ICRP Publication 38 [18]. Information concerning the calculation of absorbed fractions  $\phi_i^{(j)}$  for different dimensions of organisms is defined from [17] in the case of testing the dose module operation. Other methods can also be used to calculate  $\phi_i^{(j)}$  as described later. External doses in the present paper are calculated using the dose conversion factors tabulated in [15].

It is necessary to note that evaluation of the dose conversion factors for internal and external radiation for wide set of reference organisms is also under development in the FASSET (Framework for assessment of environmental impact) project of EC 5th Framework Programme [20] and can be easily adopted by the present methodology.

### 3. RESULTS OF CALCULATIONS: DEMONSTRATION OF METHODOLOGY

#### 3.1. Innovation features and feasibility of the dispersion module

Innovative elements of the present approach are clearly indicated by Figure 3, which corresponds to calculations of the dispersion of 1 TBq of  $^{239}\text{Pu}$  discharge into the Ob Bay of the Kara Sea.

The present version of the model predicts that dispersion of  $^{239}\text{Pu}$  is still limited by the Ob Bay boundaries, six months after the discharge of the radionuclide, whereas the dispersion of the radionuclide predicted by the traditional approach has to cover the whole of ocean space. The other feature of the present version is that the model, in contrast to the basic version [7], predicts boxes without contamination in the initial phase of dispersion from a point source (for example, for the present scenario, the activity will reach the Greenland Sea after six years, approximately). Plots in Figure 3 correspond to the seawater concentration of  $^{239}\text{Pu}$  in the Greenland Sea and indicate significant differences of up to orders of magnitude, between the traditional and newly-developed models, during the initial time of dispersion because of the instantaneous spread of radionuclides according to traditional box modelling [7].

Practical feasibility of the present approach for box modelling is demonstrated in Figure 4 by the comparison of model predictions with the experimental data for discharges of  $^{99}\text{Tc}$  from Sellafield [21, 22] for the Skagerarrak Strait of the North Seas. The solid and dashed lines correspond to the results of calculations executed by the present model and traditional box modelling, respectively.

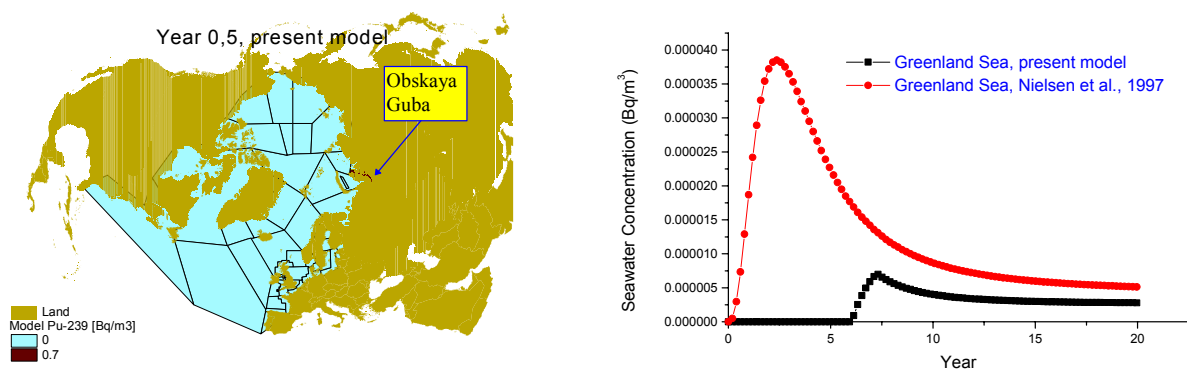


FIG. 3.  $^{239}\text{Pu}$  dispersion after discharge of 1TBq activity into the Ob Bay

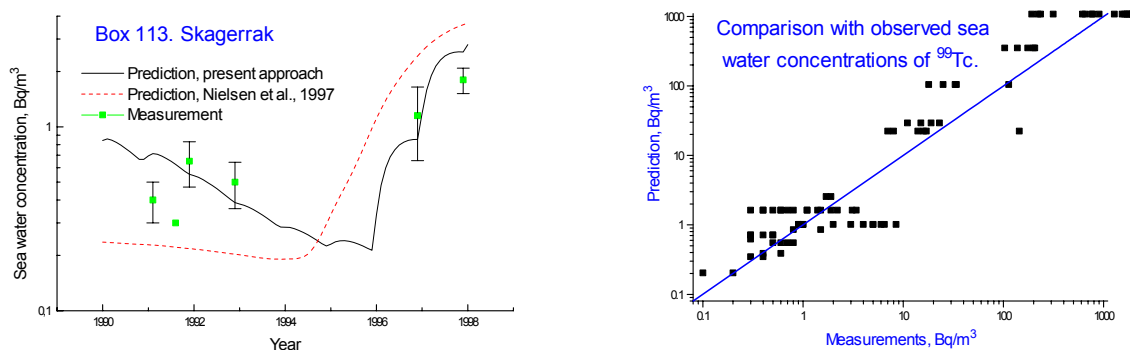


FIG. 4. Comparison of the results of calculation and experimental data.

Figure 4 shows, also, the correlation between experimental and predicted data for the Irish and North Seas, where solid lines corresponded to total agreement (1:1 relationship) between experimental data and calculations. Correlation and analysis produces the estimation for correlation coefficient  $r$  of 0.83 with the confidence interval [0.62, 1.03]. A regression analysis shows that the regression equation  $Y=K \cdot X$  is the best approximation for, at least, the class of equations of the linear type  $Y=L+K \cdot X$  (here,  $Y$  and  $X$  corresponded to experimental and predicted data, correspondently,  $L$  and  $K$  are the regression coefficients). Statistical estimation for the parameter  $K$  is 0.98 with the confidence interval [0.73, 1.23] (all calculations correspond to a significance level of 0.05). These results show that the present model provides a correct description of the relationship between prediction and experiment data (an ideal relationship corresponds to  $K=1$ ) for the present experimental data set.

### 3.2. Demonstration methodology for dose calculation

As mentioned above, dose calculations to man will not be considered in this paper, but it is interesting to stress that for some scenarios, calculations of doses to man indicate significant differences in comparison with traditional box modelling as well as it is shown in Figure 3 for concentrations of radionuclide in seawater.

Furthermore, it should be noted that selection of the set of reference organisms is not trivial [5] and could be based, amongst others, on criteria such as (a) organisms which, by virtue of environmental transfer and concentration factors, have the greatest potential for exposure (b) organisms which have a high radiosensitivity (c) organisms which are important to the healthy functioning of the community or ecosystem (d) organisms which are common. A set of reference organisms with regards to these criteria is under development [20]. Although various criteria could be applied in the selection of reference organisms, marine biota which will be under consideration in this paper were selected on the basis of available information for demonstrating the dose calculation methodology.

#### 3.2.1. Testing of the dose module operation

The operation of the computer-based model was tested using real environmental data from Norwegian marine environments and adjacent (Table 1). In such a way, preliminary information with respect to the impact of radionuclides on selected biota, would be provided in addition to the check on the program's operational performance. The data used were generalised in the sense that they were based on average levels of radionuclides monitored in Norwegian coastal waters. Lobsters (*Homarus gammarus*), seaweed (*Fucus vesiculosus*) and mussels (*Mytilus edulis*) were selected on the basis of data availability. Comprehensive data sets exist for environmental samples from Norwegian coastal waters [23, 24], however knowledge gaps are evident. Owing to this lack of data in certain areas, information was derived from other studies [25, 26] and, in some cases, generic concentration factors [10] were used, where only seawater concentrations were known. These data are presented in Table 1.

TABLE 1. TOTAL DOSES (mGy y<sup>-1</sup>) AND ACTIVITY LEVELS FOR BIOTA (Bq kg<sup>-1</sup>, w.w.), SEA WATER (Bq m<sup>-3</sup>) AND SEDIMENT (Bq kg<sup>-1</sup> d.w.)

Biota	<sup>137</sup> Cs <sup>a</sup>	<sup>90</sup> Sr <sup>c</sup>	<sup>99</sup> Tc <sup>a</sup>	<sup>238</sup> Pu <sup>a,d</sup>	<sup>239,240</sup> Pu <sup>a,d</sup>	<sup>241</sup> Am <sup>a,d</sup>	<sup>40</sup> K <sup>e</sup>	<sup>210</sup> Po	Total doses
Lobster	0.9	0.75	38.5	5 × 10 <sup>-4</sup>	7 × 10 <sup>-4</sup>	5 × 10 <sup>-4</sup>	70	17	1.35
Mussels	0.08	2.6	0.5	5 × 10 <sup>-3</sup>	7 × 10 <sup>-3</sup>	0.02	70	60	2.5
Seaweed	0.5 <sup>a,b</sup>	4.1	44.5	3 × 10 <sup>-3</sup>	4.5 × 10 <sup>-3</sup>	8 × 10 <sup>-3</sup>	400	0.35	1.8
Sea water	11.4	3.5 <sup>f</sup>	5 <sup>d</sup>	1.6 × 10 <sup>-3a</sup>	2.2 × 10 <sup>-3a</sup>	1 × 10 <sup>-3a</sup>	11 × 10 <sup>3</sup> *		
Sediment	1.9	3.5 <sup>d</sup>	0.5	2.7 × 10 <sup>-4a</sup>	2.8 × 10 <sup>-3a</sup>	0.8 × 10 <sup>-3a</sup>	500 *	20	

<sup>a</sup> data (1997) from Norwegian national surveillance report [24]; <sup>b</sup> dry weight measurement was 2.7 Bq kg<sup>-1</sup>, converted to wet weight by multiplying by 0.2 [10]; <sup>c</sup> data from MAFF [25]; <sup>d</sup> CFs/k<sub>ds</sub> [10] applied to sea water concentrations; <sup>e</sup> data from MAFF [26]; <sup>f</sup> 1993–1995 data from [23]; \* estimated – silty sediments.

Lobsters were represented by a 1 kg sphere/thick ellipsoid living on the sea bed (benthic), mussels as a 10 g small ellipsoid (surrounded by scattering medium) living on the sea bed and seaweed as a benthic 1 kg sphere/thick ellipsoid. Total doses (internal and external) were calculated and results are presented in Table 1.

For the suite of radionuclides considered, total doses varied between 1.35 mGy y<sup>-1</sup> and 2.5 mGy y<sup>-1</sup>. For present calculations, the doses were essentially attributable to <sup>40</sup>K and <sup>210</sup>Po. For mussels and seaweed, the internal component of the dose was predominant, forming 73 % and 62 % of the total dose respectively, whereas for lobster, the external component and internal component were of equal magnitude. In terms of the anthropogenic radionuclides, <sup>99</sup>Tc was the most important dose-forming radionuclide for lobsters and seaweed. Doses arising from <sup>99</sup>Tc were calculated to be 20 μGy (microGy) y<sup>-1</sup> for lobster and 23 μGy (microGy) y<sup>-1</sup> for seaweed and were almost entirely attributable to the internal component of dose. Calculations indicate, also, that the chronic dose rates for organisms in the Norwegian coastal waters are orders of magnitude below the levels where any significant effects are likely to be observed. An UNSCEAR report from 1996 [27] includes a section which provides a comprehensive overview of the studies and reviews which have considered the effects of ionising radiation on aquatic organisms. A general conclusion is that a dose rate of less than 1 mGy (milliGy) per hour is unlikely to produce significant effects in the gonads of fish which are assumed to be a highly radiosensitive end-point. Clearly, this limit, which corresponds to 8.8 Gy y<sup>-1</sup> at equilibrium, is well above the maximum of 2.5 mGy (milliGy) y<sup>-1</sup> (whole-body dose) calculated in this study.

### 3.2.2. “Kursk” submarine accident: methodology demonstration

On the morning of the 12<sup>th</sup> of August 2000, an accident involving a Russian nuclear submarine, the Kursk, occurred in international waters in the Barents Sea, approximately 250 km off the coast of Norway. The “Kursk” submarine was equipped with two pressurized water reactors each having a thermal effect of 190 megawatts, or less than 10% of that of a typical nuclear power plant reactor. All reactor and fuel configuration data concerning Russian naval, military vessels is classified due to military restrictions.

In this paper the potential worst case scenario, corresponding to an abnormal event one year after the accident where 100% of inventory from both reactors is released instantaneously, is under consideration. Additional information concerning technical and operational data is available from [28].

Calculations of <sup>137</sup>Cs dispersion in the oceanic space, corresponding to the worst case scenario show that 0.5 years after a hypothetical accidental release of 100% of the inventory, the average water concentration in the Barents Sea will be in the range 160 – 210 Bq/m<sup>3</sup> for areas in close vicinity to the submarine. <sup>137</sup>Cs activities will decrease rapidly, and after 10 years it is estimated that the average water concentration in the Barents Sea will be in the range 0.1 – 2.8 Bq/m<sup>3</sup>.

The dynamics for the <sup>137</sup>Cs concentration in fish for the Barents Sea region (corresponding to the same scenario) is shown in Figure 5. The maximum, minimum and average activity concentrations in fish correspond to areas at different distances from the Kursk. The plots displayed in the Figure 5 indicate that during the first years of potential dispersion, the <sup>137</sup>Cs activity concentration in fish varies widely

depending on the habitat of fish. During the early stages of the dispersion, the Barents Sea contains regions with relatively high contamination and regions that are not affected by the release of radionuclides. Calculations show a maximum activity concentration of  $^{137}\text{Cs}$  in fish, during the first year following a hypothetical leakage from the Kursk, in the range 0–100 Bq/kg (w.w.).

Doses to cod and Kamchatka crab corresponding the average concentration plot in Figure 5 are shown in Figure 6.

Cod was presented by 1.6kg ellipsoid with axes (m) 0.5, 0.1 and 0.06 living in seawater. Kamchatka crab was presented by the set of cylinders: a central body (radius of 0.1 m; height of 0.03 m) and six claws (radius of 0.02 m; length of 1 m) living in semi-infinite (seawater – sediment) environment.

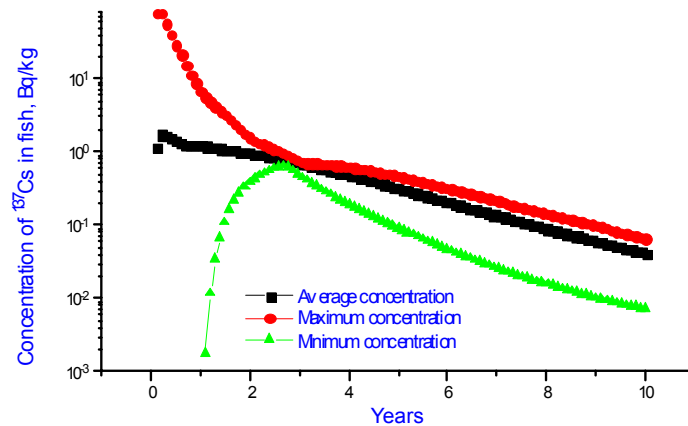


FIG. 5. Dynamics of the  $^{137}\text{Cs}$  concentration in fish (Bq/kg, w.w.) from the Barents Sea regions.

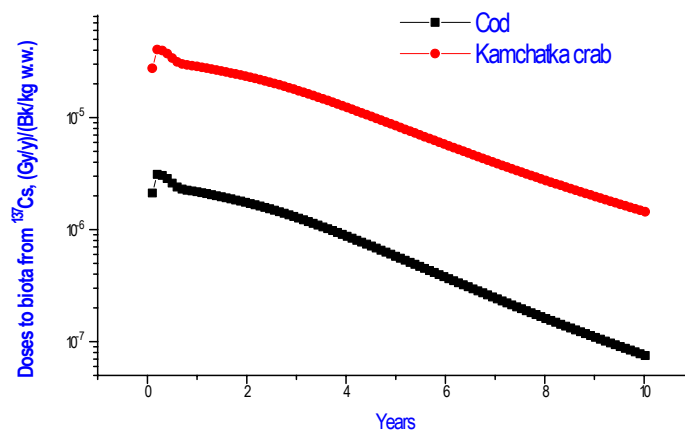


FIG. 6. Dynamics of the doses to biota ( $\text{Gy y}^{-1}/\text{Bq kg}^{-1}\text{w.w.}$ ) from  $^{137}\text{Cs}$ .



The absorbed fractions for characteristic energies of  $^{137m}\text{Ba}$  were calculated using a preliminary version of modelling tools [29], which can be easily used to calculate  $\phi$  for a wide set of geometries and energies. Preliminary intercomparison with results (for uniform distributions of photon-emitters) derived from Monte-Carlo calculations [17] provided satisfactory agreement.

Plots in Figure 6 were calculated using an assumption about negligible influence of  $\alpha$  and  $\beta$  radiation for external doses. External dose for  $\gamma$  radiation was calculated by assuming that the dose rate in the biota equaled that of the dose rate in the medium (in other words the organism is assumed to be infinitely small). The dose rate is calculated from the concentrations in the medium, seawater (cod) and seawater-sediment (crab), using equation (2) where  $\phi=1$ .

#### 4. CONCLUSIONS

The practical feasibility of the present approach for calculating dose-rates to flora and fauna is shown. The modelling approach can be used to simulate the dispersion of radionuclides in water, sediment and biota phases of the marine environment and the output can then be used to perform an “environmental” dose assessment. Information concerning the set of reference organisms for marine environment with internal and external dose conversion factors, which is under development, can be easily adopted by the present model. The potential of the present modelling approach is illustrated by calculations of doses for a selected set of the scenarios and reference organisms.

### ACKNOWLEDGEMENTS

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## **4. POSTER PRESENTATIONS**

# **Is environmental radiation protection an attempt to psychologically avoid confronting our own fear?**

**C. Seymour, C. Mothersill**

St. Luke's Institute of Cancer Research, Dublin, Ireland

**Abstract.** Fear is part of the natural human condition. It resides within all our psyches, but perhaps because we try to keep it hidden, and to pretend it doesn't exist, we do not recognize it as a powerful and motivating force. Refusal to acknowledge fear can lead to unrecognised bias in determining an acceptable paradigm.

The early pioneers of radiation work, and also those working with radiation prior to 1945 had no fear of radiation. The fear of radiation was caused by the atomic weapons deployed to end the war in Japan, and the subsequent nuclear arms race ending with the stalemate of mutually assured destruction. In order for this scenario to be convincing, horrific and often exaggerated messages of the awfulness of nuclear power had to be used.

As a result, many people have a fear of radiation. Many people who work with radiation have the same fear, and subscribe to the simplistic and unscientific belief that if large doses of radiation are demonstrably harmful all doses must be harmful. It is this which has driven the LNT model, when there is much evidence to suggest that low doses of radiation are not deleterious to man.

The suggestion is now being made in some quarters that it is necessary to examine the effects of radiation on species other than man. Why? If it is to examine mechanisms of radiation induced effects in different species, then there is scientific value. If it is to do a general trawl through different species in an attempt to find one that is singularly radiosensitive, it has propaganda but little scientific value. It has been suggested that radiation protection be less anthropocentric, but this is meaningless. We, as humans, cannot presume to decide that the greatest worry of a koala bear is radiation. The only assumptions that can be made is that as all animal behaviour involves eating, sleeping and sex, any environmental disturbance that prevents this is bad for the animal as an individual. As essentially omnivorous primates, it is not logical for humans as a species to concern themselves with the welfare of individual animals. The biggest danger to animals is human destabilization of a complex ecosystem. Chernobyl was bad for humans, but wildlife around the plant has flourished.

Advocating radiation protection policies for animals is a seriously flawed approach that is pandering to a human fear, and the logical instability that is a corollary fear, that allows serious causes of pollution and environmental destruction to be ignored.

We are all aware of what the term environment means to us. Although we like to think of it as nature, it is a nature that is particularly benevolent to us. We enjoy a controlled natural environment, the nice side of nature without the nasty. We cannot view nature with anything but an anthropocentric viewpoint. For this reason environmental protection policy cannot be anything but anthropocentric. Similarly an environmental radiation protection policy must be anthropocentric, and must reflect human aspirations and fears. We take upon ourselves the "duty" of deciding what is good for the environment and we define "harm" as deviation from our preferred state of perfection.

We all have a sense of fear. It is an emotion that is universal through all peoples, and can be useful in preserving bodily boundaries. We are wary if people "invade our space". We are naturally afraid of things that may cause damage to our boundaries. Fear is a natural survival response; although for it to work effectively for self-preservation it must be a rational fear. If I live in a small African village and am afraid of going out at night in case I get eaten by a lion, that is a rational fear. If I live in Darwin or London and avoid going out at night for the same reason, that is an irrational fear.

Irrational fear is also part of our lives. We fear the dark, or things that cannot damage our bodily integrity. We also have more vague fears – we fear the future, death, and the unknown. We fear that which we feel may overwhelm our bodily boundaries, or that which we felt as children may overwhelm us. We fear the dissolution of our boundaries, and the consequent dissolution of ourselves.

This theme is reflected in myth, for example that of Saint George and the dragon. In the story, Saint George is a valiant knight who rescues a young maiden from a hideous fire-breathing dragon. We are all aware that dragons don't exist, but it is more than just a story. The young maid represents the youthful, naïve state that is in danger from the dragon. The dragon represents those vague, horrific fears, which largely bubble undisturbed in the unconscious, but occasionally break through in dreams, or in irrational and extreme avoidance of non-dangerous objects such as spiders or mice.

Radiation has become a twenty-first century dragon. It has come to represent all the horrors of our collective unconscious. Radiation bombs were used to terminate the Second World War, but start and maintain the subsequent "cold war". The awesome destructive powers of the bombs at Hiroshima and Nagasaki became merged in the collective unconscious with all the other horrors of war, with terrible battles, with the blitz on London, the fire-bombing of Dresden, the appalling concentration camps.

This fear was consciously exacerbated by the nuclear arms race during the cold war. The public were continuously impressed by films and reports showing how amazingly destructive the nuclear weapons were. The balance of power was held using a policy of mutually assured destruction, and doomsday scenarios of nuclear winters were made. In this environment, nuclear weapons had to be so awful and so inhumane that they could never be used. The propaganda of their destructive power was much more important than an objective and scientific view.

Saint George in the myth represents a mature perspective, a rational view that can cut down to size irrational monsters. The question is whether the rational fear of radiation, which has been deliberately exaggerated and distorted as a matter of public policy, and then melded in the collective unconscious with rational fear of all aspects of war to produce the radiation dragon, can be separated.

The development of nuclear power, as a "good" use of controlled nuclear reactions always faced an uphill battle in the public psyche. To explain the production of good from evil is always a difficult philosophical problem. To confound the problem, if some kind of error occurred in the benevolent nuclear reactor, it behaved as though it were a nuclear weapon, causing damage through the force of the explosion and the resulting release of radionuclides.

Such a problem occurred at Chernobyl, where a nuclear reactor exploded during the course of an ill-advised experiment. The course of events at Chernobyl, and a measurement of the type and quantity of the radioactive particles released are now generally agreed in the literature. However, disputes are still occurring as to the effects of these releases. These problems are in part due to continuing disagreements as to the exact dosimetry of the exposed Hiroshima and Nagasaki populations, because of distance from the epicentre, type of building materials, and relative contribution of the neutron component of the dose. The population receiving the dose were all of one genetic sub group, and were physically malnourished because of prolonged starvation during the war years. In Chernobyl the population was also largely poor, but not as malnourished. Moreover, there were important psychological differences. In Japan it was considered somewhat shameful to be a radiation survivor, so any radiation effects tended to be minimised. The radiation survivors in Japan were examined each year following the bombing. Part of this examination included whole body x-rays, but the radiation dose from this procedure was not included in any calculations. Around Chernobyl, there was an incentive to exaggerate any bad health because of government compensation schemes, and generous western relief efforts. This situation led to an interesting dichotomy in the literature.

If a linear no threshold model of radiation risk is applied, there should have been more cancer cases following Chernobyl than were observed. The fact that there is a less than expected number of cancers is reported in all of the established scientific literature. The less scientifically established literature shows a large increase in what they term radiation syndrome type effects, which include any congenital abnormality or childhood genetically based disease.

The difference in radiation endpoints used reflects the general fear of radiation. The rational argument is that radiation can only be responsible for effects that have been proven clinically and experimentally to have been caused by radiation. The irrational fear says that radiation is so awful we really don't know all it can do, and so anything nasty that occurs after the radiation exposure must have been caused by the radiation. As the majority of people have an irrational fear of radiation, this perspective further fuels their fear.

As a result, the nuclear industry is one of the most tightly controlled and regulated in the world. Any radiation dose, no matter how small, is regarded as potentially harmful. This position would be regarded as absurd in the pharmaceutical and chemical industries, where the concept of threshold doses is used. The fact that even regulatory agencies are unwilling to use the concept of a threshold dose for the general public reflects the level of fear of radiation in the public psyche.

As the fear is largely unconscious, it is difficult to have a rational debate on it. The concept of relative risk only applies to the conscious mind, not the unconscious. If you have no unconscious fear of lions, you can appreciate that in Darwin you are at greater risk of being run over by a vehicle than being eaten by a lion. In your movements around Darwin you thus remain aware of the greater risk. If, however, you have an unconscious fear of lions, you remain more concerned with the imaginary risk of being eaten by a lion than the real risk of being run over by a car. If you have a fear of radiation, you are in an even worse position. As you know what a lion looks like, you can avoid it. But radiation cannot be seen, you don't know that it's there, and when you find out it may be too late to avoid the horrific consequences of a slow, lingering death. The effects of radiation, as well as radiation itself, cannot be easily observed. This complicates the fear of radiation with suspicion that radiation might be there. I can reassure myself there are no lions around, but I cannot reassure myself that there is no radiation around.

As I cannot succumb entirely to my fear of radiation I must oppose it. In doing so I am also opposing all the negative images of radiation – I am opposing war, death, destruction and pollution of the environment. Through an essentially negative action (opposition) I am doing something positive. Moreover, the fear of radiation is so psychologically embedded I can rationalise that I am doing something for the world as a whole, even though a logical analysis would suggest that more people are killed by (for example) forgotten landmines or contaminated water supplies.

There is now a suggestion that the effects of radiation on the environment should be more closely regulated. The question is why. All reasons emanating from man have to be anthropocentric – it is impossible for a fly to have the perspective of an elephant. The only logical reason is that we're happy with the amount and type of biodiversity existing on the planet, and don't wish to see it altered. This is getting very close to a fear of the unknown, of change. Logically, radiation could cause new and exotic species to bloom as it alters the environmental balance. Equally, we are all aware that the balance of the environment is constantly shifting, and that nothing can remain constant over time.

It is also logically apparent that it is the initial construction of factories and plant that destroys the local environment. By the time the plant comes into operation, much of any environmental damage has already been done.

It is obvious that man himself, and by extension his activities, is the major polluter of the environment. To concentrate exclusively on one of man's myriad activities as being the most important in terms of global pollution, because of an illogical fear of that activity, is illogical and counterproductive. It is counterproductive because it allows far greater environmental damage from less regulated industries – everyone remembers Chernobyl but nobody remembers the far larger tragedy of Bhopal.

However, as Dr Woodhead has sagely remarked, just knocking or attacking the views of others is not constructive. It is not the purpose of this paper to say that radiation has no negative environmental effects. Rather it is an attempt to put the hazards of radiation in perspective. If the environment is to be protected, it should be protected equally from all types of environmental hazard. The only way to do this is using an additive or integrative risk model. Within this model, a threshold maximum permissible environmental burden is set for a designated land use, for example amenity or residential or industrial. It is possible to set different thresholds for different land uses. The trade off is between many small polluters or one large polluter. Instead of radiation being treated as a unique polluter, it is treated as one of many polluters. Instead of having maximum permissible discharges for radiation, for arsenic, for lead and for many other chemicals, there is a blanket maximum pollution burden, or threshold. This can be fully utilised by radiation, or arsenic, or by any other pollutant. Once the threshold is reached from any single or combination of sources, then no more discharges of any pollutant are allowable.

The difficulty with this method is in agreeing comparable endpoints in measuring toxicity, or damage to the environment. The advantage of this method is that it allows a permissible level of environmental stress only, and can be further refined by allowing for synergistic interactions as well as additive ones.

To treat radiation as an environmental hazard that can be viewed in isolation is giving in to the unconscious dragons of the mind. Saint George, representing the conscious mind, protected by his armour of logic and rational thought, must do battle with the dragon. His objective though is not to kill the dragon, but to wrestle it from the unconscious to the conscious, where it can be critically examined. It is my belief that a critical examination of radiation will reveal that legislation for environmental radiation protection should not be isolated, but should be part of a systemic environmental legislative programme.

# The justification for developing a system of environmental radiation protection

A. Janssens<sup>†</sup>, G. Hunter<sup>‡</sup>

European Commission, Luxembourg

**Abstract.** This paper presents concluding points relevant to the question whether (and why) it is justified to spend resources for the development of a system of environmental radiation protection.

The paper puts the issue in historical perspective to radiation protection philosophy and to current trends in ecology and “green” politics. It also looks at the issue of scientific rationality, paradigms and beliefs.

Among the arguments for the development of a System the paper considers public concerns, integration in overall environmental policy, uncertainties and sustainable development.

The paper also examines the prospects for the development of a System within the EU and offers some general considerations on the scope of environmental protection.

## 1. SCIENTIFIC RATIONALITY

### 1.1. Radiation protection

Radiation protection has evolved over a century in response to increasing knowledge of the health detriment resulting from exposure to ionizing radiation. In the beginning of the 20<sup>th</sup> century the concern was mainly with the protection of radiologists against directly observable health effects. After the 2<sup>nd</sup> World War, with increasing application of fission energy and the use of artificial radionuclides in medicine and research, it also became apparent that low levels of radiation were a cause of cancer and possibly of genetic damage. This not only caused dose limits for workers and members of the public to be lowered, but also the development of a unique approach to the limitation of lifetime risks.

The key factor in this development was the assumption that while radiation-induced cancers were not certain to occur, the probability of occurrence is proportional to dose. There is no dose threshold for “stochastic” effects. While there was little evidence to demonstrate the absence of a threshold, the understanding of radiation effects at cellular levels and of cancer induction, prevailing at that time, made the absence of a threshold the most probable scientific hypothesis. Today, with a much-improved level of knowledge in cellular and molecular biology, the hypothesis is still defended by most radiation experts, even though it is not undisputed.

At the time of adoption of the hypothesis by ICRP this was a precautionary measure in view of the uncertainties in stochastic effects, essentially applying the “Precautionary Principle” even before it had been formulated. Similarly, while there was considerable uncertainty as to the scope of the dose-effect curve, it was agreed to adopt a linear relationship. Despite the evidence of other curve shapes (linear-quadratic) depending on biological effect, dose rate, radiation quality, the uncertainties on the “true” shape made it, for regulatory purposes, much more convenient to adopt the linear hypothesis, corrected with radiation quality factors and a dose/dose-rate correction factor (DDREF). The hypothesis indeed allowed the straightforward link between a quantity to express exposure (absorbed dose, Gy) and a quantity to express the health detriment (effective dose, Sv). It also allowed the definition of the powerful concept of collective dose, and its corollary, optimisation of protection on the basis of cost and benefit.

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<sup>†</sup> This paper reflects the authors' personal opinions as a starting point for further discussion, it does not represent in any way the position of the European Commission.

<sup>‡</sup> Seconded to the European Commission from the Scottish Environment Protection Agency until 1 June 2002.



A very good overview of the historical development of radiation protection has been presented by the Chairman of ICRP, R. Clarke, on many occasions over the past few years. It is indeed important to know the history in order to understand the rationale of new developments, and ICRP now acknowledges explicitly the societal and ethical considerations underlying the adoption of its scientific rationality for protection against a health detriment.

## 1.2. Level of protection

Among the societal factors one should also understand those used to determine the adequate level of protection, which is different for workers and for members of the public. In the system for dose limitation dose limits merely define an upperbound to the “tolerable” region, in the same way as dose constraints are set on the basis of equity considerations.

The difference between the dose limits for workers and members of the public (a factor 20) can be explained on grounds of the greater benefit of practices for workers than for members of the public, and the voluntary character of employment. There is also a difference in nature of the exposure: workers are to a large extent in control of their own exposure, and doses are monitored individually. Members of the public have very little control of their exposure, and one should not expect them to adjust their behaviour for the presence of radioactivity in the environment.

The degree of control of one’s exposure, to radiation or to other hazards, is an important factor in terms of acceptance. This also explains why some radiation experts believe children deserve better protection than adults. While individual doses are calculated for children as for other age groups on the basis of age-specific dose coefficients for internal exposure, risk coefficients for children are not expected to be significantly different from adults, even taking into account the longer integration period (“residual lifetime”) for the expression of cancer (ignoring biological repair mechanisms). Others advocate that for low levels of exposure children are not at risk as children, but later in life as aged adults, and that it is the lifetime exposure that matters, not annual dose. However, society does expect a higher level of protection for children, not necessarily because they are more vulnerable, but above all because they should not develop in an environment where they, or their parents, should fear any pollutant and should feel no need to adjust their behaviour accordingly. Adults on the other hand can accept a certain level of risk, to their own life, and can make an effort to understand the risk properly and to learn to live with it.

Fear of radiation is an important societal factor. It can be explained by historical facts such as its association with nuclear weapons, socio-political factors such as aversion to and distrust of powerful industries with high capital investment, political factors such as the need for far-reaching security measures (enhanced since 11<sup>th</sup> September). It can also be explained by the fact that radiation is not subject to observation by the senses, but on the other hand can very easily be detected by suitable equipment. The penetrating power of (gamma) radiation is also something of which we have little experience and while x-ray examinations are common today they still provoke, in many people, a sense of fear.

This is true for most people, in general with a rather poor scientific background, be it members of the public or workers in nuclear industry. Workers get to understand their radiation environment and fear decreases with familiarity. The innate fear nevertheless remains, even in educated workers, and familiarity with the risk may then be cause of dismissal. Those who have worked in nuclear industry, research, or radiology departments, will have known colleagues who developed a careless attitude towards radiation and working with radioactive substances. We believe safety culture has developed very much over the last two decades, but it is our belief that it was rather poor in the seventies. The dismissal of radiation or the repression of concern for radiation is just as irrational as the fear, sometimes exacerbated, of certain members of the public. It may even affect, for some of us, the scientific perception of the evidence on low-level radiation detriment. There is indeed no scientist whose “judgement” is free of values and emotions, even though we believe most scientific “facts”, used to form those judgements, to be free of such influences.

Radiation Protection is in part an exact science, but in part it reflects societal judgement and is thus a social (and economic) science. The societal aspect is, we think, even more prominent now that its scope is extended to biota, or the environment *per se*. Recognising this is however not admitting that

this is a “seriously flawed approach that is pandering to a human fear” (C. Seymour, this conference). It means that we must make an effort to understand the rather recent environmental concerns in our society and uncover the judgmental framework that could be applied to environmental radiation protection, in line with modern ecological thinking.

### **1.3. Environment protection**

The concern for environmental issues is today part of mainstream public opinion. While this is a fairly recent phenomenon starting some 50 years ago, the roots of this development are older and very diverse. It correlates with other changes over the last centuries in views on society, religion, and with the extraordinary impact which science and technology has had on society and on the environment. Environmentalism is often in opposition to science, ranging from belief in an irrational truth which a reductionist science cannot attain, to scepticism about natural sciences’ historical optimistic belief that they would ensure man’s supremacy over nature and that any problems resulting from science and technology would be resolved by science.

The explicit inclusion of protection of the environment in the scope of radiation protection reflects this trend, and it is amazing to see the rapid spread of the idea to abandon the “anthropocentric” paradigm focussing only on human health which has become, if not yet part of mainstream radiation protection, at least very fashionable.

Many experts are not convinced however, and view this development as a contamination of science by irrational myths and beliefs, and sentimental concern for the well being of biota, extending from pets to “loveable” wild animals. The conservative view has merits, after all there is as yet no evidence that radiation and environmental levels of radioactivity have caused any observable detriment to biota, but it ignores the recent advances made in defining the scope of environmental protection.

The idea that animals deserve a level of protection similar to man has rapidly faded. Its sentimental basis is similar to the concern for children, innocent creatures exposed to evil residues of mankind’s technology. It is true that the “koala bear’s worry is not radiation” (C. Seymour, this conference), in fact he is unaware of ionising radiation, and an animal-centered protection philosophy would indeed be an anthropocentric projection of man’s psyche.

The scientific thinking about environmental radiation protection has shifted to an ecosystem approach. This is more appealing to scientific objectivity. It cannot be disputed that mankind is part of a complex ecosystem and that the perturbation of the latter may be detrimental to man. In this way value can be assigned to the preservation of the present ecosystem, and such value can be incorporated in the overall radiation protection philosophy (e.g. optimisation on the basis of the value of human life and the value of the ecosystem).

Environmental radiation protection is thus again solidly anchored in scientific rationality. However, as we will discuss later, there is a risk that this will once more lead to a reductionist approach – giving good answers to the wrong questions. At this stage it is important to recognise what the driving forces are behind the development of a system of environmental radiation protection.

## **2. DRIVING FORCES**

### **2.1. Public concern**

Levels of radioactivity in the environment rank high among public concerns. This is often part of an overall rejection of nuclear energy, which is today a feature of mainstream public opinion, shared by an important part of the scientific community.

There are other societal grounds for this rejection, rooting in an overall “green” opposition to large-scale technology, placing an industrial, economic and political consortium beyond democratic control. Renewable (small-scale?) energy sources are on the other hand regarded as a democratically-owned technology [1]. While other grounds such as non-proliferation, safety, the absence of facilities for final disposal of radioactive waste, are also important, environmentalists focus very much on the discharges of radioactivity to the environment to dispute the justification of nuclear facilities. Levels of radioactivity in the Irish Sea from reprocessing at Sellafield envenimate the relationship between the

UK and Ireland, and even Norway is opposed to Sellafield in view of the very low levels of Tc-99 observable along their coast.

The political emphasis on environmental radioactivity builds upon widespread belief that it has dreadful consequences not only for public health but also for the environment. Leukaemia clusters around nuclear installations, and other allegations of health effects on the population, have required careful epidemiological and environmental analysis [2] which identified no causal link with environmental radioactivity. Despite these efforts people continue to believe that there must be something wrong.

Discharges of radioactivity with effluents from nuclear installations have been substantially reduced since the seventies. While many would view this as an exemplary application of the ALARA principle (doses should be As Low As Reasonably Achievable), the high cost of measures taken to clean the effluent is in fact rarely proportionate to the health detriment, whatever reasonable value is taken for the value of a human life.

Further reduction, asked for by society, can thus not be justified from a health perspective, so that the environmental perspective may become equally important.

## **2.2. Integration in overall environmental policy**

As discussed in the introductory chapter 1, radiation protection has a long history, and many experts in this field, including the authors of this paper, have been driven by the belief that their scientific and societal approach was exemplary for other environmental pollutants. In the meantime risk-based management has been developed for other pollutants and targets have been set to a level of ambition for human health comparable to or beyond the dose limits for ionizing radiation. Nevertheless it is fair to say that in terms of health protection we have a very good standing, in particular with regard to comprehensiveness and reliability of risk assessment.

For chemicals, and in particular for pesticides and other persistent organic pollutants, the health perspective has never obscured the concern for the environment. In fact, in many cases, it was the observation of detriment to biota which prompted attention for specific pollutants.

Environmental policy and radiation protection have evolved in isolation. To a certain extent this may be related to the specific features of ionizing radiation which made it essentially a discipline for (nuclear) physicists rather than biologists, but it was probably caused much more by the link with nuclear technology, for energy or weapons production, and funding being provided in this context, as well as radiation protection being subject to a regulatory framework which in most countries was autonomous and kept separate from other departments.

This “splendid isolation” has the drawback that those working in environmental protection usually ignore the achievements of radiation protection. The dialogue is difficult, not only because there is an important bias from both sides, but also because the assessment methodologies have developed in different ways. The most striking anomaly is that radiation protection ignores the concern for biota. It is important that the methodologies converge, and this implies that radiation protection of biota must be addressed

## **2.3. Precautionary principle**

Despite the outstanding scientific basis of radiation protection it is *a priori* assumed that there are uncertainties, in particular where the assessment relies on models (metabolic, ecological) and has a poor experimental and epidemiological basis. The uncertainty will vary with different radionuclides, depending on their chemical properties, decay time, type of radiation emitted. Assessments of uncertainties are rarely made in radiation protection, which is often justified by the fact that doses are so far below limits that, with any reasonable range of uncertainty, compliance with limits is ensured.

Uncertainty is in itself not a sufficient ground for invoking the precautionary principle, in such cases where there are no grounds to expect serious and irreversible detriment to result from ignoring these uncertainties. For human health the precautionary principle is by some put forward in view of new developments in the understanding of radiation detriment at cellular and molecular level (genome

instability, bystander effect, ...). While these effects are indeed not yet fully understood, the absence of epidemiological evidence is a strong argument to support the absence of severe effects.

It is true that there is no evidence of any severe impact of ionizing radiation on biota. However, uncertainties in the assessment of doses to biota are probably much more important than for doses to humans. While most biological species are, for acute exposure, much more resistant to radiation than humans, there is little or no information for chronic exposure. The focus of radioecology research on pathways of exposure to man has yielded a lot of information on edible species, but may have missed out very high concentration factors for certain nuclides in other species. Finally, the metabolism of many species is not well known, radiation-weighting factors for effects on biota need to be investigated and detriment in terms of fertility and genetic effects could be quite different from humans.

It is important that the knowledge base of radiation protection be properly and fully extended to include biota in order to be able to decide whether the precautionary principle may, or may not, apply.

## **2.4. Sustainable development**

Sustainable development has become the leading principle of environmental protection. For protection of human health we have already a powerful instrument: collective dose is not only a measure of the overall impact per year of practice, it also allows one to project the cumulative impact if the practice was to be continued over a long time period and to extrapolate e.g. over energy production.

For long-lived radionuclides collective doses can be important because the biological availability may reduce only slowly with time, as opposed to biodegradable chemicals. The persistence of availability may apply in particular to marine biota, sharing the main environmental compartment into which radionuclides are discharged. On the other hand the concept of collective dose is closely related to the no-threshold hypothesis, which for biota may not be applicable.

While there is a tendency to dismiss the concept of collective dose to members of the public (keeping it merely to optimise occupational protection or to choose between options), there is instead merit in expanding it to a new concept of “environmental collective dose”, comprising man and biota.

## **3. PROSPECTS IN THE EUROPEAN UNION**

### **3.1. Water Framework Directive**

At the time of the previous conference in Ottawa, the Council Directive “establishing a framework for Community action in the field of water policy” was still at the stage of a Commission proposal. The European Parliament had proposed an amendment to the effect that the scope of this Directive be extended to radioactive substances (as was the case for the Directive on Drinking Water). The Commission was in favour of including this amendment, but faced the problem that a legislative act could not have a dual legal basis, i.e. both the EC Treaty and the Euratom Treaty, in particular because the procedures for adoption are quite different under the two Treaties (specifically that the European Parliament has no codecision powers under the Euratom Treaty). In addition, while the Commission’s Legal Service explained that the Chapter III of the Euratom Treaty (“Health and Safety”) related to human health, not to the environment *per se*, there was concern that measures taken under the EC Treaty would not be coherent with those taken under the Euratom Basic Safety Standards, and it was felt that one way or another the Group of Experts established under Article 31 of the Euratom Treaty should be in control of both aspects.

Eventually the Directive [3] was adopted without explicit reference to radioactive substances but with the silent understanding that they could be regarded as pollutants as far as biota are concerned.

On the one hand this development does not allow us to move forward rapidly. On the other hand, even if it was argued at the time of the Ottawa conference [4] that the procedure for the setting of chemical quality standards could be applied in practical terms to radioactive substances, it was also clear that it would offer a poor scientific basis. At the conference in Ottawa the starting blocks were laid for a major research programme to develop a scientific basis for the protection of the environment, in the meantime known as the FASSET project.

### 3.2. OSPAR

The European Commission plays an active role in the OSPAR Convention (Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic). On 22–23 July 1998 (Sintra) the Ministerial meeting agreed on a strategy for radioactive substances. The goal is to achieve by the year 2020 a reduction of discharges of man-made radioactive substances so that concentrations in the marine environment are “close to zero”. To this end the OSPAR Commission will undertake the development of environmental quality criteria and report on progress by 2003.

The deadline of 2003 was very ambitious, and today it seems very difficult to meet. The definition of “close to zero” is not yet settled (concentrations, doses?) and there is at this stage no accepted methodology for taking into account “radiological impacts to man and biota, technical feasibility and legitimate uses of the sea”.

To help OSPAR reach its objectives the European Commission has launched a comprehensive study of levels of radioactivity in the North Atlantic. This MARINA II study is an update of a similar study in the eighties, and will in particular serve as a baseline for judging progress towards reaching the OSPAR objectives. The various working groups under MARINA include one looking into effects on biota. Another new feature of the project is that stakeholder NGO’s are invited into the process. The project is intended to be finalised before summer 2002.

### 3.3. Environment and Health

The Radiation Protection Unit of the European Commission is part of the Directorate General for Environment. After the DG lost the administrative competence in the area of nuclear safety and radioactive waste management, radiation protection was incorporated in a new “Environment and Health” Directorate. The objective of this new structure was to allow more emphasis to be put on the “Health” dimension of environmental policy e.g. with regard to air pollution and chemicals. For radiation protection it is an opportunity to develop its “Environment” basis.

As part of the sixth Environmental Action Plan (6 EAP) a strategy paper on Environment and Health is being elaborated. While being closely associated to this work, we are again facing the problem that the 6 EAP, adopted under the EC Treaty, does not include the activities under the Euratom Treaty (it would merely be understood that the chapter “Biodiversity” applies to radioactivity).

In order to go forward from to this uncomfortable situation we have now started drafting a “EURATOM-EAP”, which would address both health protection (workers, members of the public, medical applications) and environmental protection.

## 4. OVERALL PERSPECTIVES

The concluding remarks in this chapter are not intended to give a comprehensive view on the way forward, but merely to point to a number of specific issues and new developments which have not previously been discussed.

There is an important need to set environmental quality criteria for radioactivity. While this concept does not fit very well in the dose limitation system for radiation protection, it is an important component of environmental policy in other areas. There is political pressure (e.g. under OSPAR) to establish EQC. We would also hope that the introduction of EQC would help in communicating with the public, since the concept is much more familiar and understandable than the concept of “effective dose”.

It is worth noting that there is a new quite similar development at international level, essentially driven by IAEA, to define simple sets of “scope defining levels”. This development builds upon the earlier work on exemption and clearance levels which were incorporated in the EU Basic Safety Standards and for which guidance has been offered [5]. The SDL’s are important for international trade, but the most important objective is to define the boundaries of the scope of radiation protection. Despite the problems to incorporate commodities, food, artificial and natural radioactive substances, in a coherent set of values, there is hope that SDL’s will allow communication with the public and the industry in a less elaborate manner.

The rationality of the SDL's is based on the concept of "trivial dose", with a default value of 10  $\mu\text{Sv}$  per year for artificial radionuclides. The concept breaks down however for natural substances, for which EC guidance introduced a default value of 300  $\mu\text{Sv}$  per year, but others prefer an approach based on the natural range of concentrations rather than doses (an approach which does not make sense for foodstuffs). It is also recognised that despite SDL's for trade in food, consumers would prefer uncontaminated food, and there may be little scope to put foodstuffs from a contaminated area on the international market.

This is an important consideration also with regard to EQC. If consumers reject e.g. fish contaminated by discharges of nuclear industry this may in turn be regarded as an environmental degradation in terms of preserving "legitimate uses of the sea".

While EQC should "scientifically" be based on the impact on man and biota, societal reality is that simple consideration in terms of concentrations, not dose, will prevail. People put high value on a clean environment, irrespective of health consequences. Where pollutants are observable to human senses (smell, colour), they are not regarded acceptable. For radioactivity and radiation this is not the case, but where it is detected by technical sensors it is cause of similar aversion. This is a plea not to overemphasise scientific rationality based on the protection of biota, and to set EQC also for the physical environment ("air, water and soil" as in Chapter III of the Euratom Treaty, Articles 35–37).

The powerful concept of "effective dose", for man, and the appeal of "dose to biota" (still to be defined) should not hide the fact that while in terms of physics and biology "a Gray is a Gray", irrespective of its origin, this is not true for the Sievert. The Sievert is an expression of risk or detriment, and for the acceptability of risk there is no universal figure. This explains the distinction between natural and artificial radioactive substances, and needs to be taken into account in the scientific basis for the protection of the environment as well.

In conclusion we would like to warn against making the same mistake as with the protection of man against ionizing radiation. A scientific methodology for the protection of biota may be expected to lead to the conclusion that at current environmental levels there is no detriment. This result may by some be regarded as an important achievement, by others it may discredit the whole move towards environmental standards as being opportunistic and merely serving the interests of nuclear propaganda.

It is crucial to take people's real concerns into consideration and to answer the actual questions, not merely those for which there is a scientific answer. The involvement of stakeholders in the process is therefore essential, and the European Commission endeavours to follow this route, starting with a stakeholders conference on its policy with regard to environmental radioactivity in December 2002.

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## **Hanford's West Lake and the Biota Dose Assessment Committee's screening methodology\***

**E.J. Antonio, G.W. Patton, B.L. Tiller, T.M. Poston**

Pacific Northwest National Laboratory, Richland, WA, United States of America

**Abstract.** Radionuclide data collected at the Hanford Nuclear Reservation's West Lake (a small seepage pond) was used to test the utility of the U.S. Department of Energy's Biota Dose Assessment Screening Methodology for protection of plants and animals from ionizing radiation. The Hanford Site environmental surveillance database was queried for all 1999 data on radionuclides in surface water, soil and sediment. Maximum and mean radionuclide concentrations were determined for each combination of radionuclide + media. These data were entered into the Biota Dose Assessment Committee's "Biota Concentration Guide Calculator", a semi-automated tool used to determine compliance with proposed biota dose limits for shorebirds (riparian animals with a proposed dose limit of 0.1 rad/day). Concentrations in the aquatic environment did not pass the screen. Further review of the data indicated that a water sample taken from the lake exceeded the limiting water concentration for uranium. As a consequence of the assessment, a limited biota monitoring effort was established to determine the potential exposure of wildlife at West Lake. The effort involved determining residence time of breeding shorebirds and actual tissue burdens of radionuclides in the birds. In the final analysis, West Lake was determined to be in compliance with proposed radiological dose limits. The application of this methodology has helped to identify a potential exposure problem and provide direction for the sampling program.

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\* Only an abstract is given as the full paper was not available.

# Aboriginal participation and concerns throughout the rehabilitation of Maralinga

P.N. Johnston<sup>a</sup>, A.C. Collett<sup>b</sup>, T.J. Gara<sup>c</sup>

<sup>a</sup> Department of Applied Physics, Royal Melbourne Institute of Technology, Melbourne, Australia

<sup>b</sup> Murray Chambers, Adelaide, South Australia

<sup>c</sup> Department of History, Adelaide University, Adelaide, South Australia

**Abstract.** Maralinga was established as a permanent range for the British nuclear weapons testing program in 1955. From 1955–1963 it was contaminated by numerous toxic materials including various radioactive substances. There was a Royal Commission into the British Tests in 1985 followed by a range of technical studies (1986–1990) to recommend options for the rehabilitation of the Maralinga range and a major rehabilitation program for the site (1995–2000).

Since 1955, the Maralinga people, who are traditional owners of the land, have been excluded from parts of their land and since the early 1980's they have pursued claims to regain their lands in a safe condition. This paper addresses the contributions of the Maralinga people to rehabilitation of the site beginning with their representation at the Royal Commission into British Nuclear Tests in Australia, followed by assistance with scientific studies to determine the rehabilitation options and through the lengthy consultation process during rehabilitation. Each phase of this participation caused new issues to be addressed. Differences arose between the Australian Government and Maralinga Tjarutja but the rehabilitation is now effectively complete and it is timely to review the stakeholder issues and the consultative process. Post-rehabilitation land management issues are at the centre of ongoing negotiations.

## 1. BACKGROUND

The rehabilitation of the former nuclear test site at Maralinga is almost complete and it is timely to review the stakeholder participation issues with respect to the rehabilitation and seek to learn lessons about the consultative process.

Maralinga was established in 1955 as a permanent range for nuclear testing. The establishment of the range together with the existence of the Woomera Defence Reserve had the long-term effect of breaking the traditional communication paths between Aboriginal people isolated south of Maralinga at Yalata and those to the north.

Following realignment of US–UK co-operation in nuclear weapons development in the late 1950s and the atmospheric test ban treaty in the early 1960s, testing at Maralinga ended in 1963 and the range was permanently closed in 1967. In 1964 and 1967, British military operations Hercules and Brumby were mounted to render the range 'safe' from the viewpoint of a secured site.

In the early 1980's there were strong moves by traditional Aboriginal people from the Maralinga region to gain title to the lands adjacent to the Maralinga test site (section 400) from the South Australian Government. Substantial land rights were granted in 1984 by way of the Maralinga Tjarutja Land Rights Act.

At about the same time there was the 'discovery' that the range was not as safe as the United Kingdom had indicated to Australian Authorities. These discoveries in conjunction with strong public pressure from nuclear veterans groups and a sympathetic Government resulted in the establishment of the Royal Commission into British Nuclear Tests in Australia in July 1984. Presided over by the Hon. James McClelland, the Royal Commission conducted a broad-ranging enquiry into the conduct and management of the nuclear tests in relation to health and safety issues and, in particular, examined any adverse effects on "Aboriginals and other civilians". The Royal Commission took evidence from scientists, veterans, Aboriginal community groups and other people throughout Australia and also sat for several weeks in Great Britain.



The foreword of the Technical Assessment Group (TAG) report summarises well the outcome of the Royal Commission into British Nuclear Tests in Australia and Australian Federal Government response [1]:

*With respect to the clean-up of the Test Sites the Royal Commission concluded in part:*

*The treatment of the plutonium contaminated areas was inadequate, based on the wrong assumptions, and left the area in a more difficult state for any future clean-up.*

*The most significant hazard to Aborigines using the test sites is from plutonium contamination. The hazard from the inhalation of dust raised by wind appears to be acceptable. However, three other pathways – inhalation by children digging and playing, ingestion through bush foods and injection of plutonium – do produce unacceptable levels of risk.*

*It is clear that more information is needed on the possible Aboriginal lifestyles in the area, the dust conditions in the Aboriginal camps, the types and amounts of specific food items and amounts of plutonium in these food items. Information on the particle size distribution of plutonium contamination is also important and needs to be determined.*

*The plutonium-contaminated areas must be cleaned up. However more work is needed to develop realistic hazard assessments so that criteria can be derived for the clean-up.*

The Royal Commission into British Nuclear Tests in Australia [2] recommended a Maralinga Commission be established to determine clean-up criteria, oversee the clean-up and co-ordinate all future test site management. It suggested membership of the Maralinga Commission should include representatives of the traditional owners, the UK and Australian Governments and the South Australian Government.

The Australian Government responded to these recommendations by forming in February 1986, a Technical Assessment Group (TAG) to address the technical conclusions stemming from the Royal Commission and a Consultative Group was formed as a forum for discussion of the program. TAG's task was to provide the Australian Government with options for rehabilitation rather than a recommendation. Membership of the Consultative Group was as envisaged by the Royal Commission for the Maralinga Commission but with additional representatives of the West Australian Government.

Notably this structure which formed the basis for the entire rehabilitation project left the traditional owners and the South Australian Government out of direct decision making. It ensured that real authority remained with bureaucrats within the Department of Primary Industries and Energy which obtained advice from TAG and later the Maralinga Rehabilitation Technical Advisory Committee (MARTAC). It allowed the South Australian Government to not have any real policy regarding Maralinga for more than a decade.

The story of Aboriginal participation in radiological issues really begins through Aboriginal representation before the Royal Commission.

## 2. PARTICIPATION IN THE ROYAL COMMISSION

Aboriginal interests were represented before the Royal Commission into British Nuclear Tests in Australia by two counsel, Geoffrey Eames and Andrew Collett. They prepared a substantial submission on behalf of all Aboriginal groups [3]. Historical research into Government records undertaken as part of that submission indicated that measures taken by the Australian authorities to locate and remove Aboriginal people from the prohibited areas were very effective. During its visit to Maralinga in April 1985, the Royal Commission heard oral testimony from Aboriginal people that they had continued to live within and travel through the prohibited area during the time of the nuclear tests and the subsequent "minor trials", unaware of the potential danger from radioactive contamination. One well-documented incident involved an Aboriginal family who, in May 1957, were found by military personnel camped close to a site where a nuclear weapon had been exploded in October the previous year. The family was taken away for immediate decontamination at Maralinga Village and were then trucked south 200 km to be resettled at Yalata Reserve.

During the Royal Commission, Maralinga Tjarutja strove to obtain and have made public as much information as possible about the then residual contamination at the test sites. Maralinga Tjarutja also pressed the Royal Commission to make recommendations for compensation for the traditional owners who had been kept away from the Maralinga lands during the tests and for an appropriate assessment to be performed of the risks to Aboriginal people living their traditional lifestyle at the test sites. The Royal Commission made recommendations in these terms.

### 3. TAG STUDIES

The strategy used by the TAG was to commission five studies in the areas of anthropology, radioecology, bioavailability, inhalation hazard assessment and radiochemical and chemical analysis. All of these studies except the anthropology study were conducted by scientists who produced quantitative results and had little to do with the Aboriginal people on the Maralinga lands.

Probably the biggest obstacle to TAG estimating potential doses to Aboriginal people was a lack of good information about lifestyle and environmental setting of the people who would reoccupy the Maralinga lands. The traditional Aboriginal owners of the Maralinga lands had recently established an outstation, essentially a camp, at Oak Valley following the Maralinga Tjarutja Land Rights Act (1984). An anthropological study conducted by Kingsley Palmer and Maggie Brady was used to provide information on lifestyle diet, dust raising activities, methods of food preparation and consumption, practices for the treatment of wounds and skin sores and generally on any other practices with potential for ingestion inhalation or absorption of soils. This necessitated the anthropologist's spending a good deal of time with the Oak Valley community observing behaviour as well as making attempts at quantification of food consumption. Consumption of store foods was comparatively simple to estimate however consumption of indigenous foods "bush tucker" was far more difficult. The anthropologists were not used to performing detailed quantitative analysis of ingestion and inhalation related quantities and in many cases it was totally impractical to obtain good estimates. Red kangaroo forms a significant part of the diet of the Oak Valley people and the anthropological study estimated 0.6 kg per person per day of red kangaroo meat consumption.

More importantly than issues of ingestion were issues of inhalation. By far the largest source of dose by inhalation was due to life in the camp situation. It was essentially impossible to estimate dust loadings with much accuracy without unreasonable interference in the life of the people at Oak Valley.

The TAG report presented a wide range of options from the fencing of a large area to soil removal. The TAG report was not without controversy. It recommended that a criterion for rehabilitation be the annual risk of fatal cancer following the inhalation or ingestion of contaminated soil should not exceed one in 10,000 by the 50th year. This corresponds to an annual committed dose of 5 mSv and TAG suggested that this be the borderline between acceptable and unacceptable risk for the Aboriginal people living in a semi-traditional lifestyle who would reoccupy the Maralinga lands. This was based in part on the premise that the Oak Valley community would maintain its present size and continue to live in a semitraditional lifestyle for several generations. In that case it was estimated that the incidence of fatal cancer would amount to one or two per century from an annual committed dose of five mSv. This was also based on life expectancy data that showed that average life expectancy for Aboriginal people in South Australia was typically of the order of 50 years.

TAG acknowledged that there were substantial uncertainties in quantifying the Aboriginal lifestyle. It was considered that the acceptable level of contamination should not be reduced on this account. Rather a trade-off is seen between these uncertainties and over-estimation of the occupancy factor.

At the end of the technical assessment, Maralinga Tjarutja convened its own conference of expert advisers in 1990 and these included Professor Kohn from Harvard University, Dr Paretzky from GSF and one of the authors (PJ). The conference convened for four days and as a group had discussions with the South Australia Government, members of the TAG and the anthropologists. The advisers were in agreement that the most contaminated areas had to be remediated, discussion focussed on whether Maralinga Tjarutja should press for the stripping the soil from the 120 km<sup>2</sup> identified as exceeding the TAG criteria or whether a restricted area would be more appropriate. In many parts of this area the soil was only 10 cm deep. The environmental damage of denuding such a large area of soil was rejected as unreasonable and the Option 6 soil removal proposal was accepted.

The conference of expert advisers recommended to Maralinga Tjarutja and they accepted that Option 6(c) as it was most reasonable option presented, the option adopted by the Australian Government and advocated by TAG.

In the light of later experience, it is worth mentioning that Option 6(c) was a variant of Option 6(a) and both form part of the later story and so brief outlines are presented. Option 6(a) involved:

- removal of the soil that had been treated (ploughed during Operation Brumby in 1967) which would be transported to a burial trench . The previously treated area was 2.1 km<sup>2</sup>, and included an estimated 400,000 m<sup>3</sup> of soil,
- construction of a burial trench for disposal of soil
- exhumation of the formal pits at Taranaki, TM101 and Tietkens Plain Cemetery and burial of the contents in a sub trench [of the soil disposal trench]
- exhumation the other numbered pits and burial of the contents in a sub trench[of the soil disposal trench]
- importation of clean soil to cover and revegetation of the denuded area
- sorting through the unnumbered pits and disposal of contaminated items
- construction of a fence of length 112 km to denote the area where permanent occupation could lead to a dose in excess of 5 mSv.

Option 6(c) was similar in scope to Option 6(a), except that the pits at Taranaki, TM101, Tietkens Plain Cemetery and all other numbered pits are treated by in situ vitrification (ISV). Option 6(b) was another option involving concrete grouting of pits in situ.

This ISV technology was experimental but it offered good encapsulation of Pu buried in shallow pits and was strongly supported by TAG, especially the chairman, Des Davy.

#### 4. LONG TERM RELATIONSHIPS WITH SCIENTISTS AND PUBLIC SERVANTS

The TAG studies brought Maralinga Tjarutja into contact with scientists from the range of organizations, in particular ARL and ANSTO as well as bureaucrats from DPIE especially Pat Davoren, Rob Rawson and Members of TAG, Des Davy and Keith Lokan. This was the beginning of long standing personal relationships which were very important in the development of trust between Maralinga Tjarutja and those charged with rehabilitating Maralinga. The focus of these studies was Pu contamination at the Taranaki site where the UK had undertaken Vixen B 'single point initiation' minor trials. The trials were described as minor because they had little or no nuclear yield. These trials did cause the fissile material from nuclear weapon cores to be dispersed around the site and onto adjacent lands.

#### 5. MARTAC

Following general agreement to pursue Option 6(c) and negotiation of financial arrangements with the UK Government, the Minister for Primary Industries and Energy agreed to the formation of an independent expert group to provide high-level technical and scientific guidance to the rehabilitation project in August 1993. This became known as the Maralinga Rehabilitation Technical Advisory Committee (MARTAC) and comprised members with expertise and experience in health physics, engineering, geotechnical and nuclear site remediation, three of whom had been members of the earlier technical advisory group (TAG). MARTAC held its first meeting in September 1993. MARTAC's role was to advise the Minister on strategies for the implementation of Option 6(c). MARTAC's role was advisory and is effectively defined in the committee's terms of reference. Although MARTAC did not manage the project, MARTAC's recommendations strongly influenced operational policy in relation to, for example, clearance levels and pit treatments (including the use of ISV technology) that were implemented in the course of project.

MARTAC's views were reflected in the scope of works of the project, and MARTAC endorsed the health physics policy developed by the regulator for the project. MARTAC acted as a source of expert advice on a range of potentially sensitive matters, such as the adequacy of worker protection measures,

the extent to which the project was satisfying its objectives, and tasks that might practicably be undertaken by the Maralinga Tjarutja Aboriginal community. MARTAC formally met as a full committee on 16 occasions between September 1993 and January 2000. MARTAC held two joint meetings with members of the Consultative Group in 1999.

Members of the Aboriginal community were employed in the erection of boundary markers around the restricted parts of the Maralinga range and were sought for training positions in other aspects of the Maralinga Rehabilitation Project (MRP).

## 6. CONSULTATIVE GROUP AND PROBLEMS WITH ISV

The Maralinga Consultative Group met on an ad hoc basis during the TAG investigations in order to provide information on the progress of scientific studies, planning the rehabilitation work, during the rehabilitation work and in the preparation of the final reports. The consultative group was established to discuss and monitor the TAG studies was re-established in 1993 involving South Australian Government and Maralinga Tjarutja to discuss the MRP. It first met in March 1994. During 1997 and 1998, while a great deal of the rehabilitation work was done, there were few meetings of either MARTAC or the consultative group.

Toward the end of 1998, MARTAC members expressed the view that there were technical problems with ISV and changes in the program would be necessary. There had already been a number of changes to Option 6(c).

MARTAC had already resolved at an early stage that ISV was not warranted at locations other than Taranaki. After consultations it was also agreed to substitute boundary markers for the planned fence as maintenance of a fence was expected to be impractical. These changes had been made in a timely manner with adequate opportunity for consultation.

The decision to terminate the in situ vitrification and the change to the exhume and bury Option 6(a) for the remaining pits at Taranaki was probably the most significant deviation from Option 6(c).

It was clear that MARTAC members especially Des Davy and John Morris became unhappy with the progress of the in situ vitrification. The quantities of Pu buried in the pits at Taranaki were re-evaluated through the late 1980's and 1990's so that by 1995 it was thought to only be approximately 2 kg rather than 20 kg anticipated during the TAG studies. The latter figure was estimated by British sources at the time of the Royal Commission, while the 2 kg figure came from US data associated with Operation Rollercoaster which involved similar trials to the Vixen B trials in Nevada. Discrepancies in probable quantities of Pu at Taranaki generated significant interest in an inventory which was to be performed during the rehabilitation. This task was more difficult than was anticipated and the final estimate of 4 kg [5–8] has very large uncertainties giving a probable range of 2–10 kg.

In situ vitrification was proving much more expensive than anticipated and the expectations of MARTAC members were not being met by Geosafe. This technology was more expensive in part because the burial pits at Taranaki were much larger than expected from British documentation. Finding deficiencies in British documentation and science was not very surprising as these deficiencies were widely aired during the Royal Commission.

MARTAC members had an expectation that all material within the pits would be melted and incorporated into the glass phase. There was evidence of pit contents protruding from the glass monoliths and also it was found that there were small amounts of debris beneath the monoliths and not incorporated into the melt. Steel both solid and molten had a much higher density than molten soil and so sank to the bottom of melts, sometimes forming ingots of resolidified steel after ISV and sometimes unmelted steel protruded. This was a significant problem in the view of MARTAC members, but it was unclear that Geosafe was failing to meet its obligations as ISR conceded that no performance criteria existed in the original Geosafe contract. Indeed a draft criterion stated "There should be a high degree of confidence that the contaminated soils and debris within the pits are completely melted or encased in the vitrified product."

The problems with in situ vitrification came to a head in March 1999 when there was an explosion during the melting of pit 17. This highlighted the fact that the contents of the pits were not well characterised and it became clear very rapidly that no further in situ vitrification on uncharacterised pits would be allowed on safety grounds. This caused a crisis in the rehabilitation project because a

period of stand down began which was very expensive and the immediate solution to the problem was not obvious.

Geosafe proposed that the burial pits at Taranaki be sorted and that once the contents were characterised, the contaminated steel would be placed in a specially designed hybrid pit for in situ vitrification. This solution was predicated on the basis that the explosion in pit 17 was the result of the explosion of some material in the pit rather than a fault of the in situ vitrification process itself. Of course, the hybrid proposal negated a key attraction to ISV – the ability to immobilize the Pu contaminated material without excavation.

The investigation into the pit 17 explosion was conducted by Geosafe and it was clear from the beginning that this would both take considerable time and was unlikely to identify the cause of the explosion with absolute certainty. The Australian Government required that Geosafe conduct the investigation into the explosion and that the report be independently audited. This added to the delay in concluding the investigation. The final report, dated June 2001 [4], suggested that the most likely causes were detonation of explosives in the pit or the explosive release of a small, superheated, high-integrity container of organic liquids. The Commonwealth considered that the cause of the incident was not conclusively demonstrated.

As there was no absolute certainty that the explosion was not due to the in situ vitrification process, the Australian Government took the decision to change to Option 6(a) “Exhume and Bury” for the remainder of the Taranaki pits. This was announced to the Maralinga Consultative Group in the middle of a meeting in July 1999 without the consent of the other members of the Consultative Group, particularly Maralinga Tjarutja and South Australia. It was becoming clear at this stage to the advisers for Maralinga Tjarutja that in situ vitrification was becoming untenable and that the “Exhume and Bury” option, i.e. Option 6(a), would ultimately be taken.

The Consultative Group has continued to meet through 2000 and 2001, but there was diminished confidence in the consultative process as a result of the Commonwealth unilateral decision to abandon ISV.

The problems with ISV arose following some significant personnel changes in MARTAC and within DPIE (ISR). Alan Parkinson had been removed as a consultant to ISR and his position on MARTAC terminated in 1997 [9] and the key staff in ISR also changed at about this time. Parkinson has described this period in his report [9]. These changes in staff had a destabilizing effect on the Consultative process.

## 7. TECHNICAL RISK

ISV was always an experimental technology which involved significant technical risk. The risk was borne out by the considerations of Public Works Committee of the Australia Parliament (PWC) in 1995. In its report, the PWC called for an independent scientific audit of the ISV trials at Taranaki [10]. These concerns were primarily about the successful application of the technology, which was subject to 2 stages of preliminary trials. The use of ISV on the pits at Taranaki also entailed safety risks because the contents of the pits were uncharacterised. In retrospect this is not surprising given the poor recording of pits contents as supplied by the British Government. Its initial strong support by key advisers from TAG and MARTAC followed by ultimate abandonment demonstrated a lack of thorough consideration of the consequences of the use of ISV.

The issues with ISV highlighted that consultation became a secondary issue for the Department managing the project.

## 8. STEWARDSHIP

Toward the end of the rehabilitation project, issues of stewardship were raised by MARTAC for the future use of Section 400. This started a completely new phase of consultations that required more substantial contributions from South Australian Government and Maralinga Tjarutja. The South Australian Government had been able to ‘not have a policy’ on Maralinga for most of the period since the Royal Commission and so this prompted a major change in its attitude.

Some elements of government were quite negative to the prospect of section 400 being restored to South Australian jurisdiction because of the potential for greatly increased demand for some services.

## 9. LACK OF TECHNICAL SKILLS WITHIN THE DEPARTMENT MANAGING THE PROJECT

A considerable problem throughout the MRP was a lack of technical skills within the Department managing the project. Until 1997, engineering skills were provided by Alan Parkinson, but there was no Radiation Protection expertise. Technical advice had to come from contractors, MARTAC or ARL and on important occasions it was not sought.

This is best illustrated through 2 significant failures. Firstly, the department managing the project was determined to conclude a contract for ISV with Geosafe in 1996 at the earliest possible time. It pressed PWC for permission to go ahead with the contract. The contract did not include any performance criteria, causing numerous problems and delays later in the project. MARTAC had a clear view of what such criteria should include but clearly advice was not sought. Secondly, throughout the drafting of the Maralinga Land and Environment Management Plan numerous parts of the document were presented to Maralinga Tjarutja and the South Australian Government with technically unmeaningful terms, eg. statements referring to contamination levels of 'cps' or 'integrated surface radiation'. Australian Government officials were critical of the length of time taken in draft this plan, but the lack of technical skills within the managing Department was a major cause of delay.

The engineering company Gutteridge, Haskins & Davey Pty. Ltd. were contracted by the Department managing the project to provide both project management services to the project as well as other services. As project manager they were relied upon by ISR to provide advice on a wide range of issues. They provided engineering technical skills but were unable to directly provide specialised radiation protection services.

## 10. SUMMARY AND CONCLUSIONS

In any long and complex project there will be aspects that do not proceed as expected and as a consequence project management needs to be flexible and fully conversant with all the issues.

In the MRP, soil removal was handled successfully and within budget. It was learnt that Option 6(a) "Exhume and Bury" was not as difficult to perform as expected at the start of the project. ISV on uncharacterised pits was technically very risky and poorly handled from a project planning and management perspective.

The responsible Department failed to maintain adequate control because it did not have ready access to experts in the key disciplines and project management linkages were convoluted.

The problems with ISV highlight the fact the Australian personnel were not involved in many activities that took place at the site, there is no documentation in Australian hands about those activities and the cleanup operations 'Hercules' and 'Brumby' served to further mask what activities were undertaken by the British testing program. Despite great expense, there are many things that remain unknown about Maralinga.

Stakeholder participation was an important secondary goal of the project – but not its focus. This produced difficulties in finalising the project as there have been protracted legal disputes with a key contractor and the technology 'sold' to Maralinga Tjarutja and the South Australian Government is not the implemented solution. It makes convincing the traditional Aboriginal owners of the appropriateness of the final outcomes quite a challenge.

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# **Ecosystem modelling in exposure assessments of radioactive waste in coastal waters\***

**L. Kumblad**

Department of Systems Ecology, Stockholm University, Stockholm, Sweden

**Abstract.** Environmental transport and fate of radioactive isotopes in a brackish water bay of the Baltic Sea, was investigated in this study by using an ecosystem modelling approach. The purpose was to develop a method to assess the exposures to humans and to other organisms in the environment in case of discharges from an underground repository for radioactive operational waste. The radionuclides considered are long-lived, have high environmental mobility and bioavailability and pose a potential threat to living organisms for an extensive period of time. In the model, the radionuclide transport is tightly associated with a site-specific carbon flow model based on general ecological principles, that identifies and quantifies the main flows and storages of energy in the ecosystem, both in the physical environment and in the food web. In the model, the radionuclides are introduced into the food web via photosynthesising organisms. C-14 is assumed to assimilate in proportion to its presence compared to other carbon isotopes in the water while transfer factors are used in the initial uptake of other radionuclides. The trophic transfer in the food web are presumed to follow the flow of energy between the ecosystem compartments and to be proportional to the rate of predominant ecological processes, such as respiration, consumption and excretion. Modelling results demonstrates that this approach provides a method that allows evaluation of several scenarios. For instance, it is possible to compare the influence of different abiotic and biotic processes, such as accumulation pattern, excretion rates and water exchange, on radionuclide accumulation and transfer between the compartments of the ecosystem. This technique has potential to be a powerful tool in safety assessments of radioactive waste.

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\* Only an abstract is given as the full paper was not available.



# Some common regularities of synergistic effects display

V.G. Petin<sup>a</sup>, J.K. Kim<sup>b</sup>

<sup>a</sup> Medical Radiological Research Center, Obninsk, Kaluga Region, Russian Federation

<sup>b</sup> Korea Atomic Energy Research Institute, Taejon, Republic of Korea

**Abstract.** Our purpose here is to review and discuss some new general rules of synergistic effect display. The response of various biological objects to the simultaneous combined action of hyperthermia with ionizing radiation, ultraviolet light, ultrasound and some chemical agents was analysed using the experimental data obtained by authors. To check the universality of the regularities of synergistic effect display, the results published by others were also involved. The data presented strongly invoke the need to elaborate a new theoretical conception of the synergy which, being useful for environmental radiation protection, took into account the new regularities revealed.

## 1. INTRODUCTION

In view of the emerging importance of the combined actions of ionizing radiation with other environmental factors, the potential significance of the synergy, among other radioecological problems, has to be carefully assessed. Knowledge of common regularities of synergistic effects derived from experimental results obtained at molecular and cellular levels as well as with laboratory animals should enhance our understanding of the risks of radiation exposure and their interaction with other agents and possible strategies to reduce them. Moreover, the elaboration of the quantitative assessment of combined effects and common rules of synergistic effects display is urgently needed for many practical and theoretical reasons. First of all, to bridge the gap between differing conceptual approaches in the mechanisms of synergistic interaction. Secondly, these results will be useful to develop the unified concept and mathematical approach to predict and optimize the synergy. Thirdly, they will promote the extrapolation to low exposures. And finally, practical improvement of combined treatments in radiotherapy and sterilization methods can be expected. Our experimental and theoretical research efforts were directed towards these goals for several years mainly using the example of thermoradiation action.

Much experimental evidence and theoretical considerations demonstrated the potential importance of synergistic interaction of radiations combined with other environmental agents including heat. The combined thermoradiation action is known to result in a synergistic interaction when the final biological effects are greater than the addition of the effects from separate exposures to the individual inactivating agents. Heat is extensively utilized to enhance the biological effects induced by ionizing radiation, ultraviolet light, microwaves, ultrasound and chemicals. From many publications, it is easily recognized that the degree of synergistic effect appears to be influenced by many factors such as the biological object and end point, selected for observation, thermotolerance, recovery and repair ability, phase of cell cycle, temperature, exposure time, treatment sequence, dose and dose rate of ionizing radiations. The greatest synergistic interaction is observed when hyperthermia and another agent are applied simultaneously; i.e., the level of synergism decreases as the time interval between the two modalities increases.

Despite the potential importance of synergistic interaction, information regarding common regularities of synergistic interaction of hyperthermia with various physical and chemical inactivating agents is largely missing. Our purpose here is to review and discuss some new general rules of synergistic effect display. The response of various biological objects to the simultaneous combined action of hyperthermia with ionizing radiation, ultraviolet light, ultrasound and some chemical agents will be analysed using the experimental data obtained by authors. To check the universality of the rules obtained, the results published by others will be involved.

## 2. MATERIALS AND METHODS

The following yeast strains were used in the experiments: *Zygosaccharomyces bailii* (haploid), *Saccharomyces cerevisiae* (two diploid strains, XS800 and T1), *Endomyces magnusii* (diploid). Cells were incubated before irradiation for 3–5 days at 30°C on a complete nutrient agar layer to a stationary phase. Homogeneous cell populations were treated at stationary phase of growth.

The  $^{60}\text{Co}$   $\gamma$ -ray source was a Gammacell 220 (Atomic Energy of Canada LTD). The  $\gamma$ -ray dose-rate was 10 Gy/min. The electron beam from a 25 MeV pulsed linear accelerator was also used in these experiments. The average dose rates were 5, 10, 25 and 250 Gy/min as determined by ferrous sulphate dosimetry.

For UV exposure, cells were irradiated with germicidal lamps that emitted predominantly UV light of wavelength 254 nm at fluence rates of 0.033, 0.15, 0.25, 1.5 W/m<sup>2</sup>. Variation of the intensity was achieved by means of calibrated metal wire nets. The fluence rates were measured using a calibrated General Electrical germicidal meter.

A continuous mode of sonication was accomplished by a 20 kHz ultrasonic unit (Fisher sonic dismembrator). The ultrasonic dose rates were 0.05 and 0.2 W/cm<sup>2</sup> which were measured by the calorimetric method. For sonication, 0.1 ml of cell suspension (10<sup>8</sup> cells/ml) at room temperature was put into 9.9 ml of sterile water which was placed into a metal vessel constructed together with the transducer. To determine the effect induced by ultrasound alone, the absorbed ultrasound heat was completely removed by cooling with water.

Hyperthermia was given in a water bath where a desired temperature  $\pm 0.1$  °C was maintained. For the simultaneous action of hyperthermia and other agents, the time interval between the introduction of the cells into the preheated water and the beginning of exposure was about 0.1–0.3 min, which was significantly less than the total treatment time. At the end of the treatment, the samples were rapidly cooled to room temperature. Therefore, the duration of physical agent treatment and heat exposure were identical.

After treatment with each agent applied alone or combined simultaneously with heat, a known number of cells were plated so that 150–200 colonies per dish would form by the surviving yeast cells after 5–7 days of incubation at 30°C. All experimental series were repeated 3 to 5 times. The details were described elsewhere [1–5].

## 3. SELECTED EXPERIMENTAL RESULTS

Figure 1 provides an example of the basic experimental data used in this investigation. It is appropriate to mention here again that interactive agents were applied simultaneously, i.e. the upper scale concerns all of the curves presented in this figure. To estimate quantitatively the sensitization action of hyperthermia, one can apply the thermal enhancement ratio defined as the ratio  $D_3/D_1$  or  $t_3/t_1$  (Figure 1). This ratio indicates an increase of cell radiosensitivity by high temperature. However, it does not reflect the kind of interaction (whether it was independent or synergistic). To calculate the synergistic effect, we used the synergistic enhancement ratio ( $k$ ), defined as the ratio of the calculated radiation dose (assuming an additive effect of radiation and hyperthermia) to that observed from the experimental survival curve for the simultaneous action of radiation (or other agents employed) and hyperthermia at a fixed level of survival. For example for 1% survival,  $k = D_2/D_1 = t_2/t_1$  (Figure 1). For exponential survival curves, this parameter is independent of the survival level for which it is calculated. For sigmoidal survival curves, the synergistic enhancement ratio was calculated for 10% survival. The thermal enhancement ratio was found to increase indefinitely with increasing the exposure temperature, while the synergistic enhancement ratio at first increases, then reaches a maximum, which is followed by a decrease [1–5].

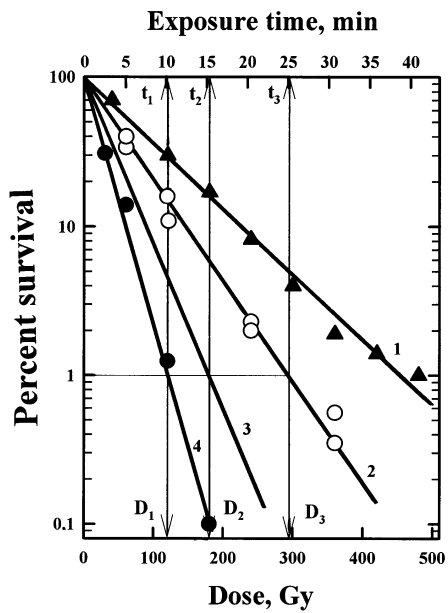


FIG. 1. Survival curves of *Zygosaccharomyces bailii* haploid yeast cell: curve 1 – heat treatment (45 °C) alone; curve 2 – ionizing radiation ( $^{60}\text{Co}$ ) at about 10 Gy/min and room temperature; curve 3 – calculated curve for independent action of ionizing radiation and heat; curve 4 – experimental curve after simultaneous thermoradiation action.

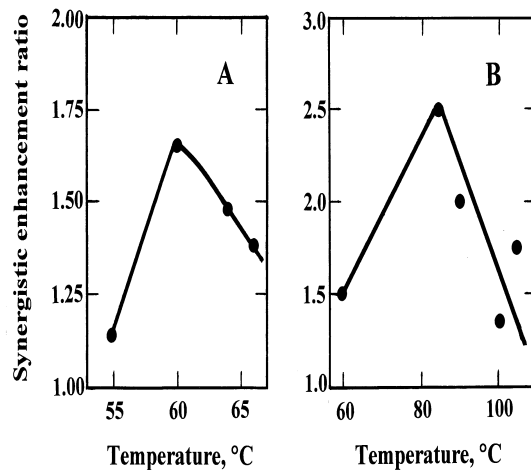


FIG. 2. The dependence of the synergistic enhancement ratio upon the exposure temperature for bacteriophage (A) and bacterial spores (B). The original survival curves data were taken from the following papers: A – [6], B – [7].

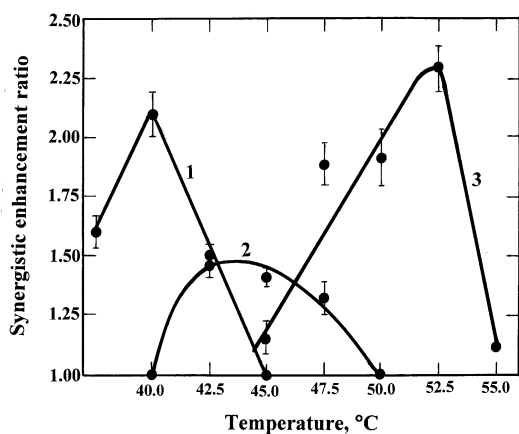


FIG. 3. The dependence of the synergistic enhancement ratio upon the exposure temperature for diploid *Endomyces magnusii* (curve 1), haploid *Zygosaccharomyces bailii* (curve 2) and diploid *Saccharomyces ellipsoideus* (curve 3) yeast cells. The original survival curves data were obtained by the authors. Error bars show interexperiment standard errors.

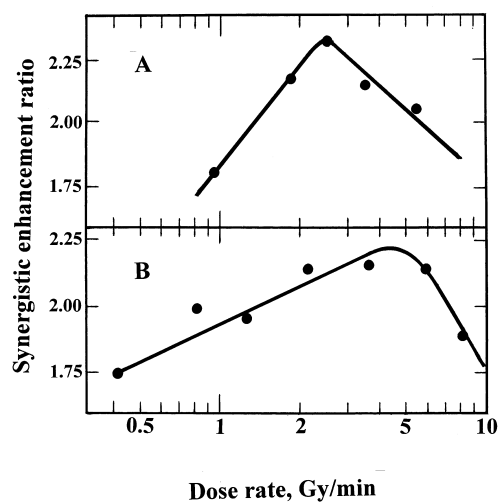


FIG. 4. The dependence of the synergistic enhancement ratio upon the dose rate for bacterial spores *Bacillus subtilis* exposed to ionizing radiation at 95 (A) and 105 °C (B). To calculate this dependence, the original survival curves data were taken from the earlier published paper [7].

Figures 2 and 3 show some experimental examples obtained with viruses (Figure 2A), bacterial spores (Figure 2B) and yeast cells of different species and ploidy (Figure 3). Data on the inactivation of viruses and dry bacterial spores exposed to ionizing radiation at different temperature were published by others [6, 7]. In these papers, the authors did not calculate the synergistic effect so we used their data to obtain results presented in Figure 2. One can see that the synergistic interaction between hyperthermia and ionizing radiation is realized only within a certain temperature range independently of the object analyzed. For temperatures below this temperature range, the synergistic effect is not observed and cell killing was mainly determined by the damages induced by ionizing radiation. For temperatures above this temperature range, the synergistic effect was also not observed but cell killing was chiefly caused by hyperthermia.

Noteworthy is the fact that such a dependence of synergistic effect on temperature under which the exposure was occurred was also obtained upon the simultaneous combination of hyperthermia with UV light [4], ultrasound [5] and some chemical inactivating agents [8, 9]. It means that hyperthermic treatment can drastically increase the cell sensitivity to different physical and chemical agents in a similar manner. Thus, one can conclude that for a given intensity of physical factors or concentration of chemical agents there would be a specific temperature that maximizes the synergistic interaction. Any deviation of the exposure temperature from optimal value results in a decrease of synergism.

The temperature range strengthening the effect of ionizing radiation has been varied with different cell systems and shifted toward lower temperatures for temperature-sensitive cell lines. Our experiments and estimations showed that this range was about 55–70°C for viruses (Figure 2A), 95–105°C for the most thermoresistant bacterial spores (Figure 2B), 50–60°C for bacterial cells [2], 45–55°C for relatively thermoresistant *S. cerevisiae* yeast cells (Figure 3, curve 3) and 37,5–45°C for *Bacillus subtilis* spores [7]. They were used to estimate the dependencies of the synergistic enhancement ratio on dose rate for simultaneous thermoradiation action. The results are depicted in Figure 4. For relatively thermosensitive *Endomyces magnusii* yeast cells (Figure 3, curve 1) and cultured mammalian cells [2]. Inside the temperature range, synergistically increasing the effect of inactivating agents, a specific temperature which maximizes the interactive effect may be found. In other words, there is a definite ratio of damages produced by the agents employed in combination that would ensure the greatest synergistic effect. Any deviation of this ratio from optimal one resulted in a decrease in the synergy. One more important conclusion can be made from the results presented in Figure 3. One can see that the effectiveness of synergistic interaction was smaller for haploid cells (curve 2) than for diploid ones (curves 1, 3). It is in agreement with the well known fact that the mechanism of synergy is related with cell ability to repair radiation damage while repair of DNA double strand breaks requires two homologous DNA duplexes.

It can be supposed on this basis that for any constant temperature there should be existed an optimal dose rate resulting in the greatest synergy. The data on the simultaneous thermoradiation action on *Bacillus subtilis* spores [7] were used to estimate the dependencies of the synergistic enhancement ratio on the dose rate for two exposure temperatures 95 and 105°C. The results are depicted in Figure 4. One can see that for a constant temperature, at which the irradiation occurs, synergy can be observed within a certain dose rate range. Inside this range an optimal dose rate of ionizing radiation may be indicated that maximizes the synergy. As the exposure temperature is reduced, the optimal dose rate decreases and *vice versa*. The universality of this important conclusion is supported by our extensive data regarding yeast cells exposed to heat combined with ionizing radiation [3], 254 nm UV light [4] and 44 kHz ultrasound [5] and some chemicals [8, 9].

To emphasize the importance of synergistic effects at low intensity of inactivating agents, we analysed the dependence of synergistic interaction on the intensity of physical factors or on the concentration of chemical agents applied in combination with hyperthermia. Using survival curves data published for simultaneous action of hyperthermia and ionizing radiation on bacteriophage [6], bacterial spores [7], yeast [1, 2] and mammalian cultured cells [10], we were able to calculate the synergistic enhancement ratio for various cell systems and irradiation condition. It allowed us to establish the correlation between the dose rate and the exposure temperature, which both provide maximum or other arbitrary levels of synergistic interaction (Figure 5). Open circles denote the results of our calculations based on experimental results published in current cited papers. The value of synergism was the highest for bacterial spores ( $k_{\max} = 2.2$ ), diploid yeast cells ( $k_{\max} = 1.7$ ) and was intermediate for bacteriophage ( $k$

= 1.3) and cultured mammalian cell ( $k = 2.2$ ). This was due to the fact that for last cell systems the highest synergistic effect was not obtained for all dose rates used in the experiments. One can see that linear relationships are found between these values for various cellular objects. This means the general importance of the dose rate of ionizing radiation in the manifestation of synergistic interaction. It can be inferred that the temperature at which ionizing radiation is delivered should be diminished to obtain the maximum or a definite synergistic effect with dose rate decreasing and *vice versa*.

To check the universality of this rule, the data on simultaneous effect of hyperthermia combined with UV light [2, 4] or ultrasound [5, 8] on diploid yeast cells, as well as with tris(1-aziridinyl)-phosphine sulfide (thio-TEPA) [10] and cis-diamminedichloroplatinum (II) (cis-DDP) [11] on cultured mammalian cells were involved. The last two set of data include the relationship between exposure temperature, concentration, and rate of cell inactivation for chemical agents used in clinical chemotherapy. Hence, they have no direct attitude toward environmental harmful agents and they used here to demonstrate that the concentration of chemical agents is also important for their synergistic interaction with heat. Using data published in the above cited works, we could obtain the relationships between the intensity of physical factors or the concentration of chemical agents with the exposure temperature which both provide the greatest synergy (Figure 6). Here again, open circles denote the results of our calculations based on the survival curve data published earlier. The value of the synergistic enhancement ratio was about 1.6 for UV light, 2.5 for ultrasound, 2.5 for thio-TEPA and 3.2 for cis-DDP combined with hyperthermia. In all cases, at a smaller intensity of the physical factor or concentration of the chemical agents, it was required to reduce the acting temperature to preserve the highest synergistic effect.

It is known [13, 14] that the decrease in the effective dose  $D_{eff}(t)$  with the recovery time  $t$  was fitted to an equation of the form:

$$D_{eff}(t) = D_1[K + (1 - K) e^{-\beta t}], \quad (1)$$

where  $D_1$  is the initial radiation dose;  $K$  is an irreversible component of radiation damage;  $e$  is the basis of the natural logarithm, and  $\beta$  is the recovery constant characterizing the probability of recovery per time unit. In other words, the recovery constant is equal to a fraction of radiation damage recovering per time unit. To determine whether the mechanism of synergistic interaction was related to the impairment of the recovery capacity *per se* or to the production of irreversible damages which cannot be repaired we estimated the recovery parameters ( $K$  and  $\beta$ ) after different combined treatments. The final results presented in Table 1. One can see that for various biological objects and different conditions of the combined action the irreversible component increased with exposure temperature and the concentration of chemicals while the probability of recovery stayed unchanged. It can be concluded on the basis that the recovery process itself is not damaged and the inhibition of recovery is entirely due to the enhanced yield of irreversibly damaged cells.

#### 4. CONCLUDING REMARKS

The main common regularities of the synergistic interaction obtained in this investigation may be summarized as follows. (1) For any constant rate of exposure, the synergy can be observed only within a certain temperature range. (2) The temperature range synergistically increasing the effects of radiations is shifted to lower temperature for thermosensitive objects. (3) Inside this range, there is a specific temperature that maximizes the synergistic effect. (4) An increase in exposure rate resulted in an increase of this specific temperature to achieve the greatest synergy and *vice versa*. (5) For a constant temperature at which the irradiation occurs, synergy can be observed within a certain dose rate range. (6) Inside this range an optimal intensity of physical agent may be indicated that maximizes the synergy. (7) As the exposure temperature is reduced, the optimal intensity decreases and *vice versa*. (8) The recovery rate after combined action is decelerated due to an increased number of irreversible damage. (9) The probability of recovery is independent of exposure temperature for yeast cells irradiated with ionizing radiation. (10) Chemical inhibitors of cell recovery act through the formation of irreversible damage but not *via* damage the recovery process itself.

The data strongly invoke the need to elaborate a new theoretical conception of the synergy which, being useful for environmental radiation protection, took into account the regularities revealed.

TABLE 1. RADIOBIOLOGICAL PARAMETERS OF CELL RECOVERY AFTER COMBINED TREATMENTS WITH IONIZING RADIATION AND PYRUVATE (PUR), NOVOBIOCIN (NOV), LACTATE (LAC) AND NALIDIXIC ACID (NAL), AS WELL AS AFTER SIMULTANEOUS THERMORADIATION ACTION

Treatments	Biological objects	Irreversible component $K$	Recovery constant $\beta$ , hr <sup>-1</sup>	Reference
$\gamma$ -rays alone	Yeast cells	0.41	0.07	[1]
$\gamma$ -rays + 45°C	Yeast cells	0.51	0.063	[1]
$\gamma$ -rays + 50°C	Yeast cells	0.75	0.07	[1]
$\gamma$ -rays + 55°C	Yeast cells	0.90	0.063	[1]
X-rays alone	Mammalian cells	0.60	0.15	[15]
X-rays + 10 mM Pur	Mammalian cells	0.75	0.16	[15]
X-rays + 20 mM Pur	Mammalian cells	0.92	0.16	[15]
X-rays + 5 $\mu$ M Nov	Mammalian cells	0.82	0.14	[15]
X-rays + 10 $\mu$ M Nov	Mammalian cells	0.90	0.14	[15]
X-rays + 10 mM Lac	Mammalian cells	0.78	0.14	[15]
X-rays + 20 mM Lac	Mammalian cells	0.99	–	[15]
X-rays + 5 $\mu$ M Nal	Mammalian cells	0.74	0.15	[15]
X-rays + 10 $\mu$ M Nal	Mammalian cells	0.82	0.15	[15]

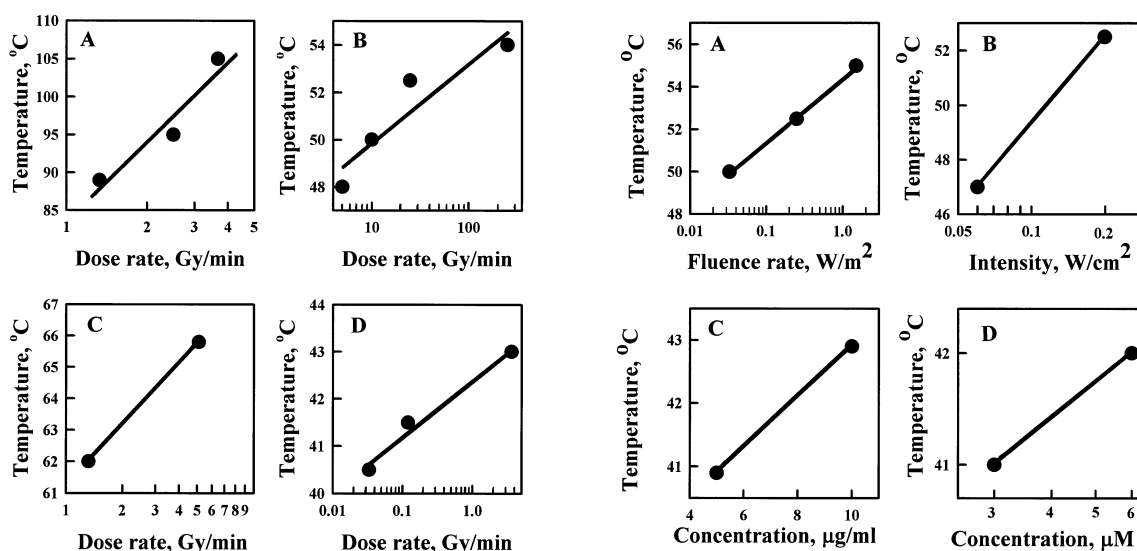


FIG. 5. Correlation of dose rate and exposure temperature providing the same synergistic interaction under simultaneous thermoradiation action: A – bacterial spores (*Bacillus subtilis*); B – diploid yeast cells (*Saccharomyces cerevisiae*, XS800); C – bacteriophage (T4); D – cultured mammalian cells (Chinese hamster cells). The original survival curves data were taken from the following papers: A – [7], B – [3], C – [6], D – [10].

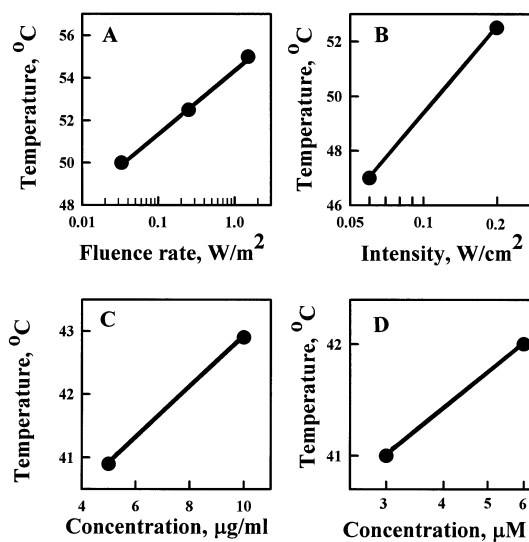


FIG. 6. Correlation of exposure temperature with UV light fluence rate (A), ultrasound intensity (B), and concentration of tris(1-aziridinyl)-phosphine sulfide (thio-TEPA) (C) and cis-diamminedichloroplatinum (II) (cis-DDP) (D) providing the highest synergistic interaction of these agents with heat. The original survival curves data were partly obtained by authors and were taken from the following papers: A – [4], B – [5], C – [12], D – [13].

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# **Towards an improved ability to estimate internal dose to non-human biota**

## ***Development of conceptual models for reference non-human biota***

**T.L. Yankovich**

Atomic Energy of Canada Limited, Chalk River, Ontario, Canada

**Abstract.** Conceptual models have been developed for Reference Fish and Amphibian based on the chemical composition of biota tissues. It was found that these non-human biota can be divided into simplified compartments, where the number of compartments depends upon the element being considered. In most cases, one of two conceptual models can be applied to assess internal partitioning patterns between tissues: 1.) Elements can be relatively uniformly-distributed throughout the body (i.e. whole organism is represented by one homogeneous compartment); or 2.) Elements can be sub-divided between two to three homogeneous compartments. Although preliminary, this work is aimed at developing realistic frameworks to facilitate estimation of internal dose to non-human biota for use in Ecological Risk Assessment (ERA) and Environmental Effects Monitoring (EEM).

### 1. INTRODUCTION

Internal radiation often contributes the major proportion of dose that is received by biota. Consequently, to assess potential risk to biota associated with exposure to radiation, a solid understanding of internal dosimetry is required. Although detailed methodologies have been developed to estimate internal dosimetry for humans and other mammalian species [e.g. 1], the assessment of internal dose to non-mammalian species is still in its infancy. Realistic quantification of internal dose to non-human biota is complex due to the large range of body sizes and geometries for different species and their tissues [2–5]. In addition, the potential risk of damage to body tissues of biota depends upon the distribution of radionuclides in the tissues, the amount of energy deposited in the tissue, the type of energy, and the radiosensitivity of each tissue type. Therefore, to further our understanding of ecodosimetry across the wide range of non-human biota species, it is necessary to simplify and compartmentalize biota so that standardized approaches can be established for estimating internal dose. This can be accomplished by developing models for Reference Non-Human Biota, which are similar to Reference Man [6–7].

Models of Reference Biota can range in complexity from simple, one-compartment models representing whole organisms to multiple-compartment models that differentiate internal tissues. For example, internal dose rates can be estimated using the “point source dose distribution” approach that was developed by the IAEA [2–5]. This involves use of simplified, empirically-derived dose rate formulas, which define organisms that fall within various size categories using ellipsoid geometries. The total energy absorbed by an organism will then be dependent upon the size of the organism, where larger organisms are assumed to absorb a greater proportion of the emitted energy than smaller organisms.

More complex dosimetric models and frameworks, which can be applied to flora and fauna, have also been developed [e.g. 6–8]. Such models sub-divide organisms by body size, geometry, and the ecological significance of tissues. For example, Pentreath and Woodhead [6] suggest that non-human biota could be sub-divided into one of six types of geometries with varying levels of complexity (ranging from whole-body to multiple-organ compartments) for the purposes of internal dose assessment. Based on the discussion in Pentreath and Woodhead [6], a key question arises: ‘*What level of complexity is appropriate/feasible when estimating internal doses to non-human biota?*’ (i.e. *how many compartments should be used to estimate internal dose to non-human biota?*). In general, compartments can be identified based on their perceived radiosensitivity, their size and/or their propensity to accumulate radionuclides. The current study focuses on compartmentalization of biota based on their chemical composition and the partitioning behaviour of radionuclides between tissues.



The **objective** of this study was to develop conceptual models for fishes and frogs based on internal partitioning patterns of stable analogues to radionuclides. This paper focuses univalent and divalent cations, which include  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{40}\text{K}$ , as well as  $^{90}\text{Sr}$  radionuclides, respectively. These radionuclides are of interest since the Cs and Sr radioisotopes represent common fission products with intermediate to long radiological half-lives, and  $^{40}\text{K}$  is a naturally-occurring Cs analogue of primordial origin and with a ubiquitous distribution [9–10].

## 2. EXPERIMENTAL APPROACH

Bullfrogs (*Rana catesbiana*), brown bullheads (*Ameiurus nebulosus*), and northern pike (*Esox lucius*) were collected from Perch Lake (Chalk River, Ontario) between 1995 and 1999. Bullfrogs ranged from 12 to 29 cm in body length, with fresh weights ranging from 28.4 to 248.9 g, and bullheads ranged from 196 to 230 cm in total length. Northern pike were sub-divided into immature (267 to 374 cm) and mature (611 to 861 cm) age categories to assess differences in internal partitioning of stable analogues to radionuclides between different life stages. Bullheads were chosen to represent benthivorous fishes, which frequently come into contact with the sediments, whereas pike were selected to represent piscivorous fishes that occupy a relatively high trophic position in aquatic foodwebs [11]. Individual fishes were dissected to remove blood, boneless hypaxial muscle, bone, liver, kidney and gonadal tissues, whereas bullfrogs were sub-divided into blood, bone, muscle, liver, kidney, gonads, and heart tissues for stable element analysis. Stable analogues to radionuclides were then measured in tissues using Inductively-Coupled Plasma Mass Spectroscopy (ICP-MS) and ICP-Atomic Emission Spectroscopy (ICP-AES).

Literature data and data from this study relating tissue biomass to organism fresh weight were compiled for teleost fishes and amphibians with varying body sizes, and predictive relationships were developed. Biomass estimates for biota and their internal compartments, and corresponding concentration measurements taken for each tissue, were utilized to estimate the expected percent loadings of univalent and divalent radionuclides in biota tissues, as follows:

$$\text{Percent Loading in Tissue} = \frac{C_{\text{tissue}} \cdot m_{\text{tissue}}}{C_{\text{whole}} \cdot m_{\text{whole}}} \cdot 100\% = \frac{CR_{\text{tissue}} \cdot C_{\text{Reference Tissue}} \cdot m_{\text{tissue}}}{C_{\text{whole}} \cdot m_{\text{whole}}} \cdot 100\%$$

where:

$C_{\text{tissue}}$  is the element concentration in a given tissue (in mg/kg fresh weight);

$m_{\text{tissue}}$  is the mass of that tissue (in kg fresh weight);

$C_{\text{whole}}$  is the element concentration in the whole organism (in mg/kg fresh weight);

$m_{\text{whole}}$  is fresh weight (in kg);

$CR_{\text{tissue}}$  is the concentration ratio of the tissue of interest for a given type of biota (based on literature data); and

$C_{\text{Reference Tissue}}$  is the concentration of the element of interest measured in the Reference Tissue (i.e. muscle).

Wherever possible, percent loads were tabulated using actual data measured during this study; however, for some tissues, data were not available, making it necessary to estimate loads based on literature data. This was accomplished by using concentration ratios for the tissue of interest in given type of biota. A concentration ratio is defined as the ratio of the concentration of a given element in the tissue of interest relative to its concentration in the Reference Tissue (in this case, muscle) for a given type of biota (e.g. freshwater fishes) [12]. For example, measurements of stable elements have been made for fish muscle tissue, but not for brain tissue, as part of this work. Despite this, however, element concentrations in brain can be estimated by taking the product of the concentration in muscle (i.e. the Reference Tissue) and the brain-to-muscle concentration ratio for freshwater fishes taken from the literature. Concentration ratio data for various types of non-human biota have been summarized by Yankovich and Beaton [12]. In general, much fewer concentration data are available for amphibian tissues than for fishes. Therefore, in some cases, it was necessary to estimate missing data by assuming that similar partitioning patterns occur between amphibian viscera.

### 3. RESULTS AND DISCUSSION

#### 3.1. Tissue-to-whole body biomass ratios

Estimation of tissue and whole body biomasses is important in internal dose calculations for two reasons. First, the potential dose received from  $\gamma$ -emitters, as well as from relatively energetic  $\beta$ -emitters, is highly sensitive to the body size of the organism under consideration when estimating whole body doses. Since non-human biota can range in body size by orders of magnitude, there is a strong likelihood that at least some of the internally-bound radiation will escape to the surrounding environment. Current practices in Ecological Risk Assessment (ERA) often assume that internal radiation in whole organisms is self-absorbing with no energy loss to the environment, regardless of the magnitude of energy under consideration or the size of the organism [e.g. 13]. Such assumptions lead to potentially over-conservative estimates of internal dose to biota, depending upon the type of radiation to which biota are being exposed and the type of organism being considered. To address this problem, it is important to establish predictive relationships relating tissue and whole body biomasses for biota with varying body sizes, which can ultimately be used to account for losses of energy from biota tissues to surrounding tissues and the environment. At a finer level, for less energetic radionuclides, this information can ultimately be used to estimate energy deposition patterns from a given *source* tissue to surrounding *target* tissues [14]. Such data are particularly important when estimating doses to specific tissues, which are expected to be relatively radiosensitive and are immersed in or adjacent to tissues which tend to accumulate certain types of radionuclides (e.g. bone).

Relationships between body sizes of fishes and frogs, as well as percent biomasses of each tissue relative to the whole body, have been estimated based on literature data, as well as measurements taken during this study (Table 1). As expected, predictive relationships between animal biomass and the biomasses of most individual tissues are fairly strong with the exception of thyroid and gonadal tissues (Table 1). This is not surprising, however, since many factors can potentially affect the weights of these tissues. For example, maturity, season, temperature, sampling location, nutritional status and other factors can influence gonad weights for different species, particularly for females, thereby contributing to the variability observed within and between studies and individuals. Consequently, the ability to predict gonad weight based on body size is limited. Similarly, variability in thyroid biomass can be related to differences in gender, age, diet, geography, climate, external stimuli, internal stimuli, and/or iodine uptake [1].

Information on tissue biomasses can be applied in conjunction with data on the relative distributions of radionuclides in different tissues to estimate the relative proportions of radionuclides in different tissues using a mass balance approach, similar to the approach used in environmental transport models. Anatomical models, similar to those discussed by Pentreath and Woodhead (2000), can then be developed, which describe source tissues in terms of their expected radionuclide load, elemental composition, size, shape and spatial relationships. Based on this information, internal radiation deposition patterns between source regions and target regions can be described using Monte Carlo simulations [15].

#### 3.2. Radionuclide partitioning in biota tissues

Concentration ratios of univalent and divalent elements have been compared for several fish and amphibian tissues as an indication of whether univalent and divalent radionuclides show a uniform or a localized internal distribution (Table 2). A uniform distribution between tissues was assumed if stable element concentration ratios fell within 5-fold of one another. In general, the chemical distribution of univalent and divalent elements between biota tissues is highly conserved, with very similar internal partitioning patterns occurring between different life stages (i.e. mature vs. immature pike), feeding modes (i.e. benthivorous vs. pelagic fishes) and biota types (i.e. fishes vs. amphibians) (e.g. Table 2). As expected, it was found that not all elements are uniformly-distributed between biota tissues. For example, divalent elements (including Ba, Ca and Sr) that become integrated in the bone matrix of teleost fishes (such as pike and bullheads) are typically 2 to 3 orders of magnitude higher in bone than observed in soft tissues in the body. By comparison, elements (such as Mg and Na) that tend to deposit on the crystal surfaces of bone were approximately 1 order of magnitude higher in bone than in soft tissues (Table 2). The trends observed for amphibians were comparable to those observed for

fishes, with relatively higher concentrations detected in bone than in soft tissues for elements that integrate in bone matrices; however, bone-to-soft tissue concentration ratios for matrix-bound bone-seeking elements tended to be lower in amphibians than in fishes, on average. By comparison, in most cases, soft tissues tended to fall within less than or equal to 5-fold of one another in terms of their concentrations of univalent and divalent elements. Concentrations of divalent elements in scale tissues, which are also expected to contain elevated levels of matrix-bound bone-seeking elements [16], tended to fall between those measured in bone and those measured in soft tissues (Table 2). Therefore, it may be reasonable to sub-divide Reference Fish into three compartments, including bone, scales and soft tissues, when estimating the internal dose received from divalent elements (Table 2). It is important to note, however, that some fish species, such as brown bullheads, lack scales, and in such cases, Reference Fish would only consist of two compartments, which include soft tissues and bone (Table 2). Similarly, Reference Frog, which also lacks scales, might also be represented by a simple, two-compartment model, consisting of bone and soft tissue.

Unlike divalent elements, such as Sr, which often tend to be localized in hard tissues, univalent elements, such as Cs, tend to show a relatively uniform distribution between biota tissues. A notable exception is Na, which was approximately 10-fold higher in bone tissue than in soft tissues. This is likely due to the tendency of Na to deposit on crystal surfaces, such as bone, as opposed to becoming incorporated in the bone matrix. In general, bone-seekers can be divided into elements that become integrated in the bone matrix (such as Sr, C, F, P, Ca, Ba and Ra) and those that are deposited on bone surfaces (such as Na, Mg, Y, La, Th, Pu and Am) [17]. Matrix-bound elements become incorporated in the skeletal matrix either because they are components of the bone matrix or are chemical analogues of matrix components [18]. Comparison of measured data for the univalent and divalent elements considered in this study suggests that concentrations of elements that integrate in the bone matrix are higher than those that deposit on bone surfaces by approximately 1-to-2 orders of magnitude (Table 2).

### **3.3. Percent loading of univalent and divalent elements in biota tissues**

Based on the compartment size and concentration data, the total percent loading (or relative amount) of stable Cs and Sr have been estimated for each compartment (or tissue) and species considered (Tables 3 and 4). The percent loading of each element in soft tissues relative to hard tissues (including bone, gills and scales) was then estimated using data for individual tissues. Overall, the distribution of Cs was quite uniform between body tissues for all species considered, as reflected by their load-to-biomass ratios. The percent loadings of Cs ranged from 78.9 to 92.2% in fish and amphibian soft tissues, and from 7.8 to 21.1% in biota hard tissues (Table 3). These values corresponded quite well with differences in percent biomasses of hard-to-soft tissues, which ranged from 87 to 90% for soft tissues, and fell between 10 and 13% for hard tissues, respectively. As a result, Cs load-to-biomass ratios were near unity, ranging from 0.77 to 1.79 for biota tissues (Table 3).

Similar calculations that were performed to assess the internal partitioning patterns of Sr, clearly showed that Sr tends to concentrate in biota hard tissues (as expected). Percent Sr loading in biota soft tissues fell between 1.19 and 2.39%, whereas loadings in hard tissues ranged from 97.6 to 98.8%, despite the much smaller biomass of hard tissues relative to soft tissues in the body. As a result, Sr load-to-biomass ratios ranged from 273 to 416 for fishes, with a value of approximately 749 for amphibians. Estimated values were slightly higher for amphibians possibly because amphibian bone mass (10%) is more than double that of fish (4.7%) (Table 4). In addition, it appears that Sr load-to-biomass ratios may be slightly lower for immature than mature pike, possibly due to lags in Sr accumulation by younger fish.

Overall, evaluation of Cs and Sr chemical loading data for fish and amphibian tissues again indicates that in most cases, stable elements can be sub-divided into two categories, which include bone-seekers and uniformly-distributed elements. It is likely that similar trends exist for other bone-seeking elements.

TABLE 1. PREDICTIVE RELATIONSHIPS BETWEEN WHOLE BODY WEIGHT (IN KG), X, AND TISSUE WEIGHT (IN G) FOR TELEOST FISHES AND AMPHIBIANS, Y [19–24]

Tissue Type	Relationship between Whole Body Weight and Tissue Biomass	Tissue-to-Body Weight (%) Geomean ± GeoSE (Min. –Max.) [n]
<b>Teleost Fishes:</b>		
Bone	$Y = 40.68 \cdot X^{1.03}$ ( $r^2 = 0.992$ )	$4.71 \pm 0.035$ (2.34–9.1) [17]
Brain	$Y = 0.960 \cdot X^{0.504}$ ( $r^2 = 0.747$ )	$0.087 \pm 0.00019$ ( $7.02 \times 10^{-5} - 2.29$ ) [183]
Eyes	$Y = 5.36 \cdot X^{0.76}$ ( $r^2 = 0.727$ )	$0.504 \pm 0.017$ (0.034–1.65) [174]
Gills	n.a.	$1.3 \pm 0.076$ (0.7 – 1.8) [4]
Gizzard	n.a.	1.80 (1.8, 1.8) [2]
Gizzard Contents	n.a.	$0.242 \pm 0.200$ (0.03–0.7) [5]
Gonads (female)	$Y = 3.67 \cdot X^{0.729}$ ( $r^2 = 0.340$ )	$1.53 \pm 0.089$ (0.040–6.41) [39]
Gonads (male)	$Y = 2.03 \cdot X^{1.13}$ ( $r^2 = 0.421$ )	$0.860 \pm 0.124$ (0.034–1.8) [35]
Heart	$Y = 1.92 \cdot X^{1.00}$ ( $r^2 = 0.915$ )	$0.192 \pm 0.012$ (0.077–2.71) [180]
Kidney	$Y = 5.16 \cdot X^{1.03}$ ( $r^2 = 0.891$ )	$0.518 \pm 0.014$ (0.155–1.44) [137]
Liver	$Y = 13.42 \cdot X^{1.08}$ ( $r^2 = 0.899$ )	$1.43 \pm 0.015$ (0.222–6.23) [216]
Muscle	n.a.	$64.3 \pm 0.051$ (55.3–76.7) [5]
Scales	n.a.	7.0 [1]
Skin	n.a.	7.1 [1]
Skin & Scales	n.a.	$12.0 \pm 0.053$ (9.3–14.1) [5]
Spleen	$Y = 1.12 \cdot X^{0.98}$ ( $r^2 = 0.856$ )	$0.112 \pm 0.014$ (0.031–0.413) [77]
Stomach/Intestine	$Y = 39.61X + 36.76$ ( $r^2 = 0.894$ )	$5.06 \pm 14.6$ (0.200–12.3) [157]
Thyroid	$Y = 0.0131X + 8 \times 10^{-5}$ ( $r^2 = 0.628$ )	$1.42 \times 10^{-3} \pm 2.03 \times 10^{-5}$ ( $2.03 \times 10^{-6} - 0.162$ ) [170]
Viscera	n.a.	$10.4 \pm 0.091$ (6.5–16.1) [3]
<b>Amphibians:</b>		
Adrenal Gland	n.a.	$0.0287 \pm 0.036$ (0.0177–0.0372) [12]
Bone	n.a.	10 [1]
Brain	n.a.	$0.200 \pm 0.076$ (0.089–0.553) [11]
Eyes	$Y = 4.124 \cdot X^{0.693}$ ( $r^2 = 0.987$ )	$0.751 \pm 0.047$ (0.494–1.57) [12]
Gonads	n.a.	0.02 (estimated)
Heart	$Y = 0.464 \cdot \ln(X) + 1.96$ ( $r^2 = 0.986$ )	$0.871 \pm 0.027$ (0.317–1.26) [41]
Kidney	n.a.	$0.978 \pm 0.027$ (0.273–2.53) [41]
Liver	n.a.	$5.70 \pm 0.025$ (1.89–7.77) [41]
Lung	$Y = 3.68 \cdot X^{0.471}$ ( $r^2 = 0.978$ )	$1.70 \pm 0.026$ (0.531–2.48) [41]
<sup>a</sup> Muscle	n.a.	0.69 (estimated)
Spleen	$Y = 0.0877 \cdot \ln(X) + 0.443$ ( $r^2 = 0.814$ )	$0.275 \pm 0.038$ (0.042–0.512) [40]
Stomach/Intestines	n.a.	$8.67 \pm 0.022$ (4.72–9.90) [35]
Thyroid	n.a.	$0.014 \pm 0.055$ (0.00646–0.0281) [12]

<sup>a</sup> Muscle biomass for amphibians was estimated as the difference between whole body weight and the masses of the remaining tissues.

TABLE 2. CONCENTRATION RATIOS MEASURED IN MATURE TISSUES FOR MATURE NORTHERN PIKE COLLECTED FROM PERCH LAKE. THE CS CONCENTRATION RATIO REPRESENTS THE MEAN CS CONCENTRATION IN A TISSUE RELATIVE TO THE REFERENCE TISSUE (IN THIS CASE, MUSCLE) AND REFLECTS THE UNIFORMITY OF CS IN THE BODY. SIMILAR TRENDS WERE OBSERVED FOR IMMATURE PIKE, MATURE BULLHEADS AND BULLFROGS

Element	Concentration Ratio (CR) in Tissues of Mature Pike				
	Blood	Kidney	Liver	Gonads	Bone
Univalent Elements:					
Cs	0.526 ± 0.184	0.579 ± 0.238	0.461 ± 0.150	0.421 ± 0.196	1.32 ± 0.401
K	0.529 ± 0.103	0.628 ± 0.144	0.529 ± 0.118	0.443 ± 0.138	1.98 ± 0.301
Na	1.45 ± 0.636	1.84 ± 0.509	1.66 ± 0.408	1.38 ± 0.452	9.66 ± 1.37
Rb	0.519 ± 0.152	0.588 ± 0.210	0.588 ± 0.163	0.470 ± 0.204	1.71 ± 0.395
Divalent Elements:					
Ba	0.408 ± 0.307	5.33 ± 3.03	0.449 ± 0.254	0.388 ± 0.255	212 ± 114
Ca	0.719 ± 0.225	1.95 ± 0.926	0.980 ± 0.407	0.641 ± 0.240	1185 ± 263
Mg	0.353 ± 0.074	0.662 ± 0.146	0.720 ± 0.166	0.604 ± 0.137	17.0 ± 2.02
Sr	0.714 ± 0.216	2.43 ± 1.19	1.43 ± 0.581	1.02 ± 0.283	1171 ± 230

TABLE 3. SUMMARY OF THE RELATIVE INFLUENCE OF INTERNAL PARTITIONING AND COMPARTMENT SIZE ON CESIUM LOADINGS IN HARD AND SOFT TISSUES OF FISHES AND AMPHIBIANS

Tissue Type	Cs Concentration Ratio (CR)		% Cesium Loading in Biota Tissues			
	Fish	Frogs	Immature Pike	Mature Pike	Brown Bullhead	Bullfrogs
Soft Tissues:						
Adrenal Gland	n.a.	<sup>a</sup> 0.629	n.a.	n.a.	n.a.	0.020%
Brain	<sup>a</sup> 0.538–0.769	<sup>a</sup> 0.629	0.068%	0.053%	0.065%	0.143%
Eyes	<sup>a</sup> 0.538–0.769	<sup>a</sup> 0.629	0.392%	0.306%	0.376%	0.536%
Gizzard & Contents	<sup>a</sup> 0.538–0.769	<sup>a</sup> 0.629	1.59%	1.24%	1.52%	n.a.
Gonads	<sup>b</sup> 0.421–0.778	<sup>b</sup> 0.769	0.833%	0.619%	1.35%	0.915%
Heart	<sup>a</sup> 0.538–0.769	<sup>b</sup> 0.488	0.149%	0.117%	0.143%	0.455%
Kidney	<sup>b</sup> 0.579–0.808	<sup>b</sup> 1.00	0.423%	0.280%	0.359%	0.852%
Liver	<sup>b</sup> 0.444–0.488	<sup>b</sup> 0.500	0.723%	0.699%	0.631%	3.48%
Lung	n.a.	<sup>a</sup> 0.629	n.a.	n.a.	n.a.	1.21%
Muscle	<sup>b</sup> 1	<sup>b</sup> 1	65.1%	72.6%	73.0%	78.2%
Skin	<sup>a</sup> 0.538–0.769	n.a.	5.53%	4.31%	5.29%	n.a.
Spleen	<sup>a</sup> 0.538–0.769	<sup>a</sup> 0.629	0.087%	0.068%	0.083%	0.196%
Thyroid	<sup>a</sup> 0.538–0.769	<sup>a</sup> 0.629	0.111%	0.086%	0.106%	0.010%
Stomach & Intestines	<sup>a</sup> 0.538–0.769	<sup>a</sup> 0.629	3.94%	3.07%	3.77%	6.18%
Hard Tissues:						
Bone	<sup>b</sup> 1.32–2.69	<sup>b</sup> 0.750	12.6%	8.18%	11.0%	7.84%
Gills	<sup>b</sup> 1.28	n.a.	2.73%	1.76%	2.37%	n.a.
Scales	<sup>a</sup> 0.81	n.a.	5.74%	6.41%	scaleless	n.a.
Data Summary:						
Soft Tissue % Load:			78.9%	83.5%	86.6%	92.2%
Hard Tissue % Load:			21.1%	16.4%	13.4%	7.8%
Hard-to-Soft Tissue Load Ratio:			26.8%	19.6%	15.4%	8.51%
Soft Tissue % Biomass:			87%	87%	87%	90%
Hard Tissue % Biomass:			13%	13%	13%	10%
Hard-to-Soft Tissue Biomass Ratio:			15.0%	15.0%	15.0%	11.1%
Load-to-Biomass Ratio:			1.79	1.31	1.03	0.766

<sup>a</sup> Estimated based on measured bone-to-soft tissue concentration ratio.

<sup>b</sup> This study.

TABLE 4. SUMMARY OF THE RELATIVE INFLUENCE OF INTERNAL PARTITIONING AND COMPARTMENT SIZE ON STRONTIUM LOADINGS IN HARD AND SOFT TISSUES OF FISHES AND AMPHIBIANS. THE SR CONCENTRATION RATIOS REPRESENTS THE MEAN SR CONCENTRATION IN A TISSUE RELATIVE TO THE REFERENCE TISSUE (IN THIS CASE, MUSCLE) AND REFLECTS THE UNIFORMITY OF SR IN THE BODY

Tissue Type	Sr Concentration Ratio (CR)		% Strontium Loading in Biota Tissues			
	Fish	Frogs	Immature Pike	Mature Pike	Brown Bullhead	Bullfrog
Soft Tissues:						
Adrenal Gland	n.a.	<sup>a</sup> 1.75	n.a.	n.a.	n.a.	0.001%
Brain	<sup>a</sup> 3	<sup>a</sup> 1.75	0.006%	0.004%	0.005%	0.004%
Eyes	<sup>a</sup> 3	<sup>a</sup> 1.75	0.032%	0.022%	0.030%	0.015%
Gizzard & Contents	<sup>a</sup> 3	n.a.	0.091%	0.063%	0.085%	n.a.
Gonads	<sup>b</sup> 0.556–1.80	<sup>b</sup> 1.80	0.021%	0.025%	0.056%	0.030%
Heart	<sup>c</sup> 3.6	<sup>b</sup> 3.31	0.018%	0.010%	0.014%	0.021%
Kidney	<sup>b</sup> 1.52–2.43	<sup>b</sup> 1.84	0.018%	0.014%	0.019%	0.019%
Liver	<sup>b</sup> 0.407–1.70	<sup>b</sup> 1.94	0.014%	0.023%	0.049%	0.109%
Lung	n.a.	<sup>a</sup> 1.75	n.a.	n.a.	n.a.	0.034%
Muscle	<sup>b</sup> 1	<sup>b</sup> 1	1.36%	0.946%	1.27%	0.779%
Skin	<sup>c</sup> 2.3	n.a.	0.426%	0.240%	0.322%	n.a.
Spleen	<sup>c</sup> 2.1	<sup>a</sup> 1.75	0.006%	0.003%	0.005%	0.005%
Thyroid	<sup>a</sup> 3	<sup>a</sup> 1.75	0.004%	0.007%	0.008%	0.0003%
Stomach & Intestines	<sup>a</sup> 3	<sup>a</sup> 1.75	0.396%	0.223%	0.299%	0.171%
Hard Tissues:						
Bone	<sup>b</sup> 741–1171	<sup>b</sup> 830	85.2%	90.9%	94.1%	98.8%
Gill	<sup>c</sup> 145	n.a.	3.99%	2.77%	3.72%	n.a.
Scales	<sup>c</sup> 46	n.a.	8.40%	4.74%	scaleless	n.a.
Data Summary:						
Soft Tissue % Load:			2.39%	1.58%	2.16%	1.19%
Hard Tissue % Load:			97.6%	98.4%	97.8%	98.8%
Hard-to-Soft Tissue Load Ratio:			4078%	6221%	4532%	8317%
Soft Tissue % Biomass:			87%	87%	87%	90%
Hard Tissue % Biomass:			13%	13%	13%	10%
Hard-to-Soft Tissue Biomass Ratio:			15.0%	15.0%	15.0%	11.1%
Load-to-Biomass Ratio:			273	416	303	749

<sup>a</sup> Estimated based on measured bone-to-soft tissue concentration ratio.

<sup>b</sup> This study.

<sup>c</sup> Estimated based on literature data, as summarized by Yankovich and Beaton [17].

#### 4. SUMMARY AND RECOMMENDATIONS

In summary, like the environment, organisms can be sub-divided into compartments based on structure and function. Compartments can be assigned based on their perceived radiosensitivity and/or ecological significance [6], as well as their chemical composition and their propensity to accumulate radionuclides (as discussed in this paper). Consequently, the number of internal compartments may change, depending upon the biological attributes of a given tissue, as well as the radionuclides being considered and their behaviour in the body. For example, the univalent and divalent elements considered here can be sub-divided into three groups: i) Uniformly-distributed elements; ii) Matrix-bound, bone-seeking elements; and iii.) Elements that tend to deposit on crystal surfaces. Based on these groups, two conceptual models can be developed for Reference Animal that relate to the chemical behaviour of the elements under consideration. The first model of Reference Fish or Frog can be assumed to consist of one homogeneous compartment to account for elements that are uniformly-distributed between tissues. In such cases, existing models for internal dose to whole organisms that account for body size [e.g. 4–5] can be applied. The second model can consist of 2 to 3 compartments, including soft tissues, bone and/or scales, depending on whether or not the species under consideration has scales. Models, similar to those discussed by Pentreath and Woodhead [6], could then be applied for multiple compartments.

The approach developed in this paper can also be applied when estimating internal doses received from other types of radionuclides with differing internal partitioning patterns. For example, iodine tends to partition in thyroid tissue. Consequently, separation of a Reference Organism into hard tissues and soft tissues is not likely a valid approach. Instead, the same general approach that is described above could be applied, where the body can be sub-divided into thyroid versus non-thyroid tissue compartments to assess the potential internal dose to biota received from iodine.

Work in this area is ongoing at AECL and preliminary evaluation indicates that similar trends are being observed for other bone-seeking elements. The work presented in this paper is primarily focused on data collected in one aquatic system for a subset of stable analogues to radionuclides. More work is required to determine if similar trends exist in other systems with varying physico-chemical attributes, for other families of elements and for other types of biota with different lifestyles than the species considered here. In addition, information presented in this paper should be assessed in the context of existing internal ecodosimetry models to facilitate the improvement of internal dose estimation for non-mammalian species.

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# Significance of the air pathway in contributing radiation dose to biota

K.A. Higley<sup>a</sup>, S.L. Domotor<sup>b</sup>

<sup>a</sup> Department of Nuclear Engineering, Oregon State University, OR, United States of America

<sup>b</sup> United States Department of Energy, Office of Environmental Policy and Guidance, Washington, DC, United States of America

**Abstract.** A screening methodology was developed as part of the U.S. Department of Energy's (DOE) graded approach for evaluating radiation doses to aquatic and terrestrial biota from contaminants released into the environment. Included in the graded approach methodology were limiting media concentrations for water, soil, and sediment for twenty-three radionuclides. These concentrations were designed to restrict potential doses to biota below biota dose limits (i.e., dose rate guidelines) specified within existing and proposed DOE regulations. While implicitly included in the derivation of limiting media concentrations, separate biota concentration guides (BCGs) for air were not provided. This paper presents BCGs for air developed within the context and framework of the DOE methodology. The same twenty-three radionuclides are examined. Three air exposure pathways are considered: external exposure (cloudshine), inhalation, and absorption. Allometric equations are used to assess exposure via inhalation, and simplifying assumptions (similar to those used in human dose calculations) are used to assess external and absorption pathways. For purposes of comparison, the air BCGs are compared to current DOE air concentration limits for humans. This analysis validated the initial assumption that the air pathway is unlikely to be a major exposure pathway for biota. In addition, limits for humans are sufficiently restrictive that at sites with active air releases no populations of terrestrial animals or plants are likely to receive significant doses from this exposure pathway.

## 1. INTRODUCTION

A screening methodology was developed as part of the U.S. Department of Energy's (DOE) graded approach for evaluating radiation doses to aquatic and terrestrial biota [1]. Included in the methodology were limiting concentrations (Biota Concentration Guides – BCGs) for radionuclides in soil, water, and sediment. However, the methodology purposely excluded the active air (i.e., continuous air emission) release pathway from routine consideration. The rationale for this exclusion was that doses to biota from exposure to contaminated air were estimated to be relatively insignificant compared to other pathways of exposure. Current U.S. Government policies restrict active air emissions so that dose to the public is less than 0.1 mSv/y [2]. These policies were a significant factor in the original decision not to include the active air pathway in the graded approach methodology. Also, unlike exposures to radionuclides in soil, water, and sediment, the exposure pathways from active air releases are generally the same for biota as they are for humans. The purpose of this paper is to examine the significance of the air pathway in contributing radiation dose to biota and the validity of the decision to exclude it from routine consideration.

## 2. METHODOLOGY

Within the context of radiation doses to biota from atmospheric releases there are three air exposure pathways of concern: external exposure (cloudshine), inhalation, and absorption. The methodology used to derive limiting air concentrations for exposure under each of these scenarios is described below. All other potential exposure pathways are a consequence of deposition of airborne radionuclides onto land or water. These other pathways are already explicitly incorporated in the graded approach methodology [1].

### 2.1. External exposure

The exposure of terrestrial animals from submersion in contaminated air occurs in a manner similar to that experienced by humans. Screening-level external dose coefficients for exposure of terrestrial animals to radionuclides in air were based on three principal assumptions. First, the radioactive cloud was assumed to be semi-infinite in extent and to contain uniform concentrations of radionuclides.

Second, the exposed organism was assumed to be infinitely small. Finally, the organism was presumed to be located 100% of the time at the air-soil interface. Thus, the assumption of exposure at the boundary of a semi-infinite medium results in an absorbed dose rate in the organism which is one-half of the dose rate in an infinite source volume. This methodology is conceptually similar to that described in the DOE technical standard [1] for the derivation of external dose factors for soil, water, and sediment.

## 2.2. Inhalation doses

The intake of radionuclides through inhalation was calculated and subsequently evaluated for terrestrial animals over a range of body mass and metabolic rates (e.g., a marsh wren; a heron; a large elk) at allowable air concentrations at DOE sites. A brief description of the approach taken to derive inhalation BCGs for biota is shown below.

Studies on wildlife indicate that the lung ventilation rate scales as a function of body mass to the  $\sim 3/4$  power [3, 4]. Values reported for mammals have shown that [3]:

$$r_{inhalation} = 0.334 M^{0.76}$$

where  $r_{inhalation}$  is the inhalation rate ( $l \text{ min}^{-1}$ ), 0.334 is a constant derived through regression analysis of the data, and  $M$  is the body mass in kg.

If an organism is exposed to a unit concentration,  $C_{air}$  of a contaminant in air (e.g.,  $Bq \text{ m}^{-3}$ ), then the daily rate of inhalation of a radionuclide ( $Bq \text{ d}^{-1}$ ) is calculated as the product of the inhalation rate adjusted to a daily rate in  $\text{m}^3 \text{ d}^{-1}$  and the near-surface air concentration (in  $Bq \text{ m}^{-3}$ ) of the nuclide. Hence, the inhalation rate ( $Bq \text{ d}^{-1}$ ) is calculated as:

$$I_{inhalation} = C_{air} 0.481 M^{0.76}$$

where all terms have been defined.

This approach to estimating intake through inhalation assumes all material inhaled is retained. However, because of differences in solubility in body fluids, material taken into the body via inhalation may (or may not) be more readily absorbed than if taken in via ingestion. Zach [5] assessed the contribution of inhalation by food animals to dose (to man), and derived a series of correction factors (PT/IT) that provided an adjustment for inhalation relative to ingestion. These factors are used to correct the inhalation rate ( $I'$ ) of an animal to that of an equivalent amount of ingested soil:

$$I'_{inhalation} = \frac{PT}{IT} C_{air} 0.481 M^{0.76}$$

where PT is the percentage transfer to body fluids from inhalation, IT is the percentage transfer to body fluids from ingestion, and all other terms have been defined.

If first order kinetics for input and loss to the animal are assumed, and the animal is treated as a single compartment, then the radionuclide concentration in the animal, at any time,  $T$  (in days) is given by:

$$C_{animal} = \frac{R_{adj}}{k_{eff} M} (1 - e^{-k_{eff} T})$$

where  $C_{animal}$  is the concentration of radionuclide in the animal ( $Bq \text{ kg}^{-1}$  of tissue) predicted to occur from continuous exposure to  $1 Bq \text{ m}^{-3}$  of contaminant in air;  $k_{eff}$  is the effective loss-rate constant for the nuclide (in  $\text{d}^{-1}$ ), and  $T$  is chosen to be equal to the maximum lifespan of the animal:

$$T_{ls,d} = 365.25 \cdot 0.69 M^{0.28}$$

where  $T_{ls,d}$  is the maximum lifespan in days; 365.25 is a conversion factor from years to days, and all other terms have been defined.

The dose-rate to the tissues of the organism can be estimated through use of the internal dose conversion factors:

$$D_{\text{animal}} = C_{\text{animal}} DCF_{\text{int}}$$

where  $DCF_{\text{int}}$  is the ( $\text{Gy y}^{-1}$  per  $\text{Bq kg}^{-1}$ ) is the dose conversion factor for radionuclides in the tissue of the organisms and  $D_{\text{animal}}$  is the dose rate ( $\text{Gy y}^{-1}$ ) from the radionuclides in the tissue. Values of DCF have been previously tabulated in the DOE technical standard [1].

That air concentration which would result in a dose rate at some predetermined dose limit can then be calculated through manipulation of the preceding equations:

$$BCG_{\text{inhalation}} = \frac{DL}{DCF_{\text{int}} C_{\text{animal}}}$$

### 2.3. Dermal absorption calculations

The absorption of gaseous airborne radionuclides into the tissues of plants and animals were examined (there is no known mechanism for significant absorption of radionuclides in particulate form). Some radionuclides in gaseous form are readily absorbed, especially  $^3\text{H}$  as tritiated water and  $^{14}\text{C}$  as carbon dioxide.

Limiting air concentrations for the dermal absorption pathway were calculated in a manner similar to that described in ICRP 30, Part 1 [6] for workers. The calculation presumes a prolonged exposure to a radioactively contaminated cloud. Under these circumstances, it can be presumed that equilibrium is reached between the concentration of radionuclide in tissue,  $C_T$  and the concentration in air ( $C_{\text{air}}$ ). This relationship can be expressed as:

$$C_T = \frac{\delta C_{\text{air}}}{\rho_T}$$

where  $\rho_T$  is the density of tissue, and  $\delta$  is the solubility of a specific gas in tissue. The solubility is measured as the volume of gas in equilibrium with the unit volume of tissue under (normal) atmospheric pressure. Solubility is a function of the atomic weight of the gas and increases with increasing atomic number. Tabulated values of solubility measured in water (at body temperature) range from 0.02 for hydrogen to 0.1 for Xe. Because these values can be increased in fatty (adipose) tissue, a default value of 1.0 was used in the derivation of  $BCG_{\text{absorption}}$ .

The BCGs were calculated as:

$$BCG_{\text{absorption}} = \frac{DL}{DCF_{\text{int}} C_{\text{Tissue}}}$$

## 3. RESULTS

### 3.1. External exposure

The exposure of terrestrial animals from submersion in contaminated air occurs at a rate similar to that experienced by people. Thus, if a DOE facility or site is in compliance with the dose limit for the general public (0.1 mSv/y), then the total dose to terrestrial animals should be far below the biota limit of 1 mGy/d. The 0.1 mSv/y (10 mrem/y) dose limit to the public is required under the National Emission Standards for Hazardous Air Pollutants (NESHAPS) [7] for air emissions from DOE facilities and specified in DOE Order 5400.5, "Radiation Protection of the Public and the

Environment” [2]. Terrestrial plants also typically receive external doses of ionizing radiation from air at rates similar to those experienced by humans, and their dose limits are even higher: 10 mGy/d. A listing of external dose conversion factors for people and biota, along with the derived biota concentration guides (BCGs) are shown in Table 1. A comparison of concentration guides in air for the general public at 0.1 mSv/d (10 mrem/y) and those derived for the 1 mGy/d (0.1 rad/d) biota dose limit is shown in Figure 1.

With the exception of  $^{60}\text{Co}$ , the concentration limits for biota are higher than for humans (skin or total body). This is to be expected, considering the conservative nature of the calculation methodology used in the derivation of the dose conversion factors.

### 3.2. Inhalation

Because the equations used to estimate the tissue concentrations from inhalation of contaminated air are mass dependent, the limiting air concentrations also are mass dependent. Calculated values of  $\text{BCG}_{\text{inhalation}}$  for six body masses for each of the 23 nuclides are shown in Table 2 and are graphically presented in Figure 2.

As can be seen by inspection of Table 2 and Figure 2, there is some mass effect, but it varies by nuclide. In general, the larger the animal, the more restrictive the air concentration. This is due to mass dependence of the lifespan and the biological elimination rate (specific values of these are listed in [1]). In general, the heavier the animal, the longer the projected lifespan which allows for greater tissue buildup of the nuclides. There are exceptions, however. For all but 5 nuclides there was no significant difference between the limiting value and that calculated for 1000kg body mass. For the elements Ce, I, and Zr, the 1000kg body weight did not generate the limiting BCG. For Ce, the limiting body mass was the 1 kg organism, for I and Zr, the 0.001 kg organism was limiting.

The  $\text{BCG}_{\text{inhalation}}$  values were compared to derived concentration guides (DCG(air)) [2] for members of the general public. The comparison is shown in Figure 3. None of the  $\text{BCG}_{\text{inhalation}}$  values are more restrictive than the DCG(air) values. It was found that the air concentrations to which populations of these terrestrial animals would need to be exposed in order to reach the dose limit for terrestrial animals at DOE sites would need to be two to three orders of magnitude greater than the allowable air concentrations for humans.

### 3.3. Dermal absorption

Because the dose limits to terrestrial animals are more restrictive than for plants or aquatic animals, a value of 0.01 Gy/d was used for DL. Figure 4 shows a comparison of the derived BCGs for absorption with the human limits [2] for skin absorption.

## 4. SUMMARY

The Biota Dose Assessment Committee (BDAC) derived BCGs for air to evaluate its potential contribution to biota dose. Active air BCGs were calculated using ecologically-based modeling approaches consistent with those used for the other media types in the DOE graded approach methodology. Inhalation and external exposure pathways were included. Allometric equations were used to assess exposure via inhalation, and did not consider other pathways of exposure (e.g., consumption of foodstuffs contaminated by deposition of radionuclides). For the active air pathway the dose limits and derived concentration guides for radiation protection of humans are more restrictive than the BCGs derived for radiation protection of biota. (Figure 5).

In summary, this paper presents air concentration limits that have been derived to be protective of biota from active discharge or radionuclides to the air. However, the active air pathway is not included in the graded approach when assessing compliance with the DOE biota dose rate guidelines. Exposure of biota to radionuclides released to the atmosphere is not likely to be significant when compared to other pathways. Also, these limits when compared to those for the general (human) public, are never more restrictive than the human limit.

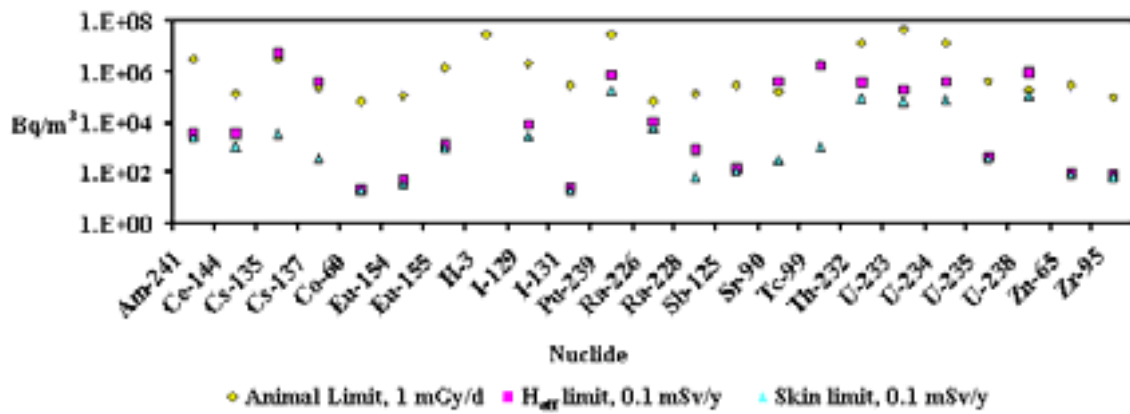


FIG. 1. Comparison of derived air concentration limits for humans and biota: air submersion pathway.

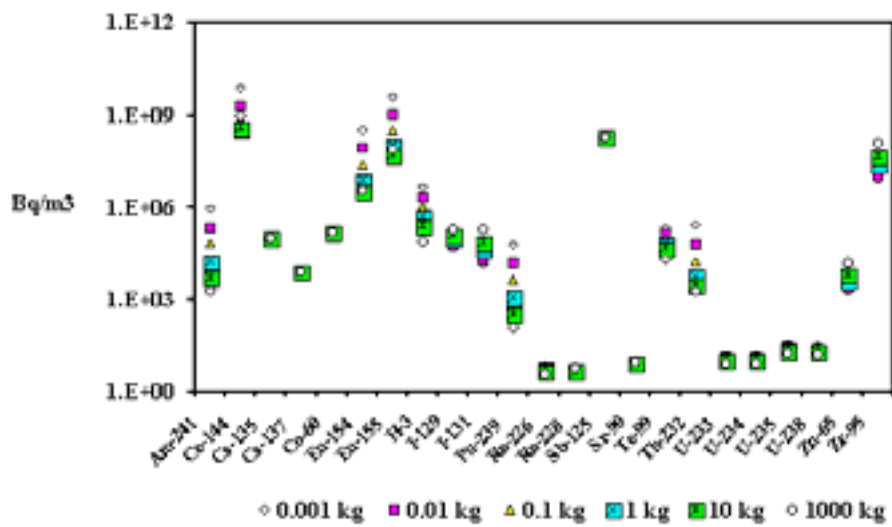


FIG. 2. Mass dependence of BCGs for the air inhalation pathway.

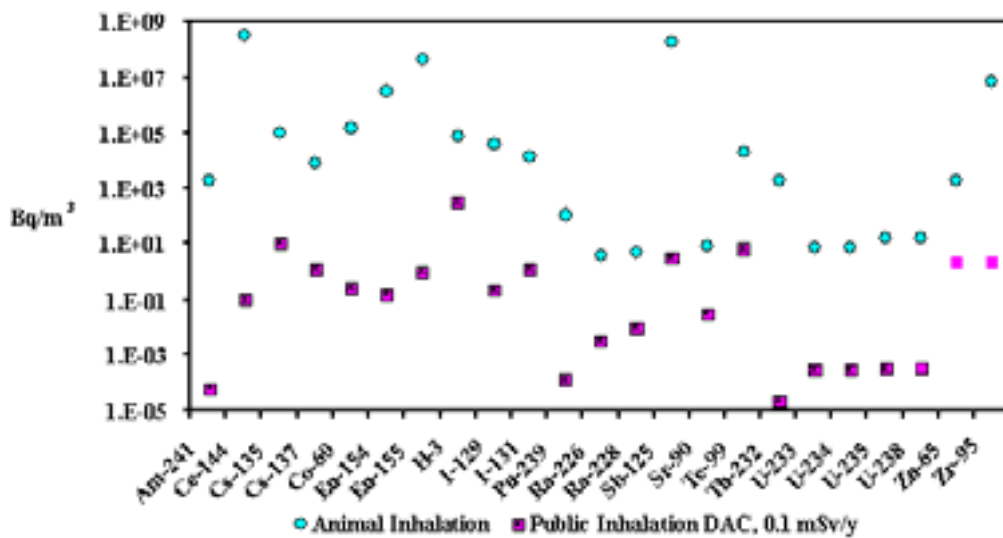


FIG. 3. Comparison of human and biota concentration limits for the air inhalation pathway.



TABLE 1. EXTERNAL DOSE CONVERSION FACTORS AND BCGs<sup>a</sup> FOR SUBMERSION IN CONTAMINATED AIR

Nuclide	Human Dose Coefficients for Submersion in Contaminated Air <sup>b</sup>		Biota Dose Coefficients for Submersion in Contaminated Air	
	External Dose Conversion Factors			
	H <sub>eff</sub> , Sv/y per Bq/m <sup>3</sup>	Skin, Sv/y per Bq/m <sup>3</sup>	Biota <sup>c</sup> , Gy/y per Bq/m <sup>3</sup>	BCG <sub>air, external</sub> , Bq m <sup>-3</sup>
Am-241	2.58E-08	4.04E-08	1.17 × 10 <sup>-7</sup>	3.13 × 10 <sup>6</sup>
Ce-144	2.69E-08	9.25E-08	2.83 × 10 <sup>-6</sup>	1.29 × 10 <sup>5</sup>
Cs-135	1.78E-11	2.86E-08	1.17 × 10 <sup>-7</sup>	3.13 × 10 <sup>6</sup>
Cs-137	2.44E-10	2.72E-07	1.67 × 10 <sup>-6</sup>	2.19 × 10 <sup>5</sup>
Co-60	3.98E-06	4.58E-06	5.50 × 10 <sup>-6</sup>	6.64 × 10 <sup>4</sup>
Eu-154	1.94E-06	2.62E-06	3.17 × 10 <sup>-6</sup>	1.15 × 10 <sup>5</sup>
Eu-155	7.86E-08	1.07E-07	2.58 × 10 <sup>-7</sup>	1.41 × 10 <sup>6</sup>
H-3	–	–	1.17 × 10 <sup>-8</sup>	3.13 × 10 <sup>7</sup>
I-129	1.20E-08	3.47E-08	1.67 × 10 <sup>-7</sup>	2.19 × 10 <sup>6</sup>
I-131	3.53E-06	4.99E-06	1.17 × 10 <sup>-6</sup>	3.13 × 10 <sup>5</sup>
Pu-239	1.34E-10	5.87E-10	1.17 × 10 <sup>-8</sup>	3.13 × 10 <sup>7</sup>
Ra-226	9.94E-09	1.51E-08	5.67 × 10 <sup>-6</sup>	6.44 × 10 <sup>4</sup>
Ra-228	1.18E-07 <sup>d</sup>	1.42E-06 <sup>d</sup>	2.83 × 10 <sup>-6</sup>	1.29 × 10 <sup>5</sup>
Sb-125	6.37E-07	8.36E-07	1.17 × 10 <sup>-6</sup>	3.13 × 10 <sup>5</sup>
Sr-90	2.38E-10	2.90E-07	2.33 × 10 <sup>-6</sup>	1.56 × 10 <sup>5</sup>
Tc-99	5.11E-11	8.65E-08	1.75 × 10 <sup>-7</sup>	2.09 × 10 <sup>6</sup>
Th-232	2.75E-10	1.09E-09	2.50 × 10 <sup>-8</sup>	1.46 × 10 <sup>7</sup>
U-233	5.14E-10	1.44E-09	7.75 × 10 <sup>-9</sup>	4.71 × 10 <sup>7</sup>
U-234	2.41E-10	1.34E-09	2.67 × 10 <sup>-8</sup>	1.37 × 10 <sup>7</sup>
U-235	2.27E-07	2.73E-07	7.83 × 10 <sup>-7</sup>	4.66 × 10 <sup>5</sup>
U-238	1.08E-10	9.18E-10	1.92 × 10 <sup>-6</sup>	1.90 × 10 <sup>5</sup>
Zn-65	9.15E-07	1.04E-06	1.25 × 10 <sup>-6</sup>	2.92 × 10 <sup>5</sup>
Zr-95	1.14E-06	1.42E-06	3.50 × 10 <sup>-6</sup>	1.04 × 10 <sup>5</sup>

<sup>a</sup> This BCG<sub>air</sub> is based on delivering a dose of 0.001 Gy/d to the target organism surrounded by a semi-infinite medium of air.

<sup>b</sup> Values taken from the Handbook of Health Physics and Radiological Health, Table 13.30.1 (Shleien, et al, 1998), after Federal Guidance Report No. 12 (EPA, 1993).

<sup>c</sup> Based on the calculations of Koehler (U.S. DOE, 2002)

TABLE 2. A COMPARISON OF RECOMMENDED BCGS (INHALATION) BY NUCLIDE AND ANIMAL BODY WEIGHT, IN Bq m<sup>3</sup>

Nuclide	0.001 kg	0.01 kg	0.1 kg	1 kg	10 kg	1000 kg
Am-241	9.E+05	2.E+05	6.E+04	2.E+04	5.E+03	2.E+03
Ce-144	7.E+09	2.E+09	7.E+08	3.E+08	4.E+08	1.E+09
Cs-135	1.E+05	1.E+05	1.E+05	1.E+05	1.E+05	1.E+05
Cs-137	8.E+03	8.E+03	8.E+03	8.E+03	8.E+03	8.E+03
Co-60	2.E+05	2.E+05	2.E+05	2.E+05	2.E+05	2.E+05
Eu-154	3.E+08	9.E+07	2.E+07	7.E+06	3.E+06	4.E+06
Eu-155	4.E+09	1.E+09	3.E+08	1.E+08	5.E+07	8.E+07
H-3	4.E+06	2.E+06	1.E+06	5.E+05	3.E+05	8.E+04
I-129	5.E+04	6.E+04	8.E+04	1.E+05	1.E+05	2.E+05
I-131	1.E+04	2.E+04	3.E+04	5.E+04	8.E+04	2.E+05
Pu-239	6.E+04	2.E+04	4.E+03	1.E+03	3.E+02	1.E+02
Ra-226	7.E+00	6.E+00	6.E+00	5.E+00	5.E+00	4.E+00
Ra-228	6.E+00	5.E+00	5.E+00	5.E+00	5.E+00	6.E+00
Sb-125	2.E+08	2.E+08	2.E+08	2.E+08	2.E+08	2.E+08
Sr-90	1.E+01	1.E+01	9.E+00	9.E+00	9.E+00	1.E+01
Tc-99	2.E+05	1.E+05	1.E+05	7.E+04	5.E+04	2.E+04
Th-232	3.E+05	7.E+04	2.E+04	6.E+03	3.E+03	2.E+03
U-233	1.E+01	1.E+01	1.E+01	1.E+01	1.E+01	8.E+00
U-234	1.E+01	1.E+01	1.E+01	1.E+01	1.E+01	8.E+00
U-235	3.E+01	3.E+01	3.E+01	2.E+01	2.E+01	2.E+01
U-238	3.E+01	3.E+01	2.E+01	2.E+01	2.E+01	2.E+01
Zn-65	2.E+03	2.E+03	3.E+03	4.E+03	6.E+03	2.E+04
Zr-95	8.E+06	1.E+07	2.E+07	3.E+07	5.E+07	1.E+08

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# The influence of solution speciation on uranium uptake by a freshwater bivalve (*Corbicula fluminea*): its implication for biomonitoring of radioactive releases within watercourses\*

F. Denison<sup>a</sup>, C. Adam<sup>a</sup>, J. Garnier-Laplace<sup>a</sup>, J. Smith<sup>b</sup>

<sup>a</sup> Institute of Protection and Nuclear Safety, Division of Environmental Protection, Laboratory of Experimental Radioecology, Saint-Paul-lez-Durance, France

<sup>b</sup> Centre for Ecology and Hydrology, Winfrith Technology Centre, Dorchester, United Kingdom

**Abstract.** Within the framework of using bivalves as radioindicators for freshwaters, one of the challenge to reduce uncertainty in predictive transfer models is to establish numerically well-defined relationship between solution speciation of radionuclides -depending on their biogeochemical behaviour in the ecosystem- and bioaccumulation. To realise such a programme for the biomonitoring of uranium contaminated sites, it is necessary to assess the contribution of each possible exposure sources (aqueous solution, mineral suspended matters, phytoplankton) in the contamination processes of bivalves (contamination level, uptake and depuration kinetics, tissue distribution). As a first step, a series of experiments have been performed to identify the most important uranium solution species (e.g.  $\text{UO}_2^{2+}$ ,  $\text{UO}_2\text{OH}^+$ , etc.) responsible for uranium uptake by a model bivalve species (*Corbicula fluminea*). The exposure experiments were performed in regulated and well characterised synthetic solutions, the compositions of which were varied in a systematic manner in order to investigate the influence of the significant inorganic ligands ( $\text{OH}^-$ ,  $\text{CO}_3^{2-}$ ,  $\text{PO}_4^{3-}$ ), and cations (competitive exchange on cationic transport sites by  $\text{H}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) on the bioavailability of uranium. The uptake kinetics of uranium were measured over relatively short time periods at both a whole body and individual tissue level to minimize behavioural changes due to the lack of a food source, any alteration of the exposure medium by the organisms and to ensure that elimination of uranium was not significant. The influence of natural organic matter (both on the solution speciation of uranium and also the possibility of co-transport of organic uranium complexes) was similarly investigated by incorporating a characterized preparation of a natural organic matter. The uptake was interpreted in terms of (i) the modelled solution speciation of uranium (JCHESS geochemical code and OECD/NEA thermodynamical database); (ii) the behavioural and physiological status of the organisms during the exposure experiments. This latter, especially with valve movement and filtration rate, is critical in determining the uptake kinetics of the bioavailable forms of uranium and may be influenced both by the extent of uranium contamination and also ancillary factors (such as pH, temperature) which have to be varied to extend the validity field of the data to environmentally realistic conditions. This global approach, completed by long term exposure in order to establish links between uranium bioaccumulation and biological effects, enables the development of a mechanistic model, avoiding some of the problems inherent in the more commonly applied approach of using empirical relationships between bioavailability and physicochemical parameters.

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\* Only an abstract is given as the full paper was not available.

# Theoretical conception, optimization and prognosis of synergistic effects

J.K. Kim<sup>a</sup>, V.G. Petin<sup>b</sup>

<sup>a</sup> Korea Atomic Energy Research Institute, Taejon, Republic of Korea

<sup>b</sup> Medical Radiological Research Center, Obninsk, Kaluga Region, Russian Federation

**Abstract.** There are two possible explanations for the increase in the overall biological effect under the combined action of ionizing radiation with another inactivating, mutagenic or carcinogenic agents. The first hypothesis is that the mechanism of the synergistic interaction of ionizing radiation with another effective agent may attribute to a reduced cellular capacity of repairing damage after their combined action. The second possible mechanism to explain and describe the basic features of synergistic interaction can be based on the supposition that synergism might be related to additional lethal or potentially lethal damage that arises from the interaction of sublesions induced by both agents. Based on this hypothesis, a simple mathematical model was employed. The model suggests that synergism takes place due to additional effective damage originated from the interaction of sublesions induced by each agent. These sublesions are considered to be ineffective when each agent is applied separately. The suggested model was shown to predict the greatest value of the synergistic effect, the condition under which it can be achieved as well as the dependence of synergy on the intensity of agents applied. The validity of the proposed model was tested for simultaneous action of hyperthermia with ionizing radiation on biological systems such as bacteriophage, bacterial spores, yeast and mammalian cells.

## 1. INTRODUCTION

There are many examples of interactions between ionizing radiation and other toxic physical or chemical agents. Biological responses obtained range from strong antagonism to strong synergism. The most effective simultaneous exposure to ionizing radiation and another contaminant may result in four possible modes of interaction: (i) no interaction – the effects observed are based on the most toxic agent; (ii) additivity – the effects produced by separate application of each agents are simply summed up; this mode can be subdivided on the additive and independent interaction; (iii) antagonism – the effect is less than expected additivity; (iv) synergism or supra-additivity the effect is great than expected additivity. A great number of experimental data obtained in this area stress the need for a mathematical approach in efforts to optimize and predict the interaction of environmental agents. Recently, a simple mathematical model was proposed to describe the synergistic effect of simultaneous action of ionizing radiation and high temperature [1]. The basic postulates underlying the model were not related to a particular cytotoxic agent, so the model may have an application to describe the synergistic interaction of other agents. Some examples have been already published. Our purpose here to discuss the simplest model to account for currently available experimental findings. This work describes a part of continuing research activity directed toward optimizing of combined action of two harmful agents by achieving some new insight into the synergistic mechanism.

## 2. MATHEMATICAL APPROACH

Synergism is defined as the combined interaction of two agents that exceeds the additive sum of their individual effects. On this definition, it might be reasonable to assume that some additional lethal lesions are produced during combined action. Our analytical approach is based on the supposition that the additional lethal lesions are arisen from the interaction of sublesions induced by both agents. These sublesions are non-lethal when each agent is applied separately. We assume that one sublesion produced for instance by ionizing radiation interacts with one sublesion from another environmental agent (for concreteness sake, let it be heat) to produce one additional lethal lesion. It would seem probable to suppose that the number of sublesions was directly proportional to the number of lethal lesions. Let  $p_1$  and  $p_2$  be the number of sublesions that occur for one lethal lesion induced by ionizing radiation and hyperthermia respectively. Let  $N_1$  and  $N_2$  be the mean numbers of lethal lesions in a cell produced by these agents. A number of additional lesions  $N_3$  arising from the interaction of ionizing radiation and hyperthermia sublesions may be written as:

$$N_3 = \min\{p_1N_1; p_2N_2\}. \quad (1)$$

Here,  $\min\{p_1N_1; p_2N_2\}$  is a minimal value from two variable quantities:  $p_1N_1$  and  $p_2N_2$ , which are the mean number of sublesions produced by ionizing radiation and hyperthermia respectively. In other words, this value is the mean number of additional lethal lesions per cell arising from the interaction between the ionizing radiation and the hyperthermia and accounting for the synergism. Thus, the model describes the yield of lethal lesions per cell as a function of ionizing radiation ( $N_1$ ), hyperthermia ( $N_2$ ), and interaction ( $\min\{p_1N_1; p_2N_2\}$ ) lethal lesions. Then the synergistic enhancement ratio  $k$  may be expressed as:

$$k = (N_1 + N_2 + N_3)/(N_1 + N_2). \quad (2)$$

Taking into account Equation (1), the last expression can be rewritten as:

$$k = 1 + \min\{p_1; p_2N_2/N_1\} / (1 + N_2/N_1). \quad (3)$$

It is evident from here that the highest synergistic interaction will be determined by the least value from the two functions:  $f_1 = 1 + p_1 / (1 + N_2/N_1)$  and  $f_2 = 1 + (p_2 N_2/N_1) / (1 + N_2/N_1)$ . Figure 1 shows the dependence of both this functions on the ratio of  $N_2/N_1$ , calculated for arbitrary chosen  $p_1$  and  $p_2$  ( $p_1 = 1, p_2 = 2$ ). The dotted line at this Figure depicted the dependence of the synergistic enhancement ratio on the ratio  $N_2/N_1$ , i.e. the ratio of the effects produced by each agent used in combination. Since  $f_1$  decreases while  $f_2$  increases with  $N_2/N_1$ , the greatest synergistic effect will be obtained when  $f_1 = f_2$ , i.e.:

$$p_1 / (1 + N_2/N_1) = (p_2 N_2/N_1) / (1 + N_2/N_1). \quad (4)$$

From here, the condition of the highest synergistic interaction achievement can be easily obtained:

$$p_1N_1 = p_2N_2. \quad (5)$$

It means that the highest synergistic interaction occurred when equal numbers of sublesions are produced by both agents. Taking into account Equations (3) and (5), the value of the greatest synergistic enhancement ratio is given by:

$$k_{max} = 1 + [p_1p_2/(p_1 + p_2)]. \quad (6)$$

Some examples of theoretically predicted dependencies of the synergistic enhancement ratio on the  $N_2/N_1$  ratio for various values of the basic model parameters  $p_1$  and  $p_2$  are depicted in Figures 2 and 3.

If the observed biological effect is mainly induced by heat ( $p_1N_1 < p_2N_2$ ) then taking into account Equations (1–3), the parameter  $p_1$  can be expressed as:

$$p_1 = (k_1 - 1)(1 + N_2/N_1), \quad (7)$$

where  $k_1$  is the value of synergistic enhancement ratio observed in experiments performed in this condition. On the contrary, if the observed biological effect is mainly induced by ionizing radiation, we have:

$$p_2 = (k_2 - 1)(1 + N_1/N_2), \quad (8)$$

where  $k_2$  is the experimental value of the synergistic enhancement ratio observed for the condition  $p_2N_2 < p_1N_1$ .

The corresponding number of lethal lesions can be calculated [4] as:

$$N = - \ln S, \quad (9)$$

where  $S$  is the surviving fraction.

Thus, based on Equations (7) and (8) and experimental data of  $k_1$  and  $k_2$ , we can estimate the basic model parameters  $p_1$  and  $p_2$  and then predict the value of the synergistic enhancement ratio for any  $N_1$  and  $N_2$  (Equation (3)), the highest value of the synergistic enhancement ratio (Equation (6)), and condition (Equation (5)) under which it can be achieved. It should be emphasized that since the postulates constituting the basis of the model are of very general character and not related to the mechanism of lethal action of the applied agents the described model can, in principle, describe and predict the synergistic interaction of various environmental factors.

### 3. SELECTED EXAMPLES

The main value of the mathematical approach presented is the possibility to predict the highest synergism and the  $N_2/N_1$  ratio at which it can be achieved. We tested the applicability of the model for quantitative description, prediction and optimization of the synergistic interaction observed for inactivation of cells of different origin subjected to simultaneous thermoradiation exposure, i.e. to ionizing radiation applied at different elevated temperatures. Figure 4 presents the experimentally obtained (circles) and theoretically predicted (solid lines) relationships between the synergistic enhancement ratio and the  $N_2/N_1$  ratio for inactivation of T4 bacteriophage (A), *Bacillus subtilis* spores (B), diploid *Saccharomyces cerevisiae* yeast cells (C) and cultured mammalian cells (D). The procedures for calculating these relationships have been described in detail in the previous section. Initial experimental data used for these calculations were taken from earlier papers on simultaneous thermoradiation inactivation of bacteriophage [5], bacterial spores [6], yeast [1] and mammalian cells [7,8]. The errors in the synergistic enhancement ratio values ( $k$ ) were calculated, if it was possible, from interexperimental variation. Predicted values of  $k$  were estimated by Equation (3) using the basic parameters  $p_1$  and  $p_2$  of the model which have been derived (Equations (7) and (8)) from real experiments. These parameters together with  $k_{max}$  (Equation (6)) as well as the ratio  $N_2/N_1$  (Equation (5)), at which it can be achieved, are collected in Table 1.

Since the main postulates of the model are not related to the particular mechanism of the action of inactivating agents, it was of interest to consider the applicability of the model for other agents. Figure 5 presents the experimentally obtained (circles) and theoretically predicted (solid lines) relationships of the synergistic enhancement ratio to the  $N_2/N_1$  ratio (hereinafter  $N_2$  refers to hyperthermia) for inactivation of cultured mammalian cells induced by simultaneous exposure to elevated temperature and chemical cytotoxic substances: A – tri(1-aziridiny)phosphine sulphide (TAPS), B – cis-diamminedichloroplatinum (cis-DDP), as well as for inactivation of diploid yeast cells of *Saccharomyces cerevisiae* after simultaneous exposure to hyperthermia and ultrasound (C) and 254 nm UV radiation (D). Initial experimental data and some previous calculation were taken from earlier publications of hyperthermia and cytostatic preparations [9–10] as well as for hyperthermia combined with either ultrasound [2] or ultraviolet light [3]. The errors in the synergistic enhancement ratio values ( $k$ ) were calculated, if it was possible, from interexperimental variation. Predicted values of  $k$  were estimated by Equation (3) using the basic parameters  $p_1$  and  $p_2$  of the model which have been derived (Equations (7) and (8)) from real experiments. These parameters together with  $k_{max}$  (Equation (6)) as well as the ratio  $N_2/N_1$  (Equation (5)), at which it can be achieved, are also included in Table 1.

TABLE 1. PARAMETERS OF THE MATHEMATICAL MODEL DESCRIBING THE INACTIVATION OF CELLS ON COMBINED EXPOSURE TO DIFFERENT ENVIRONMENTAL FACTORS AND HYPERTHERMIA (H)

Biological object	Inactivation agents	$p_1$	$p_2$	$K_{max}$	$N_2/N_1$
Bacteriophage T4	$\gamma + H$	2.4	1.6	2.0	1.5
<i>Bacillus subtilis</i> spores	$\gamma + H$	2.2	4.5	2.5	0.5
Yeast cells	$\gamma + H$	2.3	3.0	2.4	0.8
Mammalian cells	$\gamma + H$	4.3	30	4.8	0.14
Mammalian cells	TAPS + H	2.7	13.7	3.2	0.2
Mammalian cells	Cis-DDP + H	18.0	4.0	4.3	4.5
Yeast cells	Ultrasound + H	1.8	25.0	2.7	0.07
Yeast cells	UV-light + H	1.5	0.77	1.5	2.0

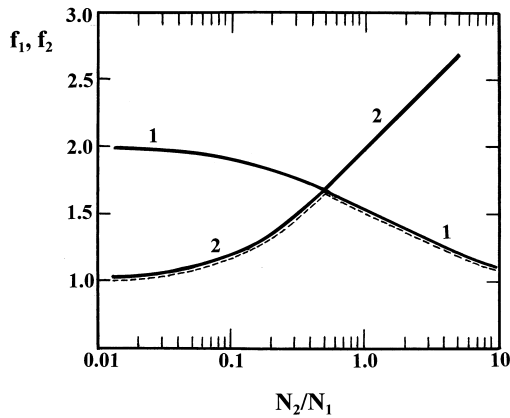


FIG. 1. The calculated dependencies of functions  $f_1$  (curve 1) and  $f_2$  (curve 2) on the  $N_2/N_1$  ratio of lethal damages induced by ionizing radiation ( $N_1$ ) and heat ( $N_2$ ) for the following values of the basic parameter:  $p_1 = 1$  and  $p_2 = 2$ . The dotted curve determines the dependence of the synergistic enhancement ratio  $k$  on the  $N_2/N_1$  ratio.

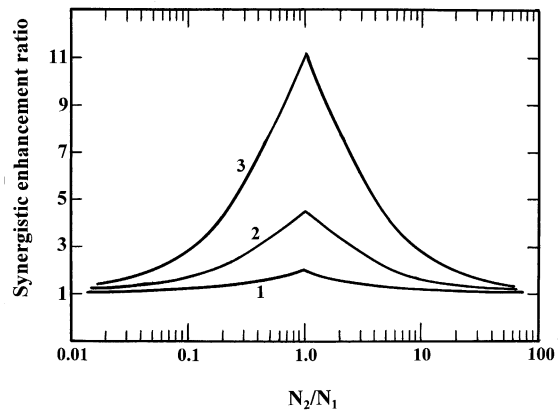


FIG. 2. Theoretically expected dependencies of the synergistic enhancement ratio on the  $N_2/N_1$  ratio of lethal damages induced by ionizing radiation ( $N_1$ ) and heat ( $N_2$ ) for the following values of the basic parameter:  $p_1 = p_2 = 2$  (curve 1), 7 (curve 2), 20 (curve 3).

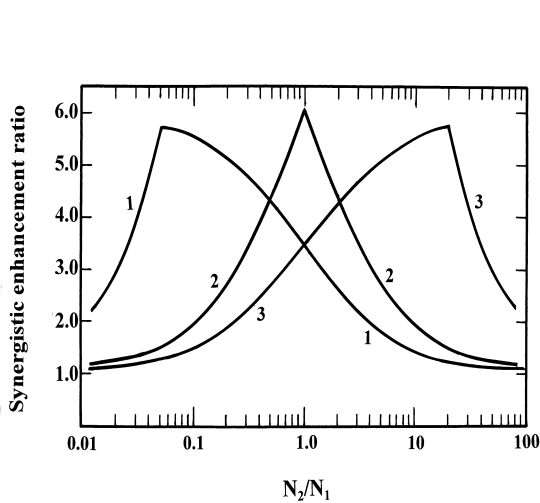


FIG. 3. Theoretically expected dependencies of the synergistic enhancement ratio on the  $N_2/N_1$  ratio of lethal damages induced by ionizing radiation ( $N_1$ ) and heat ( $N_2$ ) for the following values of the basic parameter:  $p_1 = 5$  and  $p_2 = 100$  (curve 1),  $p_1 = p_2 = 10$  (curve 2),  $p_1 = 100$  and  $p_2 = 5$  (curve 3).

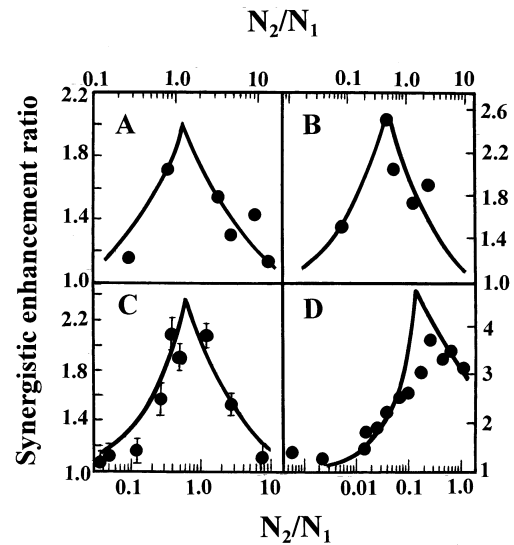


FIG. 4. Experimentally obtained (circles) and theoretically predicted (solid lines) dependencies of the synergistic enhancement ratio  $k$  on the  $N_2/N_1$  ratio for simultaneous thermoradiation inactivation of T4 bacteriophage (A), *Bacillus subtilis* spores (B), diploid *Saccharomyces cerevisiae* yeast cells (C) and cultured mammalian cells.

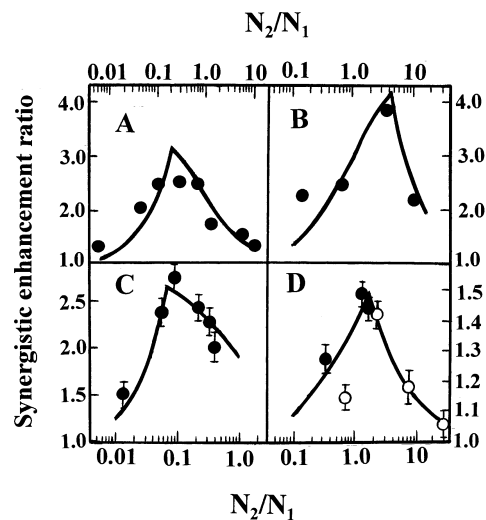


FIG. 5. Experimentally obtained (circles) and theoretically predicted (solid lines) dependencies of the synergistic enhancement ratio  $k$  on the  $N_2/N_1$  ratio for inactivation of cultured mammalian cells induced by simultaneous exposure to heat and chemical substance TAPS (A), *cis*-DDP (B) as well as for inactivation of diploid *S. cerevisiae* cells induced by simultaneous exposure to heat and 20 kHz ultrasound (C) and 254 nm UV light (D).

#### 4. DISCUSSION

The most interesting result of this paper concerns the mathematical model which has been proposed to explain the experimental data of synergistic interaction of hyperthermia with other inactivating agents. The model is based on the supposition that synergism takes place due to the additional lethal lesions arising from the interaction of non-lethal sublesions induced by both agents. These sublesions are considered noneffective after each agent taken alone. The idea of sublesions is widely used in radiobiology [11–13]. In the model, the synergistic effect is given by  $\min\{p_1N_1; p_2N_2\}$  (Equation (1)). This means that one sublesion caused by irradiation or chemicals interacts with one sublesion produced by heat. This process is assumed to proceed until the sublesions of the less frequent type is used up. However, it does not mean that the model requires some kind of long-range interaction across the whole cell or at least across the whole cell nucleus. To estimate the basic parameters  $p_1$  and  $p_2$  we have used the experimental values of the synergistic enhancement ratio  $k_1$  and  $k_2$  (Equations (7) and (8)). It means that the model takes into consideration only the actual interaction determined the synergistic effect.

The model predicts the dependence of synergistic interaction on the ratio  $N_2/N_1$  of lethal lesions produced by every agent applied (Equation (3)), the greatest value of the synergistic effect (Equation (6)) as well as the conditions under which it can be achieved (Equation (5)). The model is not concerned with the molecular nature of sublesions, and the mechanism of their interaction remains to be elucidated. In spite of the approximation used in this simplified model, it is evident from the data presented that a good agreement appears to exist between theoretical and experimental results. Moreover, the model discussed here can be used to predict conditions at which the greatest synergy can be observed and its value. A slightly greater discrepancy between the experimental and theoretical data for mammalian cells may be due to the fact that the initial data were taken from papers published by different authors. The degree of synergistic interaction was found to be dependent on the ratio of lethal damage ( $N_2/N_1$ ) induced by the two agents applied. The synergistic interaction is not observed at any  $N_2/N_1$  ratios. The important aspect of this interaction is that there appears to be a sharp dependence of the synergistic effect on the ratio  $N_2/N_1$ . One can see also that there is an optimal value of  $N_2/N_1$  for each of these cell systems at which the synergism is maximal. The effectiveness of synergistic interaction appears to decline with a deviation of the ratio  $N_2/N_1$  from optimal value.

The fact that different biological objects subjected by composite heat and ionizing radiation (or other inactivating agents) can be analyzed by the same model suggests its general validity for optimization

and prediction of synergistic effects. The degree of synergistic interaction was found to depend on the line of cell studied, their radio- and thermosensitivity. The foregoing considerations provide evidence that the highest synergy attained are smaller for microorganisms and yeast cells (1.5–2.7) and somewhat greater for mammalian cells (3.2–4.8). In accordance with the model, we might interpret this as being due to the higher number of sublesions produced for one lethal damage induced by both ionizing radiation and hyperthermia in mammalian cells.

The most remarkable feature, obtained from the model, is the conclusion that for a lower intensities of physical agents and lower concentration of chemicals lower temperature must be used to provide the greatest synergy. Actually, any decrease in the intensity of physical agents would result in a increase of the duration of thermoradiation action to achieve the same absorbed dose. Therefore, the number of thermal sublesion will also be increased resulting in the disruption of the condition at which the highest synergy should be observed (Equation (5)). Hence, to preserve an optimal  $N_2/N_1$  ratio with any decrease of the dose rate (or the intensity of other agents) the exposure temperature should be decreased. This conclusion corresponds to the published experimental results [14, 15]. It can be concluded on this basis that for a long duration of interaction, which are important for problems of the protection of environment from ionizing radiation and other physical and chemical agents, low intensities of deleterious environmental factors may synergistically interact with each other either or with environmental heat. Hence, the assessment of health or environmental risks from numerous natural and man-made agents at the level of intensities found in the biosphere should, in principle, take into account the synergistic interaction between harmful environmental stressors.

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## **Practical issues in demonstrating compliance with regulatory criteria\***

**D.B. Chambers, M. Davis**

SENES Consultants Limited, Ontario, Canada

**Abstract.** The ultimate goal of environmental protection is to ensure that communities and populations of organisms can survive and that all component parts will be self-sustaining. Thus, the focus of a strategy for environmental protection must be on the collective response of the community rather than the response of a single individual within the community.

There are many sources of uncertainty, which need to be addressed, or as a minimum acknowledged, in assessing risks to the environment. Some of these sources of uncertainty include identifying the population at risk, defining what adverse effect is to be assessed, spatial and temporal averaging of exposures, the dose calculation procedure, and the role of natural background variability, among other factors. Each of these aspects introduces uncertainty into the assessment of environmental risk. The presence of such uncertainties is an important consideration in deciding whether or not there is an environmental risk.

Two examples of tiered or graded systems for demonstrating environmental protection are discussed, namely, the “tiered” approach used by Environment Canada in their assessment of toxicity from radiation or radioactivity and the graded approach developed by the US Department of Energy. At each tier, there are two decision errors which could arise in determining whether or not a particular environment has potentially harmful levels of radiation or radioactivity. A type 1 decision error arises when it is determined that the levels are toxic when in fact they are not. In such situations, there is a high probability that impacts will be overestimated and low probability that they will be underestimated. This type of error is to be expected from (hyper-) conservative assessments. A type 2 decision error occurs when the levels of radiation or radioactivity are determined to be non-toxic when in fact they are.

The potential consequences of either type of error can be important. A type 1 error leads to the requirements for additional and unnecessary costs for monitoring and remedial efforts. A Type 2 error has the potential to result in unexpected adverse effects which might lead to future remedial expenditures and increased public concern.

The effects of various sources of uncertainty on the decision process in a tiered or graded system are discussed and illustrated with the help of an example arising from the assessment of potential environmental effects at the decommissioned Stanleigh uranium mine in Elliot Lake Ontario.

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\* Only an abstract is given as the full paper was not available.



# Ultrastructural effects of ionising radiation on primary cultures of rainbow trout skin\*

P.M. Olwell<sup>a</sup>, F.M. Lyng<sup>a</sup>, C.B. Seymour<sup>a</sup>, D.C. Cottell<sup>b</sup>, C. Mothersill<sup>a</sup>

<sup>a</sup> Radiation and Environmental Science Centre, Dublin, Ireland

<sup>b</sup> Electron Microscopy Laboratory, Dublin, Ireland

**Abstract.** There are few reports on the comparative effects of ionising radiation on different species. Published data report the use of radiation levels far in excess of what could be considered in an environmental context and are therefore difficult to assimilate into environmental research. We are engaged in a comparative study of the cellular effects of non-lethal radiation doses on tissue explants from fish and aquatic invertebrates. The aim of the present work is to investigate the ultrastructural effects of ionising radiation on primary cultures of skin from the rainbow trout.

Primary cultures were grown using tissue explants from rainbow trout skin. The cultures were irradiated with doses of cobalt 60 gamma radiation ranging from 0.5Gy to 15Gy. The cultures were fixed in glutaraldehyde, at 7 days post irradiation. They were then post-fixed in osmium tetroxide and dehydrated in ascending grades of alcohol before being embedded in Epon. Blocks of tissue were then sectioned at 1  $\mu\text{m}$ , stained with toluidine blue and surveyed using light microscopy. Ultra-thin sections of particular areas were cut at 50nm and examined by electron microscopy.

Nuclear aberrations and changes in cytoplasmic organelles particularly mitochondria, were observed in irradiated skin cultures. Mitochondria were also of a different shape in irradiated cultures. The occurrence of apoptotic bodies in the irradiated cultures suggests that of controlled cell death is involved in the cellular response to radiation. These results may have important considerations for environmental protection.

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\* Only an abstract is given as the full paper was not available.

# The application of the U.S. Department of Energy's graded approach at the Waste Isolation Pilot Plant

## *A case study*

**R.C. Morris**

TREC, Inc., Rigby, Idaho, United States of America

**Abstract.** For two consecutive years, the U.S. Department of Energy's (DOE) Graded Approach for showing compliance with proposed radiation dose limits for non-human biota was applied at the Waste Isolation Pilot Plant (WIPP) in New Mexico, USA. WIPP is an underground facility for the disposal of low-level transuranic (TRU) waste. Waste was first emplaced in 1998, and there has been no measurable TRU contamination of the environment. DOE's Graded Approach is modeled on the U.S. Environmental Protection Agency's Framework for Ecological Risk Assessment. It includes a screening phase and three-level analysis phase. Each level includes more site specific data and more complex models. Users enter the process at the screening phase and stop when compliance is demonstrated. During the initial screening phase, the highest environmental radionuclide concentrations found at the site of interest are divided by Biota Concentration Guides derived using conservative assumptions from the proposed dose limits. These quotients are combined using a sum-of-fractions methodology to determine compliance. Concentrations of radionuclides in soil, water, and sediment from the environment surrounding WIPP, measured as part of the WIPP Environmental Monitoring Program, were used in the Graded Approach screening methodology and, as expected, they passed the screen both years. The Graded Approach was successfully applied and proved easy to use. However, an important philosophical, rather than technical, problem with the method was illustrated. The method was easy to apply at this site with little thought given to what components of the environment were to be protected from what contaminants. This led to the assessment of doses from non-WIPP related radionuclides. In addition, because nothing in the method requires it, no thought has been given to whether the Environmental Monitoring Program is sampling the correct pathways in the correct locations for species other than humans. This may lead to overconfidence in the future.

### 1. THE WASTE ISOLATION PILOT PLANT (WIPP)

The Waste Isolation Pilot Plant is the world's first underground repository permitted and certified for safe, permanent disposal of transuranic (TRU) radioactive and mixed waste generated by defense-related activities. Its mission is to provide for the safe, permanent, and environmentally-sound disposal of TRU radioactive waste left from research, development, and production of nuclear weapons. Over the next 35 years, WIPP is expected to receive about 37,000 shipments of waste from locations across the United States.

On March 25, 1999, the first waste bound for WIPP departed Los Alamos National Laboratory in New Mexico; it arrived at WIPP the following morning, and the first wastes were placed underground later that day. On April 17, WIPP celebrated its official grand opening. Ten days later, on April 27, the first out-of-state shipment arrived at WIPP, from the Idaho National Engineering and Environmental Laboratory. Later in the year, on October 27, the Secretary of the New Mexico Environment Department issued a WIPP Hazardous Waste Facility Permit, which allows WIPP to manage, store, and dispose of contact-handled TRU mixed waste.

#### 1.1. Location and environment

Located in Eddy County in the remote Chihuahuan Desert of southeastern New Mexico (Figure 1), the WIPP site encompasses approximately 41.1 km<sup>2</sup>. The site is 42 km east of Carlsbad in a region known as Los Medaños. This part of New Mexico is relatively flat and is sparsely inhabited, with little surface water. The WIPP site boundary extends a minimum of 1.6 km beyond any of the WIPP underground developments.

Although sparsely populated by humans, southeastern New Mexico is home to diverse populations of plants and wildlife. Shrubs and grasses are the most prominent components of the local flora. Dominant trees include shinnery oak (*Quercus havardii*), honey mesquite (*Prosopis glandulosa*), and western soapberry (*Sapindus drummondii*). Much of the area is composed of combined dune and

grassland habitats that include perennial grasses and shrubs. The juxtaposition of shinnery oak/dune habitat with grassland habitat has resulted in a diverse wildlife population.

This portion of New Mexico supports an abundant and diverse population of mammals, including black-tailed jackrabbits (*Lepus californicus*), desert cottontails (*Sylvilagus auduboni*), desert mule deer (*Odocoileus hemionus*), coyotes (*Canis latrans*), gray foxes (*Urocyon cinereoargenteus*), badgers (*Taxidea taxus*), and striped skunks (*Mephitis mephitis*).

The habitat heterogeneity of the Los Medaños region also accounts for a wide assortment of bird species. Scaled quail (*Callipepla squamata*), mourning doves (*Zenaida macroura*), loggerhead shrikes (*Lanius ludovicianus*), black-throated sparrows (*Amphispiza bilineata*), Chihuahuan ravens (*Corvus cryptoleucus*), and a unique desert subspecies of the northern bobwhite (*Colinus virginianus*) are examples. In addition, this area supports a particularly abundant and diverse population of raptors. Harris' hawks (*Parabuteo unicinctus*), Swainson's hawks (*Buteo swainsoni*), and great horned owls (*Bubo virginianus*) are species commonly found nesting in the area. Northern harriers (*Circus cyaneus*), burrowing owls (*Athene cunicularia*), barn owls (*Tyto alba*), and American kestrels (*Falco sparverius*) are also found at the site. Due to a scarcity of surface waters in the immediate vicinity of WIPP, migrating or breeding waterfowl are not common.

Reptiles and amphibians are also found in great numbers in southeastern New Mexico. Representative amphibians include the tiger salamander (*Ambystoma tigrinum*), green toad (*Bufo debilis*), plain's spadefoot toad (*Spea bombifrons*), red-spotted toad (*Bufo punctatus*), and New Mexico spadefoot toad (*Spea multiplicata*). Characteristic reptiles in the region include the ornate box turtles (*Terrapene ornata*), side-blotched lizards (*Uta stansburiana*), western whiptails (*Cnemidophorus tigris*), bullsnakes (*Pituophis melanoleucus*), prairie rattlesnakes (*Crotalus viridis*), and Texas horned lizards (*Phrynosoma cornutum*), a federal notice-of-review species listed under the U.S. Endangered Species Act.

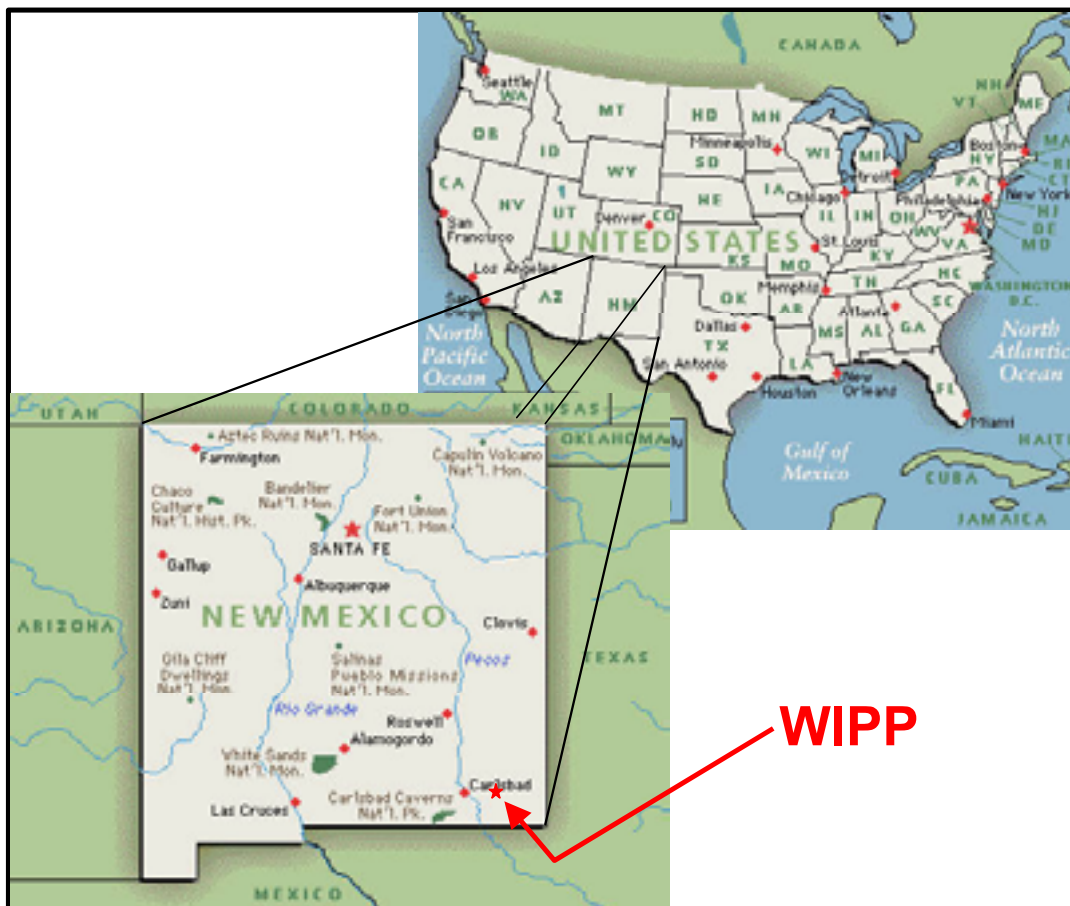


FIG. 1. Location of the Waste Isolation Pilot Plant (WIPP).

TABLE 1. THE WIPP ENVIRONMENTAL MONITORING PLAN [4] OUTLINES THE SAMPLING SCHEDULE FOR THE WIPP ENVIRONMENTAL MONITORING PROGRAM

Type of Sample	Number of Sampling Locations	Sampling Frequency
Liquid effluent	1	Semiannual (oversight)
Liquid effluent	1	Quarterly (DP 831 permit <sup>a</sup> )
Airborne effluent	3	Periodic/Confirmatory
Meteorology	2	Continuous
Atmospheric particulate	7	Weekly
Vegetation	6	Annual
Beef/Deer/Game Birds/Rabbits	3	Annual
Soil	6	Annual
Surface water	14	Annual
Groundwater	7	Semiannual
Fish	3	Annual
Sediment	12	Annual
Aerial photography	Sitewide	Annual
Volatile organic compounds (VOCs)	2	Semiweekly

<sup>a</sup> Monitoring compliance with the WIPP Sewage System Discharge Plan, DP-831.

## 1.2. Environmental sampling [1, 2]

The United States Department of Energy's (DOE) policy is to conduct effluent monitoring and environmental surveillance programs that are appropriate for determining adequate protection of the public and the environment during WIPP operations, and to ensure operations comply with DOE and other applicable federal or state radiation standards and requirements. The WIPP Environmental Monitoring Program monitors pathways by which WIPP-related radionuclides and other contaminants could reach the environment surrounding the WIPP site (Table 1). The pathways measured include air, surface water, groundwater, sediments, soils, and biota (e.g., vegetation, game birds, and fish).

In addition to monitoring for radionuclides contained in WIPP wastes, background radiation (naturally-occurring radioactivity and radioactivity associated with worldwide fallout from historic weapons testing) is monitored. The geographic scope of radiological sampling is based on projections of potential release pathways for the types of radionuclides in WIPP wastes. Also, Carlsbad, NM and local ranches are monitored, even though release scenarios involving radiation doses to residents of these population centers are improbable.

The atmospheric pathway, has been determined to be the most likely exposure pathway to the public from WIPP. Therefore, airborne particulate sampling for alpha-emitting radionuclides is emphasized. Air sampling results are used to trend environmental radiological levels and determine if there has been a deviation from established baseline concentrations.

Based on the results from this monitoring program, there is no evidence that operations at WIPP have contaminated the environment.

## 2. THE GRADED APPROACH

### 2.1. Background

Radiation dose rates below which no deleterious effects on populations of aquatic and terrestrial organisms have been observed have been discussed by the International Atomic Energy Agency [6] and the National Council on Radiation Protection and Measurements [5]. Those dose limits are:

- Aquatic Animals – 10 mGy/d (1 rad/d);
- Terrestrial Plants – 10 mGy/d (1 rad/d);
- Terrestrial Animals – 1 mGy/d (0.1 rad/d).

DOE has considered proposing these dose standards for aquatic and terrestrial biota under proposed rule 10 CFR § 834, “Radiation protection of the public and the environment” but has delayed until guidance for demonstrating compliance was developed. The DOE Interim Technical Standard, *A Graded Approach for Evaluating Radiation Doses to Aquatic and Terrestrial Biota* [3] (Graded Approach), was developed to meet this need. Although the proposed rule has not been implemented, DOE requires reporting of radiation doses to non-human biota in each facility’s Annual Site Environmental Report using the Graded Approach.

## **2.2. Screening**

The Graded Approach uses a multi-phase approach, including an initial screening phase with conservative assumptions. Software is provided with the Graded Approach to conduct the screening evaluation. In the initial screen, Biota Concentration Guides (BCG), defined as the concentration of a single radionuclide in soil, sediment, or surface water which, under the model assumptions, would result in the limiting dose to the organism of interest, are derived using very conservative assumptions for a variety of generic organisms. Maximum concentrations of radionuclides detected in soil, sediment, and water during environmental monitoring are divided by the BCGs and the results are summed for each organism [3]. If the sum of these fractions is less than 1, the site is deemed to have passed the screen and no further action is required. This screening evaluation is intended to provide a very conservative evaluation of whether the site is in compliance with the recommended limits.

## **3. APPLICATION OF THE GRADED APPROACH AT WIPP**

The Graded Approach was used to screen radionuclide concentrations observed around WIPP during 1999 and 2000 using the maximum radionuclide concentrations listed in Table 2. In both years, the sum of fractions was less than one for all media, demonstrating compliance with the proposed rule. Under the conservative assumptions used in the Graded Approach, radiation in the environment surrounding WIPP does not have a deleterious effect on populations of plants and animals. This was the expected result from this analysis because there have been no detectable releases of radioactivity from the WIPP facility [1, 2].

## **4. LESSONS LEARNED/RECOMMENDATIONS**

### **4.1. Philosophical foundation**

The Graded Approach was easy to apply to the environmental monitoring data from the WIPP facility and gave the expected results. However, this ease of application might be the greatest drawback of the approach. Although it did not occur in this case, the temptation inherent in the application was to apply the Graded Approach in a “cookbook” fashion, giving little thought to which populations or other environmental components were intended to be protected from which contaminants. Given the complexity of every ecological system, this lack of thought in the application of the approach might result in an inappropriate level of confidence in the results. While the Graded Approach was designed for ease of application, it was not intended to be applied thoughtlessly. Its documentation contains numerous warnings that ecologists and environmental monitoring specialists should collaborate to ensure the approach is appropriate and meaningful for the intended application.

Another temptation was to apply the Graded Approach with little thought about why it is important and necessary to demonstrate protection of non-human components of the environment. At WIPP, the most common reason given for using the Graded Approach was “it is a required part of the Annual Site Environmental Report.” While this ensures the approach is used, it may also ensure it is used in a *pro forma* fashion. Several international groups are currently considering the ethics and philosophy supporting the need to determine the impacts of ionizing radiation on non-human biota [7, 8], and this discussion should be followed by the environmental monitoring organization at WIPP and elsewhere.

TABLE 2. GENERAL SCREENING RESULTS FOR POTENTIAL RADIATION DOSE TO NON-HUMAN BIOTA FROM RADIONUCLIDE CONCENTRATIONS IN MEDIA NEAR WIPP. MAXIMUM DETECTED CONCENTRATIONS WERE COMPARED WITH BIOTA CONCENTRATION GUIDES<sup>a</sup> (BCG). IF THE SUM OF THE RATIOS BETWEEN MAXIMUM CONCENTRATIONS AND ASSOCIATED BCGs IS BELOW 1.0, NO ADVERSE EFFECTS ON PLANT OR ANIMAL POPULATIONS ARE EXPECTED [3]

Medium	Nuclide	Maximum concentration	BCG	Concentration/BCG
<b>1999</b>				
Aquatic system evaluation				
Sediment (Bq/g)	<sup>90</sup> Sr	$3.40 \times 10^{-2}$	$2.00 \times 10^1$	$1.70 \times 10^{-3}$
	<sup>137</sup> Cs	$1.90 \times 10^{-2}$	$1.00 \times 10^2$	$1.90 \times 10^{-4}$
	<sup>233+234</sup> U	$6.30 \times 10^{-2}$	$2.00 \times 10^2$	$3.15 \times 10^{-4}$
	<sup>235</sup> U	$2.30 \times 10^{-3}$	$1.00 \times 10^2$	$2.30 \times 10^{-5}$
	<sup>238</sup> U	$5.20 \times 10^{-2}$	$9.00 \times 10^1$	$5.78 \times 10^{-4}$
	<sup>241</sup> Am	$2.20 \times 10^{-3}$	$2.00 \times 10^2$	$1.10 \times 10^{-5}$
	Water <sup>b</sup> (Bq/l)	<sup>90</sup> Sr	ND	$1.00 \times 10^1$
<sup>137</sup> Cs		ND	$2.00 \times 10^0$	$0.00 \times 10^0$
<sup>233+234</sup> U		$1.60 \times 10^{-1}$	$7.00 \times 10^0$	$2.29 \times 10^{-2}$
<sup>235</sup> U		$3.00 \times 10^{-3}$	$8.00 \times 10^0$	$3.75 \times 10^{-4}$
<sup>238</sup> U		$8.50 \times 10^{-2}$	$8.00 \times 10^0$	$1.06 \times 10^{-2}$
<sup>241</sup> Am		ND	$2.00 \times 10^1$	$0.00 \times 10^0$
			Sum of Fractions	$3.67 \times 10^{-2}$
<b>1999</b>				
Terrestrial system evaluation				
Soil (Bq/g)	<sup>137</sup> Cs	$1.20 \times 10^{-2}$	$8.00 \times 10^{-1}$	$1.50 \times 10^{-2}$
	<sup>233+234</sup> U	$1.60 \times 10^{-2}$	$2.00 \times 10^2$	$8.00 \times 10^{-5}$
	<sup>235</sup> U	$4.40 \times 10^{-3}$	$1.00 \times 10^2$	$4.40 \times 10^{-5}$
	<sup>238</sup> U	$1.60 \times 10^{-2}$	$6.00 \times 10^4$	$2.67 \times 10^{-7}$
	<sup>239+240</sup> Pu	$1.40 \times 10^{-3}$	$2.00 \times 10^2$	$7.00 \times 10^{-6}$
Water (Bq/l)	<sup>137</sup> Cs	ND	$2.00 \times 10^4$	$0.00 \times 10^0$
	<sup>233+234</sup> U <sup>3</sup>	$1.60 \times 10^{-1}$	$1.00 \times 10^4$	$1.60 \times 10^{-5}$
	<sup>235</sup> U	$3.00 \times 10^{-3}$	$2.00 \times 10^4$	$1.50 \times 10^{-7}$
	<sup>238</sup> U	$8.50 \times 10^{-2}$	$2.00 \times 10^4$	$4.25 \times 10^{-6}$
	<sup>239+240</sup> Pu	ND	$7.00 \times 10^4$	$0.00 \times 10^0$
			Sum of Fractions	$1.52 \times 10^{-2}$
<b>2000</b>				
Aquatic system evaluation				
Sediment (Bq/g)	<sup>60</sup> Co	$7.74 \times 10^{-3}$	$5.00 \times 10^1$	$1.55 \times 10^{-4}$
	<sup>137</sup> Cs	$1.86 \times 10^{-2}$	$1.00 \times 10^2$	$1.86 \times 10^{-4}$
	<sup>234</sup> U	$4.75 \times 10^{-2}$	$2.00 \times 10^2$	$2.38 \times 10^{-4}$
	<sup>235</sup> U	$5.68 \times 10^{-3}$	$1.00 \times 10^2$	$5.68 \times 10^{-5}$
	<sup>238</sup> U	$3.38 \times 10^{-2}$	$9.00 \times 10^1$	$3.76 \times 10^{-4}$
	<sup>241</sup> Am	$3.70 \times 10^{-4}$	$2.00 \times 10^2$	$1.85 \times 10^{-6}$
	Water <sup>b</sup> (Bq/l)	<sup>60</sup> Co	$6.75 \times 10^{-1}$	$1.00 \times 10^2$
<sup>137</sup> Cs		$8.94 \times 10^{-1}$	$2.00 \times 10^0$	$4.47 \times 10^{-1}$
<sup>234</sup> U		$1.90 \times 10^{-1}$	$7.00 \times 10^0$	$2.71 \times 10^{-2}$
<sup>235</sup> U		$6.09 \times 10^{-3}$	$8.00 \times 10^0$	$7.62 \times 10^{-4}$
<sup>238</sup> U		$8.72 \times 10^{-2}$	$8.00 \times 10^0$	$1.09 \times 10^{-2}$
<sup>241</sup> Am		$4.92 \times 10^{-4}$	$2.00 \times 10^1$	$2.46 \times 10^{-5}$
			Sum of Fractions	$4.94 \times 10^{-1}$
<b>2000</b>				
Terrestrial system evaluation				
Soil (Bq/g)	<sup>60</sup> Co	$5.55 \times 10^{-3}$	$3.00 \times 10^1$	$1.85 \times 10^{-4}$
	<sup>137</sup> Cs	$1.37 \times 10^{-2}$	$8.00 \times 10^{-1}$	$1.72 \times 10^{-2}$
	<sup>234</sup> U	$2.01 \times 10^{-2}$	$2.00 \times 10^2$	$1.00 \times 10^{-4}$
	<sup>235</sup> U	$1.77 \times 10^{-3}$	$1.00 \times 10^2$	$1.77 \times 10^{-5}$
	<sup>238</sup> U	$2.35 \times 10^{-2}$	$6.00 \times 10^1$	$3.92 \times 10^{-4}$
	<sup>241</sup> Am	$3.70 \times 10^{-4}$	$1.00 \times 10^2$	$3.70 \times 10^{-6}$
	Water (Bq/l)	<sup>60</sup> Co	$6.75 \times 10^{-1}$	$4.00 \times 10^4$
<sup>137</sup> Cs		$8.94 \times 10^{-1}$	$2.00 \times 10^4$	$4.47 \times 10^{-5}$
<sup>234</sup> U		$1.90 \times 10^{-1}$	$1.00 \times 10^4$	$1.90 \times 10^{-5}$
<sup>235</sup> U		$6.09 \times 10^{-3}$	$2.00 \times 10^4$	$3.05 \times 10^{-7}$
<sup>238</sup> U		$8.72 \times 10^{-2}$	$2.00 \times 10^4$	$4.36 \times 10^{-6}$
<sup>241</sup> Am		$4.92 \times 10^{-4}$	$7.00 \times 10^3$	$7.03 \times 10^{-8}$
			Sum of Fractions	$1.80 \times 10^{-2}$

<sup>a</sup> The radionuclide concentration in the medium that would produce a radiation dose in the organism equal to the dose limit under the conservative assumptions in the model.

<sup>b</sup> Sediment and water samples were assumed to be co-located.

## 4.2. Sampling the correct pathways

The Graded Approach is intended to be cost-effective. With that in mind, users are encouraged to make use of available data rather than to generate new data specifically for this application. An important assumption of this advice, is that current monitoring programs sample radionuclide transport pathways appropriate for non-human biota. Because plants and animals frequently access pathways not normally available to humans, this requires users to develop a conceptual model of their environment, explicitly considering potential pathways to non-human biota. This has not been done at most DOE facilities, and WIPP is no exception. This is not likely a major issue at WIPP to date because of the extremely low probability of a release. In addition, the nature of the facility limits the number of potential release pathways and those pathways are routinely sampled. However, proper application of the Graded Approach cannot be assured until an appropriate conceptual model is developed and used to confirm that the correct pathways are being sampled.

## 5. CONCLUSIONS

The Graded Approach was easy to apply at WIPP and yielded the expected results. However, greater confidence in the results would result from the development of an appropriate philosophical and ethical foundation for using the approach, and the development of a conceptual model explicitly incorporating radionuclide transport pathways for non-human biota.

## ACKNOWLEDGEMENTS

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# Application of biota dose assessment committee methodology to assess radiological risk to salmonids in the Hanford reach of the Columbia River

T.M. Poston, E.J. Antonio, R.E. Peterson

Pacific Northwest National Laboratory, Richland, WA, United States of America

**Abstract.** Protective guidance for the protection of biota in the U.S. Department of Energy's Graded Approach for Evaluating Radiation Doses to Aquatic and Terrestrial Biota is based on population level protection guides of 10 or 1 mGy d<sup>-1</sup>, respectively. Several "ecologically significant units" of Pacific salmon are listed under the ESA. The Hanford Reach supports one of the largest spawning populations of fall chinook salmon in the Northwest. Listed units of steelhead are located upstream of Hanford and in an adjacent river basin. The existence of the major spawning areas in the Hanford Reach has focused considerable attention on their ecological health by the Department of Energy, state and other federal regulatory agencies, and special interest groups. Dose assessments for developing salmonid embryos were performed for the hypothetical exposure to tritium, strontium-90, technetium-99, iodine-129, and uranium isotopes at specific sites on the Hanford Reach. These early life stages are potentially exposed in some areas of the Hanford Reach to radiological contaminants that enter the river via shoreline seeps and upwelling through the river substrate. At the Tier 1 screening level, no site approached the dose guideline of 10 mGy d<sup>-1</sup> established with the RAD-BCG calculator. Special status of listed species affords these populations more consideration when assess potential impacts of exposure to radionuclides and other contaminants associated with Hanford Site operations. The evolution of dose benchmarks for aquatic organisms and consideration of precautionary principal and cumulative impacts are addressed.

## 1. INTRODUCTION

Since 1989, a massive clean up and restoration mission has been underway at the Hanford Site in south-central Washington State [1]. This mission follows 45 years of weapons material production that has left the site with numerous areas undergoing or scheduled for cleanup. Many of these sites are sources of contamination that reach the Columbia River as groundwater seepage. Most plumes that impact the river originate in the 100 Areas (plutonium production reactors), the 200 Areas (plutonium recovery and waste management), or the original fuel processing area (300 Area, see Figure 1). None of the DOE reactor areas are operational and all are in some stage of cleanup and decommissioning. This seepage is viewed as one of the more serious environmental issues at the Hanford Site. The Hanford Reach of the Columbia River supports the last major area of spawning habitat for anadromous fall chinook salmon (*Oncorhynchus tshawytscha*) in this large river [2]. The majority of the Columbia River that supported salmon and steelhead (*O. mykiss*) spawning in the past is now either inundated with impoundments behind dams or has been eliminated as spawning habitat by dams without fish passage facilities.

Pacific Coast salmonids have been organized into "ecologically significant units" (ESU) based on the river basins where they spawn. Many ESUs of Pacific coast salmon are listed by the federal government as either threatened or endangered [3]. Salmonid spawning in the Hanford Reach is viewed as a critical resource by the federal and state government, Native peoples, stakeholders, and activist groups. Steelhead spawn in the Hanford Reach, but this section of the Columbia River falls outside of areas designated as habitat for listed ESUs. The spawning population of Middle Columbia River fall chinook salmon are not listed, but represent the largest spawning population of fall chinook salmon in the Pacific Northwest. Other ESUs that spawn upstream of the Hanford Reach are also listed. Consultation between federal agencies is mandated when potential actions of an agency may impact a listed species.



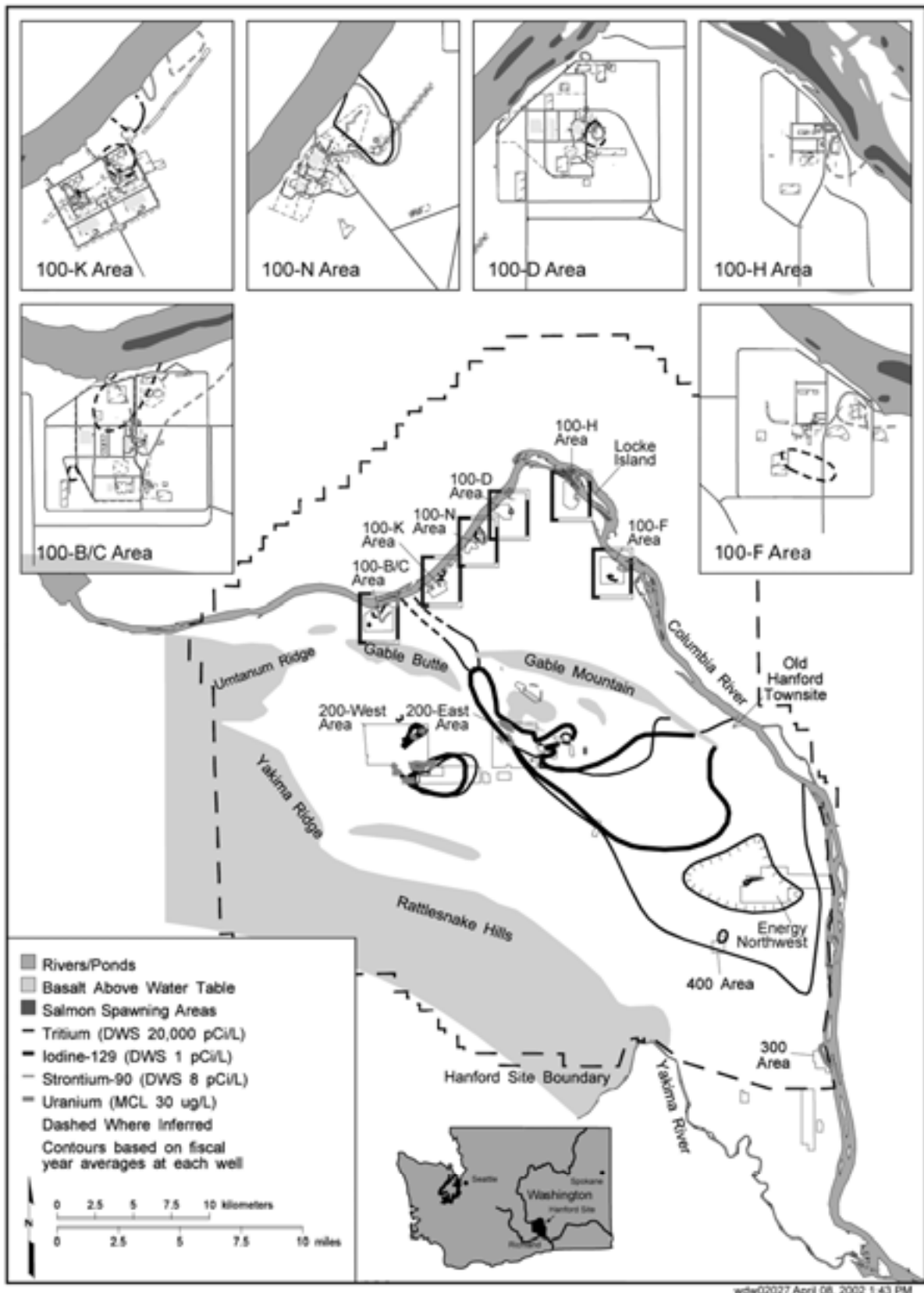


FIG. 1. Contaminant plumes on the Hanford Site.

Exposure of salmonids to contaminants entering the Columbia River has been a point of interest by all those concerned with the well being of this resource. Risk to salmonids is based on their life cycle. Although adults are potentially exposed to Hanford effluents when they return to spawn in the Hanford Reach, the developing embryos and sac-fry are the most susceptible life-stage. These early life stages are potentially exposed in some areas of the Reach to radiological contaminants that enter the river via shoreline seeps and upwelling through the river substrate. Eggs are laid in nests called “redds”. Redds are depressions in the river cobble constructed by the breeding salmonid in areas with high inter-cobble flow of water [4]. Once the developing fry adsorb their yolk sac and emerge from the cobble, the dilution provided by the Columbia River reduces exposure levels to essentially background levels. Within weeks, the emergent fish migrate downstream to enter the Pacific Ocean. The cycle begins a new in two to five years (depending on the species) when the adults return to spawn.

The most critical life stage of salmon is the egg-larval life stage that occurs in spawning redds constructed by salmon in the river cobble. This report evaluates the radiological risk to salmon embryos in the Hanford reach by utilizing a graded dose assessment process developed by the U.S. Department of Energy [5], hereafter referred to as the DOE Technical Standard. The methodology is used to evaluate dose and additional site specific assessments (i.e., graded approach) were performed to more accurately evaluate the dose to developing salmon embryos. Other factors affecting the estimated dose to salmon are evaluated. These include life cycle considerations, physiological responses of developing salmon, and spawning behavior. Last, the paper evaluates the basis for screening doses and addresses the application of the methodology to regulated species in the context of current environmental policy and appropriate dose limits.

## 2. METHODS

**Selection of Contaminant Sources.** There are numerous contaminant plumes associated with industrialized facilities found on the Hanford Site [1]. None of the DOE production reactor areas are operational and all are in some stage of cleanup and decommissioning. There are known plumes of radiological contaminants, however, only those located at 100-H, 100-D and 100-F Area are likely to result in the exposure of developing salmon embryos (Figure 1).

The river shoreline adjacent to the Old Hanford Townsite has been the location of contaminant seepage that originates from the fuel processing facilities located in the 200 Areas. Salmon spawning has been documented upstream and downstream of this area [2], so it was included in the analysis. Salmon spawning has not been observed near the 300 Area and it was not considered further in this analysis. The most contaminated plumes are associated with the 100-N Area, however, the river bottom adjacent to this area does not support spawning salmon. Furthermore, spawning areas located near the 100-D and 100-F Areas are outside the likely influence of contaminant plumes entering the river (Figure 1). The most likely area of developing salmon embryo exposure is between Locke Island and the 100-H Area shoreline.

## 3. DOSE ASSESSMENT

This assessment involves the application of the DOE Technical Standard for a two-tiered screening level assessment [5] and an embryo-specific analysis to estimate the dose to the developing salmon embryo and larval life stage. Source terms for the exposure were the maximum undiluted seep water concentrations (Tier I) associated with areas of known contaminant entry from the Hanford Site. Tier 2 use median concentrations with site-specific  $B_{iv}$  values. Seep water data used covered the period of 1995 through 2000 [1]. Locations included six inactive reactor sites along the Columbia River and the Old Hanford Townsite shoreline area due east of the central Hanford plateau. Internal concentrations of radionuclides in salmonid embryos were based on generic bioaccumulation factors ( $B_{iv}$ ) and sediment concentrations were estimated by distribution coefficients ( $K_d$ ) in Tiers 1 and 2 (Table 1). Results of the DOE Technical Standard are tabulated as a “sum of fractions” and are based on biota concentrations guidelines found in the RAD-BCG calculator. The sum of fractions is based on the dose guideline of  $10 \text{ mGy}\cdot\text{day}^{-1}$  where each fraction is the ratio of the concentration in water or sediment compared to the level that would result in the  $10 \text{ mGy}\cdot\text{d}^{-1}$  dose guideline for aquatic organisms. Summing the fractions accounts for all sources of radionuclide exposure. A sum greater than 10.0 indicates that the guideline has been exceeded. A second “precautionary” benchmark dose rate of  $2.5 \text{ mGy}\cdot\text{d}^{-1}$  was also used as a basis of comparison [6].

TABLE 1. RADIONUCLIDES AND DEFAULT PARAMETERS USED IN SCREENING ASSESSMENTS (RAD-BCG) CALCULATIONS AND EMBRYO-SPECIFIC DOSE ASSESSMENTS TO DEVELOPING SALMONIDS

Radionuclide	Areaa	RAD-BCG (Tiers 1 and 2)		Embryo-Specific Analysis	
		Aquatic Organism		(Salmon Eggs/Larvae)	
		$B_{iv}$	$K_d$	$B_{iv}$	$K_d$
$^3\text{H}$	B/C, K, N, D, H, F, OHT	0.2	0.001	1.0	NA
$^{90}\text{Sr}$ - $^{90}\text{Y}$	B/C, K, N, D, H, F	320	30	5.7b	NA
$^{99}\text{Tc}$	B/C, K, H, OHT	78	5	15	NA
$^{129}\text{I}$	OHT	220	10	50	NA
$^{234}\text{U}$	H, OHT	1000	50	10	NA
$^{235}\text{U}$	H, OHT	1000	50	10	NA
$^{238}\text{U}$	H, OHT	1000	50	10	NA

<sup>a</sup> Reactor sites (100 Areas) and Old Hanford Townsite (OHT), see Figure 1.

<sup>b</sup> Vanderploeg et al. [9].

To complement the DOE Technical Standard, embryo-specific dose assessments for salmonid eggs were performed using concentration factors specific for the Columbia River and some elements of the CRITR II code [7] within the Technical Standard platform. With the exception of  $^3\text{H}$ , the site-specific concentration factors were significantly lower than those used in the screening RAD-BCG calculator (Table 1). The “effective energy adsorbed” terms were based on a hypothetical sphere of 1.4 cm. Steelhead and salmon eggs range in diameter from 0.5 to 0.95 cm; hence, use of the greater diameter was a conservative factor. A hypothetical dilution factor of 0.22 was applied to the maximum seep water concentration used in each assessment. This factor was derived from conductivity measurements taken from pore water concentrations collected in the cobble compared to seep water measurements at the 100-H shoreline [8]. Last, because mid-Columbia River salmonids spawn in cobble, the sediment component of the source term was dismissed. There is no accumulation of fine sediment particles other than coarse sand dispersed among the cobble. This sand is likely dislodged and carried downstream during redd construction. External sources of beta and alpha radiation would not expose the developing embryo or penetrate to internal organs of the larval salmon.

#### 4. RESULTS AND DISCUSSION

Tier 1 dose estimates from the RAD-BCG screening methodology ranged from  $2.2\text{E-}05$   $\text{mGy}\cdot\text{d}^{-1}$  at 100-F Area to  $1.9\text{E}00$   $\text{mGy}\cdot\text{d}^{-1}$  at 100-N Area (Table 2). The dose estimate accounts for internal exposure to adsorbed radionuclides, hypothetical immersion in water (both captured by the “Water Partial Fraction” column), and sediment exposure. Estimated doses at all locations were below the sum of fractions screen that corresponds to  $10$   $\text{mGy}\cdot\text{d}^{-1}$  for aquatic organisms. The maximum dose at 100-N was a factor of 5 below the sum of fraction’s trigger value of 1.0 (corresponds to  $10$   $\text{mGy}\cdot\text{d}^{-1}$  for aquatic organisms). In this RAD-BCG analysis (Tier 1 and 2), the aquatic target organism is not defined.

Strontium-90 was the major contributor to dose at 100-N, 100-B/C, 100-K, 100-F, and 100-D Areas. At 100-H, and the Old Hanford Townsite, RAD-BCG dose estimates were dominated by uranium isotopes. Uranium was included in the assessments at 100-H and the Old Hanford Townsite because of its known presence in seeps from Hanford operations, however, background uranium was not measured at the remaining locations. Tritium,  $^{99}\text{Tc}$ , and  $^{129}\text{I}$  were also measure in some locations, and in some cases, contributed as much as 15% to the estimated Tier 1 dose rate. The percentage of the estimated dose attributable to immersion in water was very low; immersion dose conversion factors ranged from  $3.8\text{E-}14$  to  $7.7\text{E-}12$   $\text{mGy}\cdot\text{d}^{-1}$  per  $\text{Bq}\cdot\text{m}^3$  [5]. The percentage dose attributed to exposure to sediment ranged from 1 to 3 orders of magnitude less than the water pathway (Table 2). The majority of the estimated dose was due to internal dosimetry based on the generic bioaccumulation factors ( $B_{iv}$ ) used in the RAD-BCG analysis.

TABLE 2. DOSE ESTIMATES OF DOE “GRADED APPROACH” [5] FROM UNDILUTED MAXIMUM SEEP WATER CONCENTRATIONS OF RADIONUCLIDES (TIER 1 SUM OF FRACTIONS REFERENCED TO 10 mGy·d<sup>-1</sup>)

Radionuclide	Water Partial Fraction	Sediment Partial Fraction	Dose <sup>a</sup>	Percentage Dose
100-B/C Seep Water [Total Dose = 1.04E-03 mGy·d <sup>-1</sup> ]				
<sup>3</sup> H	4.38E-05	2.92E-08	4.39E-05	4.2%
<sup>90</sup> Sr- <sup>90</sup> Y	8.38E-04	5.03E-05	8.88E-04	85.5%
<sup>99</sup> Tc	1.04E-04	2.34E-06	1.06E-04	10.2%
100-K Seep Water [Total Dose = 4.5E-04 mGy·d <sup>-1</sup> ]				
<sup>3</sup> H	3.64E-05	2.43E-08	3.65E-05	8.0%
<sup>90</sup> Sr- <sup>90</sup> Y	3.92E-04	2.35E-05	4.16E-04	91.7%
<sup>99</sup> Tc	1.12E-06	2.52E-08	1.14E-06	0.3%
100-N Seep Water [Total Dose = 1.9E-00 mGy·d <sup>-1</sup> ]				
<sup>3</sup> H	4.51E-05	3.01E-08	4.52E-05	<0.01%
<sup>90</sup> Sr- <sup>90</sup> Y	1.83E+00	1.10E-01	1.94E+00	100.00%
100-D Seep Water [Total Dose = 1.1E-03 mGy·d <sup>-1</sup> ]				
<sup>3</sup> H	1.09E-05	7.24E-09	1.09E-05	1.0%
<sup>90</sup> Sr- <sup>90</sup> Y	9.77E-04	5.86E-05	1.04E-03	99.0%
100-H Seep Water [Total Dose = 4.7E-01 mGy·d <sup>-1</sup> ]				
<sup>3</sup> H	4.22E-06	2.81E-09	4.22E-06	<0.01%
<sup>90</sup> Sr- <sup>90</sup> Y	3.22E-03	1.93E-04	3.41E-03	0.73%
<sup>99</sup> Tc	5.59E-04	1.26E-05	5.72E-04	0.12%
<sup>234,235,238</sup> U	4.61E-01	3.88E-05	4.61E-01	99.1%
100-F Seep Water [Total Dose = 2.2E-05 mGy·d <sup>-1</sup> ]				
<sup>3</sup> H	3.27E-06	2.18E-09	3.28E-06	15.1%
<sup>90</sup> Sr- <sup>90</sup> Y	1.74E-05	1.04E-06	1.84E-05	84.9%
Old Hanford Townsite Seep Water [Total Dose = 4.3E-01 mGy·d <sup>-1</sup> ]				
<sup>3</sup> H	2.22E-04	1.48E-07	2.22E-04	0.05%
<sup>129</sup> I	4.93E-04	1.11E-05	5.04E-04	<0.01%
<sup>99</sup> Tc	3.76E-06	7.51E-08	3.83E-06	0.12%
<sup>234,235,238</sup> U	4.32E-01	3.32E-05	4.32E-01	99.8%

<sup>a</sup> Sum of water and sediment dose rates.

Salmonid spawning has not been observed near the 100-N shoreline. At the 100-B/C, 100-K, 100-D, and 100-F Areas, some spawning has been observed in the vicinity of these seeps, however, most of observed spawning areas are located across the river channel and are not directly impacted by the discharges or potential near-shore upwelling of dilute groundwater [2].

All study areas were taken to a Tier 2 analysis with the RAD-BCG and an embryo-specific dose assessment salmonid exposure. Generally, the embryo-specific dose estimates for salmonid embryos were >1 to 4 orders of magnitude lower than the Tier 1 RAD-BCG dose estimates (Figure 2). The Tier 2 dose rate estimates were intermediate between the Tier 1 and embryo-specific dose estimates. All were well below the DOE 10-mGy·d<sup>-1</sup> guideline and the NCRP precautionary guideline of 2.5 mGy·d<sup>-1</sup>. The embryo-specific 100-N Area dose estimate was 1.6E-05 mGy·d<sup>-1</sup>, but was less than the 100-H and Old Hanford Townsite estimates because they included uranium.

There are several significant environmental variables that effected dose estimates for salmonid embryos. As contaminated ground water approaches the shoreline, mixing with riverbank recharge occurs along the shoreline and with interstitial flow in the cobble substrate in the river channel. The hydrology of this phenomenon is very complex and variable depending on river stage, hydraulic head, and variation in the underlying substrata [10]. These factors dilute the groundwater and reduce the concentration of contaminants that reach the eggs. For this assessment, flow through the hypothetical redds was reduced to 22% seep water with a corresponding reduction in contaminant concentration for all sites. Actual dilution may be considerably greater.

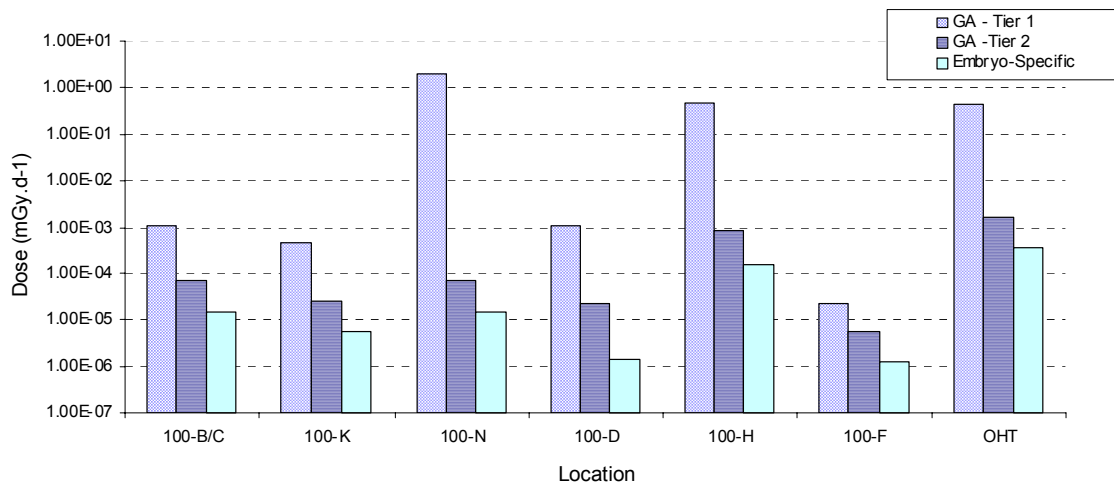


FIG 2. Comparison of DOE RAD-BCG Methodology (graded approach – GA) for Aquatic Organisms and Embryo-Specific Dose Assessment of Salmonid Embryos

Bioaccumulation factors ( $B_{iv}$ ) were also adjusted from the default values to values more reflective of the Columbia River. The concentration factor for  $^{90}\text{Sr}$  was based on Ca concentration [9]. The concentration factor for uranium isotopes was reduced based on observations of uranium accumulation in liver and bone of sculpin (*Cottus* sp.), a small bottom dwelling fish found in the Hanford Reach. The  $B_{iv}$  were also adjusted for  $^3\text{H}$ ,  $^{99}\text{Tc}$ , or  $^{129}\text{I}$  (see Table 1). The  $B_{iv}$  had the greatest influence on the reduction of dose estimates between Tier 1 and the embryo-specific dose estimates.

There is some uncertainty as to whether the developing embryo would accumulate radionuclides to levels found in free swimming fish. The chorion (egg shell) has micro-pores (visible with a low power microscope) that can exchange electrolytes and water, however, uptake rates may be attributed to simple diffusion. There is no metabolic requirement for uranium or strontium, the major contributors to dose, however,  $^{90}\text{Sr}$  is expected to accumulate in inverse proportion to the Ca content of the river. Calcium is critical for bone development, however, ossification does not occur until the developing embryo has hatched. There is no established dietary requirement of calcium in fish; i.e., fish can absorb all dietary requirements of calcium from the water column. Hence, there is no mechanism for accumulation of Ca for bone production until after the fish hatches. As such, adsorption of  $^{90}\text{Sr}$  by developing embryos is not expected to be significant until ossification occurs. This may reduce the risk associated with  $^{90}\text{Sr}$ - $^{90}\text{Y}$  exposure as the embryo is developing and through early pre-swim up life stages in river cobble.

The embryo develops on the outside of the yolk in direct contact with the chorion. The estimated thickness of the salmonid chorion is  $\sim 0.050$  mm [11]. Hence, a particle of ionizing radiation that travels further than 0.050 mm could intercept the developing embryo. The chance that an embryo would be exposed increases as the embryo grows.

Generally, external alpha and beta particles are not expected to penetrate the chorion of the salmonid egg. The expected travel distance for a 6 MeV alpha is 0.060 mm through water. Assuming similar rates of penetration in the chorion as water, it is unlikely that an alpha particle emanating from an atom of uranium ( $<4.8$  MeV) would reach the embryo. The same logic applies to the beta emission of  $^3\text{H}$  with a penetration distance of 0.025 mm. Penetration distances based on the maximum beta energy for  $^{90}\text{Sr}$ ,  $^{90}\text{Y}$ ,  $^{99}\text{Tc}$  and  $^{129}\text{I}$  are 1.78, 16.5, 0.78 and 0.30 mm in water [12]. Consequently, an atom of any of these beta emitters that decays within the penetration distance could irradiate the developing embryo. The likelihood of a randomly directed beta particle interacting with an embryo of 0.1mm diameter is on the order of  $<0.0001$  and can be considered insignificant compared to internal dose sources. This simplistic example supports the guidance provided in IAEA [13] for dismissing the contribution of external beta and alpha emitters to dose estimates of aquatic organisms.

External exposure resulting from immersion in water is another source that was dismissed with alpha and beta emitters in the site-specific analysis. Thermo-luminescent dosimeters (TLD) have been used to determine immersion dose rates in the Columbia River. When TLDs were last used for surveillance in 1992, TLD immersion dose rates were  $1.7E-03 \text{ mGy}\cdot\text{d}^{-1}$  [14]. TLDs would only detect gamma sources in the river. The location was slightly downstream of the inactive 100 B/C Area. Seepage from the in-active reactor area may have influenced TLD results. The TLD results are probably representative of background external dose rates in the water that salmonid eggs would experience. However, dose rates in the cobble could be slightly higher due to naturally occurring radionuclides in the rocks.

Another important variable is the composition of the salmonid redds. Salmon nests are constructed in medium to large cobble in areas with high interstitial water flow. The act of nest construction likely dislodges most sand that is then carried away from the nest by the currents. The eggs, once deposited in the nest, are covered by cobble to protect them from predators after the parents die. There is no significant vector for sediment exposure to the salmon embryos. From this standpoint, the site-specific analysis did not incorporate a sediment exposure vector. Application of sediment dose conversion factors (see [5]) for the embryo-specific evaluation would have overestimated the dose to salmonid embryos, however, the contribution is not large because the grade approach accounts for attenuation of beta and alpha emissions in water.

## 5. REGULATORY FRAMEWORK OF DOSE ASSESSMENT TO LISTED SPECIES

In the United States, environmental assessments performed under applicable federal and state regulations need to address the potential for cumulative impacts. Cumulative impacts were originally defined by the Council of Environmental Quality (CEQ) to simply address the interaction of impacts associated with one activity on another activity. The intent of the CEQ was to assure that an impact of low or questionable significance needed to be evaluated if the impact was exacerbated by other activities. Though simplistic in its original narrative form, the application of this principle has grown in terms of addressing time, space and synergistic effects. The Council of Environmental Quality [15] has identified four types of cumulative effects based on number of actions and how they interact.

These 4 types of cumulative effects demonstrate increasing complexity of impacts and interactions:

- Type 1. Single effect – repeated additive effects.
- Type 2. Interactions between stressors and receptors that are non-linear (sum is greater than the parts).
- Type 3. Multiple actions – additive effects (this is the classical type of cumulative effect originally and simplistically defined by the CEQ).
- Type 4. Interactive effects from multiple stressors, there are multiple stressors operating with multiple effects that are interactive.

Quantifying cumulative impacts of increasing complexity is difficult and carries a high degree of uncertainty, particularly when assessed against high natural variation and seasonal or annual fluctuations in habitat characteristics. In these situations where exposure to contaminants and estimates of risk are inherently uncertain, it is both realistic and desirable to invoke additional conservatism in the assessment process (i.e., precautionary principle). Precautionary principle has been applied in most dose assessments by incorporating conservative variables where uncertainty is high in model parameters; some times to the point of creating very unrealistic exposure conditions or overly conservative guidelines. The recommendation by the NRC [6] to re-evaluate dose assessments when estimated doses reach  $2.5 \text{ mGy}\cdot\text{d}^{-1}$  is an application of precautionary principle.

The RAD-BCG guidelines are couched with the understanding that there may be some level of impact to members of the exposed population, but that the amount of impact will not compromise the survival or overall well being of that population. There is no allowance for other stressors on the population sans an acknowledgement that special consideration may be given to listed species and that other stresses may also be significant. Under Section 7 of the Threatened and Endangered Species Act, any action that is judged to impact a listed species is identified as “take” under Section 4(d) must be reviewed and approved by the appropriate regulating agency. In administration of this law, the focus is

directed towards the protection of individuals within the population, rather than the population as a managed unit. As such, it follows that additional consideration should be given to the application of the DOE Technical Standard (DOE 2001) when a threatened population of organisms is being evaluated. Clearly, the concept of precautionary principal is appropriate to this situation when other stressors may be present. At the Hanford Site, DOE has implemented a Salmon and Steelhead Management Plan to address these issues [17].

Scientific progress for understanding the mechanisms of radio-toxicity and ecologically relevant endpoints, as well as environmental fate and exposure of contaminants, have reduced some of the uncertainty associated with environmental dose assessments. Still, a need exists to establish a lower guideline for those species that require additional protection. This need has been recognized since the inception of the  $10 \text{ mGy}\cdot\text{d}^{-1}$  “guideline” by the IAEA in 1976 [13].

## 6. EVOLUTION OF DOSE LIMIT FOR AQUATIC ORGANISMS

The dose rate of  $10 \text{ mGy}\cdot\text{d}^{-1}$  was initially reported by the International Atomic Energy Agency [13]. The guidance was based on the fact that the most radio-sensitive aquatic organisms were teleost (i.e., bony) fish, and that the most sensitive life stage was the developing eggs and larvae. IAEA notes that some mortality was observed at acute doses of 1 Gy, and that there were some minor physiological or metabolic effects associated with chronic dose rates of  $10 \text{ mGy}\cdot\text{d}^{-1}$ . IAEA cautioned that most of the studies involved acute high dose exposures and that data for low dose chronic exposures were lacking. Their evaluation involved the review of 384 scientific studies dating from 1958 to 1974.

A parallel “guiding principal of environmental dosimetry” also surfaced around 1977 declaring that standards and limits that are protective of humans in the environment are also protective of other organisms present in the environment. To that end, IAEA [18] initiated a review in 1986 to pursue this question. The 1976 IAEA assessment [13] was a baseline for that study as well as a review of subsequent research on radiological dose effects of aquatic organisms. Their results, published in 1992, agreed that a dose rate for the aquatic “maximally exposed individual” organism of  $<10 \text{ mGy}\cdot\text{d}^{-1}$  would be protective at the population level for aquatic organisms.

In a third and somewhat related activity, the U.S. Department of Energy requested that the National Council on Radiation Protection and Measurements convened a committee (64-9 Scientific Committee on Effects of Radiation on Aquatic Organisms) to review the effects of ionizing radiation on aquatic organisms [6]. Over half of the NCRP committee members also participated in the 1976 review by IAEA [13]. The committee reiterated the emphasis for protection of aquatic organisms was focused on populations rather than individuals. In general terms, dose rates of  $0.4$  to  $1 \text{ mGy}\cdot\text{h}^{-1}$  ( $9.6$  to  $24 \text{ mGy}\cdot\text{d}^{-1}$ ) were viewed as the lower range that could cause adverse effects in the “most sensitive” aquatic organisms. Points particularly germane to the exposure of salmonid embryos include:

- consistently damaging effects appeared at  $4 \text{ mGy}\cdot\text{h}^{-1}$  [ $96 \text{ mGy}\cdot\text{d}^{-1}$ ] for developing chinook salmon eggs;
- highest environmental dose rate with no observed effects was  $0.02 \text{ mGy}\cdot\text{h}^{-1}$  – [ $0.48 \text{ mGy}\cdot\text{d}^{-1}$ ];
- Lowest dose rate at which some adverse effects have been observed was  $0.4 \text{ mGy}\cdot\text{h}^{-1}$  ( $9.6 \text{ mGy}\cdot\text{d}^{-1}$ ).

The NCRP committee concluded that the application of the  $10 \text{ mGy}\cdot\text{d}^{-1}$  guideline to situations for a point source release of contaminants would be protective of “presumed healthy” endemic populations. The committee acknowledged that the standard may not be appropriate for special situations such as endangered species or species with low fecundity, a consideration also identified in the DOE Technical Standard [5]. However, recommendations for acceptable dose levels where exacerbating conditions prevail as is the case for listed species were not made and would have to be evaluated on a case-by-case basis. As a result of this review, DOE adopted as guidance (under DOE Order 5400.5) an administrative dose rate limit of  $10 \text{ mGy}\cdot\text{d}^{-1}$  for aquatic organisms. NCRP had also recommended that when doses approach  $0.1 \text{ mGy}\cdot\text{hr}^{-1}$  [ $2.5 \text{ mGy}\cdot\text{d}^{-1}$ ], that additional study and review is warranted. There is no agreed upon standard for developing salmon embryos or consideration for special situations.

This study on salmonid embryos addressed the most sensitive life stage of resident salmonids. The study has demonstrated that doses are well below any plausible dose level that could have an adverse

impact on developing salmon embryos. However, the assessment does not address potential cumulative impacts of other stressors. Other contaminants also are released to the Columbia River via seeps including chromium and nitrate. Uranium, was assessed as a radiological contaminant at two locations, yet it more likely acts as a chemical toxicant due to its low specific activity. The Columbia River is classified as a Class A stream of very high water quality, yet it is the recipient of agricultural and mining effluents upstream of the Hanford Site. No attempt has been made to assess the cumulative impacts of these additional stressors, however, the risk is likely quite small due to the dilution afforded by the river and surveillance data that indicates that the quality of the Columbia River is good and within applicable water quality standards [1]. Moreover, the viability of spawning population of salmonids in the Hanford Reach continues to be one of the “success” stories in what has been a dismal performance for most other ESUs of this resource in the Pacific Northwest.

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# Implementation and validation of the USDOE graded approach for evaluating radiation impacts on biota at long-term stewardship sites

D.S. Jones<sup>a</sup>, P.A. Scofield<sup>b</sup>, S.L. Domotor<sup>c</sup>

<sup>a</sup> Oak Ridge National Laboratory, Environmental Sciences Division, Oak Ridge, TN, United States of America

<sup>b</sup> Oak Ridge National Laboratory, Environmental Protection and Waste Services Division, Oak Ridge, TN, United States of America

<sup>c</sup> United States Department of Energy, Office of Environmental Policy and Guidance, Washington, DC, United States of America

**Abstract.** The DOE Technical Standard, “A Graded Approach for Evaluating Radiation Doses to Aquatic and Terrestrial Biota” (the Graded Approach) was used to evaluate two geographically discrete DOE sites, Bear Creek Valley (BCV) in Oak Ridge, Tennessee and the Test Reactor Area (TRA) ponds in Idaho Falls, Idaho. The first step entailed reviewing existing ecological risk assessments (ERAs) of contaminated waste sites and selecting the most appropriate sites for testing the Graded Approach, based on the existence of measurable radiological contamination and biological data. Ambient media (soil) data were evaluated using the initial (conservative) screening protocol in the Graded Approach. The next step entailed comparing the results from the Graded Approach with those of the existing ERAs for each site, which were used as the primary standard of performance. The default (conservative) screening protocol correctly classified the positive control site (TRA) as posing potential risks to biota and the negative control site (BCV) as not posing potential risks to biota, based on exposures to ionizing radiation. Future evaluations will use both ambient media and the available biota data to test the more realistic tiers (i.e., the Analysis Phase) of the Graded Approach.

## 1. INTRODUCTION

The United States (U.S.) Department of Energy (DOE) has determined that long-term stewardship (protection of human health and the environment from residual contamination following site cleanup) will be required for over 100 of the 144 DOE waste sites [1]. Long-term stewardship programs will need to be responsive to increasing stakeholder concerns and regulatory requirements for demonstrating protection of the environment (non-human biota) from the effects of ionizing radiation during the pre-closure and post-closure phases. The purpose of this project is to conduct a pilot implementation and validation of the DOE’s Graded Approach and its companion software. The objective is to test the method’s utility as an acceptable, cost-effective tool that can be used for evaluating radiation doses to biota as part of routine long-term surveillance and monitoring activities. Local stakeholders have been briefed and are keenly interested in this project.

## 2. METHODOLOGY

The DOE Graded Approach for evaluating radiation doses to biota, documented in a DOE technical standard [2] and in a series of technical papers [3–6], was applied at two test sites. First, contaminated waste sites were evaluated for their appropriateness for testing the Graded Approach. Biota Dose Assessment Committee (BDAC) representatives at major DOE facilities in the U.S. were consulted and project records for the BDAC-recommended waste sites were reviewed. Ideal waste sites would have:

- been the subject of a base line ecological risk assessment (BERA);
- measurable radiological contamination; and
- existing biological data available.

Two sites were sought:

- a negative control = BERA indicated no radiological risk to biota; and
- a positive control = BERA indicated a radiological risk to biota.

Unfortunately, an ideal positive control site was not available.

## 2.1. Site descriptions

### 2.1.1. Negative Control

- Location: Bear Creek Valley (BCV) watershed at Y-12 Plant, Oak Ridge, TN.
- Data: Concentrations in surface soils, plants, earthworms, and small mammals from floodplain transects and hazardous waste sites [7].
- Assessment: BERA available, indicates risks to ecological receptors are unlikely.

### 2.1.2. Positive Control

- Location: Test Reactor Area (TRA) ponds at Idaho National Engineering and Environmental Laboratory, Idaho Falls, ID
- Data: Concentrations in surface soil, sediment, water, and birds [8].
- Assessment: BERA not available, TLD-measured dose rates to small mammals exceeded 0.1 rad/day (1 mGy/day) level.

## 3. ANALYSIS

Surface soil concentrations were evaluated using the Data Assembly and General Screening Phases from the Graded Approach (Figure 1). Maximum radionuclide concentrations were compared with the soil Biota Concentration Guides (BCGs) using the RAD-BCG Calculator. Each sampling location at both sites were evaluated separately. Future analyses will include site-specific biota data and more realistic protocols from the Graded Approach.

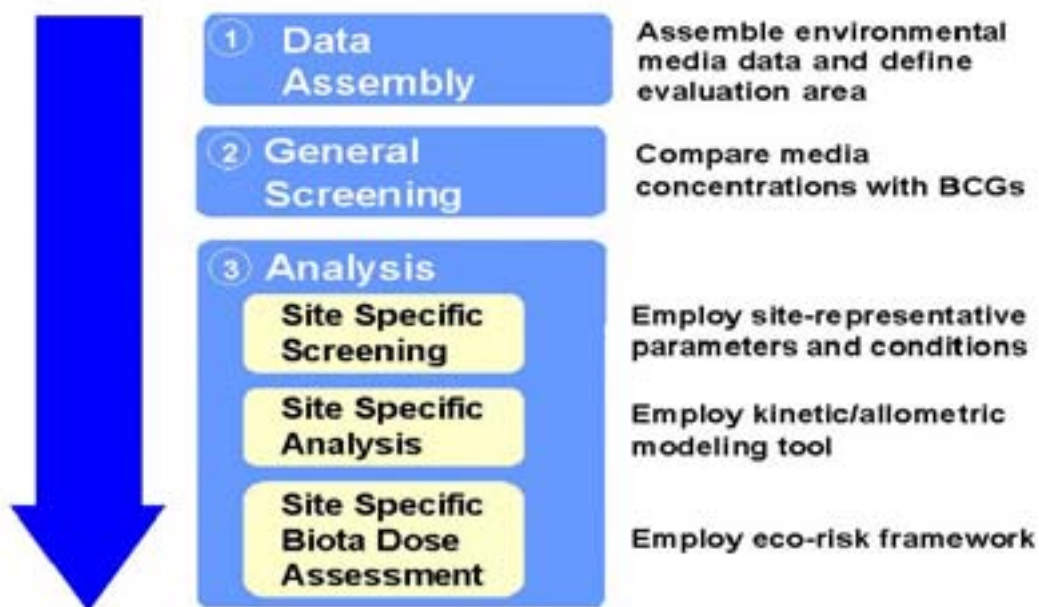


FIG. 1. DOE's Graded Approach for Evaluating Radiation Doses to Biota.

#### 4. RESULTS

The partial fraction (isotope concentration to BCG ratio) and sum of fractions for the most contaminated sites are shown in Table 1 and Table 2. The sampling location with the highest sum of fractions at the Negative Control site was the “burn yard” waste pit, with U-238 accounting for most of the estimated exposure. This location is not expected to pose a risk to biota. Surface soil concentrations from around the TRA waste ponds were used for the Positive Control site, where Cs-137 accounted for most of the estimated exposure. This site appears to pose a risk to biota.

#### 5. DISCUSSION

Suitable control sites were difficult to find. Biological data were often not collected at highly contaminated sites because interim corrective measures were taken to protect human health. Also, true BERAs were not performed at sites assessed before the mid-1990s and there is a paucity of radiological data for biota.

BCG values are not yet available for all isotopes of concern. They will need to be developed to allow for proper evaluation of waste sites. DOE intends to generate more BCGs in the near future.

TABLE 1. SCREENING RESULTS FOR MOST CONTAMINATED SAMPLING LOCATION AT THE NEGATIVE CONTROL SITE: SUM OF FRACTION SUMMARY<sup>a</sup>

Isotope	Site Data (Bq/Kg)	BCG (Bq/Kg)	Partial Fraction
<sup>90</sup> Sr	70	800	0.085
<sup>99</sup> Tc	2852	200000	0.017
<sup>232</sup> Th	70	60000	0.0013
<sup>234</sup> U	9630	200000	0.051
<sup>235</sup> U	630	100000	0.006
<sup>238</sup> U	32593	60000	0.56
		SUM	0.72

<sup>a</sup> BCG soil limits were unavailable for <sup>228</sup>Th and <sup>230</sup>Th.

TABLE 2. SCREENING RESULTS FOR MOST CONTAMINATED SAMPLING LOCATION AT THE POSITIVE CONTROL SITE: SUM OF FRACTION SUMMARY<sup>b</sup>

Isotope	Site Data (Bq/Kg)	BCG (Bq/Kg)	Partial Fraction
<sup>241</sup> Am	0.59	100000	0.0000041
<sup>144</sup> Ce	37.0	50000	0.00069
<sup>137</sup> Cs	8148.2	800	10.6
<sup>60</sup> Co	2518.5	30000	0.097
<sup>155</sup> Eu	4.07	600000	0.0000069
<sup>239</sup> Pu	2.4	200000	0.000011
<sup>125</sup> Sb	8.2	100000	0.000065
<sup>90</sup> Sr	211.1	800	0.25
<sup>95</sup> Zr	3.0	40000	0.000069
		SUM	11

<sup>b</sup> BCG soil limits were unavailable for <sup>7</sup>Be, <sup>54</sup>Mn, <sup>95</sup>Nb, <sup>141</sup>Ce, <sup>134</sup>Cs, <sup>152</sup>Eu, and <sup>238</sup>Pu.

## 6. CONCLUSIONS

The Graded Approach appears to be relatively easy to use and generally reliable. The general screening protocol in the Graded Approach correctly classified the two test sites:

- The Negative Control site had a maximum sum of fractions  $< 1.0$ ;
- The Positive Control Site had a maximum sum of fractions  $> 1.0$ .

Screening results can be used to focus realistic assessments on the most important radionuclides, pathways, and receptors.

Pilot implementation of the Graded Approach identified several issues for which guidance or further development are warranted.

Subsequent evaluations will focus on the more realistic assessment protocols in the Graded Approach (i.e., the Analysis Phase).

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# Investigations on the mechanism of terrestrial transport of radionuclides in a complex terrain

P.M. Ravi, R.P. Gurg, G.S. Jauhri

Health Physics Division, Bhabha Atomic Research Centre, Mumbai, India

**Abstract.** Indian Atomic Energy program ensures that the operations of nuclear facilities do not lead to any adverse effect on the surrounding environment. Environmental Survey Laboratories (ESLs) are setup at major nuclear sites to assess the impact of the operating nuclear facilities. The laboratories are involved in R&D programs leading to improved understanding of the mechanism of interaction of radiation with the biota and that of the transport of radionuclides through different environmental compartments. The Kaiga site, being part of Western Ghats, is ecologically complex with the presence of hills and valleys and a forest cover inhabiting several biological species. This paper presents the results of the investigation on the mechanism involved in the terrestrial transport of radioactivity in Kaiga region. In view of the heavy rainfall in this area, wet deposition is the significant pathway of transfer of radionuclides from air to soil. Air to rain water transfer and wet deposition rate were estimated using  $^7\text{Be}$  as a tracer. Influence of Kd on the soil-to-soil solution transfer was studied. The data on the distribution of some trace and major elements in plant system is studied. It was observed that, when a competition arises between chemically similar nuclides ( $^{40}\text{K}$  and  $^{137}\text{Cs}$ ), biologically necessary elements are preferentially absorbed by the plant. This data will be an input to the calculation of radiation risk to the plant as well as animal feeding on them.

## 1. INTRODUCTION

Two PHWR type nuclear reactors are operating and two are under construction at Kaiga, located in Western Ghats of southern Indian peninsula. The site is ecologically sensitive with hills and forests, inhabiting variety of species (Figure 1). Environmental survey laboratory at Kaiga has initiated R&D programs to assess the radiological impact on overall eco-system. A radionuclide released to the atmosphere gets attached to the air particles and forms aerosols. These aerosols undergo foliar deposition on part of vegetation and also get deposited on the ground. The extent of deposition depends upon the physicochemical properties of aerosols. Ground deposited nuclides find its way to plants by root absorption. Root absorption depends upon the extent of transfer from soil matrix to soil solution, which in turn depends upon Kd of the soil, moisture content of the soil, etc. Root absorption by plant is also influenced by the chemical and biological competition from major elements especially chemically similar to the nuclide of interest. Once absorbed in the plant, the radionuclides get redistributed in the plant organs to meet the biological requirements of the plant. The radiological impact from a radionuclide released from a nuclear power plant depends upon the deposition rate, trace and major elements present in the soil, Kd of the soil, biological affinity and redistribution pattern of radionuclides within the plant.

This paper presents the results of estimation of site-specific wet deposition rate, investigation on the element distribution within the plant, influence of Kd and chemical congeners on root absorption.

## 2. $^7\text{Be}$ AS A TRACER

$^7\text{Be}$  is produced by cosmic ray interaction [1, 2] and is widely used as a tracer for atmospheric studies. In view of its similarity to fission products such as  $^{137}\text{Cs}$  and  $^{131}\text{I}$  (Water solubility, particle size, etc) we have used it as a tracer for wet deposition rate estimation. It has a convenient half-life of 53.3 days (very useful for retention half life estimation) and convenient gamma energy of 477.6 keV suitable for gamma ray spectrometric estimation.

## 3. MATERIALS AND METHODS

Standard methods for environmental analysis were used [3]. Air particulates are sampled through a filter paper (flow rate  $1\text{m}^3/\text{min}$ ).  $^7\text{Be}$  activity in monthly cumulative air sample is estimated by  $\gamma$  ray spectrometry using HPGe.

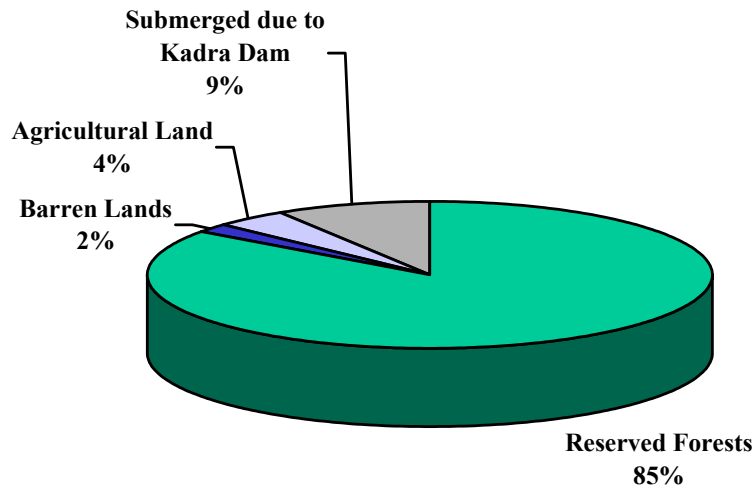


FIG. 1. Utility of Land around Kaiga Site (10 km Radial Zone).

$^7\text{Be}$  in monthly cumulative rainwater sample was estimated by gamma ray spectrometry. Leaf samples of some plants commonly available in this area are collected, dried and analyzed for  $^7\text{Be}$  by  $\gamma$  ray spectrometry.

Plant parts (root, stem and leaf) are ashed using standard procedure. Ash and dried soil were digested with  $\text{HNO}_3/\text{HClO}_4$  mixture. Concentrations of Na, K and Ca were estimated in the extract by flame photometry. Concentrations of trace elements such as Sr, Zn, Mn, Mg and Fe in the extract were estimated by Atomic Absorption Spectrophotometry.

#### 4. RESULTS AND DISCUSSIONS

##### 4.1. Major intake processes

Plants may pick up radioactivity by two pathways namely foliar deposition and root uptake [4].

##### 4.2. Foliar uptake

Release  $\rightarrow$  Atmosphere  $\rightarrow$  Foliar deposition (wet or dry)  $\rightarrow$  Assimilation and distribution

Radionuclide concentration in vegetation, due to foliar deposition, C (Bq/kg), is given by the equation [2]:

$$C = \{d\alpha[1-e(-\lambda_E T_E)]\} / \lambda_E \quad (1)$$

where:

- d is the deposition rate in wet and dry process;
- $\alpha$  is the fraction of intercepted activity of the organ of interest per unit mass;
- $\lambda_E$  is the effective rate constant for reduction of activity; and
- $T_E$  is the period that plants are exposed to contamination.

$$\text{Wet deposition rate} = \text{Scavenging ratio} * \text{Rainfall rate}$$

$$\text{Scavenging ratio} = [\text{Activity in rain (Bq.m}^{-3}\text{)}] / [\text{Activity in air (Bq.m}^{-3}\text{)}]$$

Rainfall rate is expressed as  $\text{m.s}^{-1}$  and wet deposition rate is expressed as  $(\text{Bq.m}^{-2}.\text{s}^{-1})$  Per  $(\text{Bq.m}^{-3})$  or simplify  $\text{m.s}^{-1}$ .

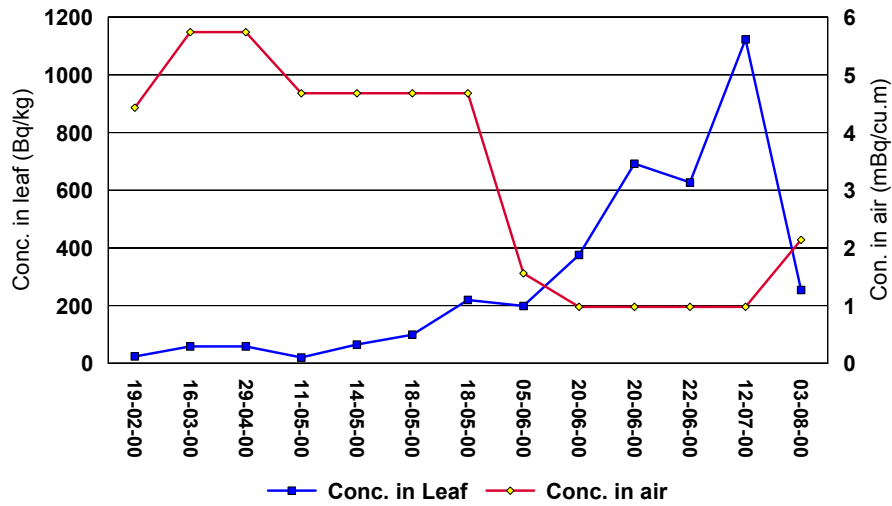


FIG. 2. Air to plant transfer pattern of <sup>7</sup>Be observed at Kaiga.

Figure 2 shows the distribution of <sup>7</sup>Be in air and some plants and its influence over the season. It can be clearly seen that the concentration of <sup>7</sup>Be in leaf samples during non-rainy seasons (February to May) is comparatively less as compared to that during monsoon seasons (June, July). During summer season, foliar deposition is only due to dry deposition, while it is enhanced by the rainfall in monsoon season. During monsoon season, the activity is scavenged by the atmospheric water and carried down by rainfall.

Wet deposition rate calculated using the above equation is shown in Table 1. In view of physicochemical similarity of <sup>7</sup>Be aerosols with <sup>137</sup>Cs and <sup>131</sup>I, the presently estimated deposition can be used for estimation foliar uptake of <sup>137</sup>Cs and <sup>131</sup>I in the plants.

### 4.3. Root uptake

Radionuclide concentration in vegetation, c(Bq/kg), due to root uptake is given by [4]:

$$C = F_V * C_S \quad (2)$$

where:

$F_V$  is the concentration factor for the uptake; and  
 $C_S$  is the concentration in soil.

The equation does not take into account the non-availability of one fraction of element due to retention by soil. In the present work, we consider the influence of soil characteristics also by considering the soil to soil solution transfer. Accordingly, the pathway may be modified as:

Release → Atmosphere → Soil deposition (wet or dry) → Soil solution → Root →  
 Assimilation and distribution

Transfer from soil to soil solution depends upon the chemical characteristics of the soil and the element of interest. The Concentration in soil solution ( $C_{SS}$ ) is governed by the equation:

$$C_{SS} = C_{TOT}/(Kd+M_c) \quad (3)$$

where:

$Kd$  is the Distribution ratio and is equal to  $C_{EQ}/C_{AQ}$ ;  
 $C_{EQ}$  (mg/kg) = Concentration of the element in soil in equilibrium with aqueous phase;  
 $C_{AQ}$  (mg/l) = Concentration of the element in aqueous phase in equilibrium soil;  
 $C_{TOT}$  represents elemental concentration in soil (mg/Kg); and  
 $M_c$  represents the water content in the soil.

TABLE 1. WET DEPOSITION RATE USING 7BE

Month	Activity in air (Bq.m <sup>-3</sup> )	Activity in rain water (Bq.m <sup>-3</sup> )	Scavenging ratio	Rain fall rate (m.s <sup>-1</sup> )	Deposition rate (m.s <sup>-1</sup> )
May, June	1.6E-3	175.2	1.1E05	7.6E-07	0.09
June, July	1.0E-03	208.5	2.1E05	6.6E-07	0.14
July–Aug	2.1E-03	79.9	3.7E04	5.3E-07	0.02
Aug–Sept	1.6E-03	199.0	1.3E05	7.8E-07	0.09

K<sub>d</sub> is the most important parameter, which influences the transfer. Its influence can be clearly understood by simulating C<sub>ss</sub> values for different K<sub>d</sub> values. Figure 3 shows the influence of K<sub>d</sub> on the transfer of element from soil to soil solution.

As K<sub>d</sub> increases, extent of transfer to soil solution decreases considerably. K<sub>d</sub> depends upon the chemical state of radionuclide, soil particle size and organic content, pH etc. For a given radionuclide, transfer to soil solution decreases with increase in size of soil particle and decrease in organic content.

Transfer from soil solution to root depends upon the cation exchange capacity of root and ion exchange affinity of radionuclide. In general, root act as the storage location of minerals and will be distributed depending upon the biological necessity of the plant. The radiation dose received by the plant depends upon the distribution of radionuclide within the plant.

#### 4.4. Distribution of radionuclide within the plant

The relative distribution within the plant varies from element to element depending upon the chemical properties and biological necessity of the plant. Figure 4 shows the distribution of some major and trace elements within the plant *Chromoleana sps* observed commonly in this area. In most of the elements, major accumulation takes place in leaf of the plant. However in the case of Na and Fe, considerable fraction was absorbed in root also. Distribution pattern of Ca and Sr, which are chemically similar, is qualitatively similar. In view of its chemical similarity with K, <sup>137</sup>Cs distribution pattern is expected to be similar to that of K. This distribution data will be very useful in the dosimetry of plants burdened with nuclides <sup>137</sup>Cs and <sup>90</sup>Sr.

Due to the biological necessity of the plant, distribution pattern of chemically similar elements in the plant can differ considerably. Table 2 shows the concentrations of Ca and Sr in soil and in parts of some commonly available plants in this area. The Ca/Sr ratio is shown the last column. It can be seen that Ca/Sr ratio is comparatively high in plant parts as compared to soil. This leads to the conclusion that Ca is selectively absorbed by the plant as compared to Sr. Similar observation was made in our earlier studies pertaining to the chemically similar pair Cs and K [5]. Among the chemically similar elements, biological necessity of the plant imparts selectivity. This factor has to be considered in the uptake studies.

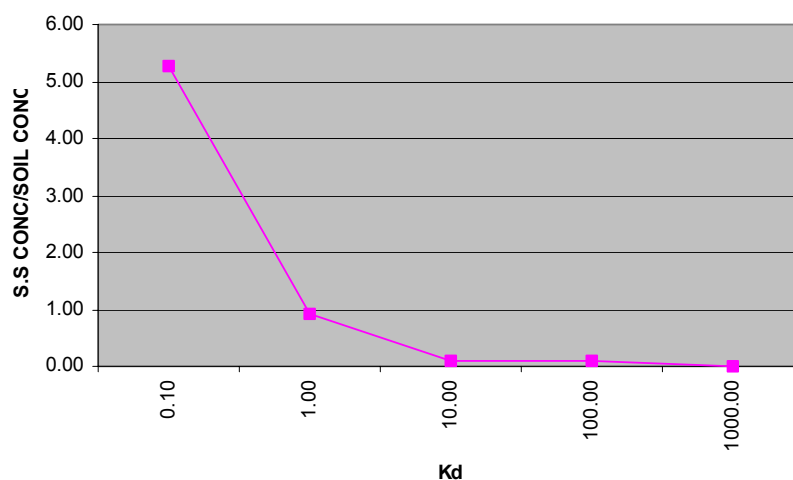


FIG. 3. Theoretical curve showing the effect of K<sub>d</sub> on relative transfer to Soil Solution (SS) from soil.



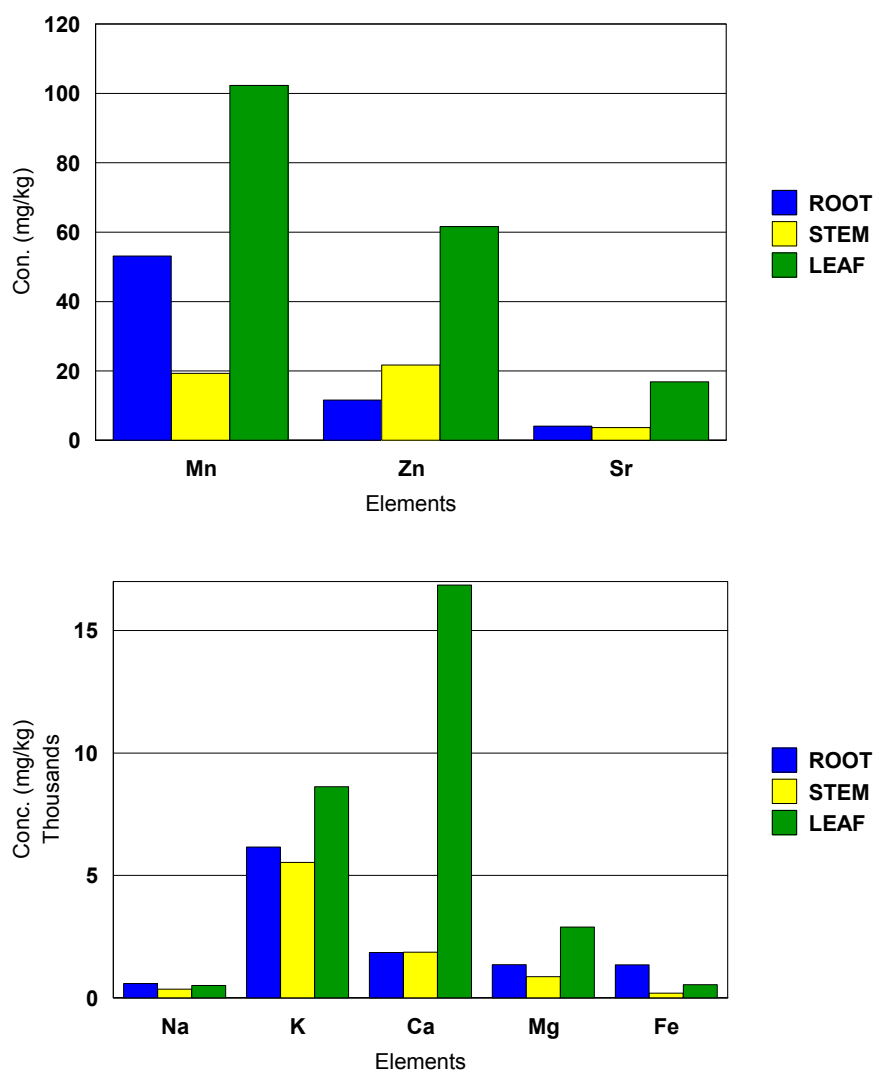


FIG 4. Relative distribution of elements in plant organs.

TABLE 2. Ca/Sr RATIO IN SOIL AND DIFFERENT ORGANS OF PLANTS

Type of Plant Species	Part of the Plant	Sr conc. (mg/kg Dry wt)	Ca conc. (mg/kg Dry wt)	Ca/Sr Ratio
Soil	–	9.3 to 12.1	1275.3 to 2067.5	105.4 to 222.3
<i>Chromolaena sps</i>	Root	4.1	1852.7	451.9
<i>Chromolaena sps</i>	Stem	3.7	1865.9	504.3
<i>Calotropis sps</i>	Leaf	16.9	16858.1	997.5
<i>Calotropis sps</i>	Root	3.3	2233.1	676.7
<i>Calotropis sps</i>	Stem	5.4	2758.3	510.8
<i>Calotropis sps</i>	Leaf	24.0	18130.7	755.4
<i>Elephantopus sps</i>	Root	6.3	2012.2	319.4
<i>Elephantopus sps</i>	Stem	11.7	2823.8	241.4
<i>Elephantopus sps</i>	Leaf	13.6	9565.6	703.4
<i>Terminalia sps</i>	Root	27.3	13369.9	489.7
<i>Terminalia sps</i>	Stem	13.8	5639.0	408.6
<i>Terminalia sps</i>	Leaf	27.4	14705.5	536.7

## 5. FINAL CONCLUSIONS

Concentration of  $^7\text{Be}$  in leaf samples during non-rainy seasons (February to May) is comparatively less as compared to that during monsoon seasons (June, July). During summer season foliar deposition is due to only dry deposition while it is enhanced by the rainfall in monsoon season. During monsoon season, the activity is scavenged by the atmospheric water and carried down by rainfall. It can be seen that Ca/Sr ratio is comparatively high in plant parts as compared to soil. This leads to the conclusion that Ca is selectively absorbed by the plant as compared to Sr. Among the chemically similar elements; biological necessity of the plant imparts selectivity.

This factor has to be considered in the uptake studies. Influence of Kd on soil to soil solution transfer is estimated. As Kd increases, extent of transfer to soil solution decreases considerably. In most of the elements, major accumulation takes place in leaf of the plant. However in the case of Na and Fe, considerable fraction was absorbed in root also. Distribution pattern of Ca and Sr, which are chemically similar, is qualitatively similar. In view of its chemical similarity with K,  $^{137}\text{Cs}$  distribution pattern is expected to be similar to that of K. This distribution data will be very useful in the dosimetry of plants burdened with nuclides  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . A fairly good idea of the factors influencing air to plant and soil to plant transfer of radionuclides were obtained by this study and useful for the radiological impact assessment of plants due to the release from a nuclear installation.

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## 5. SUMMARY OF WORKSHOP DISCUSSIONS\*

### 5.1. INTRODUCTION

On the fourth day of the Symposium, three Workshops were held to provide a more extensive opportunity for discussion of the main issues covered in the previous presentations and poster sessions. The subject of the Workshops mirrored those of the preceding days' sessions, as follows:

- Workshop 1: Ionizing Radiation and Biota: Effects, Responses and Mechanisms (Leader: Ward Whicker);
- Workshop 2: Frameworks for Environmental Radiation Protection (Leader: Lars-Erik Holm);
- Workshop 3: Methods and Models for Evaluating Radiation as a Stressor to the Environment (Leader: Jan Pentreath).

These Workshops were led by the Keynote Speaker for the relevant session, identified above, and supported by a member of the International Organising Committee (IOC). In advance of the Symposium, the IOC had identified the topics for discussion with the help of a review of key topics from the submitted abstracts, and the reports of the Specialists Meetings, held by the International Atomic Energy Agency in the years 2000 and 2001. These suggested topics were distributed to the Symposium participants and formed the initial basis for discussion, and they are included at the end of this paper for ease of reference.

The objectives of Workshops 1 and 3 were similar, and were to:

- Determine what is known;
- Determine what needs to be known;
- Set priorities for filling the missing knowledge and information gaps;
- Ascertain what work is already being done.

The objectives for Workshop 2 were to:

- Identify the underlying policy and philosophical bases for regulating possible impacts of radiation on the environment;
- Explore similarities and differences amongst existing policies and frameworks (national and international);

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\* Prepared by: Carol Robinson (International Atomic Energy Agency) and Alex Zapantis (Supervising Scientist Division, Environment Australia), with contributions from the Keynote Speakers and Members of the International Organising Committee.

- Consider the need for an international system of environmental radiation protection, and how it would relate to the existing, human-centred system;
- Identify the principal issues which need to be overcome to develop a national or international system of environmental radiation protection.

Many issues were discussed in more than one Workshop and this paper presents an overall summary arranged by subject area rather than by Workshop.

## 5.2. THE NEED FOR AN ENVIRONMENTAL PROTECTION SYSTEM

It was recognised that there are situations in which the environment may not be sufficiently protected under the current system of radiological protection; for example in situations where humans are absent from the environment into which radionuclides are released or are otherwise present (e.g., where there are disposal areas remote from human habitation and in some remediation situations, respectively). Furthermore, the radiation protection community has, until now, not taken full account of the implications of the UNCED Rio Declaration of 1992, and the consequent range of legal obligations for some countries relating to protection of the environment. It was however recognized that radiation is only one of many environmental stressors and, although there is some information to indicate that it is usually a minor issue compared with other environmental pollutants, the public often perceives the risk associated with radiation differently.

There is therefore now a need for an international approach that will allow regulators and operators to address environmental protection explicitly, rather than assume that the protection of man will ensure the protection of the environment.

## 5.3. THE PROCESS OF DEVELOPING AN ENVIRONMENTAL RADIATION PROTECTION SYSTEM

There is value in the development of an internationally agreed approach for environmental radiation protection. This is essential for many reasons, but particularly to maximise the level of acceptance within different countries, and to ensure that one country does not have a competitive advantage over another as a result of being bound by different rules. It was agreed that the ICRP and IAEA have a very important role in coordinating and achieving an international consensus, but that their function should be in developing the overall policy framework and the objectives of protection, but without being overly prescriptive. Detailed regulatory requirements and criteria should be the subject of local legislation or procedures. A mechanism to allow periodic international review should be established.

## 5.4. PRINCIPLES OF ENVIRONMENTAL PROTECTION

It was concluded that it is in man's interests to protect the environment, but it was also acknowledged that a healthy environment is valued, in its own right, irrespective of the direct benefits it affords humans.

It was considered that the principles of environmental protection are well defined and internationally agreed. The challenge now is to develop an approach for environmental radiation protection that conforms to these principles. The existing responsibilities of regulators and operators, relating to radiation, may need to be revised accordingly.

## 5.5. COMPONENTS OF A PROTECTION SYSTEM

A broad range of expertise is required to develop a system for environmental radiation protection. It was recognised that powerful assessment and management tools already exist, within the radiation protection community, but that these may be enhanced by the incorporation of aspects of those used in a wider environmental protection context.

The overall protection approach adopted for radiation protection should be consistent, where possible, with those applied for other pollutants. There are similarities, at the conceptual level, between radiation effects and the effects of some chemicals, and the principles and objectives for protection should be identical. However, it was appreciated that different pollutants are often regulated by different agencies within any one country and that different protection endpoints may be applied.

The applicability of the existing principles of radiological protection in an environmental context was considered. While the principle of *Justification* has a wider scope than radiological protection, and relates to decisions made by society as a whole, it would appear to be equally applicable to environmental and human radiation protection. In its broadest sense, *Optimisation* — the aim of achieving an optimum level of protection, taking account of the various costs involved — is also relevant to environmental radiation protection. It was, however, noted that the concept of collective dose has no corresponding application. It was agreed that absorbed dose is likely to be the ultimate basis for establishing environmental protection criteria, but that *Dose Limitation* should be applied differently. Rather than set mandatory dose limits, it would be preferable to derive a set of ‘action levels’ of dose that trigger an escalating scale of considerations, investigations and responses. It may also be more practical to place regulatory requirements on directly measurable quantities, such as concentrations in effluents, environmental media, etc. These maximum concentrations could be derived on the basis of the appropriate dose ‘action level’.

In developing an environmental protection system, it will be necessary to incorporate the best scientific information available. There is already a great deal of information available on responses, mechanisms and the effects of ionising radiation on biota, including reports by the IAEA and NCRP in the 1990s the 1996 UNSCEAR report. It is therefore important to ensure that the available information is reviewed in a structured way, in order to apply it appropriately and to identify significant data gaps. As an example, the EC FASSET project will facilitate this process in a European context.

A number of issues requiring early resolution in the development of a protection system were identified:

- The treatment of natural background radiation — whether background doses are included in possible criteria and corresponding compliance assessments, given that protection will probably be aimed at preventing deterministic effects rather than minimising the probability of stochastic effects;
- The derivation of a quantity for biota assessment that performs a similar function to the radiation weighting factor, particularly for alpha particles.

## 5.6. STAKEHOLDER INVOLVEMENT IN ENVIRONMENTAL DECISION-MAKING

Stakeholder consultation should form an element of any system of environmental radiation protection. For many projects (e.g., major clean-up projects or facility siting decisions), stakeholders are already likely to be involved in the environmental impact assessment process, and the justification process. In the case of environmental radiation protection, stakeholder participation may, instead, focus on the optimisation of protection. In order to facilitate stakeholder involvement, transparency of process and agreement on the conditions under which it is conducted are essential. Ideally, stakeholders, taking part in a consultation process, should have access to appropriate expertise. There should also be a recognition that the result of this process may be that the stakeholders agree to disagree.

## 5.7. THE REFERENCE ORGANISM APPROACH

Participants recognised the practical value of the reference organism approach. This approach is included in the current ICRP Task Group Report and the work of the EC FASSET programme and it is consistent with approaches applied in several countries (e.g., USA). This approach provides a practical basis for transparent assessments, by a clear identification of the parameters involved, and by providing a basis for comparisons between different situations and management options. The level of simplification involved in this approach was recognized, as was the existence of alternative approaches. The simplified nature of the reference organism approach corresponds with the current availability of parameter information, and the need to augment it as more information becomes available, was noted.

The reference organism would fulfil the same role as reference man within the current system of radiological protection. As such, it would provide a *phantom* for dosimetry, and for the interpretation of dose/effect relationships which are, in most cases, available only at higher dose rates than those likely to be generally encountered. The approach would, however, be much simpler than that adopted for man, e.g., in terms of the metabolic models underlying the uptake of radionuclides by the reference organisms.

The choice of appropriate reference organisms should consider radiosensitivity, ecological significance and exposure potential. An overall environmental assessment and management approach that made use of 3 levels of reference organism was discussed; primary, secondary, and tertiary, utilizing models and data of different complexities, with the following attributes:

- Primary: Highly detailed characterisation of a small range of specific fauna and flora, at all stages of their life cycle, e.g., a benthic type of fish, a carnivorous terrestrial mammal (perhaps developed by ICRP);
- Secondary: A more superficial approximation for a larger range of biota in particular ecosystems, using parameters based upon local conditions (perhaps developed by national authorities);
- Tertiary: Models referring to particular species in particular environments.. These may also include species that legally have to be assessed.

Work that would enable the definition of *reference organisms* was identified as a high priority. For example, relevant dose models will be needed to calculate doses to reference organisms for the most ecologically-relevant endpoints. In applying such models, and interpreting effects data, the most relevant stages in the life cycle of biota need to be considered; the adult stage is rarely the most susceptible to radiation damage.

Whilst the task of defining reference organisms, for the full range of environments, appears onerous, it was recognised that there is already a significant amount of data available. The data gaps are not so great as to prevent the development and implementation of a system of environmental radiation protection. Indeed, some countries and organizations have already applied similar methodologies, that could be considered in the development of standardized reference organisms.

## 5.8. METHODS AND MODELS FOR EVALUATING RADIATION AS A STRESSOR TO THE ENVIRONMENT

Operational environmental radiation protection systems should include explicit definitions and delineation of such aspects as *Problem Formulation, Context, Assessment/Evaluation and Management*. The detailed application of these elements may differ from one country to another, but the overall procedure and the significant issues for consideration are likely to be similar. Consideration will be given to: compliance with regulatory requirements; research, particularly on the linkages between individual radionuclides, dose to biota from those nuclides, and the effects arising from that dose. Other general considerations may include:

- Identification of the most exposed and most radio-sensitive species affected, or likely to be affected, by a proposed or actual industrial operation;
- The application of probabilistic (including Monte Carlo) dose modelling and risk assessments;
- The identification of requirements and limitations on the accuracy and precision of measurements, especially those relating to dose and effects;
- Clarification of where intervention is necessary, using an holistic approach that will be able to determine where intervention may do more harm than good;
- Allowance for accommodation of new and changing scientific information;
- The application of screening approaches within an overall protection system, to help focus resources appropriately.

One of the key steps in this process is to establish the *effluent toxicity*, relevant to the specific site and species where appropriate. In many cases, ecotoxicological methods and models are available to facilitate this process. Environmental transfer models exist, as part of the system of radiological protection for humans, but these may require modifications to allow biota dose assessments, and relevant experience. In many cases, lack of data places a limit on the extent to which detailed models of environmental transfer can be constructed, with the result that simple conservative models are likely to have continuing application. Furthermore, while the environmental monitoring programmes currently in place will provide information of value in such assessments, other biological monitoring approaches may also be useful, in the longer term.

## 5.9. FUTURE DIRECTIONS AND RESEARCH NEEDS

Research to fill the gaps should be prioritised and coordinated, and it was suggested that there was a significant role for the IAEA and ICRP in this capacity.

Scenarios based on uranium mining would provide excellent case studies to assist in the development and testing of methods and models of potential environmental impacts using reference organisms.

Environmental models have been developed to determine distribution of contaminants in the environment. At present, the models applied for radionuclide migration are designed for the purpose of estimating doses to humans. In the future these will need to be modified to take account of animal life histories etc. that may result in the exposure of fauna and flora. Furthermore, models will need to be developed to include the influences of chemical speciation, physical and biological half-lives.

A system of environmental radiation protection may also require some form of biological monitoring regime to measure, directly, change or damage to ecosystems. Population statistics such as population size, density or dose distribution, reproduction rates and information on genetic diversity could be used, as well as *biomarkers*, in order to obtain direct and quantifiable indicator of impact in the affected environment.

The complexity of ecosystems requires that a multidisciplinary approach be adopted in research into the mechanisms underlying the effects of, and responses to, ionising radiation in biota. Particular issues that warrant further work were identified:

- Stress biomarkers for biological monitoring of contaminated systems;
- Effects of ionising radiation at the molecular and cellular level, (where effects must occur first) and the propagation of those effects up through the individual, population, community and ecosystem;
- Practical models and parameters to improve endpoint-specific dosimetry and effects information;
- Experimental micro and mesocosms should be used to approximate the natural environment to address potential interactive effects with other environmental stressors either of natural origin, or arising from human activities;
- Population genetics and the role of gene pool isolation or mixing and natural selection.



## ANNEX

### PRELIMINARY ISSUES FOR DISCUSSION

*Developed by the International Organising Committee*

#### WORKSHOP 1

##### **Ionizing Radiation and Biota: Effects, Responses and Mechanisms**

- What biological effects are attributable to ionising radiation – and how are these manifested in individuals and populations?
- Is the development of a Reference Non-Human Biota (analogous to Reference Man) approach the most effective use of effects data – what are the advantages of this approach and what alternatives exist?
- What priority should be given to research of radiological impacts under low dose-rate chronic exposure conditions – and how should it be conducted?
- To what extent can the results of research conducted in a laboratory environment be interpreted in a broader environmental context?
- To what extent is it possible to link individual and population responses?
- In view of the possibility of synergistic effects, is it essential that radiological impacts on biota are considered in conjunction with non-radiological impacts – and how should this be achieved?
- How should biota-specific RBE values for different types of radiation (especially high-LET) be determined => biota-specific weighting factors (cellular and ecological)?
- To what extent is it necessary to develop more detailed internal dosimetry models for biota in order to interpret dose-effect relationships?
- How should cell self repairing and hormetic effects be considered?
- Do the effects of specific radionuclides need to be researched in more detail and why (e.g. tritium, U) and are biota exposed to a broader range of radionuclides than humans?

#### WORKSHOP 2

##### **Frameworks for Environmental Radiation Protection Example**

- To what extent is the human-orientated system of radiological protection generally applicable to the environment?
- Is there a consensus on identifiable environmental protection principles and, if so, how may these be related to the impact of radiation in a quantifiable way?
- To what extent should an approach for environmental radiation protection be consistent with policies and methods adopted for chemical toxins and other non-radiological hazards?
- Is the development of a Reference Organism approach the most effective basis for developing an environmental protection framework – what are the implications of this and are there alternatives?
- Should a system of radiological protection for the environment be part of or separate from the approach for man?
- Are the concepts of justification, optimisation and dose limitation applicable as principles in environmental radiological protection?
- What mechanisms exist to ensure adequate stakeholder involvement in environmental radiation protection policy making, and what are the implications for regulator independence?
- What are the main elements of a protection framework and to what degree is it necessary and feasible to develop an international approach (e.g. practicality, suitable endpoints)? In what form should criteria for environmental protection be specified (dose rate/environmental concentration)?
- What are the most suitable compliance assessment endpoints (dose rate/environmental concentration)?
- How should natural background be addressed in the development of an assessment approach.

### **WORKSHOP 3**

#### **Methods & Models for Evaluating Radiation as a Stressor to the Environment**

- What models currently exist for impact assessment and what are the key similarities and differences?
- Is there a need for a global approach to constructing models for the assessment of impacts on biota?
- Under what conditions would national or regional approaches to environmental radiation assessment be applicable?
- Under what circumstances is it sufficient to apply a generic modelling approach and when is it necessary to be more realistic?
- What factors should be considered in identifying a suitable target for assessment – for example reference organisms relevant to particular environments and radiological exposure scenarios?
- What are the most suitable compliance assessment endpoints (dose rate/environmental concentration), and what are the implications for compliance monitoring?
- How should natural background be addressed in compliance assessments?
- How can impact assessments address multi-pollution impacts?
- What other factors arise in assessing the impact of radiation on biota – for example averaging issues – lifetime/spatial?
- What research and development work is needed to improve assessment methodologies – e.g. biota-specific transfer information?

## INTERNATIONAL ORGANIZING COMMITTEE

Alexakhin, R.M.	Russian Institute of Agricultural Radiology and Agroecology / International Commission on Radiological Protection, Russian Federation
Burns, P. ( <i>Treasurer</i> )	Australian Radiation Protection and Nuclear Safety Agency, Australia
Carvalho, F.	Radiological Protection and Nuclear Safety Department, Instituto Tecnológico e Nuclear, Portugal
Domotor, S.	United States Department of Energy, United States of America
Janssens, A.	European Commission, Luxembourg
Larsson, C.-M.	Swedish Radiation Protection Institute, Sweden
Mothersill, C.	Radiation and Environmental Science Centre / Dublin Institute of Technology, Ireland
Robinson, C.A.	International Atomic Energy Agency
Strand, P.	Norwegian Radiation Protection Authority, Norway
Thompson, P.	Canadian Nuclear Safety Commission, Canada
Woodhead, D.	Centre for Environment, Fisheries and Aquaculture Science, Lowestoft Laboratory, United Kingdom
Zapantis, A. ( <i>Chairman</i> )	Supervising Scientist Division, Environment Australia, Australia

## DOMESTIC ORGANIZING COMMITTEE

Burns, P. ( <i>Treasurer</i> )	Australian Radiation Protection and Nuclear Safety Agency, Australia
Colgan, P.	Australian Radiation Protection and Nuclear Safety Agency, Australia
Devine, S. ( <i>Secretariat</i> )	Supervising Scientist Division, Environment Australia, Australia
Dooley, B.	Australian Radiation Protection and Nuclear Safety Agency, Australia
Higgins, L.	Australian Radiation Protection and Nuclear Safety Agency, Australia
Partridge, M.	Australian Radiation Protection and Nuclear Safety Agency, Australia
Waggitt, P.	Supervising Scientist Division, Environment Australia, Australia
Williams, G.	Australian Radiation Protection and Nuclear Safety Agency, Australia
Zapantis, A. ( <i>Chairman</i> )	Supervising Scientist Division, Environment Australia, Australia

## LIST OF PARTICIPANTS

- Adam, C. Institut de Radioprotection et de Sûreté Nucléaire (IRSN)  
Centre de Cadarache, B.P. 3  
F-131115 Saint Paul-lez-Durance, Cedex, France
- Andreuccetti, F. CISAM  
Via Bicatti, San Piero A Grado, 5610 Pisa, Italy
- Antonio, E. Pacific Northwest National Laboratory  
P.O.Box 999, 902 Battelle Boulevard  
WA 99352 Richland, United States of America
- Barescut, J.-C. Institut de Radioprotection et de Sûreté Nucléaire (IRSN)  
Rue Auguste Lemaire, B.P. 17  
F-92262 Fontenay-aux-Roses, Cedex, France
- Benedetti, A. CISAM  
Via Bicatti, San Piero A Grado, 5610 Pisa, Italy
- Brechignac, F. Institut de Radioprotection et de Sûreté Nucléaire (IRSN)  
Centre de Cadarache, B.P. 3  
F-131115 Saint Paul-lez-Durance, Cedex, France
- Burns, P. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
619 Lower Plenty Road  
3085 Yallambie, Victoria, Australia
- Calmet, D. Commissariat à l'Energie Atomique (CEA)  
Direction Matières, Surveillance, Environnement (DMSE)  
Centre de Bruyères le Chatel, B.P. 12  
F-91680 Bruyeres le Chatel, France
- Carroll, S. Greenpeace International, Political & Science Division  
Keizersgracht 176, NL-1016 DW, Amsterdam, Netherlands
- Chambers, D. SENES Consultants Limited  
121 Granton Drive, Unit 12  
Richmond Hill L4B 3N4, Ontario, Canada
- Colgan, P. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
P.O. Box 655, 2228 Miranda NSW, Australia
- Crawford, J. Australian Nuclear Science & Technology Organisation (ANSTO)  
PMB 1, NSW 2234 Menai, Australia
- Dawson, F. Directorate of Safety, Environment & Fire Policy, Ministry of Defence,  
St. Giles Court, 1-13 St. Giles Street  
London WC2H 8JD, United Kingdom
- Denison, F. Institut de Radioprotection et de Sûreté Nucléaire (IRSN)  
Centre de Cadarache, B.P. 3  
F-131115 Saint Paul-lez-Durance, Cedex, France
- Dicus, G. U.S. Nuclear Regulatory Commission (USNRC)  
11555 Rockville Pike  
MD 20852-2738 Rockville-Maryland, United States of America

Domel, R. Australian Nuclear Science & Technology Organisation (ANSTO)  
PMB 1, NSW 2234 Menai, Australia

Domotor, S. U.S. Department of Energy (USDOE)  
Headquarters Forrestal Building, 1000 Independence Avenue, S.W.  
20585-0001 Washington, D.C., United States of America

Fairchild, R. Nabrasca Wesleyan University, Department of Physics  
68504-2794 Lincoln, Nebraska, United States of America

Ferris, J. Australian Nuclear Science & Technology Organisation (ANSTO)  
PMB 1, NSW 2234 Menai, Australia

Fredericksen, J. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
P.O. Box 655, 2228 Miranda NSW, Australia

Fujimoto, K. National Institute of Radiological Sciences (NIRS)  
Environmental Radiation Protection Research Group  
9-1, Anagawa 4-chome, Inage-ku, 263-8555 Chiba-shi, Japan

Gariel, J.-C. Institut de Radioprotection et de Sûreté Nucléaire (IRSN)  
Centre de Cadarache, B.P. 3  
F-131115 Saint Paul-lez-Durance, Cedex, France

Garnier-Laplace, J. Institut de Radioprotection et de Sûreté Nucléaire  
Centre de Cadarache, B.P. 3  
F-131115 Saint Paul-lez-Durance, Cedex, France

Glaubitz, U. Sachisishces Landesamt für Umwelt und Geologie  
Zur Wetterwarte 11, D-01109 Dresden, Germany

González, A.J. International Atomic Energy Agency, Vienna

Gonze, M.-A. Institut de Radioprotection et de Sûreté Nucléaire (IRSN)  
Centre de Cadarache, B.P. 3  
F-131115 Saint Paul-lez-Durance, Cedex, France

Graham, R. U.S. Environmental Protection Agency (EPA)  
999 18th Street, 80202 Denver  
Colorado, United States of America

Gudkov, D. Institute of Hydrobiology, Department of Radioecology  
Geroyev Stalingrada Ave. 12  
UA-04210 Kiev, Ukraine

Hardege, L. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
619 Lower Plenty Road, 3085 Yallambie, Victoria, Australia

Harris, T. U.S. Nuclear Regulatory Commission (USNRC), Mail Stop T-8 F5  
DC 20555-0001 Washington, D.C., United States of America

Hingston, J. Westlakes Scientific Consulting Limited  
The Princess Royal Building, Westlakes Science & Technology Park  
Moor Row, Whitehaven, Cumbria CA24 3LN, United Kingdom

Hinton, T. University of Georgia, Savannah River Ecology Laboratory  
Drawer E, SC 29802 Aiken, United States of America

Holland, B. Australian Nuclear Science & Technology Organisation (ANSTO)  
PMB 1, NSW 2234 Menai, Australia

Holm, L.-E., Swedish Radiation Protection Authority (SSI)  
SE-171 16 Stockholm, Sweden

Hughes, A. Department of Business, Industry & Resource Development (DBIRD)  
GPO Box 3000, NT 0801 Darwin, Australia

Iosjpe, M. Norwegian Radiation Protection Authority (Statens Strålevern)  
Environmental Protection Department  
Grini Naeringspark 13, P.O. Box 55, N-1332 Østerås, Norway

Jackson, D. Enviros Limited  
Brierbank, Wellington, Gosforth, Cumbria CA20 1BL, United Kingdom

Janssens, A. Directorate C - Environment & Health, ENV.C4 - Radiation Protection  
European Commission (EC), Bâtiment Wagner, Rue Alcide de Gasperi  
Plateau de Kirchberg, L-2920 Luxembourg

Johnston, A. Environment Australia, Office of the Supervising Scientist  
GPO Box 461, NT 0801 Darwin, Australia

Johnston, A. Environment Protection Authority (EPA), Radiation Protection Branch  
GPO Box 2607, SA 5001 Adelaide, Australia

Johnston, P. Royal Melbourne Institute of Technology, Department of Applied Physics  
GPO Box 2476V, 3001 Melbourne, Victoria, Australia

Jones, B. U.S. Nuclear Regulatory Commission (USNRC)  
11555 Rockville Pike  
MD 20852-2738 Rockville-Maryland, United States of America

Jones, S. Westlakes Scientific Consulting Limited  
The Princess Royal Building, Westlakes Science & Technology Park  
Moor Row, Whitehaven, Cumbria CA24 3LN, United Kingdom

Kautsky, U. Swedish Nuclear Fuel & Waste Management Company (SKB)  
P.O. Box 5864, S-102 40 Stockholm, Sweden

Kim, J.K. Korea Atomic Energy Research Institute (KAERI)  
P.O. Box 105, 150 Dukjin-dong, Yusong  
305-600 Taejon, Republic of Korea

Koblinger, L. Hungarian Atomic Energy Authority  
Fenyés Adolf u. 4, P.O. Box 676, H-1539 Budapest, Hungary

Koperski, G. Northern Territory Department of Health & Community Services  
PO Box 40596, NT 0811 Casuarina, Australia

Koteng, A. Radiation Protection Board, Ministry of Health  
22nd Floor, Western Wing, Block A, NSSDF Building, Bishops Road  
P.O. Box 19841, Nairobi 00100, Kenya

Kumblad, L. Stockholm University, Department of Systems Ecology  
S-106 91 Stockholm, Sweden

Lambotte, J. Agence Fédérale de Contrôle Nucléaire (AFCN)  
Rue Ravenstein, 36, 1000 Bruxelles, Belgium

Larsson, C.-M. Swedish Radiation Protection Authority (SSI)  
Department of Waste Management & Environmental Protection  
SE-171 16 Stockholm, Sweden

Loy, J. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
P.O. Box 655, 2228 Miranda NSW, Australia

Lyng, F. Dublin Institute of Technology  
Radiation & Environmental Science Centre  
Kevin Street, Dublin 8, Ireland

Lyons, R. University of Auckland, Department of Physics  
Private Bag, Auckland, New Zealand

Marshman, I. Energy Resources of Australia, Ranger Uranium Mine  
Locked Bag 1, NT 0866 Jabiru, Australia

Mbambo, Z. National Nuclear Regulator (NNR)  
P.O. Box 7106, ZA-0046 Centurion, South Africa

McGill, T. Department of Business, Industry & Resource Development (DBIRD)  
GPO Box 3000, NT 0801 Darwin, Australia

Metzke, R. TÜV Süddeutschland Bau und Betrieb GmbH  
Westendstrasse 199, D-80686 Munich, Germany

Morris, R. TREC Incorporated  
101 South Park Avenue, Suite 5, Idaho Falls, United States of America

Mothersill, C. Dublin Institute of Technology  
Radiation & Environmental Science Centre  
Kevin Street, Dublin 8, Ireland

Mudd, G. Environment Center of the Northern Territory  
P.O. Box 245, Jabiru, NT, Australia

Olwell, P. Dublin Institute of Technology  
Radiation & Environmental Science Centre  
Kevin Street, Dublin 8, Ireland

Oughton, D. Agricultural University of Norway  
Department of Chemistry & Biotechnology  
P.O. Box 5028, N-1432 Aas, Norway

Pentreath, R.J. University of Reading, Environmental Systems Science Centre  
Harry Pitt Building, Whiteknights, PO Box 238  
Reading, Berkshire RG6 6AL, United Kingdom

Poston, T. Pacific Northwest National Laboratory  
P.O.Box 999, 902 Battelle Boulevard  
WA 99352 Richland, United States of America

Prosser, S. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
P.O. Box 655, 2228 Miranda NSW, Australia

Ravi, P.M. Bhabha Atomic Research Centre (BARC)  
Environmental Survey Laboratory, KGS, Health Physics Division  
Kaiga Generating Station, Kaiga, Karwar, Utta Kannada District  
581400 Karnataka, India

Robinson, C. International Atomic Energy Agency, Vienna

Rogers, B. New South Wales Environmental Protection Authority  
PO Box A290, NSW 1232 Sydney South, Australia

Saint-Pierre, S. COGEMA  
2, rue Paul Dautier, B.P. 4  
F-78141 Velizy-Villacoublay, Cedex, France

Sendin, P. Consejo de Seguridad Nuclear (CSN)  
Justo Dorado 11, E-28040 Madrid, Spain

Sdraulig, S. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
619 Lower Plenty Road, 3085 Yallambie, Victoria, Australia

Seymour, C. St. Luke's Institute of Cancer Research, Radiation Research Laboratory  
Rathgar, Dublin 6, Ireland

Shields, B. Department of Health and Human Services (DHHS)  
7000 Hobart, Tasmania, Australia

Shpyth, A. CAMECO Corporation, Regulatory & Environmental Affairs  
130 Albert Street, Suite 1610, Ottawa K1P 5G4, Ontario, Canada

Smith, C. Westlakes Scientific Consulting Limited  
The Princess Royal Building, Westlakes Science & Technology Park  
Moor Row, Whitehaven, Cumbria CA24 3LN, United Kingdom

Stark, K. Swedish Radiation Protection Authority (SSI)  
SE-171 16 Stockholm, Sweden

Sugiura, N. The University of Tokyo  
Research Center for Nuclear Science and Technology  
7-3-1 Hongo (Center: Yayoi 2-11-16), Bunkyo-ku  
113-0032 Tokyo, Japan

Thompson, P. Canadian Nuclear Safety Commission (CNSC)  
Environmental Protection & Audit Division  
280 Slater Street, P.O. Box 1046, Station "B"  
Ottawa K1P 5S9, Ontario, Canada

Tsela, A. National Nuclear Regulator (NNR)  
P.O. Box 7106, ZA-0046 Centurion, South Africa

VanHorn, R. Idaho National Engineering & Environmental Laboratory (INEEL)  
2525 Fremont Avenue, P.O. Box 1625  
ID 83415-3860 Idaho Falls, United States of America

Waggitt, P. Office of the Supervising Scientist, Environment Australia  
P. O. Box 461, NT 0801 Darwin, Australia

Wallberg, P. Swedish Radiation Protection Authority (SSI)  
SE-171 16 Stockholm, Sweden

Whicker, F.W. Colorado State University, Department of Radiological Health Sciences  
CO 80523 Fort Collins, United States of America

Woodhead, D. Centre for Environment, Fisheries & Aquaculture Science (CEFAS)  
Lowestoft Laboratory, Pakefield Road  
Lowestoft, Suffolk NR33 0HT, United Kingdom

Woollett, S. Australian Radiation Protection & Nuclear Safety Agency (ARPANSA)  
619 Lower Plenty Road, 3085 Yallambie, Victoria, Australia



Wright, B. U.S. Nuclear Regulatory Commission (USNRC)  
Mail Stop T-8 F5 (Australian Desk Office)  
DC 20555-0001 Washington, D.C., United States of America

Yankovich, T. Atomic Energy of Canada Limited (AECL)  
Environmental Technologies Branch, Building 513  
Station 51A, Chalk River Laboratories  
Chalk River K0J 1J0, Ontario, Canada

Yu, C. Argonne National Laboratory  
Building 900, 9700 South Cass Avenue  
IL 60439 Argonne, United States of America

Zapantis, A. Environment Australia, Supervising Scientist Division  
Office of the Supervising Scientist  
GPO Box 461, NT 0801 Darwin, Australia

Zinger-Gize, I. The Environment Agency  
P.O. Box 12, Richard Fairclough House, Knutsford Road  
Warrington, Cheshire WA4 1HG, United Kingdom