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ABSTRACTS

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## SOME ASPECTS OF THE ENVIRONMENTAL POLICY IN RUSSIA

A.V. Yablokov

*The Russian Federation Security Council*

In the early days of Gorbachev's perestroika (1984-1985), the main problems of environmental protection were probably departmentalization, secrecy, and the ecological illiteracy of officials making decisions. This, in turn, resulted from the totalitarian character of our society and its excessive militarization. About 70-75% of all industry was military oriented. Even in Moscow, more than 50% of all enterprises were related to MICs (military-industrial complexes), in St. Petersburg (Leningrad) this figure exceeded 75%, etc.

Today's situation in environmental protection is marked by:

- disastrous cuts in the state budget allocations aimed at solving the problems of environmental conservation and protecting the population from adverse environmental factors;
- disruption of federal management in this field (after the law "On local self-government" was adopted in 1991);
- unprecedented plundering of natural resources;
- persistent atmospheric pollution;
- the almost universal deterioration of fresh water quality and increasing contamination of water reservoirs;
- spreading of diseases of an ecological nature; and
- the historically unprecedented reduction in the average life expectancy (also showing a strong ecological influence).

I will attempt to outline several principal problems in the area of Russia's environmental protection policy.

The first and foremost objective, in my opinion, is to conclude the restructuring of the former Soviet society. The point is that half of the employable population of the totalitarian Soviet state were industrial workers. For comparison: in Western countries no more than 18-19% of the population are occupied in industry. It so happened in the USSR because 70-75% of our industry was militarized, while the services sector comprised only 18% of the population. Today, the primary objective is to most promptly accomplish, as painlessly as possible, the reshaping of the public structure. This process will be accompanied by a drastic reduction in the consumption of natural resources, materials and energy, as well as a similar reduction in environmental pollution. Even today, as many large-scale military enterprises are closing down, the total atmospheric emission by stationary sources in Russia has notably reduced.

The resources and energy supply in Russia are also problems of great concern. Today, the specific consumption of materials to our national income is 3-4 times higher, and the power intensity is 2-3 times higher as compared to those of Japan and Germany. A new law is currently being prepared, which will hopefully render unprofitable today's reckless spending of natural resources. In this process, account should be taken of both the positive and negative experiences gained in the sphere of governmental and economic regulation of environmental use in Western countries.

Among the proper environmental protection objectives of the transition period, the first one, to my mind, is to preserve all of the positive accomplishments accumulated in the course of Russia's difficult history in the 20th Century. The second is to make a complete inventory of the ecological consequences of our recent past: to determine the location, amount and type of radioactive, chemical and other hazardous wastes buried or concealed in the waters of this country.

To conclude, Russia's policy in the sphere of environmental conservation should comprise the following tasks: a) stopping the environmental deterioration and plundering of natural resources, and b) transition to a sustainable development (in the sense of the 1992 UNO Conference in Rio de Janeiro). This last problem may eventually be more easily solved than in most Western countries. Today we are at the cross-roads and it is our concern that we choose the proper direction.

# NUCLEAR ANALYTICAL TECHNIQUES IN ENVIRONMENTAL STUDIES

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Within the wide variety of analytical methods available for environmental studies various analytical methods are based on nuclear physical principles. A survey is given of the various categories of nuclear analytical methods, the associated characteristic features, and the information obtainable from these techniques.

Main characteristic features are: physically independent basis of the analytical method, isotopic rather than elemental determination, almost no interfering effect of electrons and molecular structure, and penetrating character of nuclear radiation.

Nuclear analytical techniques are facing today various constraints. There is a growing competition with other, non-nuclear, techniques. Further, governmental funds for nuclear research and thus also for application of nuclear methods are generally decreasing, while public concern about nuclear facilities and radioactivity is increasing. Nevertheless, nuclear analytical techniques remain potential techniques, also in the field of environmental studies.

In the frame of this situation suggestions are given for a full exploitation of the potentials of nuclear analytical methods, particularly when requiring considerable investment for equipment, supporting facilities, and specialized staff. In this survey emphasis is given to nuclear analytical techniques dealing with radioactivity, e.g. radiotracer techniques, activation analysis, radioisotope dilution and related techniques.



# STRATEGY AND POLICY OF UNIVERSITY RESEARCH REACTORS

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Since the 1970's, an appreciable number of research reactors at universities as well as national institutes has been closed. If this trend is further continued, it will become detrimental for maintaining the scientific competence in reactor associated fields of sciences and the further development and application of reactor-based techniques such as neutron activation analysis for protection of the environment.

Therefore, the facilities still in operation have to develop strategies aiming at assuring their activities and at further development of reactor and associated experimental facilities. Key aspects of such strategies will be discussed, using the developments observed at a number of reactors and in particular at the Interfacultair Reactor Instituut in Delft and its reactor, as an example.

Such key aspects are:

- well defined, accepted and realistic medium- and long-term policy and program;
- clear internal and external organizational structure with clear authorities and responsibilities;
- effective representation of the "outside world" at policy level;
- predictability of subsequent annual budgets;
- a program of carefully selected investments in reactor and experimental facilities;
- careful personnel management with particular attention for the age distribution;

- early planning of licensing procedures, acquisition of fresh fuel and disposal of spent fuel;
- attention for public relations.

Paying attention to these aspects and taking the necessary actions in time may not assure the future of a research reactor, but it will certainly increase the chances for a healthy development. Some examples will be presented to illustrate the potential role of large- and small-sample neutron activation analysis in solving environmental problems.

## THE FUTURE OF NAA IN THE ENVIRONMENTAL SCIENCES

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Public attitudes and governmental policies worldwide with respect to the nuclear industry have resulted in a decrease in the availability of nuclear reactors and their associated analytical facilities. The unique advantages provided by neutron activation analysis must be promoted; there is little point in illustrating their potential for analyses which can more readily undertaken, just as accurately and by far less expensive alternative methods. It is desirable to conserve the expertise which is available albeit the number of facilities will be reduced, perhaps even to the level of one centre of expertise for a country. Scientific papers need to illustrate the merits of the methods in solving defined problems, rather than just providing catalogues of analytical data. While a large number of elements can be determined by activation analysis many are of little interest in environmental studies; often data are lacking for more important minor and major elements.

# NUCLEAR AND ALTERNATIVE ANALYTICAL TECHNIQUES

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Neutron activation analysis (NAA) is a powerful technique for monitoring trace contaminants in the environment, particularly when radiochemical separation procedures are applied to improve the limits of detection. The developments in newer techniques for trace inorganic contaminants, such as inductively coupled plasma-atomic emission spectrometry (ICP-AES) and inductively coupled plasma-mass spectrometry (ICP-MS), are complementary to NAA and the three methods can be used to measure a comprehensive range of trace elements and sample types, over a broad range of concentrations. At CARE the ICP techniques are used to detect the elements which are difficult by NAA, to improve the sensitivity for other elements and to analyse liquid samples containing very low concentrations of the elements of interest. Conversely, NAA is used to analyse materials which are difficult to dissolve and volatile elements which are lost on dissolution. This paper describes environmental applications of NAA and ICP techniques, in particular for a range of sample types, such as air particulates, vegetation, sediments, food and water.

# APPLICABILITY OF THE POISSON DISTRIBUTION FOR QUALITY ASSURANCE IN ENVIRONMENTAL MEASUREMENTS BY NEUTRON ACTIVATION ANALYSIS

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The use of nuclear techniques in the study of environmental problems has many advantages, such as high sensitivity to many elements, low interference, and freedom from matrix effects. Furthermore, the final measurement is based on the process of counting radioactive decays, which are known to obey the Poisson distribution with identical mean and variance. In the absence of other significant sources of variability, the uncertainty of a measurement can therefore be predicted accurately from the so-called counting statistics.

In neutron activation analysis it has been found that excellent agreement between the predicted and actual variability of analytical results can be achieved under practical conditions, and this forms the basis for a powerful tool in the quality assurance of environmental measurements. Careful validation of the computer programs used for processing counting data, however is a prerequisite for achieving statistical control, and a highly reproducible counting geometry is essential. Special emphasis must be put on the reproducibility of the neutron irradiation conditions that is so often neglected in the evaluation of uncertainties.

Examples are presented to demonstrate the feasibility of utilizing such statistical control for the determination of trace elements in a series of environmental reference materials from the European Measurement and Testing Programme.

# QUALITY MANAGEMENT AT THE ANALYTICAL LABORATORY

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Monitoring programs of regional, national or international scope may involve measurements made by several analysts and/or laboratories which need to be interrelated for use in a decision process. Regulatory governmental bodies sometimes sub-contract analyses in the framework of law-enforcement to (private) testing laboratories and universities. Several countries and states have defined their own standards with respect to the quality or composition of products to be imported. All of these developments involve stringent requirements for the reliability and compatibility of data. Often, each laboratory introduced its own procedures for quality assurance. Establishing and maintaining a quality assurance program is a management task, and implies more than just analyzing quality control samples : careful documentation, sample control, traceability of calibration parameters but also of experimental conditions, non-conformance management etc.

Internationally accepted reference documents -the ISO guides- have been developed with criteria to evaluate and to verify the laboratory's competence to perform a given task, to establish its analytical capabilities and its system for quality assurance. Such an evaluation can lead to an accreditation for the work that is doing.

Quality management has been introduced at the laboratory for Instrumental Neutron Activation Analysis of the Interfaculty Reactor Institute at the Delft University of Technology. The laboratory has received accreditation for its quality system for compliance with the accreditation criteria of EN45001 closely following the ISO-25 guide. The laboratory's motivations for introducing quality management vary from the need for a systematic system of documentation and prevention of repetition of work, to thorough traceability of experimental conditions, enhancement of respect for and trustworthiness of the laboratory and improved chances for acquiring third party contracts. The route to accreditation will be briefly described. The experience with the quality system will be illustrated with examples of the consequences for research and routine operations, non-conformance management and analytical quality.

# MONITORING AIR POLLUTION BY NUCLEAR AND RELATED ANALYTICAL TECHNIQUES: A REVIEW OF CURRENT IAEA PROGRAMMES

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The use of nuclear and related analytical techniques has become widespread in major areas of science and technology. In the environmental area, there is a growing interest in many of the IAEA's member states in the monitoring and control of airborne pollutants arising from industrial processes and from the burning of fossil fuels. Nuclear analytical techniques have important applications in such work since they can be used not only for the determination of specific individual pollutants (e.g. toxic heavy metals) but also, via multielement analyses and chemometric evaluation, for source identification and apportionment purposes.

Work of this kind has recently become a major focus of the IAEA's programmes. It involves co-ordinated research, technical co-operation, analytical quality control and laboratory services. Emphasis is being placed on the use of a standard design of air sampler for collection of airborne particulate matter (a low volume air sampler comprising stacked filter units capable of providing separation of airborne particulate matter into two size ranges below 10 microns). The more widespread use of biomonitors of air pollution (mainly lichens) is also being promoted, as is the development and certification of appropriate analytical reference materials.

This presentation reviews the current status of nuclear analytical techniques in the IAEA's programmes on air pollution, with emphasis on applications of neutron activation analysis (NAA), energy dispersive X-ray fluorescence (ED-XRF) and proton-induced X-ray emission (PIXE).

# THE DETECTION OF ALPHA-PARTICLES USING PHOTO-DIODES, SSNTD AND PROPORTIONAL COUNTER FOR THE PURPOSE OF ENVIRONMENTAL INVESTIGATIONS

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For the past 10 years, the Universite de Franche-Comte has been develop-

ing complementary techniques to analyse alpha-particles particularly in the detection of radon and its decay products. The two main techniques - proportional counter and solid state nuclear track detectors - have been described in various papers. A methodology has also been developed to analyze the radon concentration in large and varied areas. This analysis is effected in time or in space. The analysis in time uses the proportional counter in just a few points because of its high cost. The latter analysis uses the less expensive SSNTD in many points. Thus, the measurements reveal the distribution of radon emanation and the range of variation. The method is used routinely for the following purposes: in homes in the aim of radioprotection, in geological applications for prospecting, studying the movement of the earth crust and analyzing the effect of alpha pollution in the environment.

Regarding the latter application, we are, at the moment, developing a portable alpha spectrometry using a silicon diode. A portable electronic device, coupled with a computer, is made to analyze the alpha radio-elements in the atmosphere collected on the surface of a filter through which a measured quantity of air has already passed. Thus, the spectrometry of the filter reveals the concentration of alpha emitters in the environment as well as the equilibrium factor for radon and the alpha potential energy. Filter measurements are performed over the time of the disintegration of the radon daughters ( $^{218}\text{Po}$  and  $^{214}\text{Po}$ ) using the Thomas process or an indetical method. This new method, associated with the one already presented, gives us quite a fair knowledge of the state of alpha pollution in the environment.



## STRATEGIES FOR SAMPLING AND SAMPLE HANDLING IN LARGE SCALE BIOMONITORING PROJECTS

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At the Interfaculty Reactor Institute (IRI), Instrumental Neutron Activation Analysis (INAA) has been used routinely for trace element determinations in a large variety of plant species, among which grasses, tomato plants, soy beans, spinach, pea, and various plant reference materials. A 15 years' IRI experience with INAA of lichens, mosses and tree bark in the frame of biomonitoring surveys forms the basis for a discussion of a number of analytical aspects of sampling, sample preparation and pre-treatment, and needs for standardized methods in plant monitoring.

Although not always recognized, conditions and precautions in "trivial" steps like interspecies calibration, washing and cleaning, homogenization, sample size reduction, and analytical uncertainties all have to be related to the measured degree of local variation per sampling site. Here, key problems involve the effects of leaching during washing, disturbance of equilibria, or the lack of sample material for homogenization tests. Selecting INAA as analysis technique, problems related to incomplete digestion, or losses by filtering need not be considered. Some of the problems may be overcome by analysis of large samples (kilogram size), and, in surveys, by the application of factor analysis techniques.

# ON ESTIMATING THE VALUE OF A REPRESENTATIVE PROBE USING NAA

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Previously, when discussing the material submitted for publication in the Journal of Analytical Chemistry, referees doubted the correctness of some calculations in deriving a formula for estimating the value of a representative probe,  $q_{rep.}$ , from NAA experimental estimates on relative standard deviations in homogeneity ( $s_{r,h.,i}$ ) and reproducibility ( $s_{r,b.,i}$ ) for a weighed portion,  $q_i$  :

$$q_{rep.} \geq q_i \sqrt[n/2+1]{(3s_{r,h.,i}/s_{r,b.,i})^n \cdot (1/F_{p,i})} \quad (1)$$

B (1)  $n$  is  $\leq 2$ , and  $F_{p,i}$  is the Fisher criterion for the confidence probability  $P$  and the degrees of freedom,  $f_1 = m - 1$  and  $f_2 = \infty$  ( $m$  is the number of probes used in the NAA determination of  $s_{r,h.,i}$  and  $s_{r,b.,i}$ ).

These circumstances have inspired the author to give a more detailed consideration to some stages in deriving formula (1) and determining the limits of its application.

# VERY ACCURATE METHODS FOR THE DETERMINATION OF SELECTED ELEMENTS IN BIOLOGICAL MATERIALS BY NEUTRON ACTIVATION ANALYSIS AND COLUMN CHROMATOGRAPHY

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Determination of very small amounts of some trace elements still poses problems as evidenced by the results of numerous interlaboratory comparisons in which differences by orders of magnitude occur quite frequently. So, there is a need for very accurate methods, which being more laborious and time-consuming than routine methods, would however, guarantee more accurate results. In this paper the principles set forth in our Laboratory for devising such methods based on a combination of neutron activation analysis with selective and quantitative post-irradiation isolation of the radionuclides in question by column chromatography, followed by interference-type measurement by  $\gamma$ -ray spectrometry, are described. The methods are constructed, in principle, as single element methods in order to optimize all conditions for achieving the best detection limits and maximum accuracy and precision. The recently elaborated methods for the determination of trace amounts of copper [1], cobalt [2], cadmium [3], and molybdenum and uranium [4] in biological materials are briefly discussed.

The significance of such methods for the certification of candidate reference materials and possibly also for the verification of some "information" or "consensus" data is pointed out.

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## NEW DEVELOPMENTS IN NIST ENVIRONMENTAL STANDARD REFERENCE MATERIALS

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Well characterized, certified reference materials (CRMs) are required for quality assurance purposes when analyzing environmental samples. To meet this need, the National Institute of Standards and Technology issues a wide variety of environmental Standard Reference Materials (SRMs) which are certified for elemental content and can be used for such purposes. Neutron activation analysis (NAA) often plays an important role in the certification of inorganic constituents in many environmental SRMs in view of its accuracy, multielemental capability, ability to assess homogeneity, high sensitivity for many elements, and essentially blank-free nature. In addition, INAA requires no chemical dissolution prior to analysis, unlike most other analytical techniques. Since the concentration of an element certified in a NIST SRM is usually determined by at least two independent analytical methods, the use of INAA as one method eliminates the possibility of a common error sources resulting from sample dissolution. Over the last several years NIST has issued new or renewal SRMs in such environmental matrices as Coal Fly Ash, Estuarine Sediment, Soil, and Domestic Sludge. This presentation will describe the role that NAA played in the certification of these materials, as well as describe the advantages of these new SRMs.

## ENVIRONMENTAL POLLUTION STUDIES AT THE BUCHAREST U-120 CYCLOTRON

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To determine the concentration of impurities (especially metal) in liquid and solid samples related to environmental pollution control, in-air PIXE analysis is very advantageous. We have used 3-4 MeV proton beams obtained from 6.5 MeV Cyclotron's nominal regime. Protons, extracted through a pressure-air cooled 100  $\mu\text{m}$  aluminum foil into the air, strike the sample 8-10 cm away. To avoid the strong straggling effects, large sized samples were used (50-80 mm diameter), X-rays are detected through reflection using a horizontal Si(Li) detector with a 4.3 mm diameter active area (1 mm plexiglass absorber). Maximum sensitivities have been obtained in the  $22 \leq z \leq 30$  region (down to 1 ppm for Fe and Cu at  $q = 40\text{nA} \times 600\text{s} = 24\mu\text{C}$ ). Obtained spectra for various water and soil samples are also presented.

Some results, related to different aspects of environmental radioactivity are reported. A synthesis of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  post-Chernobyl measurements of foodstuffs and human subjects is presented. Natural radioactivity (U, Th,  $^{40}\text{K}$ ) level determination in various samples (phosphates, gold and copper ores), including the use of a Phoswich NaI(Tl)-CsI(Tl) Lung Counter, are also summarized.

# PEAK-TO-TOTAL RATIO FOR VOLUMINOUS SOURCES: DETECTOR CALIBRATION AND COMPUTATION OF TRUE COINCIDENCE CORRECTIONS

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Determination of the peak-to-total (PT) ratio is necessary for a correct estimation of absolute radioactivity of a source in the presence of the true coincidence effect for its analytical gamma-lines in the case of experimental geometry close to detector .

A detailed study of different factors affecting PT-calibration has been carried out. The computer program for PT-calibration creates the calibration curve using gamma-lines of the point sources free of coincidence ( $^{241}\text{Am}$ ,  $^{57}\text{Co}$ ,  $^{139}\text{Ce}$ ,  $^{113}\text{Sn}$ ,  $^{137}\text{Cs}$ ,  $^{54}\text{Mn}$ ,  $^{65}\text{Zn}$ ).

In the case of voluminous sources a special procedure has been developed. It regards the source as consisting of a set of smaller subsamples with the final summation of the weighted coincidence coefficients for each subsample.

# NEUTRON ACTIVATION ANALYSIS FOR ENVIRONMENTAL RESEARCH AT THE LATVIAN NUCLEAR REACTOR

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During 30 years in the Nuclear Research Centre, Laboratory of Neutron activation analysis has developed and used a number of neutron activation analysis (NAA) techniques (including a channels with pneumatic system for short time irradiation).

Instrumental methods of NAA have been elaborated at the nuclear reactor, Salaspils, Latvia, to analyse the environmental objects and industrial wastes. Activation was carried out by use of thermal and resonance neutrons. More than 30 elements were established (including toxic elements - As, Se, V, Cd, Hg) on the basis of short-, middle- and long-lived radionuclides. These methods were applied for such environmental objects: forest litter and needles, atmospheric aerosols and precipitations, children food (milk, eggs, grains, vegetables), natural and waste waters, sediments, soils and also biological samples. To found correlation between pollution and condition of forest ecosystems the content of macro and micro elements in forest litter was determined. Thirty elements have been established in wide range of concentration ( $1 \cdot 10^{-3}$  -  $1 \cdot 10^4$  mg/kg) in the ashed samples. The samples were taken from 300 places throughout Latvia.

Instrumental NAA was used to analyze the drinking water of Riga. Preliminary the water samples were evaporated. From 21 different places of Riga, from the nearest surroundings and from the water supply places, water samples were analyzed. In the drinking water 26 chemical elements in wide range of concentrations were established. The chemical composition of water between different sources varied considerably. The greatest differences were established for zinc and aluminium: the minimal concentrations of zinc were  $1.0 \mu\text{g/l}$  and  $590 \mu\text{g/l}$  corresponding. The concentration of elements in the



drinking water, with a few exceptions, is near but not exceeding the Sweden normative.

Preconcentration of the elements under determination is, as a rule, applied, in some cases it is preconditioned by deficient sensitivity of the direct instrumental NAA methods itself, as well as by a strong interfering effect of sodium, chlorine, bromine.

An effective method for metal concentration is precipitation of metals by organic co-precipitants. Our studies have shown disulphides to be efficient co-precipitants. In the present work 8-mercaptoquinoline (thiooxine) is suggested as a chelating agent and its oxidation product 8,8,- diquinolyldisulphide - as a coprecipitant. In our experimental conditions element detections limits are g:

Cu -  $0.4 \cdot 10^{-6}$ , Mn -  $2.1 \cdot 10^{-8}$ , V -  $4.1 \cdot 10^{-9}$ , Fe -  $1.1 \cdot 10^{-6}$ , Co -  $1.1 \cdot 10^{-9}$ , Zn -  $1.1 \cdot 10^{-7}$ , Ag -  $5.1 \cdot 10^{-9}$ , Au -  $5.1 \cdot 10^{-11}$ , Sb -  $1.1 \cdot 10^{-9}$ , Hg -  $4.1 \cdot 10^{-9}$ . Series of analysis of 27 chemical elements contained in aerosol filters exposed in biosphere reserves and in clean areas such as Antarctic were performed using instrumental NAA. These results make possible to investigate the process of transference of aerosols and estimate their sources, involving anthropogenic.

We have worked out sensitive methods of NAA of Se, Hg, Sc, Cr, Co, Fe, Zn, Sb in human hair and blood. Analysis were carried out for clinical investigation among the population of Latvia. INAA is one of the express-type analytical methods due to application of short-lived radionuclides. We can establish such elements as Mg, Al, V, Ti, Cu, Mn, Se, F, Cl, J with detections limits of  $10^{-5}$  -  $10^{-7}$  %.

The staff of our laboratory has successfully participated in standard intercalibration organized by International Atomic Energy Agency

**SECONDARY ION MASS SPECTROMETRY : A METHOD  
FOR RAPID DETECTION OF LONG LIVED ISOTOPES IN  
BIOLOGICAL TISSUES AND ENVIRONMENTAL  
POLLUTION**

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Secondary ion Mass Spectrometry, a method proposed and developed in 1960 by R.Castaing and G.Slodzian, allows a rapid detection of stable or radioactive nuclides and an accurate measurement of isotopic ratios. As applied to radiation protection and dosimetry this method can be used either as a pure mass spectrometer, or as a ion microscope. By mass spectrometry of thin foils, the detection of radionuclides and the measurement of isotopic ratios can be obtained more rapidly and with a better sensitivity than with alpha or beta spectrometry, particularly in the case of long lived emitters. Using the instrument in the imaging mode for the study of tissue sections, the intracellular distribution of radionuclides allows useful data to be obtained for microdosimetric studies.

# PRECONCENTRATION OF TRACE ELEMENTS IN DRINKING WATER WITH TAN BY REVERSED-PHASE EXTRACTION CHROMATOGRAPHY AND NEUTRON ACTIVATION

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The concentrations of most trace elements of public health interest in drinking water are very low and are difficult to measure reliably. A method has been developed for the simultaneous preconcentration of Co, Cu, Mn, Th and U in drinking water samples and their subsequent determination by neutron activation analysis (NAA). A ligand impregnated resin has been prepared by equilibrating Amberlite XAD-4 resin with a solution of 1-(2-thiazolylazo)-2-naphthol, called TAN, in methanol. The TAN-XAD-4 resin is very effective for the quantitative extraction of the above elements from water samples. The method involves passing the water through a column of TAN-XAD-4 resin and determining the elements exchanged on the resin by neutron activation. This procedure is superior to others where the metal is stripped from the resin using an acid solution and where problems involving irreproducible recoveries and reagent blanks are frequently encountered. Detailed investigations have been done on the optimization of pH, flow rate, resin mesh size, column height, reagent blanks, and effects of interfering elements such as Na, Cl, Mg, K and Ca. The precision and accuracy of the measurements have also been evaluated. Details of the development of methodology together with the elemental content of drinking water will be presented.

## ENVIRONMENTAL TRACE ELEMENT ACTIVATION ANALYSIS WITH ENHANCED COUNTING STATISTICS

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For trace element analysis required in environmental pollution control, neutron activation analysis is a well established method. For certain short-lived nuclides, however, such as  $^{77}\text{Se}^m$  ( $t_{1/2} = 17.7$  s,  $E_\gamma = 162$  keV), and  $^{207}\text{Pb}^m$  ( $t_{1/2} = 0.8$  s,  $E_\gamma = 570$  keV), significant in high throughput environmental sample analysis, the analytical sensitivity and accuracy is rather low because of the limited counting period due to rapid radioactive decay. On the other hand, certain relatively long-lived nuclides, such as most rare earth elements, encountered in environmental studies, could be analyzed more accurately with prolonged counting times and constant count rates. Measurements could then be performed unattended overnight. In both cases better results can be obtained by enhancing the counting statistics. This is achieved by the following novel technique [1]. The irradiated sample is moved toward the detector during the counting period with variable motion velocity, produced by a step motor. This compensates for the radioactive decay by gradually increasing the counting geometry factor. To keep the count rate constant, the step motor rotation speed is adjusted by a computer programmed by a controller. Constant radiation sources and reference materials are used for calibration of the system. Initial measurements have been performed with a delayed fission neutron counting system for uranium analysis. From the theoretical and experimental data it was found that, by using the above radioactive decay compensation technique, the counting period can be prolonged with a constant count rate, improving the total counts by a factor of more than 12.

If, in addition, cumulative and cyclic activation [1], limited by the sample availability and the plastic container strength, respectively, is applied at least 6 times each, then the total counts can be increased more than 400 times, i.e., the counting statistics can be improved by a factor of more than 20.

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# DETERMINATION OF NONMETALS IN HIGH PURITY METALS, ALLOYS AND SALTS BY PHOTON ACTIVATION WITH 30 MeV MICROTRON

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The photo-activation method for determining oxygen, nitrogen and carbon contents in high-purity materials was developed with a detection limit of  $(2-6)10^{-6}$  at.%. The fast techniques of radiochemical separation of  $^{15}\text{O}$ ,  $^{11}\text{C}$  and  $^{13}\text{N}$  were used to solve the following problems:

- nonmetal impurity determination in Fe, Nb, Mo, Cu, Sn, Cd, In, Ga, Na, Li, Al, Si;
- influence of the production technique on the level of nitrogen and carbon content in Al-Mg-Li alloys;
- detection of  $\text{NO}_2$  content in TlBr and its influence on the absorption of material in the IR range ( $1373\text{ cm}^{-1}$ );
- nitrogen solubility in basalt and albite glasses at a temperature of  $1250\text{ }^\circ\text{C}$  and pressure of up to 3 kbar.

The specifics of photo-activation analysis are the following:

- the determination of the bulk content of the impurity;
- the 0.5-4 g sample to be analyzed can have any shape and this peculiarity does not limit the depth of surface etching; in particular, the content of nonmetal impurities in alloyed metals, silicon, alloys, threefold compounds, salts, low melting temperature metals, and radiative unstable compounds can be determined;
- the relatively low cost of analysis of high-purity materials.

The investigations discussed indicate that the 30 MeV microtron can be successfully applied to solve problems of gaseous impurity determination in various substances, and fully satisfies modern scientific and industrial requirements.

# THE INTERFERENCE OF PLATINUM-NAA BY GOLD: SOME NEW ASPECT OF AN OLD STORY

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$^{199}\text{Au}$  [ $^{198}\text{Pt}(n,\gamma)(\beta^-)$ ] is the most suitable reference nuclide for Pt in NAA. Its use might be limited by the "gold interference":  $^{197}\text{Au}(n,\gamma)$   $^{198}\text{Au}(n,\gamma)$   $^{199}\text{Au}$ ;  $\sigma_{n,\gamma}$  of  $^{198}\text{Au}$  is 25800 b. This situation was first described in Ref. [1] and reexamined several times e.g. [2, 3, 4]. All papers show that  $(m_{L,Pt})_{Au}/m_{Au}$  [ $(m_{L,Pt})_{Au}$  = detection limit of Pt in the presence of  $m_{Au}$ ] increases with increasing irradiation time ( $t_i$ ) at a fixed neutron flux ( $\phi$ ) and with increasing  $\phi$  at fixed  $t_i$ . The common correction for the gold interference is:

$N_{^{199}\text{Au},Pt} = N_{^{199}\text{Au},sp} - N_{^{199}\text{Au},Au}$ ;  $N_{^{199}\text{Au}} =$  net peak area of the  $^{199}\text{Au}$   $\gamma$  - line chosen for evaluation;  $N_{^{199}\text{Au},sp} = N_{^{199}\text{Au}}$  in the spectrum of the sample (analogous:  $N_{^{198}\text{Au},sp}$ );  $N_{^{199}\text{Au},Pt}$  resp.  $N_{^{199}\text{Au},Au} = N_{^{199}\text{Au}}$  caused by  $^{199}\text{Au}$  generated from Pt resp. from Au.  $N_{^{199}\text{Au},Au}$  is obtained from  $N_{^{199}\text{Au},sp}$  and the  $^{199}\text{Au}/^{198}\text{Au}$  ratio found in a gold standard irradiated together with the sample.

In Ref.[2] the aspect was introduced to take into account the relative counting statistics errors ( $s_{c,r}$ ) of  $N_{^{199}\text{Au},Pt}$  in the calculations of estimates of  $(m_{L,Pt})_{Au}$ . Estimates of  $(m_{L,Pt})_{Au}$  vs.  $t_i$  are described by one function which goes through a minimum at a  $t_i$  which is characteristic for each Pt/Au ratio.

We calculated estimates of  $(m_{L,Pt})_{Au}$  under new view-points: Criterium 1 for  $(m_{L,Pt})_{Au}$ :  $(N_{^{199}\text{Au},Pt}/N_{^{199}\text{Au},sp}) = 0.1 - m_{L,Pt,c1}$ ; criterium 2 for  $(m_{L,Pt})_{Au}$ :  $N_{^{199}\text{Au},sp}$  obtained within 15 h counting time ( $t_c$ ) is such that its  $s_{c,r} = 0.1 - m_{L,Pt,c2}$ .  $m_{L,Pt,c1}$  increases with  $t_i$  to a limiting value.  $m_{L,Pt,c2}$  decreases with increasing  $t_i$ . Consideration of both criteria results in the following course of  $(m_{L,Pt})_{Au}$  vs.  $t_i$  ( $m_{Au},\phi$ , and  $t_c$  preset):  $(m_{L,Pt})_{Au} = m_{L,Pt,c2}$  at small  $t_i$ . The decreasing  $m_{L,Pt,c2}$  intersects  $m_{L,Pt,c1}$  vs.  $t_i$  at a  $t_i$  which is characteristic for the preset  $m_{Au}$ . At the intersection:  $(m_{L,Pt})_{Au} = m_{L,Pt,c2} = m_{L,Pt,c1}$ , minimum of  $(m_{L,Pt})_{Au}$ . From then  $(m_{L,Pt})_{Au} = m_{L,Pt,c1}$ , increasing with increasing  $t_i$ . For each preset  $m_{Au}$  a specific set of the two functions

results. In the series of such sets  $t_i$  of the intersections increase with decreasing  $m_{Au}$ .

Main result:  $(m_{L,Pt})_{Au}$  vs.  $t_i$  is not one continuous function but it is represented by the described succession of two functions. The practical handling of the gold interference problem is discussed.

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# AUTORADIOGRAPHY AND NEUTRON-ACTIVATION RADIOGRAPHY IN GEOCHEMICAL STUDIES OF THE ENVIRONMENT

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Different types of autoradiography are widely used in geochemical investigations for detecting and mapping the distribution of many elements in ores, rocks and the environment. This technique has a high sensitivity and resolution owing to an application of solid state nuclear track detectors (SSNTD), nuclear emulsion films and optical microscopes.

The next types may now be recognized: (1) autoradiography of natural radioactive elements and decay products (U, Th, Ra); (2) neutron activation-induced radiography of some stable elements that may be detected after neutron flux irradiation and (3) autoradiography by radioactive tracer methods.

For detecting and mapping the distribution of natural radioactive elements the following techniques are used: macroautoradiography to study large samples of rocks and ores with high U and Th concentrations (>100 ppm); microautoradiography (alpha-autoradiography) - for alpha-particle detecting of "hot particles" and other  $\alpha$ -emitters with total concentrations of 10-100 ppm; fission-track analysis is used to locate uranium- and thorium-bearing minerals and organic phases in quantities of 0.01-10.0 ppm after irradiation in a neutron flux.

Neutron activation-induced beta radiography makes it possible to bring the advantages of radioactivity to many stable elements for detecting and mapping them in geochemical investigations. Neutron radiation induces activity by capture-type nuclear reactions in a wide range of elements such as Na, Ca, Cr, Fe, As, Sb, Ag, REE, Au and others. Radioisotopes of these elements decay by emitting gamma, X-ray and beta particle radiation and may be detected after irradiation using nuclear-track emulsions exposed in contact with a sample.

In the radioactive tracer method, beta autoradiography is used to detect synthetic radioisotopes introduced into a system studied under experimental modelling of natural processes.

Thus the autoradiographical technique provides new information which is difficult or impossible to secure by other methods. This relates to both radioactive and stable elements in geochemical studies of the environment.

# COMPLEX ESTIMATE OF THE ECOLOGICAL STATE OF TERRITORIES ON THE BASIS OF THE EXAMPLE OF THE TOWN OF CHAPAEVSK

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Complex estimate of the ecological state of territories (of a town, region) is performed:

- to determine the degree of ecological comfort to man;
- to determine the state of ecological systems and the level of changes in them in comparison with the regions under the lowest anthropogenic impact;
- to perform zoning of territories by the value of anthropogenic impact and determine allowed loads for separate regions;
- to reveal and explain the dependence of people health on the pollution level of the environment;
- to elaborate urgent measures for rehabilitating the environment and people health;
- to develop a strategy for the efficient use of natural resources;
- etc.

A complex of works approved by the nature protecting authorities of Russia and carried out to make the complex estimate of the ecological state of a territory is schematically illustrated in the Figure.

The paper discusses the results of the investigations performed in the town of Chapaevsk in the Samarskaya region (the mid-Volga region).

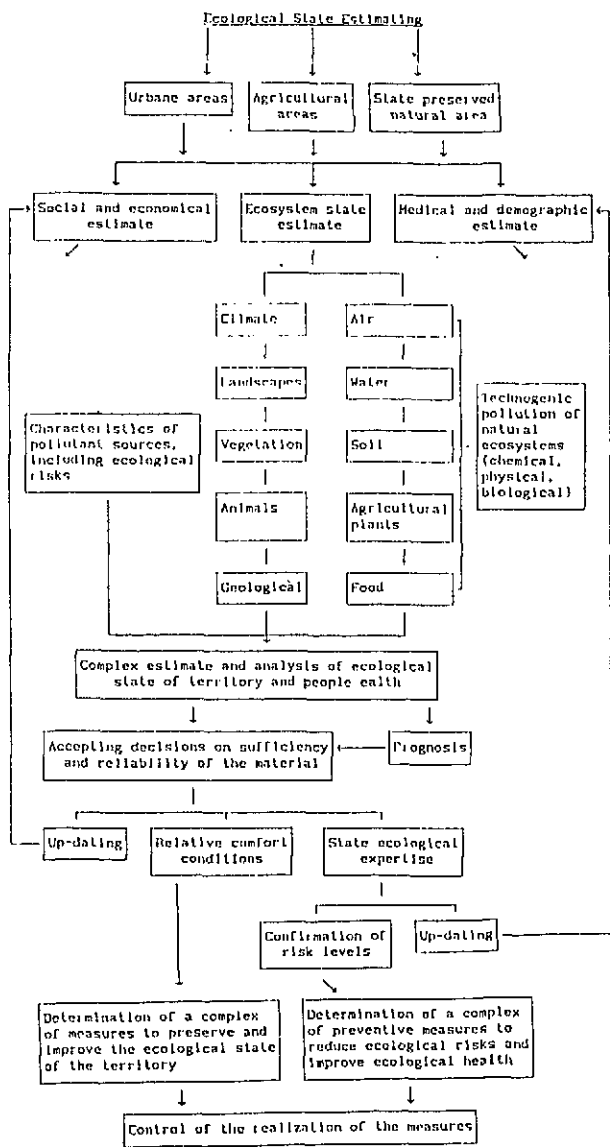


Figure. Works carried out for a complex estimate of the ecological state of territories.

The heavy ecological situation in the town was formed due to the uncontrollable use of old technologies in the chemical production of chlorine- and chlorine-organic-compounds, including pesticide, and toxins, explosives, etc. produced in the region till 1950. Dioxine, chlorine- and nitro-organic-compounds, and heavy metals make a far from complete list of chemical pollutants in the ecosystem of the town.

The work was performed following the resolution of the Council of Ministers of the Russian Federation.

POLLUTION OF AQUATIC LANDSCAPES: ASSESSMENT  
CRITERIA  
(ON THE BASIS OF EPITHERMAL INAA)

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In present technogenic conditions, aquatic landscapes of the Earth's northern hemisphere are in the ecological stress state demonstrated by stage-like violations of geochemical and biogeochemical homeostasis of the landscapes. The violations are accompanied by reversible and irreversible changes in natural self-regulation of the landscapes. The characteristic feature of the second half of the XX Century is the global-regional type of the violations.

Violations of the stress situation are caused by:

- significant expansion of the spectrum of pollutants of both technogenic and natural origin;
- increase in the emission of pollutants into natural medium;
- increased importance of a trans-regional transport of pollutants in atmosphere;
- acid fallouts as an immobilizing agent.

Modern situation corresponds to a new stage in the geochemistry of landscapes and is characterized by: formation of stable technogenic anomalies of Be, Se, and As toxic compounds; appearance of the REE class pollutants (in mining areas of strategic raw materials and in regions of developing hydrometallurgic and civil conversion industries; increase in the concentration of secondary radionuclides (Mo, Zr, Ru, Cd, Ba, Ce, Nd).

In this connection, nuclear physics methods acquire a leading role in providing a reliable estimate of pollution of natural landscapes with the aim of identifying violations in geochemical and biogeochemical homeostasis of the

# ACCUMULATION AND PROFILE DISTRIBUTION OF HEAVY METALS AND RARE-EARTH ELEMENTS IN SOIL OF THE NORTH AFFECTED BY A COPPER-NICKEL SMELTER COMPLEX AT THE KOLA PENINSULA

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Podzolic Al-Fe-humus soils are the predominant soil type in the North. They are characterized by soil-formation process that have been going on for hundreds and thousands of years, which have resulted in the formation of a shallow A0-A2-Bhfa-BC-C type soil profile with sharp differentiation as regards structure and properties. New, relatively short-term processes (years - ten of years), caused by air-borne industrial pollution by heavy metals, sulphur compounds and rare-earth elements, are proceeding in these soils. These new processes are changing the properties of the Al-Fe-humus soils.

The eluvial-illuvial character of distribution within the soil profile of so called typomorphic elements such as Al, Fe, Si, C is well-known. For more than 50 years a large territory in the Kola Peninsula have been subjected to industrial air pollution by heavy metals, sulphur compounds and rare-earth elements. Under air pollution organic horizon acts as a biogeochemical barrier to heavy metals and rare-earth elements since the amounts of these elements migrating downwards in the surface soil were small. Destruction of vegetation and soil leads to the increase of the mobility of these elements in the soil profile.

Attention has been paid to the insertion of these elements into biological migration. The dominant plants in the forests on the tree line exhibited a high capacity for pollutants accumulation in their photosynthetic organs at the sites subjected to pollution.

## THE TRACER SYSTEM THAT ALMOST GOT AWAY

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Who knows what scientific treasures lie buried in the stacks of underinterpreted data that modern analytical tools produce? Who has not experienced the frustration of rushing through one project and on to the next, knowing that the data had not been fully exploited and would remain that way?

Eighteen months ago I was able to revisit my past by organizing and computerizing my neutron-activation data on aerosols from 25 years and three continents. Recent data were already computerized, but older data lay in paper printouts or handwritten notebooks. With a dedicated hard disk and magneto-optical archiving device, I set about bringing them into a common format. I reasoned that the thousands of samples, each analyzed for 40-50 elements, had to contain exciting new ideas.

Although not finished, the effort has already yielded substantial results. First came a new graphical technique for isolating the pure crustal, marine, and pollution components from mixed aerosol. Then came a broad picture of elements in aerosol over much of the Northern Hemisphere, showing unexpected patterns of concentration and composition. Last came a new elemental tracer system to deal with these patterns, by using 20-25 elements rather than our previous seven. My presentation will describe the rich geographical patterns, the large number of elements involved, and the new tracer system. It will show that elements determined by NAA allow us to trace aerosols with far-greater power than anyone ever thought possible.



But suppose I couldn't have gone back? Chances are that none of this would have happened, because NAA of aerosols is fast becoming a casualty of funding pressures and reactor closings. Quicker and cheaper-not better-analytical methods are being demanded. We came within a hairbreadth of missing the true tracer power of NAA elements forever. How many similar opportunities are about be buried elsewhere? Perhaps we should give more weight to extracting the fullest from existing data, not just to generating new data. Perhaps we should push harder for the appropriate technique, even though it may be slower or more expensive.

ATMOSPHERIC AEROSOL MONITORING IN THE  
SIBERIAN REGION.  
"SIBERIAN AEROSOL" PROJECT

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In the frame of the "Siberian Aerosol" project on the basis of the observation stations of the Siberian Division of the Russian Academy Science the monitoring of the atmospheric aerosol characteristics was organized in the Siberian region.

The aims of the project are the following:

- Investigation of the formation, transformation, and transport of aerosols in Siberia on local, regional and global scales;
- Determination of sources and sinks of atmospheric aerosols;
- Estimation of the quality of atmospheric air, the level of contamination of vegetation, soil and water, and the fate of different substances and elements;
- Study the impact of various aerosols on the health of people and animals;
- Definition of the role of atmospheric aerosols in atmospheric processes and climate.

The project consists of five phases:

- Aerosol monitoring in different Siberian regions and by special expeditions.
- The development of methods and equipment for sampling and measuring the size distribution, number and mass concentrations, and chemical composition of aerosol particles.

- Mathematical modelling.
- Laboratory studies of the elementary stages of aerosol formation and the regularities of the interaction between aerosols and biological objects and systems.
- The data bases on atmospheric aerosols.

The particle size distribution, number and mass concentrations, and elemental and ionic compositions of the aerosol particle near Lake Baikal in the Novosibirsk region and different Siberian regions are available [1,2].

## References

- [1] SIBERIAN HAZE-2. International Workshop, September 14-18, 1993, Abstracts, Novosibirsk, 1994, 40p.
- [2] Atmospheric and Oceanic Optics. 1994, v.7, N 8. Aerosols of Siberia. Thematic issue.

## INAA FOR INVESTIGATING AEROSOLS IN SIBERIA

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Atmospheric aerosols play a decisive role in many atmospheric processes (clouds - rainfall forming, radiation heat transfer, visual range). They affect climate and the quality of the environment. In spite of an important contribution to global atmospheric pollution of aerosols formed in Siberia, they are hardly studied yet.

Earlier, we investigated aerosol fall-outs on the surface of the ground cover, as well as their role in the formation of new anthropogenic geochemical provinces in the surroundings of an industrial centre by analyzing natural maps. In the present paper INAA is used to analyse aerosols forming in the suburbs of the Norilsk city in the south of the Novosibirsk region. The aerosols were selected by the aspiration method. The microelements: Na, Mg, Al, Cl, Ca, Sc, V, Mo, Cr, Mn, Zn, Cu, As, Se, Br, Rb, Ag, Cd, I, Sb, Cs, Ba, REE, K, U, Th, Hg, Pb, Be, Si, Fe, on filters were identified by a number of methods, including INAA, X-ray fluorescence with synchrotron radiation, atomic emission and atomic absorption spectroscopy, in different laboratories.

Our INAA results obtained using the base of INR of SRI NP TPU and the results by K.A.Rahn (USA) are in good agreement with each other for a number of elements, including Al, V, Cu, Sb, Au, Cl, Mn, Cr, and to a less degree for rare-earth elements. These results allow speaking about high informativity of the method in investigating aerosols. In our opinion there is no point in using other methods to analyze aerosol filters, at least to obtain massive primary information. For example, this opinion is confirmed by the dynamics of daily changes in the composition of aerosols during one month.

We plan to use the results of the determination of the daily and seasonal dynamics of changes in the chemical composition of aerosols for developing algorithms and programs for modelling their formation, transformation and distribution, for calculating pollution levels of the atmosphere, plants, and soil, as well as for optimizing systems of monitoring and operative control of the air medium under anthropogenic impact.

## DETERMINATION OF HEAVY METALS IN THE AIR OF THE CITY OF ULAN-BATOR

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The atmospheric air of the city of Ulan-Bator is polluted as a result of the dispersion of waste products from several anthropogenic sources. These sources are coal and oil fires from 3 thermal power stations, about 300 boilers, more than 40000 stoves and a few ten thousand cars.

During a year, especially from November to April, the powerful Asian anti-cyclone does not promote an effective mixing of air due to the high pressure region near Ulan-Bator.

In recent years a number of instrumental activation and X-ray fluorescence (XRF) methods for multi-element analysis of atmospheric air samples have been developed at the Nuclear Research Laboratory (NRL) of the National University of Mongolia (NUM).

The samples collected from 3 different sites in the Ulan-Bator area were irradiated by thermal and resonance neutrons in the biophysical channel of the BRJ-2 reactor of FLNP, JINR and by bremsstrahlung beams of the MT-22 microtron of FLNR, JINR. Also,  $^{109}\text{Cd}$  and  $^{241}\text{Am}$  radioactive sources and an X-ray tube for XRF analysis in NRL of NUM were used.

In this work results of the analysis of Co, Zn, Sc, As, Sb, La, Sm, Au, Pb and U by INAA, Mg, Ca, Ti, Fe, Sc, Sr, Zr by IGAA and Fe, Zn, Sr and Br, Pb by XRF analysis in the air of Ulan-Bator are presented.

We have compared the obtained results with environmental background data.

# METHOD OF EXPRESS ANALYSIS OF SOIL FOR URANIUM AND PLUTONIUM

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The most hazardous of radioactive wastes of nuclear plants is plutonium whose depositing pollutes soil and other objects of the environment. An express method for determining the  $^{239}\text{Pu}$  content in soil on the level of  $\sim 2 \cdot 10^{-10}$  g/g without specially prepared probes has been proposed.

The method is based on the dependence of the fission cross section on the neutron energy.

Neutrons from the (d,T) reaction moderated in a soil probe were used to produce quasimonochromatic neutrons. Fission events were registered by fission gamma-rays by a  $4\pi$ -multisectional liquid detector and discriminated from radioactive capture events and inelastic process by their multiplicities. The quark content of  $^{235}\text{U}$  was over  $10^{-10}$  g/g, and this content also experienced fission under the action of soil moderated neutrons.

By measuring  $^{239}\text{Pu}$  and  $^{235}\text{U}$  fission in a polluted soil probe alternately with measuring these rates in the same soil probe but containing the known and considerably larger amounts of  $^{239}\text{Pu}$  and  $^{235}\text{U}$  in dependence on the moderation time, one can determine  $^{239}\text{Pu}$  and  $^{235}\text{U}$  contents in the probe of polluted soil.

In spite of the fact that soil consists mainly of light elements and the spectrum of soil moderated neutrons is considerably broader than the known spectrum of neutrons moderated in lead, the performed Monte-Carlo calculations have shown that using 14 MeV neutrons from a pulsed source with an average neutron yield of  $\langle Q_n \rangle = 10^8$  n/s, one can determine the  $^{239}\text{Pu}$  content on the level of  $2 \cdot 10^{-10}$  g/g with a statistical accuracy of  $\sim 5\%$  during a period of 5-10 min. At the same time the optimal weight of the probe has a very representative value of  $\sim 25$  kg.

## ON DETERMINATION OF MAN-MADE PLUTONIUM CONTENT IN NATURAL SAMPLES, PLANTS AND LIVING SPECIES

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The method of  $^{239}\text{Pu}$  minor content determination in the soil, water, air, living species is developed. It is based on chemical extraction of Plutonium from the specimens. The  $^{236}\text{Pu}$   $\alpha$ -active tracer chosen to control the chemical yield of Pu from samples being investigated was obtained with accelerated helium ions in the reaction  $^{235}\text{U}(^4\text{He}, 3n)^{236}\text{Pu}$ . After chemical separation the foils with Pu were put in contact with polyterephthalate foils and irradiated on pulsed nuclear reactor IBR-2 JINR with thermal neutron fluence  $\approx 10^{15} \text{ cm}^{-2}$  together with calibrated U and Pu etalons.

After proper etching and scanning the Pu content in soil specimens from some regions of Ukraina, Bielorussia and Ural were determined to be  $3 \cdot 10^{-12} \text{ g/g}$ . To check the possible Th-U minor admixtures of U and Th in Pu fractions the control experiments with heavy flux of 20 MeV  $\gamma$ -rays were carried out. It provides an upper limit of admixture of these nuclei at the level of  $\leq 10^8$  per sample. The method proposed provides the sensitivity of routine Pu determinations at the level of  $10^{-13} \text{ g/g}$ . By using higher neutron fluences up to  $10^{18} - 10^{19} \text{ cm}^{-2}$  and some non-organic glasses, crystals with a very low U content  $\leq 10^{-14} \text{ g/g}$  it is possible to reach the sensitivity of  $10^{-14} - 10^{-15} \text{ g/g}$  of Pu in specimens.



**ON PROBLEM OF DETERMINATION OF MAN-MADE PLUTONIUM IN  
SPECIMENS AT THE LEVEL OF SENSITIVITY UP TO  $10^{-15}$ g/g**

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The new approach to the problem of determination of subpicogram quantities of  $^{239}\text{Pu}$  in the environment and living species based on chemical separation of Plutonium and neutron activation analysis was suggested. For checking the possible Uranium impurities in Pu fraction the photofission effect of heavy nuclei was first proposed instead of application of well-known method of  $\alpha$ -radioactive Uranium traces.

It was shown that at the high degree of leaching of man-made Plutonium from the solid matrix of environmental samples the degree of extraction of chemical element Uranium out of these specimens was much lower. Nevertheless even taking into account the incorporation of Uranium in silicate matrix of natural samples being investigated (Uranium clark of about  $10^{-6}$  g/g), the needed Pu separation coefficient for Uranium is about  $10^5 - 10^6$ . It is stated that such a high rate of Plutonium purification should be possible only by applying oxidation- reduction chemical reactions which allow to convert Plutonium and Uranium in solutions in the different oxidation states. For this purpose the most perspective will be the electrochemical methods of change the oxidation states of chemical elements because such a procedure did not require any additional reagents which can introduce additional trace amount of Uranium in the system. Our preliminary study demonstrates that such problem could be solved in spite of some additional requirements and conditions for chemical separation procedures.

Taking into account that cross section of photofission of Thorium is rather high (by a factor 2-3 lower than that for Uranium), the presence of Th in Plutonium fraction can imitate rather high Uranium content. That is why it is necessary to provide rather high separation rate of purification from Thorium - at the level  $n \times 10^5$  times.

# SIZE-SPECTRA AND GROWTH OF URBAN AEROSOL PARTICLES BEARING TRACE ELEMENTS BY INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

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Aerosol particles smaller than 1.8 micrometers were size-fractionated using micro-orifice impactors at two urban sites in the heavily-industrialized Camden/Philadelphia area and College Park and two other sites in the nonindustrial Washington, DC area, and were analyzed by instrumental neutron activation for 44 elements including As, Se, Sb, and Zn, i.e., elements strongly associated with coal combustion, incineration, and regionally transported secondary aerosols. Distribution parameters, i.e., geometric mass mean and mass median aerodynamic diameters (gmmad and mmad), were determined nonparametrically and by fitting individual peaks in the spectra with a log-normal distribution function, respectively. Size spectra were influenced by local sources and strongly influenced by the relative humidity (RH) in College Park, but the influence of RH was much less for the Camden/Philadelphia aerosol. At Camden, the fresh accumulation aerosol from an incinerator plume was clearly identified by peaks at 0.2 micrometers in the spectra for Zn, Cd, and Cr. Emissions from a nearby Sb roaster were detected in 1.3 micrometer particles. In College Park, gmmad increased with increasing RH along separate curves for locally-emitted and regionally transported aerosol. At 60% RH, gmmads were: As,  $0.30 \pm 0.03$  and  $0.46 \pm 0.04$ ; Se,  $0.33 \pm 0.06$  and  $0.54 \pm 0.04$ ; Sb,  $0.39 \pm 0.03$  and  $0.53 \pm 0.04$ ; and Zn,  $0.39 \pm 0.06$  and  $0.53 \pm 0.08$ , respectively. In the Camden/Philadelphia area, mmads at  $RH < 60\%$  were: As,  $0.31 \pm 0.04$ ; Se,  $0.28 \pm 0.04$ ; Sb,  $0.31 \pm 0.03$ ; and Zn,  $0.53 \pm 0.04$  micrometers, and except for Zn, were, therefore, comparable to values associated with air influenced by local sources at College Park.

# TWENTY-YEAR STUDY OF RADIONUCLIDE DISTRIBUTIONS IN THE TROPOSPHERE AND LOWER STRATOSPHERE

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Since 1973 vertical distributions of fission products and natural radionuclides (radium-226 and lead-210) have been measured in the troposphere and lower stratosphere in north-east Poland at altitudes between 0 and 15 km. Samples of atmospheric particles were collected from filters on the ground level and at altitudes of 1, 3, 6, 9, 12 and 15 km using special high altitude samplers installed inside fuel tanks of MIG-type aircraft. The samples were analyzed by  $\gamma$ -spectrometry and radiochemical methods.

Between 1973 and 1993 we collected 638 radioactive aerosol samples forming 108 vertical concentration profiles for strontium-90, cesium-134,137, lead-210 and radium-226. In these 20 years several natural and man-made events took place which significantly influenced the concentration levels of some of these radionuclides at various altitudes. Vertical distributions of these radionuclides may change by 2 or 3 orders of magnitude dependencing on the radionuclide source location and meteorological situation.

Natural radionuclides are produced by two types of sources. The first type is the quiescent, continuous ground level and near ground level sources. The second type is the short-term, violent stratospheric source, i.e., volcanic eruptions. The combined effect of stratospheric and ground level sources, and the transport of contamination in the atmosphere produces typical vertical parabolic profiles for long-term mean concentrations of radium-226 and lead-210. Our long term observations have indicated that there is a strong correlation between volcanic eruptions and the content of natural radionuclides in the global atmosphere.

The second type of isotopes which we measured in the atmosphere are fission products. In the last twenty years three different periods can be noted:

- the period of less intense Chinese and French nuclear weapon tests in the atmosphere, which ended in 1980;
- the quiet period of no atmospheric nuclear events;
- the Chernobyl accident and the period after it.

In the first period fission products were brought to higher levels of the atmosphere by nuclear explosions, and the source was in the stratosphere. Mean vertical distributions of fission products increased with altitude, and the increase was fast in the stratospheric air. Between 1973 and 1980 many Chinese nuclear explosions were conducted in the atmosphere, and the effects of megaton tests explosion were detected over Poland.

In April 1986, the reactor accident at the Chernobyl nuclear power station also changed vertical distributions of fission products in the atmosphere, and the mean vertical concentration profiles of radiocesium demonstrated an opposite shape to that following a nuclear explosion: the highest concentration was detected near the ground surface and the lowest in the stratosphere.

By mid-summer of 1986, activity concentrations were homogeneous over all levels in the atmosphere but were higher than in the corresponding period of 1985.

Since 1987 we have been observing higher concentrations of radiocesium at 1 km of altitude. This is probably connected with an atmospheric mass transport from Chernobyl regions in the boundary layer.

# NEUTRON ACTIVATION INVESTIGATION OF COMPOSITION AND GEOCHEMICAL PECULIARITIES OF LAKE BAIKAL BOTTOM SEDIMENTS

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In connection with the problem of anthropogenic pollution of the Lake Baikal ecosystem 18 trace elements and iron were determined in samples of bottom sediments by Instrumental Neutron Activation Analysis (INAA). For the INAA 40 samples of bottom sediments from the southern part of the lake, the shore having maximal industrial development, were obtained. Two international geochemical reference samples, Andesite AGV-1 (USA) and Granite GM (Germany) were used as multielement monitors.

Precisely weighed aliquants of each sample of bottom sediment and monitor samples were placed into vials of spectrally pure quartz and sealed. Each sample portion was about 50 - 100 mg. The sealed vials were packed into a standard aluminum container for placement into a nuclear reactor for irradiation. The irradiation time was 20 hours at a thermal neutron flux density of about  $5 \times 10^{13}$  n/(cm<sup>2</sup>s).

To carry out the INAA a high precision spectrometer complex was used including two semiconducting detectors: a BDER-2K detector from high purity germanium (HPGe) with an energy resolution of 0.530 keV at the 121.8 keV <sup>152</sup>Eu gamma-line and a DGDK-40B type coaxial Ge(Li)-detector with an energy resolution of 2 keV at the 1332.5 keV <sup>60</sup>Co line. The HPGe-detector has a good efficiency in the energy range from 20 to 150 keV and the Ge(Li)-detector covers the energy range from 150 keV to 2 MeV. To measure the spectra of activated bottom sediments and monitors, a 4096-channel NTA-1024 pulse-height analyzer equipped with a magnetic tape drive was used.

The main result of the INAA investigation of Baikal bottom sediments is absence of anthropogenic pollution by trace elements.

Concentrations of 18 elements (Ba, Ce, Co, Cr, Cs, Eu, Gd, Hf, Ho, Lu, Nd, Sm, Ta, Tb, Tm, Th, U, and Yb) in the surface stratum of bottom sediments of southern Baikal are on the level of corresponding clarks for sedimentary rocks. It gives us the possibility of using the INAA data for geochemical interpretation. The main geochemical peculiarities of Lake Baikal bottom sediments are their enrichment by light REE and a deficiency of caesium and tantalum. Distribution of rare-earth elements in Lake Baikal pelitic sediments is of a specific type which is intermediate between riverine and sea types.

# ANALYSIS OF ARSENIC AND URANIUM DISTRIBUTION AND SPECIATION IN ENVIRONMENTAL SAMPLES FROM A LOW LEVEL WASTE MANAGEMENT SITE

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Mine tailings from radium and uranium ore refinement from 1933 to 1988 were buried at waste management facilities one hour east of Toronto. Over a period of fifty years radioactive waste and heavy metals have contaminated the surrounding soil. The stability of one site is under question due to its location beside the bluffs overlooking Lake Ontario. Erosion of the bluffs, surface runoff, and groundwater seepage into Lake Ontario from the site is monitored by Cameco (the company overseeing the waste management site), the Atomic Energy Control Board (AECB) and the Ontario Ministry of the Environment.

Water, soil and plant samples collected from a low level waste management site were analyzed for uranium and arsenic using epithermal neutron activation analysis (ENAA). Using ENAA eliminated most of the dominant interferences (such as sodium and chlorine peaks), allowing concentrations of these elements to easily be determined through 5 minute irradiation in a flux of  $2.5 \cdot 10^{11} \text{ n}/(\text{cm}^2\text{s})$ . The arsenic concentrations in the water samples were relatively elevated, despite efforts at the site to decontaminate it through precipitation with ferric chloride. It is suspected that the arsenic is present in a complexed form, possibly involving fluoride, resulting in its high solubility. Plant and soil samples collected nearby the water were also found to have elevated arsenic. The concentration of arsenic in the water effluent was found to be  $0.7 \mu/\text{g}$ ,  $11 \pm 0.5 \mu/\text{g}$  in field horsetail and  $231 \pm 4 \mu/\text{g}$  in soil. The normal concentration of arsenic in land plants varies from  $0.02 - 7 \mu/\text{g}$ ,  $1 - 121 \mu/\text{g}$  in Ontario soils, and less than  $0.01 \mu/\text{g}$  in surface and groundwaters.

The retention characteristics of the uranium and arsenic by the soil was evaluated using various sequential extraction techniques. These procedures involved reagents that attacked different components of the soil matrix in order to differentiate between readily exchangeable heavy metals and heavy metals bonded to the carbonate, aluminum, iron, organic, or sulfide fractions of the soil. Harsher reagents were used in each consecutive step to remove more strongly bonded metal ions. Determination of the speciation of arsenic and uranium in soils is required for the development of a remediation process for contaminated soils.



## SELECTIVE TRACK RADIOGRAPHY OF ELEMENTS Pb-Bi IN SOLIDS WITH ACCELERATED HEAVY IONS

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The problem of low concentration selective measurements of the elements Bi, Pb and Au, Pt with the aid of accelerated heavy ions in minerals is discussed. In many cases it is necessary to determine not only the average contents of the heavy elements in a sample but also to determine its local distribution with spatial resolution at about 10  $\mu\text{m}$ .

Our approach in solving that problem is based on the measurements of the yield of prompt fission fragments and induced  $\alpha$ -activity produced at the interaction of accelerated  $^{12}\text{C}$  ions with Bi, Pt, Au and Pb nuclei. The  $^{12}\text{C}$  ion exposures of metal foil targets of these elements and polished mineral samples were performed at the external beams of the accelerators U-200, U-400 of Flerov Laboratory of Nuclear Reactions, JINR at the energies 9.1 and 13.2 MeV/nucleon correspondingly. The specimens were placed perpendicularly to the  $^{12}\text{C}$  beam. Before the exposure they were covered with 15-20  $\mu\text{m}$  thick muscovite mica; the total  $^{12}\text{C}$  ion fluences were between  $3 \cdot 10^{12}$  and  $10^{14}$  ions/cm $^2$ .

After carbon ion exposure induced  $\alpha$ -activity of Po isotopes was registered by CR-39 plastic track detectors. Exposure time was 1.5 hour and 144 hours to detect short-lived and long-lived components, respectively for Bi, Pb, Au and Pt ethalons and for mineral samples.

It was established, that the yield of fission fragments in "2 $\pi$ -backward" geometry was within  $\pm 10\%$  the same for all the heavy elements being investigated. The observed local microdistribution of fission fragment tracks in muscovite mica was determined with the precision up to  $\pm 10 \mu\text{m}$ . By examining of  $\alpha$ -sensitive CR-39 plastic track detectors it was found, that the yield of  $\alpha$ -particles for metallic Au and Pt targets was  $\leq 10$  tr/cm $^2$ , for Bi and Pb targets  $\geq 10^6$  tr/cm $^2$ .

Thus we can resolve the local microdistributions due to Bi, Pb and due to Au, Pt inclusions by comparing of fission fragment and  $\alpha$ -particle tracks microdistributions.

## DETERMINATION OF Th CONCENTRATION IN SPECIMENS WITH GAMMA-RAYS AND ACCELERATED HELIUM IONS

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Experiments on improving track technique of Th content determination in natural samples have been performed. For Th concentration measurements 18 and 20 MeV  $\gamma$ -rays were used. For  $\gamma$ -rays energy range from 6 MeV upto 23.5 MeV, the ratio of fission fragment track densities  $N_U/N_{Th}$  for thick targets of these elements has been obtained. This ratio ranges from 1.7 to 3.2 with the maximum at  $\gamma$ -rays energy of 15 MeV.

The determined Th concentrations in the calibrated Th standards and iron-manganese nodules are in agreement within experimental error bar with the known values.

For 9.1 MeV/nucleon accelerated helium ions the track density ratio  $N_U/N_{Th}$  has been found equal to  $(1.06 \pm 0.07)$ , but the determined Th content was lowered down possibly due to the vertical inhomogeneity of helium beam and also due to partial thermal fading of the fission fragment tracks in mica.

The investigations have been performed at the Microtron MT-25, FLNR.

## EPITERMAL NEUTRON ACTIVATION ANALYSIS FOR MONITORING NORTHERN TERRESTRIAL ECOSYSTEMS

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New experimental data has been obtained on heavy metal and rareearth element concentration in environmental objects, namely pine needles and soils, caused by atmospheric pollution in different regions of the Kola peninsula. The investigation was performed with use of epithermal neutron activation analysis at the IBR-2 fast pulsed reactor. Analysis of nearly 40 elements distributions in pine-needles and solids from the studied geographical points testifies to the existence of a strong contamination source - the nickel smelting complex in Monchegorsk. The contamination levels for Ni, Co, Cr, Se and others are also high and may be hazardous for this region's population, because some of these elements are cancerogenic.

# NEUTRON ACTIVATION METHOD IN THE ANALYTICAL CONTROL SYSTEM OF ENVIRONMENTAL OBJECTS

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The overall problem of environmental protection can be formulated as an essential working principle of man's interaction with natural biosphere processes. In this situation the question arises concerning the immediate study of anthropogenic factors affecting the stability of biological systems and the ways anthropogenic loads interact with these systems. This is an important environmental problem. The situation is linked with landscapes (continent as a whole), water levels (rivers, lakes, ocean), air (atmosphere) and in general all natural systems.

These systems are mainly affected by technogenic processes, transportation, pollution growth, but also by natural events such as volcanic explosions, earthquakes, etc. These situations need analytical control of the activities.

The analytical control means the use of a complex of methods providing information about the chemical and phase compositions of above mentioned objects and their changes under the influence of technogenic, man-made and natural processes.

Estimation of elements (the influence is mainly environmental: As, Be, Cl, Mn, Pb, Sb, Se, Tl, V; Al, Ba, Fe, Zr, REE; Cd, Cr, Hg, Ni, etc.) essentially determines the selection of the method of analysis. Generally, in chemical analysis and in the analysis of environmental objects, particularly ore, the highest priority is given to the methods with the following parameters: Low detection limits (on the order of  $n < 0.1$  ppm), small analysis errors, high selectivity, minimum sample mass (of  $< 10^{-3}$  g), the possibility of analysis in the presence of high fluxes, which is important for water and air sample analysis, instrumentation and automatization possibilities, etc.

At present, large nuclear-physical methods fulfill the quantities and requisites of the analysis. Among them, activation analysis using neutrons (NAA) has many advantages.

As an example, different NAA-methods were examined and the data of many objects were presented. The possibility of using this method for solving some scientific and socioeconomic problems has been also discussed.

# MAUNA LOA OBSERVATORY AEROSOL CHEMISTRY 1979-1993

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Atmospheric aerosol samples have been collected at the Mauna Loa Meteorological Observatory between 1979 and 1993 and have been analyzed for the elemental composition by Instrumental Neutron Activation Analysis. The samples collected during "down slope" conditions at the observatory yield aerosol samples from the "free troposphere" at an average altitude of approximately 12,000 ft. The collections have been based upon wind direction, speed and time of day. The results of chemical analyses have shown the presence of marine aerosols, long range transport of crustal elements from the continents around the Pacific Ocean, most significantly Asian Dust. Combustion of oil from the Kuwaiti fires (Gulf War) and the burning of oil from the Exxon Valdez accident in Alaska have also been identified by their vanadium chemical and air mass trajectories, and the unique chemical signatures of the sources themselves. The vanadium signal from the burning of oil cleaned up after the Exxon Valdez accident in the Gulf of Alaska and was the largest vanadium signal in the entire Mauna Loa vanadium data record. Not only that, but the analysis of oil samples from the Exxon Valdez itself showed the presence of tungsten and other heavy metals that also are seen in the samples collected in Hawaii, and act as a chemical fingerprint of the oil from the incident. To make the signal measured in Hawaii even more unique, the presence of excess bromine (above normal sea salt contributions) shows that the that was burned in combustion barges that Exxon had in the area was mixed with sea water.

MICRODISTRIBUTION OF FISSILE RADIONUCLIDES IN  
THYROIDS OF INHABITANTS OF THE GOMEL REGION,  
BELARUS

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\* \* \*

## HIGHLY-SENSITIVE ANALYSIS OF THORIUM

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Determination of the ultrasmall thorium contents in the natural objects is the important task. The thorium determination limit for majority of the instrumental methods does not exceed  $10^{-10}$  -  $10^{-11}$  gram.

The present report is devoted to studying the possibility of determination of the ultrasmall thorium concentration by the track method with the use of the  $(\gamma, f)$  reaction.

The solution with the known thorium content was deposited on a  $175 \mu\text{m}$  thick lamsan foil and evaporated. The lexandetector was placed above the foil and fixed.

A packet consisting of 10 - 15 samples was irradiated by photons of the MT-25 microtron with an initial energy of 25McV and  $15 \mu\text{A}$  beam current for 4 hours. After the irradiation the detectors were chemically etched and determined by scanning their fission fragment tracks.

The proposed method provides to determine thorium with detection limit at the level of  $5 \cdot 10^{-12}$  g. When increasing of the irradiation time to 10 h the detection limit equals  $10^{-12}$  g.



# INVESTIGATION OF RADIOACTIVITY AND SIZE DISTRIBUTION OF HOT PARTICLES BY MEANS OF SSNTD

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By means of solid detectors the structure of track "stars" created by "hot" particles trapped by filters from atmosphere near Chernobyl PS has been studied.

Thin filters (about 1 mkm thickness) containing "hot" particles were driven into a contact with a detector (mica fluorlogopit ) and they were irradiated by the flux of thermal neutrons. Separate tracks and stars of tracks of hot particles were observed in the optical microscope after a chemical etching of the detector. Some filters were studied by means of detector CR-39. The structures of track stars created by fragments and alpha-particles were compared.

Track stars were used for defining sizes of hot particles and content of atoms of the disintegrating impurity in them.

We have established the following:

- Sizes of hot particles change in wide ranges - from fractions of microne up to 0.1 cm.
- Both free particles and particles attached to carriers have been observed.
- Concentration of the disintegrating impurity changes from 10-6% to 10-3%.
- Atoms of disintegrating impurity are concentrated mainly on the surface of particles. Only separate particles contain disintegrating impurity not only on the surface but in the bulk.

- About 10% of particles have a non-spherical form. As a rule in such particles a disintegrating impurity is distributed along the whole bulk.
- Sometimes localized clusters of hot particles joined by the common carrier are met.
- By means of local micro-X-ray spectral analysis there was found established the material of separate large particles-carriers.

# ACTUAL PROBLEMS OF RADIOECOLOGY: LONG-LIVED RADIONUCLIDES. NATURAL AND MAN-MADE TRACERS IN THE ENVIRONMENT

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Long-lived radioactive nuclides occur in nature from various sources. Primordial radionuclides have sufficiently long half-time and survived since the closing of the solar system 4.55 Ga ago. Nuclides with shorter half-lives are continuously produced by decay or spontaneous fission of primordial radionuclides, by nuclear reactions induced by neutrons and  $\alpha$ -particles from radioactive decay and spontaneous fission and by interactions of primary and secondary solar and galactic cosmic ray particles with matter. Man-made long-lived radionuclides in the environment originate from sources such as nuclear explosions and from accidental or routine releases of nuclear installations. There exist distinct emission patterns for man-made nuclides such as the bomb peak in the nineteen-sixties as well as single events or continuous emission.

Radioecology provides the scientific basis for our understanding of the pathways of radioactivity from release sources through the environment to man. Though most long-lived radionuclides are of minor importance for present day's human radiation exposure, they are of outstanding scientific importance for several reasons. They allow to detect long-term man-made changes in our radiation environment early enough for decision making in politics. They are important indicators of transport processes in radioactive waste depositories. They can be used to reconstruct radiation exposures due to fall-out of short-lived radionuclides long after accidental emissions. Last, but not least, they provide extraordinary tracers which allow for investigation of long-time and large-scale environmental processes.

Primordial radionuclides and their short-lived successors provide the wide range of equilibrium and disequilibrium dating techniques. Cosmogenic nuclides in extraterrestrial matter such as lunar surface samples, meteorites and cosmic dust particles, provide unique tools to study the spectral distribution and intensities of solar and galactic cosmic ray particles over time scales up to one billion of years. Moreover, they allow to investigate the exposure history of the irradiated objects in space and to draw conclusions about the collision history of small bodies in the solar system and about terrestrial ages of meteorites. In terrestrial matter, cosmogenic nuclides produced in the atmosphere are involved into the large natural physico-chemical cycles and finally brought in living matter or they are introduced in the hydrosphere and thus are constituents of all form of wet precipitation. Thus, they become natural tracers in hydrology, glaciology and oceanography. In the surface of the earth's lithosphere cosmogenic nuclides are produced *in situ*. These nuclides allow to study erosion ages as well as to determine deglaciation ages.

In this paper, a survey is given on the sources of long-lived radionuclides in nature, the nuclear analytical methods needed to investigate them and on their applications as man-made and natural tracers for the scientific description of our environment.

## RECENT ADVANCES IN TERRESTRIAL RADIOECOLOGY

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Before the Chernobyl accident, terrestrial radioecology was largely limited to agricultural systems. The specific physics physicochemical characteristics, radionuclide composition and geographical distribution of the Chernobyl fallout however has initiated extensive research related to natural environments such as forest and mountain ecosystems. The present paper will review some of these investigations related to the behaviour of radiocaesium in natural soils, uptake in vegetation, transfer in different foodchains, and exposure to domestic animals (cattle, sheep, goats, reindeer) grazing in these ecosystems. The reindeer population in Central Scandinavia was particularly affected. Various countermeasures employed in order to reduce the radiocaesium content in meat are briefly discussed.

## FIRST RESULTS OF THE RADON SURVEY IN MONTENEGRO (YUGOSLAVIA)

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This paper presents the first results of a joint Montenegro-Russian project of indoor radon measurements.

We used the Russian RRG-01M radiometer with a surface barrier semiconductor detector for express measurements and passive radiometers with cellulose nitrate detectors for integral (3 months) measurements. The main part of the results we obtained from the Podgorica and Nikshich regions. Measurements were also taken in the region to the North of Montenegro (Zabliak, Kolashin, Berane) and at the seacoast (from Hersog-Novi to Ada).

We observed high levels of indoor radon concentration in some private houses in Podgorica (more than 400 Bq/m<sup>3</sup>) and in the houses situated near the ore quarry in the Nikshich region. The concentration of radon in hotels at seacoast is rather low.

## HIGH PURE $^{237}\text{Pu}$ AND $^{236}\text{Pu}$ FOR BIOMEDICAL AND ENVIRONMENTAL RESEARCH

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Plutonium-237 is the only Pu isotope which meets the medical requirements for the metabolism research *in vivo* ( $T_{1/2}=45.6$  days,  $EC=99.99\%$ ,  $\alpha=4.2\cdot 10^{-5}$ ,  $E=100$  keV). A  $^{237}\text{Pu}$  preparation used for injection into human body must be essentially free from alpha-emitting plutonium isotopes. The  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  detection limits depend on the isotopic purity of  $^{236}\text{Pu}$  which is one of the convenient tracers for the radioecological surveys of the contamination of the environment by plutonium.

For example for the precise analysis of  $^{238}\text{Pu}$  its admixture in the tracer should not exceed  $10^{-4}$  Bq/Bq. The usage of the high pure  $^{236}\text{Pu}$  allows to determine the origin of Pu in the sample under investigation.

The experimental results of producing radiochemically and isotopically ultra pure  $^{236}\text{Pu}$  and  $^{237}\text{Pu}$  are presented. Optimal conditions for the irradiating the  $^{235}\text{U}(99,993\%)$  targets with  $^4\text{He}$ -ions and for the subsequent separation of Pu isotopes by the electromagnetic mass separator were determined. The ratio of  $^{238}\text{Pu}$  to  $^{236}\text{Pu}$  in the final  $^{236}\text{Pu}$  preparation was equal to  $10^{-5}$  Bq/Bq. The advantages and disadvantages of producing  $^{236}\text{Pu}$  via  $^{235}\text{U}(d,n)^{236}\text{Np}(T_{1/2}=22.5\text{ h})\rightarrow^{236}\text{Pu}$  reaction ( $E_d=18$  MeV) are discussed. The purest reported to date  $^{237}\text{Pu}$  preparation with  $^{236}\text{Pu}/^{237}\text{Pu}/^{238}\text{Pu}$  ratio as low as  $2\cdot 10^{-8}\text{Pu}/1/\leq 3\cdot 10^{-7}$  Bq/Bq was obtained after two successive mass-separations. The results of using these isotopes in environmental and biomedical research are discussed.

# GAMMA RAY SPECTROMETRY MEASUREMENTS RELATED TO THE DECOMMISSIONING OF NUCLEAR FACILITIES

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The paper deals with two aspects of gamma ray spectrometry techniques applied to the area of nuclear decommissioning:

- X ray and low energy gamma ray measurements for the determination of uranium levels on the surface of walls of buildings scheduled for decommissioning. It will be shown that, if the uranium has penetrated the wall surface or is covered by a coating of paint, the mean depth of uranium contamination and the level of contamination may be determined.
- techniques for surveying 'a green field site' following the decommissioning of a nuclear facility to ensure that no significant point sources of gamma rays remain on or below the surface of such a site. Detailed calculations, backed up by field measurements, have been carried out to determine detection limits as functions of gamma ray energy, source depth and survey time for a mobile gamma ray spectrometry survey system.



## ACTIVATION ANALYSIS OF "HOT" PARTICLES FROM CHERNOBYL ATOMIC POWERION

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Hot particles forming in result of explosion or accidents at atomic facilities are very unusual radioactive formations and the real mechanism of their formation is not firmly established yet.

It is assumed that in a majority of cases these particles are the fragments of mechanically crashed fuels containing a whole set or part of fission products. Condensing on aerosols of some fission products is also assumed [1].

Following accidents the temperature is increasing largely, and the environment is characterized by high humidity, a considerable increase in the concentration of different chemical elements, and large temperature gradients. In these conditions not only evaporation of some elements from fuel fragments is possible, but chemical reactions with the formation of complex gas and solid compounds can also take place. It is known [2] that about 70% per cent of radioactive iod burst into the atmosphere exist in the form of  $\text{CH}_3\text{J}$ , and free cesium and iod react with each other and fall out in the form of caesium iodide [3].

Hence, the determination of the composition of hot particles would help establishing the mechanism of their formation and tracing the interaction of fission products with natural elements.

In this work the instrumental version of NAA was used to achieve the purpose.

Hot particle containing samples were made from the probes of soil taken at a distance of 20 to 150 km from the Chernobyl atomic power station or the probes of leave surfaces taken at a distance of 30 to 35 km in 1986 and kept in the laboratory room in a dried form. The irradiation load per sample by

short irradiations (up to 30 min duration) did not exceed  $5 \times 10^7$  rad which is the upper limit for the radiation-stimulated processes of the redistribution of elements between a hot particle and a packing material [4]. At the same time the number of fission events of the uranium in the sample is still insignificant.

The obtained results for some samples are summarized in the Table.

Table.

	a isotope content, g										
	Al $\times 10^{-10}$	Mo $^{100} \times 10^{-7}$	Te $^{130} \times 10^{-8}$	Br $\times 10^{-8}$	V $\times 10^{-10}$	Rh $^{103} \times 10^{-8}$	Cl $\times 10^{-9}$	Na $\times 10^{-8}$	Mn $\times 10^{-9}$	Au $\times 10^{-8}$	U $^{238} \times 10^{-8}$
N 2	6.9	5.7	3.8	2.9	2.4	2.5	1.4	6.7	1.5	2.7	5.29
N 5	4.0	1.8	3.8	1.9	3.2	0.51	1.2	2.8	3.1	1.9	2.26
N 11	5.4	0.56	2.3	1.1	1.6	>0.01	1.0	2.8	0.86	1.6	0.803
N H2	3.2	2.9	3.0	3.4	0.34	0.36	0.55	8.9	1.0	2.0	3.09
H 1	13.0	0.52	1.0	0.98	1.8	0.27	0.26	3.8	2.2	0.79	0.138
H 7	23.0	1.3	1.9	2.4	3.3	0.29	0.37	11.0	1.2	0.07	1.07
H 9	12.0	<0.01	<0.01	0.68	1.2	1.2	0.33	8.5	2.0	<0.01	0.296
H 11	130.0	1.9	1.3	2.9	2.1	1.4	0.27	13.0	11.0	1.1	0.364
H 15	36.0	0.13	2.4	2.0	13.0	0.23	0.36	9.3	0.4	0.23	0.293
eplant particles - N 2, 5, 11, H2 soil particles - N 1, 7, 11, 15											

The summarized data evidence for a considerable enrichment of hot particles with elements having a melting temperature in the metallic or oxide state (Mo, Te) lower than that of uranium by  $10^6 - 10^7$  times.

At the same time an increase in the concentrations of natural elements not forming in the fission of uranium is so large that these elements start determining the mass of the particle. This property is characteristic of the hot particles of soil samples to a larger extent.

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# ACTIVATION ANALYSIS IN RADIOACTIVE WASTE MANAGEMENT STUDIES

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Over the past several years we have employed various aspects in activation analysis in experimental radioactive waste management studies, primarily in environmental restoration and laboratory clean-up. These investigations have included the use of tracers in the study of zirconium and chromium, evaporation technology, magnetically assisted chemical separation and separation of americium in liquid scintillation cocktails. A detailed talk will focus on the various methods used.

# A DOSE AND RISK ASSESSEMENT OF THE NORMAL OPERATION OF A CANDU-600 TYPE NUCLEAR POWER PLANT

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Any human activity involves both advantages and risks. As for Nuclear Power Plants (NPP), where radioactive materials are handled, a special attention to the risk to health due to radioactive exposure is paid. In this paper an assessment of the risk to health in the area surrounding a CANDU-600 NPP under normal operating conditions is presented. The first step in risk assessment is the estimation of the radiation dose received by the public. The radioactive releases predicted in design were taken into account. The estimation of the radiation dose was performed for the following reception means: external exposure, inhalation, food ingestion. Theoretical models for these estimations together with the results from concrete situations are presented. External exposure is considered as due to the radioactive cloud and to radioactive isotopes deposited on the earth. For food ingestion we have taken the food channels into consideration which are important for the studied zone and the characteristics of the zone from an agricultural point of view. The risk assessment was performed using an exponential correlation of dose to health effect, and was expressed in numbers of sickness per year in per cent. The parameters were taken from literature. The multiplicative model, which is briefly presented in the paper, was used for the calculation. Finally, the estimated risks have been compared with the normal numbers of sickness and with other risks from industrial and voluntary practices.

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ENVIROMENTAL MONITORING IN THE URANIUM  
MINING AND MILLING FACILITY AT POCOS DE CALDAS

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\* \* \*

## Natural Occurring Radioactive Material in the Oil Industry

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Scale formation in hydrocarbon plants is a well-known problem. Sometimes it may occur that natural radioactive elements (especially  $^{226}\text{Ra}$ ) contained in the Earth's crust are transported by fluids, coming from an oil reservoir.

In particular cases, chemical and physical mechanisms may cause deposition of radioactive scale, called Low Specific Activity scale (L.S.A. scale). The presence of radioactive substances in tubing, piping, desalters, separators and in other components of plants may reach levels high enough to raise radiation protection problems and to require particular care when performing maintenance operations.

From a radiation protection point of view, two aspects must be considered:

- external exposure of personnel passing through or remaining in the plant areas;
- contamination of workers and work areas during plant maintenance operations.

The results obtained in investigations developed by AGIP S.p.A. in plants located in Italy, North and Central Africa show a wide variety of situations.

In the plants considered, only in a few cases did the external exposure rate reach a level high enough to require adoption of restricted access areas. Nevertheless, the critical aspect concerned the risk of ingestion or inhalation of radioactive materials during maintenance operations.

Dose rate levels in the proximity of the plant's components ranged from background to  $40\mu\text{Sv/h}$ . Analysis performed on L.S.A. scale samples showed  $^{226}\text{Ra}$  concentrations as high as 1.2 kBq/gr and the significant concentrations of  $^{232}\text{Th}$  were found in the same samples.



## PHENOMENOLOGY OF METAL TOXICITY

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The development of analytical techniques leads to the accumulation of new data about the abundance of elements in Nature. New levels of knowledge periodically require a new level of generalization which leads to new attempts to search for some general regularities of the biological role of such elements. Wide application of nuclear physical methods, and especially activation analysis, allowed much new data to be obtained for elements which had earlier been determined in natural (and especially, biological) samples only rather infrequently. That includes such elements as Sc, Cr, Mn, Fe, Co, Zn, As, Se, Br, Sr, Cd, Sb, Cs, La, Ce, J, Au, Th, and U, because using INAA in most kinds of natural samples, these elements are determined independently of the list of studied elements.

The fact that the elemental composition of living organisms is determined by element availability seems to be obvious. Actually, a living organism's elemental composition is given by the composition of the environment (nutrition) with some corrections. For example, for algae, the accumulation coefficient (ratio of concentration of an element in an organism to its concentration in water) increases with the decrease of the element concentration in water. The elemental composition of plants correlates with the composition of the soil solution rather than with soils.

This regularity allows one to try to compare the element toxicity with its concentration in the organism. The concentration coefficient for humans and animals is significantly higher than for other fundamental characteristics. Such a regularity was shown for rats also, as well as for the toxic limitations of water and the atmosphere.

# IN VIVO X-RAY FLUORESCENCE OF HUMAN BONE LEAD: CUMULATIVE EXPOSURE AND ENDOGENOUS RELEASE OF LEAD

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It has long been known that exposure to high doses of lead produces toxic effects in humans. However, the effects of low to moderate exposures are less easily characterized. So it is necessary to develop reliable indicators of exposure in order to determine possible dose response relationships of the health effects of lead. The concentration of lead in whole blood has commonly been used as the indicator of an individual's exposure to lead. Blood lead, however, cannot provide a direct index of either long term exposure or of that component of lead which is most readily available to diffuse through extracellular spaces and present immediate toxic insult to most organs in the body.

Since more than 90% of the lead in an adult is in bone, it was reasonable to presume that bone lead would reflect long term exposure. This was the motivation in developing in vivo measurements of bone lead. Lead in plasma or blood serum is readily diffusible and represents that part of the body lead which presents the most immediate toxic hazard. However, serum lead concentrations are typically of the order of only 1% to 3% of whole blood lead concentrations. Thus, sensitivity of the analytical instrument and possible concentration present difficulties in analyzing serum lead.

Bone lead was first measured in vivo in Sweden by Ahlgren, et al., in 1972, using gamma-rays from  $^{57}\text{Co}$  to excite lead K X-rays. In a subsequent development, Sommerville, et al., showed that a more precise measurement could be obtained using  $^{109}\text{Cd}$  to excite the K X-rays. The  $^{109}\text{Cd}$  system also presented the very significant advantage that the ratio of lead X-rays to coherently scattered 88 keV gamma-rays from the  $^{109}\text{Cd}$  source was independent of source-sample positioning, sample size or shape, thickness of tissue overlying the bone and source strength or duration of measurement. An improved version of this system has been implemented at McMaster. A typical measurement error of 3 microgram per gram of bone mineral can be

been measured by the geochemical technique of isotope dilution inductively coupled mass spectrometry.

A number of studies of lead workers have shown that bone lead does relate strongly to an individual's cumulative exposure to lead. A person's whole blood lead is taken as indicative of their exposure at one point in time. Their cumulative exposure is taken as the integral under the exposure versus time curve, using the blood lead measured at different times during the person's employment history. The integral is approximated using simple numerical algorithms. Bone lead correlates strongly with this cumulative blood lead index.

Subjects who have ceased working with lead commonly have higher blood lead concentrations than otherwise similar people who have never worked with lead. The amount by which their blood lead exceeds that of the referent group correlates with their bone lead. This is interpreted as indicating the release of lead stored in bone into blood. This endogenous exposure can be the dominant source of lead for someone who has worked with lead for several decades, to the extent that up to 80% of a retired lead worker's blood lead can be lead released from bone stores rather than new environmental exposure.

A recent study in which serum lead was measured, as well as lead in the tibia and calcaneus and the whole blood lead, found that serum lead correlated surprisingly strongly with bone lead and that the ratio of serum lead to whole blood lead increased with increasing bone lead. This ratio of serum lead to whole blood lead was seen to depend on bone lead rather than on blood lead. The implication of this observation is that the partition of lead between serum and red cells depends on whether the source of the lead is internal or external to the body. For internal sources of lead, the ratio of serum to whole blood lead concentration was about 3%. For external exposure the ratio was a little over 1%. If serum lead is taken as indicative of the most readily toxic component of lead, then this finding may mean that endogenous exposure is toxicologically more significant than has previously been supposed.

## PECULIARITIES OF AIR POLLUTION IN INDUSTRIAL SITES OF RUSSIA AND TASKS OF ENVIRONMENTAL EPIDEMIOLOGY

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The average annual concentrations of widespread pollutants in air of Russian industrial sites are close to those characteristics of developed countries (TSP 200-500  $\mu\text{g}/\text{m}^3$  in ambient air of metallurgical cities and 100-200  $\mu\text{g}/\text{m}^3$  in other cities and towns; sulfur dioxide - 30-50 and 10-30; nitrogen dioxide - 40-100 and 10-30  $\mu\text{g}/\text{m}^3$ , respectively). Average annual concentrations of carcinogens are higher than the MACs: the level of BaP is 7-20  $\mu\text{g}/\text{m}^3$  in atmosphere of the cities containing steel-casting and aluminum industries, that of vinylchloride - 5  $\mu\text{g}/\text{m}^3$  near mining chemistry enterprises; that of benzene is 100-1,000  $\mu\text{g}/\text{m}^3$  near synthetic rubber production; that of nickel - 0.5-2.3  $\mu\text{g}/\text{m}^3$  and that of lead 0.3-0.7  $\mu\text{g}/\text{m}^3$  near smelting enterprises. The MACs were demonstrated for the pollutants released by pulp industry (hydrogen sulfide level - 15-20  $\mu\text{g}/\text{m}^3$ ), by the aluminium industry (hydrogen fluoride amounts - 6-8-  $\mu\text{g}/\text{m}^3$ ), by petrochemical plants (hydrogen sulfide concentrations - 8-64  $\mu\text{g}/\text{m}^3$  and acetaldehyde level - 15-5-  $\mu\text{g}/\text{m}^3$ ).

Environmental epidemiologic studies (geographic, group, and case-control studies) evaluate mostly the main changes in the health of children (morbidity, physical development, respiratory functions, levels of Ig A, M, G and other parameters). For example, respiratory morbidity of the children in some 20 towns situated near metallurgy and chemical enterprises appeared to be 1.5-2 times higher. The number of oncoepidemiological studies is few, and those are mostly geographic. Increased mortality from lung cancer was revealed in towns and cities with copper and nickel enterprises (Norilsk, Karabash, Ufa-ley, Rezh) and aluminium production (Kamensk-Uralsky, Krasnoturyinsk). Analytic epidemiologic studies covering the carcinogenic effects of vinylchloride, benzene and asbestos on the general pollution are absent. Application

of the European Register Eurocat is planned for evaluation of the incidence of congenital abnormalities. Internationally unified methods should be applied to the studies concerning accumulation of toxic elements (dioxines, BaP, heavy elements) in humans.

## MULTIELEMENT ANALYSIS OF HAIR FROM METALLURGY INDUSTRY WORKERS

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The concentration of Al, Co, Cu, Fe, Mg, Mn, Sb, Se, V and Zn in scalp hair samples from 20 metallurgy industry workers (10 men and 10 women) as well as from 59 normal control subjects (32 men and 27 women) were determined by the INAA method.

The hair samples, washed according to the IAEA recommended procedure, were irradiated together with appropriate standards at the VVR-S reactor in Bucharest. Neutron fluxes for long and short irradiations were of  $1.4 \cdot 10^{13}$  and  $3 \cdot 10^{12} \text{ cm}^{-2}\text{s}^{-1}$ , respectively. Gamma ray measurements were carried out using a high resolution HPGe detector connected to a multichannel analyzer.

The normality of the statistical data distributions was checked for all elements: Zn and Se concentrations follow a normal frequency distribution, while for the other elements the concentration values are distributed log-normally, to a fair degree of approximation. Therefore, geometrical means were used to describe the average element content, except for Zn and Se.

The average values of the element concentrations in the hair from the metallurgy industry group (exposed subjects) and for the normal control group are presented in Table 1.

Table 1. Element concentrations in human hair samples

Element	Al[ppm]	Co[ppb]	Cu[ppm]	Fe[ppm]	Mg[ppm]
Exposed	22.8/1.50	148/3.06	15.3/1.53	76/1.87	193/1.90
Control	14.7/1.50	20/2.21	11.5/1.43	24/1.80	123/2.10
Element	Mn[ppm]	Sb[ppb]	Se[ppb]	V[ppb]	Zn[ppm]
Exposed	1.72/2.24	69/2.01	469+/-93	54/1.81	239+/-90
Control	0.75/2.25	46/1.96	489+/-150	30.6/1.94	172+/-29

Except for Se, all the elements investigated have significantly higher concentrations in the scalp hair of the exposed group in comparison with those of the control group.

For Al, Mg, Se and Zn the concentration levels have been found to be higher in the hair of younger persons, but these differences were not statistically significant.

Except for Se, all of the elements present higher concentration values for the female group than for the male group, but only Co, Fe, Mg and Zn differences are statistically significant.

TOTAL AND METHYL MERCURY LEVELS IN SCALP  
HAIRS OF LYING-IN MOTHERS AND THEIR NEWBORNS  
LIVING IN THE SECOND SONG-HUA-JIANG DISTRICT BY  
NUCLEAR AND OTHER TECHNIQUES

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The Second Song-Hua-Jiang river system is one of the mercury-polluted areas in China in the 1970's. In 1982 the Hg content in fish there was high up to  $0.98 \mu\text{g/g}$ , significantly exceeding the allowance level of  $0.5 \mu\text{g/g}$  set by WHO and Chinese Hygiene Ministry. Since then, a number of measures had been taken to control and eliminate the Hg pollution in this district. In order to examine the present level of Hg pollution after the long-term environmental protection activities, a well-planned survey of total and methyl Hg contents in scalp hair samples from thousands local fishermen and their family members were carried out in 1993 and 1994. In this work the total and methyl Hg contents in hair samples from 29 pairs lying-in women and their newborns living there were determined by INAA and gas chromatography, along with 17 pairs maternal and infant hair samples taken from a Hg-unpolluted area as a control.

Our results indicate that the average Hg contents in the maternal and infant hair samples from the Second Song-Hua-Jiang district are  $0.79 \pm 0.41 \mu\text{g/g}$  and  $0.78 \pm 0.56 \mu\text{g/g}$ , respectively, while from the unpolluted area  $0.38 \pm 0.23 \mu\text{mg/g}$  and  $0.51 \pm 0.18 \mu\text{g/g}$ . Thus, a conclusion can be drawn that the Hg pollution still remains, although the pollution level has been substantially alleviated. The average methyl Hg contents in the hair samples show the similar tendency to the total Hg.



Meanwhile, we have also determined the Se contents in the hair samples from the above two areas by INAA. Statistically, the Se contents in infant hair samples are higher than those in maternals and the rations of Se/Hg in almost all samples are close to one, which indicates a certain combination of Se with Hg in biological tissue.

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**INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS OF  
ESSENTIAL AND TOXIC ELEMENT CONTENTS IN THE  
WHOLE-DAY DIETS OF CHILDREN AND ADOLESCENTS  
WITHIN THE KALUGA REGION TERRITORIES WHICH  
SUFFERED FROM THE CHERNOBYL ACCIDENT**

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It is shown that the main etiological factor of various diseases, syndromes and pathologic states is an excess, deficiency or imbalance of the trace element intake by a human body. Children seem to be the most sensitive to each change in trace element homeostasis. The cases of undesirable trace element intake consequences can apparently increase against a background of additional, unfavourable environmental influences (availability of different organic and inorganic toxicants, high radiation level, etc.). So, the quality control of children's whole-day diets assumes an urgent importance within the regions which suffered from the Chernobyl accident.

Instrumental neutron activation analysis was used to estimate the amounts of Ag, Br, Ca, Cl, Co, Cr, Cs, Fe, Hg, K, Mg, Mn, Na, Rb, Sb, Sc, Se, Sr, and Zn in the whole-day diets of children and adolescents. Diets were chosen from the canteens of kindergartens, boarding schools and technical colleges within the south and south-west territories of the Kaluga Region where the radionuclide contamination ranged up to 15 Ku/km<sup>2</sup>.

A deficiency of Ca and Zn was found in the whole-day diets of children and adolescents aged 7-18 years. The Ca intake is only 212 mg/day which is about 5 times lower than that for developed countries. The Zn intake is about 6.8 mg/day - 2 times lower than the level given by WHO.

# AN ACCURATE DETERMINATION OF CADMIUM IN BIOLOGICAL MATERIALS BY A RADIOCHEMICAL VERSION OF NEUTRON ACTIVATION ANALYSIS

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A new improved version of the method for determining the presence of cadmium in biological materials by radiochemical NAA, based on selective post irradiation separation of Cd using the ion exchange resin, Retardion 11As, has been elaborated. Retardion 11AS is an amphoteric type exchanger. It contains anionic, strongly basic quaternary ammonium groups and an equivalent amount of cationic, weakly acidic carboxylate groups. Cadmium, depending on the composition of the external solution, exists either as anionic complexes or cationic ammine complexes and can be taken up both by cation and anion exchange groups of the resin. Cadmium is retained by the resin from 2M HCl and remains on the column during washing with 2M HCl, 1M NH<sub>3</sub> + 0.1 M NH<sub>4</sub>Cl solutions while most of accompanying elements are eluted. Zinc which also stays on the column during the separation steps is separated from Cd by elution with 0.1M HCl and/or 0.05M NH<sub>3</sub> + 2.0M NH<sub>4</sub>Cl solutions. Finally, cadmium is rapidly and quantitatively stripped with the aid of an 8M NH<sub>3</sub> + 1.0M NH<sub>4</sub>Cl solution. The elaborated method was further verified by determining the cadmium content in six certified biological materials using neutron activation analysis. The above described procedure assures a high radiochemical purity of the cadmium fraction. Determination factors obtained for Mo, Sb, Na, Co, Zn amounted to 10<sup>3</sup> - 10<sup>5</sup>. Detection limit for Cd was 0.5 μg/kg. Analytical results show good agreement with the certified data.

# Assay of Radionuclides in Ceramics by Neutron Activation Analysis and Low-level Gamma-ray Counting.

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Ceramics contain trace amounts of radionuclides. If such material is used as medical implant the activity should be limited to a maximum value and it is a necessity to measure the radioactivity. Neutron activation analysis was carried out for the determination of Uranium, Thorium and some other elements. The activities of Potassium and the decay products of U and Th were determined by direct gamma-ray spectrometry using a Ge-NaI(Tl) detector system with passive shielding and anticoincidence counting mode for background reduction. The methods are described and results are reported.

**Method of the Investigation of the Air-Borne Radioactive  
Contamination Using Ass-500 Station**

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# SELECTION AND TREATMENT OF BIOLOGICAL OBJECTS: INFLUENCE ON THE RESULTS OF INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

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The analysis of medical and biological samples depends on several specific requirements. Those basically concern the regulation of the set of instruments used for taking samples of fluids and tissues, the conditions for conducting biopsy, operation of autopsy, the mass limit of the material obtained, and the necessity for extremely high accuracy of the results.

There are different stages of selection, conservation and treatment for human fluid and tissue samples obtained under clinical conditions. We studied the influence of these stages on the results of instrumental neutron activation analysis (INAA). The aims of study were to evaluate:

- the contamination degree of a biological object during its sample selection using an ordinary medical set of instruments;
- the dynamics of the sample mass losses from the moment of sample taking to its weighing, at the expense of evaporation;
- the dynamics of increasing the dried sample month at the expense of air moisture absorption;
- the levels of possible element loss during frozen sample drying for 100° C and 150° C drying conditions;
- the levels of possible element loss during dried sample ashing;
- the levels of possible sample dry mass losses during long-term irradiation in a nuclear reactor at the expense of thermal and radiation influence;

- the degree of influence of polyethylene and different kinds of absorbing and packing material papers on the trace analysis;
- the representativeness of biological samples weighing from several milligrams to several tens of milligrams.

The data was obtained to provide a basis for developing special medical instruments for selecting samples, choosing optimum drying conditions for conserving and treating samples of medical and biological objects for INAA.

# USE OF ACCELERATORS IN ECOLOGICALLY SAFE INDUSTRIAL TECHNOLOGIES THE NATIONAL PROJECT

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Ionizing radiation is an effective help in developing ecologically safe technologies. Accelerators play an important role in the creation of such technologies. There is large experience in this field, but mostly in the industrialized countries (USA, Japan, Germany). Unfortunately, the countries surrounding closed seas (The Mediterranean, Black and Azov seas) do not use modern accelerator based technologies.

The aim of this Project is to unify the possibilities available in those countries to use accelerators for environmental protection, in particular, for protection of the seas from technological releases. The solution of this problem for the internal seas will be an important contribution to the protection of the world's oceans at the same time.



# MONITORING OF $\alpha$ -EMITTERS BY NUCLEAR TRACK DETECTORS

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The national project for monitoring  $\alpha$ -emitters and fissile heavy elements is based on the use of nuclear track detectors - a universal and very useful tool for  $\alpha$ -particle and fission fragment registration. These detectors were very helpful in the preliminary investigation of pollution from the Chernobyl accident.

The project includes the following stages:

- development of industrial production of nuclear track detectors in the Ukraine from diethylglycolbisallicarbonate polymers;
- creation of a chemical laboratory for etching irradiated detectors;
- organization of the mass viewing of detectors by optical microscopes;
- creation of the program for processing the track information.

# MEASUREMENTS OF LOW BORON CONCENTRATIONS IN RIVER SEDIMENT SAMPLES (ARGENTINA) BY THERMAL-NEUTRON-PROMPT-GAMMA-RAY SPECTROSCOPY METHOD

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The twenty four river sediment samples from Argentina were measured on the external thermal neutron beam from the horizontal channel of WWR-M reactor (PNPI, Gatchina) according to the method described in [1].

The samples with weights about 140 mg and the boron reference standard were placed as targets in the thermal neutron beam with the intensity of  $2 \cdot 10^7$  n/(cm<sup>2</sup>)s formed by the neutron guide. The sample holder provides the identity of irradiated volumes of the targets.

The  $\gamma$ -spectra were measured with coaxial Ge(Li) detector. A measuring time for one sample was about 1-2 hours. The boron standard was prepared in *Isotope Chemistry Group* of PNPI as the mixture of dry  $H_3BO_3$  and  $MgCO_3$  ( $93.5 \pm 2.3$  ppm of boron).

The 478 keV  $\gamma$ -line from  $^{10}B(n,\alpha)^7Li$  reaction was used as analytical one. The boron concentration in the samples were calculated as

$$C_x^B = C_{st}^B (S_x^B P_{st} h_x) / (S_{st}^B P_x h_{st}),$$

where  $C_x^B$  and  $C_{st}^B$ ,  $S_x^B$  and  $S_{st}^B$ ,  $P_x$  and  $P_{st}$ ,  $h_x$  and  $h_{st}$  are the boron concentrations, the analytical line counting rates, the weights and heights of the samples under investigation and the standard, respectively.

The values of the concentrations measured were ranged from 10 to 70 ppm with errors from 1.5 to 8 ppm. The sensitivity of our installation is very close to value published in the Grenoble group paper [2].

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# INVESTIGATION OF THE EAST DOBROGEA WATER RESOURCE POLLUTION USING NEUTRON ACTIVATION ANALYSIS

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The distribution of some elements in the ground and surface water of the East Dobrogea has been investigated for two years.

Twenty six representative points (wells, catchments, springs surface courses and Lake Siutghiol - the main discharge zone of the aquifer) located in two kinds of carboniferous rocks - Mesozoic and Sarmatian - were selected for the analysis.

Instrumental Neutron Activation Analysis was used for the trace element concentration determination. Seventeen elements have been found: Sc, Cr, Fe, Co, Zn, As, Se, Br, Sr, Zr, Sb, Cs, Ba, Hg, Ce, La, Ag.

The results of the analysis show that the ground water from the Mesozoic rocks has a smaller contact with the polluting agents compared to the Sarmatian one; the element concentrations do not exceed the maximum allowed values for drinking water.

For two of the surface water courses - Tatlageac Brook and Albesti Brook - the results obtained showed increased values for Cr, Co, As, and Se due to human activities. No connection between these surface courses and the ground water has been found.

Neutron Activation Analysis proved to be a useful tool in water pollution study.

# ON INAA MONITORING THE MICROELEMENT COMPOSITION OF THE NATURAL SYSTEM: RESERVOIR - BOTTOM SEDIMENT - PLANKTON - OCCUPANTS OF THE RESERVOIR

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The content of different elements in the samples of water, bottom sediment, plankton, *Dreissena* mollusks, and fish taken from a disposal zone of cooling water for a hydro-power station and from the control zone was determined by the INAA method. Young and grown-up mollusks were investigated separately. Investigations of fish samples were carried out in comparison with samples of same fish from other reservoirs of Belarus.

In spite of the fact that the content of microelements in water samples from the disposal and control zones differed slightly and was close to the content of these elements in open reservoirs, an increased content of Fe, Sc, Co, Hf, Cr was detected in bottom sediment samples and, especially, in plankton samples taken from the disposal zone. The Hf content in plankton in the disposal zone was an order of magnitude larger than in the control zone. In the tissues and shells of mollusks from the disposal zone an increase in the content of already ten elements (Fe, Sc, Co, Eu, Cs, Cr, Hg, Ba, Rb, Hf) was detected. Among these elements were Cr and Hg which are the toxic elements. It should be noted that Cr, Hg, Ba and Rb were accumulated in shells only, Hf in tissues only, and Fe, Sc, Co, Eu, Cs were accumulated both in the tissues and shells of mollusks. For a majority of these elements the increase was by 1.5 to 2 times. A much higher increase in the content of Rb (by 3 times), and Hg (by 4 times) in the shells, and of Cs (by 3 times) in the tissues of mollusks was noted.

Age variations in the content of microelements were also noted for mollusks. With age the concentration of Fe, Sc, Co, Ba, Hg, Cs is increasing in their shells, and of Fe, Sc, Co, Hg, Eu, Cr, Cs in tissues. It should be noted that this tendency has a more expressed character for the occupants of the disposal zone. On the contrary, the content of Se decreases with age, especially, for the occupants of the disposal zone. The concentration of Zn, Cr, Sb remains constant.

An analysis of fresh-water fish whose activity is higher has noted no noticeable changes in the microelement content in comparison with the same fish from other reservoirs.

APPLICATION OF THE NAA METHOD IN OCEANOLOGY ON  
THE BASIS OF THE EXAMPLE OF THE "KOMSOMOLETS"  
ATOMIC SUBMARINE

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# INFLUENCE OF BIOCHEMICAL BARRIERS ON THE HEAVY METAL AND RARE EARTH ELEMENT FLUX IN ESTUARY ZONES OF THE PACIFIC SHORE

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Ocean bottom sediments represent the closing link in contemporary heavy-metal (HM) and rare earth element (REE) biochemical cycles. The elemental composition of these sediments in marginal-sea shelf zones is a product of natural and man-made factors. The content of elements in the surface layer of the bottom sediments depends on the relation between natural and man-made components of the liquid and solid runoff, on geochemical features of the source basin, on biochemical processes at the river-sea barrier, on the mobilization conditions and on the grain size composition of suspensate.

In the case of estuaries in the temperature zone (Razdolnaya R. - Gulf of Amur), in the intermediate zone (shelf of the East China Sea) and in the equatorial climatic zone (Mecong R. - South China Sea), some main processes have been studied that cause spatial variations in the element concentrations in the suspensate and bottom sediments.

Neutron activation analysis and also atomic absorption spectrometry, atomic emission with ICP and X-ray fluorescence methods have been used for this aim.

Several interesting facts have been discovered and it was found, for example, that zones of impact on the sediment composition (Razdolnaya R. and Gulf of Amur) have identified that the decisive factor in this region is biological processes; the bottom sediment on the shelf of the East China Sea near the Yangtse are represented mainly by weathering products from terrigenous rocks, whose compositions have been appreciably altered by diagenesis;



the main process at the mouth of the Mecong and the adjacent part of the South China Sea is the stirring of the upper layer of the bottom sediment due to tides, etc.

The main conclusion was made that when one evaluates the effects of human action on suspensate compositions in estuaries, one should allow for the extensive transformation occurring during passage through the biochemical barrier in the river-sea system.

# DEVELOPMENT AND APPLICATION OF INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS TO ATMOSPHERIC AEROSOLS, SOILS AND BOTTOM SEDIMENTS

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Monitoring the state of the environment and its protection are coupled with the processing and development of new analytical techniques for detecting pollution. The role of neutron activation analysis (NAA), among other methods, is important due to its low detection limits, small sample masses taken for analysis and sample survival without a chemical destruction.

For more complete and accurate determinations of specified groups of elements, the processing by special techniques of instrumental NAA was carried out in the framework of the environment protection problem.

The proposed method enables estimation of the main factors, by instrumental NAA, of environmental samples such as the components of aerosols, soils and bottom sediments. The technique is based on increasing the ratio of signal/background by the means of calculating the optimal conditions of the experiment in model gamma-spectra, including irradiation conditions in the reactor (with thermal neutrons and neutrons passing through the cadmium and boron filters), samples mass, time of cooling and measuring. The technique was tested by the analysis of standard samples of soils (SO), bottom sediments (SDO) and can be used for INAA of a variety of compositions of environmental samples including unknown samples.

The optimal conditions of INAA are determined by using the program package developed by the authors which applies numerical gamma spectra modeling to specified composition samples.

## MATTER FLOWS FROM TECTONIC ACTIVITY ZONES BY USING NUCLEAR PHYSICS METHODS

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The aerosol and radon flows in tectonic activity zones from magmatic, mud and other sources are very significant as natural processes of Earth degassing. The difficulty of their estimation is connected with small sized particles, and irregular distribution over the Earth's surface and time.

The deeply generated aerosols and gas flows are connected with the total dynamic of the Earth, and are natural consequences of the metastable Earth model.

The aerosol fraction flows of organic and inorganic substances are related to the nature of the tectonically deformed regions. It is very difficult to estimate the contents of the flows at present.

Modelling experiments were carried out for hot gas flows. Gas composition was similar to volcano gases and included dispersed basalt dust. The distributions of microelements and some organic compounds have been studied. It was shown that in hot gas flows the microelement composition of the gas changes, and a significant accumulation of organic compounds was observed.

Therefore, the experiment results show that it may predict the presence of significant contents of organic compounds which may be detected in an aerosol fraction related directly to gas flows in volcano. The detection of these organic compounds is possible with the use of specific sampling probes, placed on airborne carriers. The aerosol flow distributions may be used for the determination of spread regions and ring structures.

The many deformations of the Earth are concerned directly with the origins of the earth crust fracture sites. It is expedient to carry out a districting of the aerosol flow sources on the basis of seismic districting using the data on magma and mud volcanoes, underwater and surface thermal sources. For all the groups of aerosol sources, we present the distributions of metals for a microdisperse fraction, obtained by NAA and RFA methods.

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**MONITORING OF METAL ENRICHED AEROSOLS FROM  
THE AVACHA VOLCANO (1990-1994). AEROSOL  
PRECURSORS OF VOLCANIC ERUPTION**

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To study the composition of aerosols accumulated by snow on the Koselskii and Zavaritskii glaciers near the volcano crater, an analysis of soluble and insoluble aerosol fractions was made for the long sampling period from 1989 to 1994. Concentrations of microelements in solution have been determined by the method of atomic emission spectroscopy. Graphs of microelement concentrations in dependence on snow layer have been prepared. It was shown that besides the natural concentration maximum, related to the volcanic eruption in January 1991, another concentration peak for most of the elements was observed in the snow layer which had fallen a month before the volcano erupted. It should be noted that this volcanic eruption was not predicted with the help of ordinary seismic methods. Microelement analysis was made for Mg, Mn, Sr, Cr, Cd, Co, Be, Fe, Cu, Ag, Ti, Zr, Ba, Ni, V, Si, Au, Sn, Sc, Ta, B, Nb, and others. These data are compared with measured data on the amounts of metals in aerosol fractions emitted by volcanoes, obtained in 1981 (Alekseev V.A. and Alekseeva N.G., *Volcanology Seismology*, 1989, 3, p.30-37.

## DETERMINATION OF MICROQUANTITIES OF Be IN SOLIDS WITH GAMMA-RAYS

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A new technique of determining Be content in samples using  $4.6 \div 6$  MeV  $\gamma$ -rays is proposed. This energy range is sufficient the neutron from the  $^9\text{Be}$  nucleus to be knocked out and the 2.9 MeV level of the  $^8\text{Be}$  nucleus to be excited. 1.5 MeV  $\alpha$ -particles are registered by means of CR-39 or cellulose nitrate detectors. Alpha particles produced in the reaction of  $\gamma$ -rays with Li nuclei which are contained in samples may cause the background. The latter is excluded by heating of samples before irradiation for Li molecules to be evaporated. B nuclei do not produce  $\alpha$ -particles at the energy range of  $\gamma$ -rays presented above.

The results obtained in the experiments of determining Be content in natural beryl crystal samples are in good agreement with their known chemical composition. Microparticles containing Beryllium ( $10^{-3} - 10^{-4}$  g/g) were found in soil samples from the Ust-Kamenogorsk ore deposit region. The sensitivity of the method is  $10^{-6}$  g/g of Be in solids.

# THE INTEGRAL BIOLOGICAL ESTIMATE OF THE STATE OF THE ENVIRONMENT AROUND THE TOWN OF CHAPAEVSK

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The essence of the approach to estimating the health of an ecosystem by the health of the living beings inhabiting it consists of an integral (using a complex of different methods) estimation of homeostasis whose violation initiates the breaking down of the entire survival system of organisms (including the appearance of terragenic, mutagenic, and cancerogenic effects). The obtained data can be also used to characterize the favorableness of the environment to man.

To estimate the health of the environment around the town of Chapaevsk, the ground and water ecosystems were differentiated. Characterization of the state of the ground ecosystem was performed with model plants (warty birch and meadow clover) and mammals (red field-vole and small mouse) at 9 basic sites. The analysis of plants involved investigation of the evolution stability and photosynthesis intensity. The mammals' homeostasis was estimated by the complex of morphological (evolution stability), genetic (cytogenetic homeostasis characterizing the mutagenic activity by the frequency of chromosome aberrations and micro-nuclei), physiological (growth rhythms) and immune factors. The analysis allowed us to characterize the situation in the resort areas near Jasnaya Polyana and the Lake Ilmen as satisfying the norm. At the same time the situation near the main industrial plants of the town (ChZKhU and Polymer) was estimated to be on the level of 3-4 on the 5-point scale of deviations from the norm.

Characterization of the water ecosystem state was carried out at 5 basic sites using model fish (bream and roach) and amphibians (lake frog). The integral estimation of homeostasis included an analysis of the evolution stability (with the pathologic and histogenic characteristics of the reproductive systems) cytogenetic homeostasis (with the characterization of mutagenic activity on the genetic level, the Aims test, and on the chromosome level, the micronuclear test) and of the immune status. In addition to natural investigations, bio-tests of water probes were carried out (by means of a general test of crustacea for toxicity and a test for mutagenic activity by analysing the frequency of sister chromosome exchanges in fish). The analysis has demonstrated strong deviations from the norm as a result of pollution by the town's plants (against the general background of the unfavorable ecological situation in the investigated region). In between dams and near dumping areas the state of the water ecosystem corresponds to 4-5 on the 5-point scale of deviations from the norm.

The observed serious deviations from the norm, which are evidence of great changes in the health of the ecosystem, were observed against the background of the same or even increased bio-diversity in the vicinity closest to the pollution sources.

The obtained data are evidence of good prospects for the use of the method of integral biological estimates of the medium health (by homeostasis of representative living organisms) to reveal the influence of an entire complex of impacts (including the effect of the general anthropogenic load, effect of different chemical compounds, etc.).

The work was performed by a collaboration of specialists from 4 institutes of the Russian Academy of Sciences, 2 institutes of the Russian Academy of Medical Sciences and from the Moscow State University.



## NEUTRON ACTIVATION AND OTHER NUCLEAR PHYSICS METHODS FOR ANALYSING NATURAL MEDIA FOR TOXIC ELEMENTS

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Estimation of the current pollution level of natural media (water, air basin, soil, biomaterials) by toxic elements was a component of the complex investigation of the ecological state of the territory occupied by the town of Chapaevsk. In natural media, distributions and migration paths of toxic elements of the 1st hazardous class, such as arsenic, mercury, lead, cadmium, selenium, etc., were investigated first.

Neutron activation analysis was used to determine an extensive group of elements: arsenic, mercury, selenium, cadmium, antimony, zinc, chromium, cobalt, lanthanides, thorium, uranium, tungsten, scandium, bromine, etc., from aerosol filters, atmospheric fallouts (snow cover), water suspensions, soils, agricultural products, vegetation, children hair.

X-ray fluorescence analysis was applied to estimate the pollution of soil and soil cuts by arsenic. To perform an express multi-element analysis of water and the liquid phase of fallouts (snow cover), a method for concentrating "heavy" metals: vanadium, nickel, chromium, cobalt, lead, copper, zinc, iron, cadmium, on the DETATA absorbing filters in field conditions was developed and used in combination with an analysis of absorbed elements by the X-ray fluorescence method.

On the basis of the data obtained by nuclear physics methods distribution maps of separate elements over the air basin of Chapaevsk and the ground layer in the town and adjacent territories were plotted. Element migration paths and accumulating sites were also traced, and seasonal changes in the pollution intensity of air basin, water, etc. were established.

## CHEMICAL AND ANALYTICAL BASIS FOR COMPLEX ESTIMATING THE ECOLOGICAL STATE OF URBANE TERRITORIES

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Chemical and analytical basis for carrying out the complex estimate of the ecological state of urbane territories includes all methods from the arsenal of modern analytical chemistry. Their particular use depends, to a great extent, on the character and composition of dominating pollutant sources, as well as on the intensity of these sources. In urbane territories the following main sources of pollutants can be indicated:

- industrial plants in the town or around it;
- town survival systems (thermal power stations, boilers, etc.);
- transport industry and roads;
- regional background pollution.

The specificity of the industry in Chapaevsk, as well as the fact that residential areas are located in the close vicinity to industrial areas, determine priorities in contamination of the residential and resort areas of the town.

To have the complex estimate of the current state of pollution of the town's territory all natural media were tested: atmospheric air, soil, water (surface and drinking), and vegetation.

At present, 95% of an extensive list of ingredients under control are the organic compounds with a sufficiently complicated structure. Volatile organic compounds, including chlorine-, sulfur-, and nitrogen-containing compounds, were identified and analysed using the methods of gas chromatography and chromatomass spectrometry. Benzaperene was determined by the method of low temperature luminescence. Chlorine-organic pesticides were determined

by the method of gas-and-liquid chromatography with electron capture detection. Petroleum products were analysed using the weighing and extraction spectrophotometry method in the ultra-violet interval.

Over 100 organic compounds of different hazardous classes were found and identified.

The content of the usual components of the air basin: oxides, nitrogen, carbon, sulfur, formaldehyde, chlorine, etc. was controlled using a standard set of methods of chemical analysis.

Estimation of the pollution level of all natural media, vegetation and some biosubstrates by toxic microelements: arsenic, cadmium, lead, antimony, mercury, etc., was performed using a complex of physical, physical and chemical, and chemical methods of analysis. Gas-mercury and gamma-spectrometric surveying of the town area and adjacent territories was carried out.

The analytical material is presented in the form of maps, diagrams and cuts which characterize the ecological state and pollution level over the town.

ESTIMATES OF BACKGROUND CHANGES IN THE  
MICROELEMENT COMPOSITION OF  
BIOLOGICAL OBJECTS

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The main factors which provoke changes in the microelement contents of biological objects were determined. The influence of the landscape, climate, and seasonal factors, as well as analytical method uncertainties, on the microelement composition of biological objects was estimated. Regularity of changes in the microelement content of plants, and human hair and nails was demonstrated.

## TIME-SPATIAL CHARACTERISTICS OF RARE-EARTH DISTRIBUTION IN ATMOSPHERIC AIR

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Information on concentration levels of rare-earth elements (REE) in aerosols of the air of background areas and in zones under industrial waste impact was generalized and systematized. We considered:

- REE distributions by the size of the aerosol particles they are concentrated on;
- peculiarities of REE distributions from different types of pollution sources;
- changes in the REE concentrations in time by using different calculation coefficients.

# RADIOACTIVE FALLOUT AFTER THE CHERNOBYL ACCIDENT: ELEMENTAL AND RADIONUCLIDE COMPOSITIONS OF HOT PARTICLES

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Hot particles with high specific activity are an essential fraction of radioactive fallout after the Chernobyl accident. The degree of radioactive contamination of the environment is determined mainly by the activity level and physico-chemical properties of particles. Knowledge about these characteristics is needed for a correct evaluation of the ecological state in the contaminated regions. The purpose of this study is the simultaneous determination of the hot particles' elemental composition and the isotopic ratio of uranium using INAA and radionuclide composition by gamma-spectrometry.

It was found that the uranium content in the analyzed particles varied from 0.0006 to 26%. As admixture elements, the particles also contained Na, Sc, Fe, Co, Cr, La and Sm. These elements are typical for the soil from which the particles were extracted. In some particles technological elements, such as Ga, Ba and Sb, have been detected at a level up to hundreds of mg/g. The uranium isotopic composition in the hot particles (0.4 - 2.6 %  $^{235}\text{U}$ ) corresponds to that of nuclear fuel with different degrees of burn-up. Two particles with anomalously high enrichment of  $^{235}\text{U}$  (7.9 and 22%) have been found. These values are close to those for the nuclear fuel ultrafine fraction detected in the soil by some investigators. From summary data, an empirical formula for calculating the  $^{235}\text{U}$  content in particles without standards has been derived.

At present only the activity of the long-lived radionuclides,  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{125}\text{Sb}$ ,  $^{154}\text{Eu}$  and  $^{144}\text{Ce}$  ( $T_{1/2} = 284\text{ D}$ ), can be measured. These data were also used for determining some additional information about the conditions and processes of hot particle formation.

## URANIUM WASTE-DUMPS - POLLUTION SOURCES OF THE ENVIRONMENT

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The exploitation process of uranium deposits implies a storage stage, at the surface, in the form of uranium waste-dumps. Being stored at the surface, in conditions characterized by low temperature and low pressure, by the free movement of aqueous solutions and by the presence of large quantities of free  $O_2$  and  $CO_2$ , the uranium areas undergo changes; as a result, the uranium in these areas will be redistributed in various dispersion media: soils, waters, sediments, vegetation. The extension of the area exposed to the environment agents mentioned above, an extension caused by the exploitation process, amplifies the phenomenon of uranium dispersion from the waste-dumps to the surrounding areas. For protection, some waste-dumps were covered by a concrete layer; others, were covered only by a dead rock layer. The analysis performed on the samples from the area of the Tulghes uranium deposit, in the Oriental Carpathians, showed an accentuated uranium dispersion from the waste-dumps to the surrounding areas and to the populated areas. In the case of waste-dumps covered only by a deadrock layer, the uranium dispersion process is considerably slowed down, almost to a stop. The possibility of a soil strata formation on the waste-dumps allowed the growth of vegetation, such as fir-trees. The analysis clearly showed the role played as a protection barrier by this vegetation (fir-trees) against the uranium dispersion from the waste-dumps to the environment.

# ENVIRONMENTAL PROTECTION AGAINST NUCLEAR ACCIDENTS AT THE DEMOLITION OF BLAST FURNACES LABELLED WITH RADIOACTIVE ISOTOPES

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The monitoring of blast furnace refractory lining wear is an interesting method for extending the furnace operating period. This method has two stages: the first, the labeling of strategic locations inside the furnace with sealed trace sources such as  $^{60}\text{Co}$ ,  $^{110\text{m}}\text{As}$ ,  $^{204}\text{Tl}$ ; and the second, the detection of radiation emitted by these sources outside the furnace (radiodosimetry) or by samples taken of pig iron or slag (spectrometry) produced when these sources are dissolved in metal or slag, which can appear as a contamination of these materials. In Romania, the legally allowed contamination of pig iron is 0.74 kBq/kg for radionuclides having a half-life of  $T_{1/2} < 1$  y, and 0.074 kBq/kg in the case of  $^{60}\text{Co}$ . The profile of the state of wear is thus determined into two ways: by a diminution of externally detected radiation and by its appearance in the pig iron or slag samples.

When the furnace working period has expired, the remaining used brickwork is destroyed by mechanical tools and other means, and finally, by explosives. At this moment, it is necessary to reseal the residual sources, initially sealed, but after many years of work in heavy conditions (high temperatures, mechanical stresses, atmospheric corrosion) these sources become opened. Consequently, during the resealing operation it is possible for these traces to appear as contamination to the environment and to people.

This paper refers to the use of methods and means to avoid the dispersion of the radioactive powder and to respect the legally admissible residual activity values (1.11 Bq/g for debris and 0.37 Bq/cm<sup>2</sup> for contamination of surfaces).



# DISTORTION IN CHEMICAL ELEMENT RELATIONSHIPS IN LIVE ORGANISMS BY CHANGES IN THE ENVIRONMENT

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Instrumental neutron activation analysis was used to study the chemical element behaviour in different live organisms under and under action of insignificant increase in the environment the concentrations of some chemical elements (Cd, Hg, Cr, Se, Th, Ag, and Cs).

As the subject of investigation were used some species of seaweeds and indoor plants. The experiments on element accumulation in live organisms extended over ten days, then all samples were washed by water and analysed by means of INAA.

It was found that:

1. The increase in environment the concentration of any single element leads to increase in plants the concentrations not only this element, but changes (both increase and decrease) in concentrations of many other elements.
2. Simultaneous action of increase in the environment the concentrations of more than one chemical element caused the competition between ions as they in plant tissues. In this case the concentrations of these ions in plants are not as high as in the above example. Moreover, in some cases combined action of several "heavy metals" on the plants even leads to decrease of concentrations of these "added" elements in experimental plants (in comparison with control samples).
3. Compared to control plants, in experimental samples the relationships between elements are more closed and besides, the total number of statistically significant correlation relationships is increased.

## NEUTRON ACTIVATION STUDY OF ELEMENT COMPOSITION IN THE AIR A LARGE CITY

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Trace element composition of atmospheric pollutants in a large city was studied. Continuous simultaneous sampling for two weeks at multiple points was performed using passive sorption on polyethylene collectors. In addition, samples of snow as well as soils were collected. Instrumental neutron activation analysis was used to determine trace element composition of the samples. Multielement patterns and high sensitivity, as well as good productivity, make this method very applicable for environmental studies and allows the determination of concentrations of about 30 elements in the majority of samples. The scheme of the method will be described. Enrichment factors were calculated using Sc as a reference element, and were used to compare the element composition of the objects under study. Distribution of atmospheric pollution over the city was studied. Some relationships between the element composition of atmospheric pollutants collected on sorbents and in snow water were established. The data obtained allowed calculation of the soil constituents of atmospheric element pollution. Speciation of elements in the air and their distribution between different phases was assessed.

## INAA FOR INVESTIGATING ECOLOGICAL EFFECTS DUE TO THE USE OF SOLID FUELS

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In West Siberia one of the main sources of environmental pollution are the thermal power plants using solid fuels (coal), especially small plants and private boiler works equipped with low efficient gas purifiers or having none at all. In addition to fuel gas outbursts in the atmosphere, huge ash dumps and slag storage pits of large thermal power plants contribute considerably to anthropogenic impact. At the same time these slag storage pits, where dozens of millions tons of ashy products of coal burning are accumulated, can also be a source of useful minerals. The processing of dumps not only helps rehabilitating occupied areas, but can also supply useful raw materials.

The INAA method was applied to investigate the composition of non-carbon composites of coals used by thermal power stations of industrial centres during a long period of time. The investigated coal was mostly from the Kuznetsk coal field, and partly from KATEK. The main attention was paid to the determination of rare-earth elements whose contents in low-ash coals can amount to 3000 g/t. These coal ashes can have industrial importance even at the present level of processing technologies.

The method included short-term and long-term irradiations of samples, sample "cooling" during different periods of time, activity measurements with co-axial and planar detectors. In addition to REE (La, Ce, Nd, Sm, Eu, Tb, Dy, Yb, Lu) some rock-forming elements (Al, Fe, Co, Se, Si, U, Cu, Ni, etc.) were also determined. All probes were checked for radioactive elements by using a low-background chamber. In this case it is extremely important because the coal from Kuzbas, and especially from KATEK, is rich in uranium and thorium.

Ashes, slag and aerosol outbursts following coal burning were analyzed simultaneously with the coal. In this way an attempt was made to estimate in complex the ecological impact produced by coal burning and suggest a scheme for utilizing products of burning.

## INVESTIGATION OF SEASONAL CHANGES IN THE BE 7 CONTENT IN THE GROUND COVER

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Studies of the radioactivity of the environment induced by artificial and natural radioactive elements have both scientific and practical importance. An increase in the radioactive isotope content in some parts of the biosphere or a change in their qualitative composition produce unfavorable impact on the living processes. It is therefore necessary to know the radioactivity level of the environment.

This paper presents the results of measurements of the Be 7 content in samples of grasses (*Carex Hirta* L.) taken in May - September 1992 to 1994. The probes were taken at one and the same site in the Minsk city. The samples were dried, and then they were turned to ash by 5 hour heating at  $T=400$ . The gamma-radioactivity of the ash was determined by a low-background gamma-spectrometer with a Ge(Li) detector whose energy resolution for the Co 60 line at 1333 keV was 3.2 keV. The gamma-spectrum measurement time varied from 10 to 20 hours. The Be 7 and K 40 activities were determined with respect to the 0.478 meV and 1.461 meV reference lines, respectively.

The investigations demonstrated a discrete character of the Be 7 content variation in the samples, which could be possibly explained by an irregularity of rain and snow falls and changing air flow directions. At the same time an increase in the Be 7 content by the end of the plant's vegetation period has to be noted. The Be 7 content in grasses varied from 0.618 to 2.559 Bc/g in 1992, from 0.484 to 0.794 Bc/g in 1993; from 0.027 to 4.987 Bc/g in 1994 (Fig. 1). The investigations have also indicated the presence in probes of the radionuclides belonging to the thorium row:  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$ , and  $^{208}\text{Tl}$  (0.2 - 0.4 Bc/g). Thus our investigations proved the possibility of using samples of grasses in environmental monitoring.

NAA of Vegetation Samples

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ANALYSIS OF THE ELEMENTAL COMPOSITION IN  
DIFFERENT COASTAL ZONES OF KEY COCO ACCORDING  
TO THE VEGETATION

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The influence of vegetation can affect the dynamics of the nutrimental contents in each plot of beach of Key Coco at the Northern part of the island of Cuba. Twenty one samples of coastal vegetal soils were taken to determine if there is a correlation between the vegetation and the multi-element composition of the soil. The analytical technique employed was Instrumental Neutron Activation Analysis (INAA) with the specific application of the Ko Standardization method for some elements. In this paper we show the dependence of the concentration of different minority and trace elements with the samples of the coastal soils, taken in a direction perpendicular to the beach, arriving at important ecological conclusions.

## STUDY OF AIR DUST FROM THE "ERDENET" COPPER-MOLYBDENUM PLANT

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Intensive development of the extractive industry and ore mining and processing enterprises in Mongolia creates the necessity for control of the environment.

"Edernet" is one of the largest copper-molybdenum mines in the world. In this work we have attempted to study the distribution of the dust spread into the air from this plant in the process of ore extraction and enrichment by analysing the snow cover.

The samples of snow were collected from different distances from 5 to 50 kilometers in 8 directions from the centre of the explosions at the opening of the copper-molybdenum mine. The concentrations of the main and some accompanying elements were determined in soluble and insoluble fractions of snow samples by  $\chi$ -ray fluorescence and other methods.

The distribution of Cu in the filter papers, deposited with particles insoluble in water has been studied in dependence on the distance and wind directions. In comparison with the background values, the concentration of Cu is 2-13 times greater in the main wind direction. The content of elements sharply decreases with the distance from the source, the influence of which was traced up to 100 km.

We have compared the obtained results with the environmental background data provided by the other authors.

# MAPPING OF ECOLOGICALLY UNFAVORABLE TERRITORIES ON THE BASIS OF HUMAN HAIR COMPOSITION

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As has been known [1] analysis of human hair on the population level and the mapping of large territories using hair elemental composition data is a promising approach for estimating both the environmental situation and population health status. In [1], the map of Uzbekistan (sampling in 1981) is discussed. Ten years later (1991) samples from the territory in the vicinity of drying-up Aral Sea were taken again. Samples were analyzed for 24 elements using instrumental neutron activation analysis. Comparison of the data and maps drawn for 1981 and 1991 and their comparison with changes in health status have shown that repeated mapping of territories using data on human hair elemental composition could be used in medical geography, especially for predicting health status changes in ecologically unfavorable areas.

## References

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## THE DISTRIBUTION STUDY OF TRITIUM LABELED TORTEZIN IN MOUSE TISSUE

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It is known that the use of radioactively labeled compounds provides high sensitivity and allows minor quantities of compounds to be located and traced. To study the distribution of a new compound - tortezin - in mouse tissues, tritium-labeled tortezin was used. Tortezins are purified tortoise spleen water extracts that stimulate stem cell proliferation. Tortezin was labeled with tritium using the method of labeling with thermally activated tritium. The method of tritium labeling as well as tortezin will be discussed. Tritium labeled tortezin with a specific radioactivity of about 2 mCi/mg was obtained. Purification of the final product was performed by HPLC. A water solution of tritium labeled tortezin was injected intravenously into mice. After 24 hours the mouse organs were removed, including the blood, liver, thymus, spleen, and bone marrow. Liquid scintillation counting in a dioxane cocktail was used to measure tritium radioactivity in the organs. The data obtained have allowed us to evaluate the tortezin distribution in mouse tissues. The method permits the tracing of tortezin in minor quantities with a sensitivity up to  $10^{-9}$ , using simple and fast methods for tritium radioactivity counting.

## HUMAN HAIR ELEMENTAL COMPOSITION IN MEDICAL DIAGNOSIS

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It is commonly known that the elemental status of the human body changes with changes in physiological status, as in some diseases. There are many studies which detected specific changes in the elemental composition of human tissues and liquids. Human hair is very significant in addition to other tissues. In the last decade, studies have discovered specific and statistically significant changes in hair element composition in some diseases (especially if multielement relations are considered). It was shown that in some cases these changes could be detected from 1 to 1.5 years earlier than classical diagnostic indications.

Comparing the data for groups with various diagnoses and hair compositions and comparing the medical statistical data with hair composition on a population level, we tried to elaborate criteria for the detection of samples belonging to the groups at risk. In order to justify the assumptions made on the basis of hair composition, we collected 250 samples from inhabitants of two different cities. These samples were analyzed for 24 elements using instrumental neutron activation analysis. The data obtained were grouped according to previously elaborated criteria for detection of certain risk groups. Then, we compared our assumptions with the medical records for each person and the results of personal reviews.

In most cases, the reliability of the elaborated procedure was very high (93% for kidney diseases, close to 100% for thyroid gland and nervous system diseases, diabetes, malignant neoplasm, and 90% for anemia). In gastrointestinal diseases the reliability was low (less than 50%), probably because selenium (which is one of the elements used in this case) is elevated in the entire population.

The preliminary data evidence once again that hair elemental compositions can be used for medical diagnostic purposes. Because of the simplicity and productivity of the proposed procedure, this approach could be very fruitful in routine screening of human health on the population level.

## EARLY DIAGNOSTICS OPTIMIZATION AND BIOINDICATION OF URBAN TERRITORY CONTAMINATIONS

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Urban territories are, as a rule, characterized by a high technogenic stress in modern technogenic conditions. High contrast heavy metal anomalies (As, Se, F, etc.) are typical for these territories. Because of extreme toxicity of the majority of heavy metals, early diagnostics and bioindication of contamination by heavy metals are of great importance for settlement and recreational zones.

A criterion for revealing first signs of environmental contamination (water, soil, biota) using REE key elements has been elaborated on the basis of a concept of heavy metal vicissitude accumulation in the environment. A group of selective bioindicators has been determined. This group permits bioindication to be optimized using combinations of selective elements.

It has been shown that the use of INAA in combination with RFA allows the considered problem to be solved with high reliability and on a good sensitivity level.

On the Radioactivity of Bed Load Sediments of Danube River  
During 1994

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**Artificial Radioactivity in the Environmental Samples as IAEA  
Reference Materials**

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# THE DYNAMICS OF $^{137}$ -CAESIUM ACCUMULATING IN NUCLEAR POWER COMPLEX-IMPACT ZONES

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Gas and aerosol outbursts of the nuclear power complex are the main source of environmental pollution by technogenic radionuclides. Radiation monitoring of the environment of developed atomic industry regions is therefore extremely important. For this purpose new objective methods of determining radionuclides in different objects of the ecosystem are necessary.

In the reported work gas and aerosol depositions accumulated in the garrets of living and industrial constructions, where they avoid atmospheric impact and retain nativeness, were chosen for estimating the accumulation rate of technogenic radionuclides. On the other hand, by choosing buildings constructed in different times one can follow the character of radionuclide accumulation over a long period of time. Probes were taken from the inhabited areas located in the direction of the most probable impact of the Seversk plant in a 30 km zone around the plant. They were samples of the surface layer (1.0 - 2.0 cm thick) and warmth-keeping materials (earth, sawdust). Specific activities of the trace elements were measured with a special, low-background radiometric device on the basis of a DGDK-80V semiconducting detector and a multi-channel amplitude analyzer with a program packet.

The obtained data are evidence of  $^{137}$ -caesium accumulating with time up to the maximum density of 100-150 Bk/m<sup>2</sup> in all investigated places. These data agree well with the results of an earlier analysis of earth probes taken in the investigated zone.

In general, the distribution obtained for the investigated trace radionuclide coincides with the prevailing wind rose (north-east) in the investigated district with some anomaly in the opposite direction.

In this way, analysis of gas and aerosol depositions can be a good tool for obtaining objective information about the level of radioactive pollution in a region and a good basis for radiation monitoring of the environment.

## LICHENS AS INDICATORS OF METAL AEROSOL DEPOSITION OVER BACKGROUND LEVEL

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Lichens receive their mineral food from atmospheric precipitation and grow very slowly. Lichens are integral indicators of the natural metal deposition on the Earth's surface. The analysis of lichen samples from alpine zones, far from industrial regions, may give information of the atmospheric deposition of metal in global as well as historical aspects.

Neutron activation analysis has been made of "Lecanora muralis" and "Aspicilia" lichen samples from the high mountain (1200-3500 m above sea level) regions of the Pamirs. The samples were taken at different altitudes and various substrates (granites, sands).

Concentrations of the following elements:Na, Fe, Co, Br, Zn, Rb, Ag, Sb, Cs, La, Ce, Sm, Eu, Lu, Hf, Ta have been determined. The analysis results show the highest metal contents are in the lichens from earth cracks, and the metal deposition in these zones is abnormal in comparison with the background level.

## INSIDE MOLECULAR SIEVES

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Zeolites and zeolite-like materials show interesting physico-chemical properties which result from their characteristic channelled crystal structure. Zeolites adsorptive and ion-exchange capabilities have motivated their use in nuclear waste immobilization as part of environmental conservation programs. The present talk is dedicated to characterizing recent trends in zeolite uses and investigations.

As a representative environmental application, the working principles of a zeolite-based nuclear waste treatment plant are discussed.

Several zeolite structure determination methods are analyzed. Diffraction-based techniques (X-rays, neutrons and electrons) are discussed and illustrated by means of practical examples. Considered problems include framework and extra-framework cation distributions, hydrogen site determination, order-disorder phenomena and morphology. The analytical capabilities of nuclear magnetic resonance studies are reported. Some proper results on cation site determination by the Rietveld procedure are presented.

A brief review of the theoretical microscopic characterization of zeolites is given. Quantum mechanical elementary cell structure optimization are discussed. Molecular level (ab initio and semi-empirical) and periodic Hartree-Fock treatments are considered. Recent calculations of interaction energies between zeolites and adsorbable molecules (water, ammonia) are described. Cation hopping between different favorable sites is modelled by means of the absolute rate theory which allows the calculation of complex permittivity as function of frequency.



# HEAVY METAL CONCENTRATIONS IN CORAL SKELETONS FROM THE CUBAN PLATFORM RECORDED BY NUCLEAR ANALYTICAL METHODS

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Instrumental Neutron Activation Analysis performed on pulverized coral skeleton samples was used to determine concentrations of heavy metals and other trace elements. Samples were prepared from several scleractinian coral specimens extracted from the Northern Havana reef. Specimens were cleaned, crushed and sieved. Eventually they were sectioned into "foot", "center" and "head" parts before the pulverization procedure. The containers for samples were irradiated separately for 1 min, 2 hours and 30 hours in the thermal flux of the Triga Mark III reactor in Mexico. Induced high-resolution gamma-spectra, recorded after irradiation at proper decay times, allowed the identification of Na, Cl, Al, Mg, Sr, Zn, Sc and other analytical peaks. X-ray Fluorescence and Atomic Absorption spectrometry analyses applied to the same objects complemented the elemental content information. The Sr, Mg, Zn and other content ratios relative to Ca of the described skeleton samples were obtained. Alternative conclusions about the possible influence of sediment and marine pollution.

## NEUTRON ACTIVATION ANALYSIS OF MARINE SEDIMENTS AT CAYO MOA BAY, CUBA

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Instrumental Neutron Activation Analysis was used to determine the concentration of heavy metals and other trace and minor elements in superficial marine sediments from the Cuban platform, at Cayo Moa Bay on the North-East side of the Island. Relative and KO standardization methods of INAA were applied. The irradiation was performed at the Triga Mark III (Mexico) reactor. ACTAN and Spectrum Analyzer PC codes were used for data processing and the high resolution HPGe induced gamma spectra. Relatively high contents of Fe, Ni, Cr, Co, Zn and Sc were found, as well as unexpected concentrations of several rare earth elements. The results complete the information about marine pollution in that region, obtained by energy dispersive X-Ray Fluorescence and Atomic Absorption Spectrometry applied before to those samples.

# THE NEUTRON TRANSPORT METHOD FOR DETERMINATION OF THE RESPONSE MATRIX IN SPHERICAL GEOMETRY

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This paper presents a new method for computation of the response matrix, to be applied primarily to proton-recoil detectors. Proton-recoil spectrometry methods, particularly scintillation spectrometry, are applied in the development of fast reactor and fusion technology. An accurate method is necessary for fast neutron spectra measurement, both the existing fusion reaction and that behind the radiation shield. The characteristics of fertile materials and transmission properties of shielding materials can then be determined, as well as neutron doses at certain positions around the fusion reactor. This knowledge is of significance for the design of radiation protection and dosimetry system of the fusion reactor.

The neutron spectrum which produces a measured pulse-height distribution at the output of a detector, can be unscrambled by unfolding computer codes. For the purpose of the unfolding procedure, the detector response function (counts per incident neutron of known energy, versus pulse amplitude) has to be determined, either by calculation or measurement. This is then used to construct a response matrix.

In the proposed algorithm for determination of the response matrix, the integral neutron transport theory is used for modeling all neutron interaction effects inside the detector volume. The neutron flux distribution in a detector, which necessary for determination of the detector response function, is calculated by the collision probability method. The anisotropic elastic neutron scattering on hydrogen and carbon is modeled by transport correction because the equation would otherwise become unduly complicated. Also, this approximation is well suited to the detector response function calculation. The multiple scattering on hydrogen and carbon, including reactions above 7 MeV is accounted as creating a rise to the detector response.

In order to apply the collision probability method, it is necessary to discretize the spherical detector volume into I subzones (shells) of equal width. The number of subzones I is chosen to allow the assumption that the neutron flux in each subzone is a flat function. The cross section is assumed to be constant per subzone, as well. For computing the collision probability in spherical geometry, Carlvik's method is used.

There are several advantages of the proposed method compared to existing methods for computing the response matrix. Some of these advantages are:

- the possibility of overcoming the "ray effect" often present in measurements with neutron beams;
- the possibility of determining the neutron flux perturbation caused by the presence of a detector in the case of and in-core neutron detection in a nuclear reactor;
- computation of the fine neutron flux distribution in the detector volume (in contrast to other methods, assuming the neutron flux is a flat function within the detector volume) which enables a more accurate proton-escape prediction.

Based on the above algorithm the SOFAK computer program for calculating the response matrix of a spherical detector was developed.

A neutron spectrum behind the silicon filter on the RD reactor at the "Vinca" Institute of Nuclear Sciences was measured by an SP2 proportional counter. Measuring results were unfolded by the standard SPEC4 code, utilizing the response matrix obtained by SOFAK code. The neutron spectrum obtained in this way was then compared with the spectrum calculated by SPEC4 code but using the response matrix produced by the Snidow analytical method normally used in the SPEC4 code.

## NUCLEAR ANALYTICAL TECHNIQUES IN ENVIRONMENTAL STUDIES

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Within the wide variety of analytical methods available for environmental studies various analytical methods are based on nuclear physical principles. A survey is given of the various categories of nuclear analytical methods, the associated characteristic features, and the information obtainable from these techniques.

Main characteristic features are: physically independent basis of the analytical method, isotopic rather than elemental determination, almost no interfering effect of electrons and molecular structure, and penetrating character of nuclear radiation.

Nuclear analytical techniques are facing today various constraints. There is a growing competition with other, non-nuclear, techniques. Further, governmental funds for nuclear research and thus also for application of nuclear methods are generally decreasing, while public concern about nuclear facilities and radioactivity is increasing. Nevertheless, nuclear analytical techniques remain potential techniques, also in the field of environmental studies.

In the frame of this situation suggestions are given for a full exploitation of the potentials of nuclear analytical methods, particularly when requiring considerable investment for equipment, supporting facilities, and specialized staff. In this survey emphasis is given to nuclear analytical techniques dealing with radioactivity, e.g. radiotracer techniques, activation analysis, radioisotope dilution and related techniques.

## A SPECTROMETRIC DEVICE FOR EXPRESS ANALYSIS FOR STRONTIUM-90 CONTENT IN SOIL

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One of the aims of radioecology is express determination of Strontium-90 content in soil against Cesium-137 background, whose content exceeds that of Strontium by a factor of 100-200. At present, the solution of this problem is important for regions polluted in result of the Chernobyl accident. Various known radiochemical methods for determining Strontium-90 content have high precision, sensitivity and selectivity. But these methods, however, have a very serious disadvantage, i.e. they are extremely labour- and time-consuming. On the basis of special plastic scintillators developed by the Sukhumi Physico-Technical Institute and Standard radiometrical equipment we assembled and tested a simple spectrometric device for measuring Strontium-90 in soil samples, if the Strontium content is not less than  $2 \cdot 10^{-9}$  Cu/kg against the Cesium-137 background of up to  $5 \cdot 10^{-6}$  Cu/kg. The device consisted of a plastic scintillator conjugated with an EMI 9531B multiplier, BUS2-94 and BUS2-95 amplifiers, a delay line, a BAS2-95 amplitude discriminator and an AI 1024-95 multichannel analyzer switched on to in the coincidence mode.

By setting up proper discrimination modes pulses from the beta spectrum section of interest were selected. The optimal thickness of the plastic scintillator was determined experimentally, and this thickness was 2.8-3 Ohm.in for the diameter of 60-65 mm in measurements of the Strontium-90+Yttrium-90 content against Cesium background.

The device was calibrated using OSGI standard sources and special radionuclide activity standard measures (OMACH) containing either Strontium-90 or Cesium-137 deposited on quartz sand. Dependence of the counting rate of the Strontium-90-based OMACH source on the amount of contained standard radioactive substance was measured for given sizes of the cuvette and scintillator and given their mutual arrangement.

Table 1.

Probe number	1	2	3	4	5	6	7
Strontium-90 content $10^{-8}\text{Cu/kg}$	2.84	3.02	3.80	2.78	3.44	3.68	3.60

In Table 1 results of measurements of 7 soil probes taken in one of the regions of the town of Sukhumi are presented.

To check the direct measuring of Strontium-90 content in soil by the proposed method an arbitrary measurement of probe 4 by radiochemical method was performed at the "Pripiat" UDK NPO. The measured content of Strontium in the probe was  $2.40 \cdot 10^{-8}$  Cu/kg. The Cesium-137 content in the same probe determined by the spectrometric method was  $2.90 \cdot 10^{-6}$  Cu/kg according to our measurements, and according to arbitrary measurements at "Taifun" NPO this content was  $2.20 \cdot 10^{-6}$  Cu/kg.

The time of measuring Strontium-90 under condition of the equilibrium between Strontium-90 and daughter Yttrium-80 with consideration for the preparation time of the probe does not exceed 4-5 hours.

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