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Toxicity of irradiated advanced heavy water reactor fuels

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ABSTRACT

The good neutron economy and online refuelling capability of the CANDU®[*] heavy water moderated reactor (HWR) enable it to use many different fuel types such as: low enriched uranium (LEU), plutonium, or thorium, in addition to its traditional natural uranium (NU) fuel. The toxicity and radiological protection methods for these proposed fuels, unlike those for NU, are not well established. This study uses software to compare fuel composition and toxicity of irradiated NU fuel against those of two irradiated advanced HWR fuel bundles as a function of post-irradiation time. The first bundle investigated is a CANFLEX®[†] low void reactor fuel (LVRF), of which only the dysprosium-poisoned central element, and not the outer 42 LEU elements, is specifically analyzed. The second bundle investigated is a heterogeneous high-burnup (LEU,Th)O_2 fuelled bundle, whose two components: LEU in the outer 35 elements, and thorium in the central 8 elements, are analyzed separately. The LVRF central element was estimated to have a much lower toxicity than that of NU at all times after shutdown. Both the high burnup LEU and the thorium fuel had similar toxicity to NU at shutdown, but due to the creation of such inhalation hazards as: ^{238}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm and ^{244}Cm (in high burnup LEU), and ^{232}U and ^{228}Th (in irradiated thorium), the toxicity of these fuels was almost double that of irradiated NU after 2700 days of cooling. New urine bioassay methods for higher actinoids, and the analysis of thorium in faecal samples, are recommended to assess the internal dose from these two fuels.

1. INTRODUCTION

HWR reactors, such as the CANDU reactor, have a good neutron economy, and, as a consequence, considerable flexibility in the range of potential fuel types (Whitlock, 2000). In addition to the normal natural uranium (NU, as UO_2) fuel, CANDU reactors can potentially use many other oxide fuels[‡]. In general, a reactor fuel will contain fertile constituents such as ^{238}U or ^{232}Th, together with a fissile component which can be a combination of ^{235}U, ^{233}U or plutonium isotopes. Such fuels may be heterogeneous (having different fertile/fissile combinations in different bundles and elements) or homogeneous. The use of slight enrichment in the outer elements, and neutron absorbing lanthanoids in the center, allows any value of coolant void reactivity and discharge burnup to be designed. Such bundles, called Low Void Reactivity Fuels (LVRF) (Boczar, 2004), mitigate the reactivity response of the reactor (the coolant void reactivity, or CVR) to a reduction in coolant flow, and have been proposed as a way to increase the passive safety in the CANDU design. Each of these fuels produces unique radiological protection problems if workers are exposed to the irradiated fuel following its discharge from the reactor. These problems are both a function of the radionuclide inventory of the fuel and of the cooling time (time in reactor spent-fuel pool) after discharge from the reactor. In general, for fuels of similar burnup[§], the radioactive fission products produced during irradiation are broadly similar for all the fissile fuel components of potential use in CANDU fuels (principally: ^{239}Pu, ^{241}Pu, ^{233}U, ^{235}U), and do not serve to greatly differentiate them. However the activity of individual fission products can increase significantly for higher burnup fuels because long-lived products build up throughout irradiation. In addition, the composition of irradiated fuels may be expected to vary depending on the nature of their fertile actinoid constituent.

Plutonium isotopes are a normal constituent of irradiated NU fuels. Indeed, more than half of the energy output of a full burn-up (7.5 MWd kg-1 of initial heavy elements) CANDU NU fuel bundle results from the fission of 239Pu that is produced during irradiation by neutron capture (and subsequent beta decays) on 238U. However, plutonium is normally present for insufficient time during a typical HWR fuel cycle to generate significant quantities of transplutonic actinoids. If plutonium is either added to the initial fuel charge (creating a so-called Mixed Oxide or MOX fuel), or if NU or enriched uranium is subjected to a longer irradiation time, the production of higher mass actinoids from ^{239}Pu will increase. Of the radionuclides produced in Pu-MOX (or high burnup uranium) fuels, alpha emitters are the most important with respect to radiological protection (Lochard, J. et al., 1997).

In many countries, India and China being well known examples, supplies of thorium are more abundant than those of uranium. The principle constituent of thorium is ^{232}Th. The only other thorium isotope present is ^{230}Th, which is a daughter of ^{234}U and hence present at a level depending on the initial uranium to thorium ratio of the ore. ^{232}Th is not fissile and cannot support a neutron chain reaction by itself, but it is fertile and under neutron irradiation produces the fissile nuclide ^{233}U. The presence of thorium in reactor fuels may therefore create additional long-term reactivity as the initial fissile constituents added to the fuel are depleted (IAEA 2002). The irradiation of ^{230}Th and ^{232}Th produces a range of radionuclides that have high toxicity and are unique to these Th-MOX fuels.

As previously noted, adding neutron absorbing poisons into the center element of a fuel bundle, while enriching the outer elements, allows the CVR and discharge burnup to be tuned. In LVRF bundles, dysprosium, a lanthanoid with seven stable isotopes; including ^{164}Dy (28.25%) which has a very large neutron capture cross-section, is the poison added. In these bundles, the non-center elements are then slightly enriched (1% ^{235}U in U) uranium instead of NU to counter the effect of the dysprosium on reactivity under normal reactor conditions and regain normal burnup. The impact of the inclusion of dysprosium isotopes on the toxicity of the irradiated center element is assessed in this article.

In order to work safely with irradiated fuels, it is important to know the radionuclide composition of the nuclear materials to ensure appropriate bio-monitoring procedures are in situ and to allow doses to be evaluated following exposure. Such data are well established for normal-burnup conventional nuclear fuels containing isotopes of uranium and plutonium. As described above, new fuels may include isotopes that are not normally present in conventional CANDU fuel. Consequently, bio-monitoring and dosimetric procedures have not yet been established for personnel working with some of the new or proposed fuels. In some cases, e.g., for some lanthanoids, bio-monitoring and dosimetry procedures may have to be developed. In other cases, e.g. for thorium, older procedures developed by AECL for experimental fuels may need to be revised. For this reason, the present study evaluates the radionuclide compositions of novel fuels, as a function of burnup and cooling period, in order to identify potential bioassay and dosimetry problems. Relevant questions are: Do we have sufficient information on the composition of novel fuels to estimate their bioavailability following exposure? Do we have sufficient information on the new fuels to generate a comprehensive ‘Reference Hazard’ list of the radionuclide activities depending on burnup and cooling? Which radionuclides will present the greatest health risk following intakes, and do we have the technologies to assay radionuclides for internal dosimetry? Do the computer codes used to estimate internal doses following an intake (e.g., GenmodPC developed by Richardson and Dunford, 1998) have the radionuclide specific data required to allow dose estimations?

In this paper, fuel composition data under different irradiation and cooling conditions have been generated for NU fuel, and for three components of advanced HWR fuels that may present unique radiological hazards and protective measures. Fuel code data at discharge and for different periods of post-discharge cooling have been used to list Reference Hazard radionuclides and to calculate the dosimetry metric ‘committed effective dose’ ($\tilde{E}(50)$) per kilogram** of fuel inhaled as particulate material. The radionuclides present in each fuel component were studied to determine the status of current knowledge regarding bioavailability, biokinetics, the suitability of International Commission on Radiological Protection (ICRP) recommended radionuclide models and whether appropriate bioassay and dosimetric tools are already in place if required.

2. METHODS

2.1 Fuels Analyzed

Three fuel components from two novel HWR reactor fuel bundles were selected for comparison with the standard NU fuel. The first is an LVRF fuel bundle that has a central element containing dysprosium in NU. The other 42 elements in the LVRF bundle are composed of LEU (1% ^{235}U in U) to compensate for the negative reactivity of the neutron-poisoned central element (Table 2). The post-irradiation composition of these outer elements was not analyzed as it is very similar to that of normal NU fuel, and the fuel achieves approximately the same burnup. The radiological hazards should therefore be very similar. The second fuel bundle studied here is a heterogeneous (LEU,Th)O_2 bundle (Boczar et al., 2010). It is analyzed as two separate components, a) the LEU in 8 elements located in the outer two rings of elements of the bundle, and b) the thorium in 35 elements located in the inner two rings of the bundle. In this case, the LEU may present novel radiological challenges, despite being similar to NU in composition, because it is taken to a burnup of more than twice that of conventional NU fuel. These fuels have been chosen for this paper because they have been, or may be, introduced to HWRs in the near future. Thus, in this paper, only natural uranium (with and without Dy), enriched uranium, and pure thorium fuels are considered. A follow-up paper is planned to look at various kinds of homogeneous Pu- and Th-MOX fuels which may be introduced into HWRs in the future.

2.2 Fuel code calculations

Fuel data were calculated using an AECL designed code ‘WOBI’ (WIMS-ORIGEN Burnup Integration) version 2.3.0 (Edwards, 2010). This code, which alternates neutron flux calculations from the 2D transport code WIMS-AECL 2.5d (Irish and Douglas, 2002) and fuel depletion calculations in ORIGEN-S 4.4 (ORNL, 1998) has been validated for NU assemblies in CANDU reactors and LEU assemblies in light water reactors (LWR) (Edwards, 2010).

For each fuel bundle, an input data file describing the fuel bundle, pressure tube and calandria tube was constructed. Inside the calandria tube, fuel temperature, coolant temperatures and coolant densities were set at typical CANDU reactor values. The calandria tube assembly was modelled as being located inside a unit cell of heavy water with a moderator to fuel ratio equal to that of a CANDU reactor. At the unit cell edges, partially reflective boundary conditions were imposed on the neutron flux to simulate the influence of the wider reactor environment.

The neutron leakage from the unit cell was adjusted continuously as the simulated burnup progressed to keep the ratio of neutron loss and production nearly equal (as is the case globally when a reactor is running at steady power). This approach simulates the gradual shift of the bundle from being a source to a sink of excess neutrons in the reactor during aging, and slightly affects the actinoid production per unit burnup.

The fresh fuel isotopic compositions and bundle configurations of NU fuel, LVRF, and heterogeneous (LEU,Th)O_2 bundles are given in Table 1 to Table 3. For each bundle, the selected burnup and days of (continuous) irradiation given correspond to the upper end of their expected operating life. Final fuel compositions are affected by the bundle power at discharge (for cooling times short compared to the total irradiation time) and total burnup (for longer cooling times) and by details of the in-reactor history of the fuel (for intermediate cooling times similar to the total irradiation time). This analysis uses a continuous-irradiation history to full burnup which assumes no reactor downtime and a high flux throughout. This approximation is conservative for the creation of most radionuclides in the fuel[††] and allows an assessment of the relative toxicity of the different fuels to which workers may be exposed.

Following irradiation, the WOBI code was used to model the changes in fuel composition as a function of cooling time. For this analysis, the fuel composition at discharge (0 days), and for log-spaced cooling times afterwards (100 days, 300 days, 900 days and 2700 days), was calculated. For each fuel and each time of cooling, the WOBI code produced the activity of radionuclides present in the irradiated fuel. The in-reactor simulation was normalized to an axial slice of a fuel bundle containing 1 kg of initial heavy elements (any actinoid, but excluding oxygen), but the final specific activities are normalized per kg of the fuel component being analyzed.

2.3 Initial Compositions

The initial fuel composition of the uranium used in NU fuel was a pure $^{235}U/^{238}U$ mix. In nature, 0.0054 % mass of uranium is ^{234}U, in equilibrium with the natural decay of ^{238}U at the top of the radium decay chain:

$$^{238}U \rightarrow \text{alpha} + {}^{234}Th \qquad (1)$$

$$^{234}Th \rightarrow \text{beta} + {}^{234m}Pa \qquad (2)$$

$$^{234m}Pa \rightarrow \text{beta} + {}^{234}U. \qquad (3)$$

The lack of ^{234}U in the NU model was of little importance in assessing the toxicity, as it is present only in small quantities and the capture of neutrons by ^{234}U only serves to increase the amount of ^{235}U slightly.

However, mined thorium contains some ^{230}Th created by ^{234}U decay because uranium and thorium are found in the same ore deposits. ^{230}Th fractions of 5 ppm (0.0005%) or greater are seen where there are approximately equal amounts of uranium and thorium in the initial ore, and when the deposit is old enough for ^{230}Th decay to come into equilibrium with ^{234}U decay (Sims, 2008). Ores with a significant amount of uranium may have ^{230}Th fractions of ten times this value (50 ppm). The thorium in this study was assumed to have a relatively high ^{230}Th content of 25 ppm (0.0025% ^{230}Th in total thorium).

2.4 Toxicity calculations

For each fuel, the radionuclide specific activity output by WOBI (in Ci kg^{-1}) was assessed for 0 days (discharge), 100, 300, 900 and 2700 days of cooling and ranked according to the radionuclide atomic number (Z). Subsequently, the radionuclide was allocated to one of three categories:

1. Fission products and their progeny,
2. Fuel beta emitters (actinoid activation products only) and,
3. Fuel alpha emitters (actinoid activation products only).

Activated neutron poisons initially present in the fuel were included with the fission products.

For each radionuclide for which the appropriate data were available, the specific committed effective dose for the inhalation of 1 kg of fuel, $\tilde{E}(50)$ (Sv kg^{-1}), referred to as 'toxicity' was calculated as:

$$\widetilde{E}(50) = \sum_{j} \left(e_{j,inh}(50) \cdot \widetilde{A}_{j,inh} \right) \tag{4}$$

where $e_{j,inh}(50)$ (Sv Bq^{-1}) is the committed effective dose coefficient for activity intake by inhalation (ICRP, 2007) of a radionuclide j, and $\widetilde{A}_{j,inh}$ is the activity per unit mass for the same radionuclide $\left(\mathrm{Bq\ kg^{-1}} = \mathrm{Ci\ kg^{-1}} \times 3.7\mathrm{x}10^{10} (\mathrm{Bq\ Ci^{-1}})\right)$.

For each fuel component, the activity data were analyzed by first eliminating all radionuclides not contributing at least 0.1% to the total activity within their category, then calculating the $\tilde{E}(50)$ for the remaining nuclides. Finally, the data were tabulated eliminating all nuclides not contributing at least 1% to $\tilde{E}(50)$. The single exception to this procedure was the inclusion of the $\tilde{E}(50)$ of ^{241}Pu at discharge. ^{241}Pu has a very large inhalation dose coefficient, larger than most beta emitters, and was found to contribute more than 1% to $\tilde{E}(50)$ at discharge even when falling below the 0.1% of total activity cut.

The dose coefficient of a radionuclide depends on, among other factors: its physical half life, the energy of the emitted radiation and the energy deposited per unit track length, the intake route, the lung and gut solubility, the residence time in radiosensitive tissues and organs, and the excretion characteristics. Radionuclides with half lives of the same magnitude as the 50 year integration time will have little change in decay rate and hence a large number of nuclear decays occur for each initial becquerel, increasing the dose coefficient. Alpha decays increase the dose coefficient by about a factor of 100, due to the high energy density of their tracks and the consequent possibility of unrepairable cell damage. Long lived alpha-emitters tend to have inhalation dose coefficients of a few times 10^{-6} while long-lived beta/gamma emitters tend to have inhalation dose coefficients of a few times 10^{-8}.

For the calculation of $\tilde{E}(50)$, values of the inhalation dose coefficient were obtained principally from ICRP Publication 68 (1994), assuming that the radionuclides were inhaled within an insoluble fuel matrix – ICRP solubility type S (slow) - as particles with an activity median aerodynamic diameter (AMAD) of 5 μm (geometric standard deviation = 2.5 μm). For radionuclides with more soluble elemental oxides (or an F (fast) or M (moderate) solubility type for all compounds) this required a non-ICRP solubility S-type value of the committed effective dose coefficient to be specified for the radionuclide. These soluble contaminants are minor in parts of the fuels' mass and it is reasonable to assume that they dissolve as the insoluble oxide fuel matrix dissolves. This reallocation of solubility type (e.g., for ^{241}Am, which the ICRP classifies as solubility type M) is possible within the AECL GenmodPC v5 internal dosimetry code, but not within IMBA (an alternative code marketed by the Health Protection Agency, UK) and for this reason GenmodPC v5 generated inhalation dose coefficients were used for non-ICRP type S calculations.

For each fuel component and cooling time, the specific activity and $\tilde{E}(50)$ for all fission products, fuel beta-emitting and fuel alpha-emitting radionuclides contributing ≥1% of the total $\tilde{E}(50)$) were tabulated as the ‘Reference Hazard’. The Reference Hazard Tables also include values for the total specific fuel activity and committed effective dose of all nuclides in the fuel, irrespective of whether or not a radionuclide met the ‘≥1% of total $\tilde{E}(50)$’ cut. Finally, for each fuel, graphs were prepared showing changes in the total specific activity and $\tilde{E}(50)$ as a function of fuel cooling time to 2700 days.

3. RESULTS - FUEL ACTIVITY

3.1 NU Fuel

The radionuclides calculated to be present in the Reference Hazard for this fuel at discharge and after cooling times of 100, 300, 900 and 2700 days are shown in Table 4 to Table 8. These tables show only those isotopes that were calculated to contribute ≥1% of the total $\tilde{E}(50)$ from the inhalation of NU fuel particles. Fig. 1A and Fig. 1B show the changing proportion of the total specific activity and $\tilde{E}(50)$, respectively, from fission products (dominated by beta-gamma emitters), heavy-element beta emitters and heavy-element alpha emitters, from discharge to 2700 days of cooling. The total specific activity of the irradiated NU fuel falls by more than three orders of magnitude between discharge and 2700 days post-irradiation (Fig. 1A), mostly as a consequence of the loss of short-lived fission products and ^{239}Np. In contrast, the $\tilde{E}(50)$ resulting from the inhalation of this fuel only falls by just over one order of magnitude (Fig. 1B). At early times after discharge, most of the inhalation dose results from the fission products and fuel beta emitters, but by 900 days the fuel alpha emitters, which provide only a few percent of $\tilde{E}(50)$ at discharge, have become the most important contributors to toxicity because of their relatively slow decay.

At discharge, the most important fission products are ^{144}Ce and ^{131}I contributing ~10% and ~8%, respectively. The contribution of ^{131}I to $\tilde{E}(50)$ ($T½$ = 8.023 days) declines quickly with cooling, while that of ^{144}Ce ($T½$ = 284.6 days) grows in importance (Table 4 to Table 6) for a few hundred days as the shorter-lived fission products (such as ^{131}I) decay more quickly than it does. However, by the end of a 2700 day cooling period (Table 8), the majority of the fission products have decayed and only two, ^{90}Sr and ^{137}Cs, (having half lives of many decades), significantly influence the toxicity.

At discharge, the fuel beta emitter ^{239}Np is important, contributing ~20% of $\tilde{E}(50)$, but it much less important after a few days of cooling, as ^{239}Np decays rapidly ($T½$ = 2.4 days). After 100 days (Table 5), ^{241}Pu is effectively the only actinoid beta emitter left, and its relatively slow decay ($T½$ = 14.4 years) results in it obtaining an increasing share of the total toxicity over the 2700 day cooling period studied (Table 8).

Of the actinoids at discharge, ^{242}Cm ($T½$ = 162.8 days) initially contributes the most to the $\tilde{E}(50)$ of the NU fuel, but its relatively short half life compared to most of the other alpha emitters renders it insignificant after a few hundred days. Thereafter, the longer lived alpha emitters gain in significance because their individual dose contributions are high and remain stable over the decay period considered, while the other contributors to toxicity mostly decay to insignificance. By 2700 days cooling, most of the $\tilde{E}(50)$ is from four alpha emitters: ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Am, each contributing ≥6.8% of the total toxicity (Table 8).

The biokinetic models for all the important radionuclides of NU fuel are coded within the GenmodPC v5 internal dosimetry code. From a radiological protection point of view the monitoring of workers exposed to irradiated and aged (≥2700 days) NU fuel presents few challenges. Bioassay regimens are firmly established to monitor intakes of the hallmark radionuclides of long-term cooling, i.e., ^{137}Cs, ^{90}Sr, ^{239}Pu, ^{240}Pu and ^{241}Am. However, confirming the exposure of workers to fuel cooled for shorter periods of 100 days to 900 days could present difficulties since bioassay methods are not fully established for some significant radionuclides, including ^{144}Ce, ^{241}Pu and ^{242}Cm. Nevertheless, at these cooling times, sufficient ‘indicator’ radionuclides, such as ^{239}Pu and ^{240}Pu, are present in the radionuclide mix to enable estimates of the total $\tilde{E}(50)$ using bioassays for these Pu alpha emitters and knowing the full spectrum of radionuclides present (i.e., the Reference Hazard). Moreover, the detection limits for ^{239}Pu and ^{240}Pu, by urine bioassay analysis using thermal ionisation mass spectrometry (TIMS) or accelerator mass spectrometry (AMS), are sufficiently low to allow the quantification of doses as low as 5% of the 20 mSv radiation worker dose limit.

3.2 LVRF fuel bundle

The addition of dysprosium to the central element reduces the thermal neutron flux by about a factor of five at the center of the LVRF fuel bundle compared with NU fuel. The fission rate, the total number of fissions and the total number of neutron captures are reduced by a similar amount. However, at discharge the decay of the short lived activation products ^{165m}Dy ($T_{1/2}$ = 1.26 minutes), ^{165}Dy ($T_{1/2}$ = 2.33 hours) and ^{166}Ho ($T_{1/2}$ = 1.12 days) largely compensates for the reduced fission product and actinoid activity (compare Table 9 with Table 4, and Fig. 2A with Fig. 1A) so that the overall activity in the LVRF fuel is about ¾ that of NU fuel. The very short half-life of ^{165m}Dy means that it decays to insignificance after a few minutes, reducing LVRF activity to about ½ that of NU fuel. After a few days, and certainly by 100 days cooling, the isotopes from activated dysprosium have all decayed and the fission product profile at later cooling times, e.g. 2700 days, resembles that of NU fuel but with only 20% of its activity and toxicity (compare Table 10 with Table 8 and Fig. 2B with Fig. 1B).

At discharge, the toxicity of the LVRF fuel is dominated by the beta emitters, ^{239}Np, ^{165}Dy and ^{166}Ho (Table 9). After 2700 days cooling the toxicity of the fuel is dominated by alpha emitters, as for NU fuel, but the proportion of ^{239}Pu is much higher. The reason for this is that in an LVRF central element the production of ^{240}Pu lags the production of ^{239}Pu, since the production of the former requires that the latter be present in the flux for a reasonable amount of time. On the other hand, in the higher flux of an average NU element, the high fission cross-section of ^{239}Pu means that its production comes into equilibrium with its removal about halfway through the irradiation. This allows the production of ^{240}Pu in CANDU NU fuel to catch up with and surpass that of ^{239}Pu.

The overall suppression of the higher mass actinoids (most of which have relatively long half-lives) in the LVRF central elements means the $\tilde{E}(50)$ of LVRF fuel decays more rapidly than that of NU fuel. For the center pin of LVRF, $\tilde{E}(50)$ declines from 1/4th of that of NU fuel at discharge to 1/8th that of NU fuel at 2700 days cooling (Table 9 and Table 10).

3.3 (LEU,Th)O_2 fuel bundle

In the (LEU,Th)O_2 bundle, the LEU and thorium oxides are separated into different elements, with the inner 8 elements containing ThO_2 and the outer 35 elements containing UO_2. Hence, when considering the hazard presented by this heterogeneous MOX fuel, it is most realistic to consider the uranium-containing and thorium-containing elements separately – since the simultaneous exposures to radionuclides from both fuels, under normal fuel storage conditions, would seem unlikely. Only when the hazard is from orphan fuel that is exposed during reactor maintenance (and in this case the code data will be inappropriate since the assumed burnup is likely to be too low) or if fuel has deteriorated and broken down during storage, would all radionuclides present a simultaneous hazard.

3.3.1 Thorium dioxide component of (LEU,Th)O_2 fuel bundle

The radionuclides included in the Reference Hazard for this fuel bundle at discharge and after 2700 days of cooling are given in Table 11 and Table 12. Fig. 3A shows the changes in the activity of fission products, fuel beta emitters and fuel alpha emitters of the ThO_2 fuel as a function of cooling time. Similarly, Fig. 3B shows the changes in *E(50)* of the fuel as a function of time.

The 8 fuel elements containing ThO_2 in the heterogenous $(LEU,Th)O_2$ bundle have a similar burnup to that of NU fuel, and therefore, because burnup is approximately proportional to the number of fissions occurring in the fuel, a very similar spectrum of fission products that contribute (a major part at discharge and a minor part at 2700 days) to the toxicity (compare Table 11 and Table 12 with Table 4 and Table 8). However, irradiated thorium produces a significantly different spectrum of actinoids having a significant influence on $\tilde{E}(50)$. The fuel beta emitter ^{233}Pa (the short lived daughter of ^{233}Th, which in turn decays to the fissile nuclide ^{233}U) is by far the most important fuel-derived radionuclide for short cooling periods. At discharge, ^{233}Pa provides ~50% of the total activity and $\tilde{E}(50)$, although its contribution drops rapidly to 18% of $\tilde{E}(50)$ at 100 days post-irradiation. It is assumed that the initial thorium fuel has been chemically processed during fuel manufacture to remove the naturally occurring radioisotopes of the thorium series[‡‡]. After 510 days of irradiation and a subsequent cooling time of 300 days, enough time has passed that some of the actinoid products near the end of the thorium series, e.g. ^{212}Pb and ^{212}Bi, reappear to dominate the $\tilde{E}(50)$ from beta-emitting fuel radionuclides. However, at no time during the interval from discharge to 2700 days cooling do any of the beta emitters in the thorium series contribute as much as 1% of the total $\tilde{E}(50)$ and therefore they do not appear in the Reference Hazard for this material (Table 11 and Table 12).

At all times after discharge the $\tilde{E}(50)$ portion from the alpha-emitting radionuclides is dominated by the products of neutron capture on ^{232}Th, namely ^{232}U,its daughter ^{228}Th, and, to a lesser extent, ^{233}U (Table 12). The toxicity of ^{232}U and, in particular, its daughter ^{228}Th, is augmented by the presence of the decay chain under ^{228}Th, so that every ^{228}Th decay results in several subsequent decays. This effect is much smaller for ^{233}U, which has a very long half life and whose daughter (^{229}Th) is also relatively long-lived. The importance of ^{233}U to the toxicity is due to the relatively large amounts of it produced by irradiating ^{232}Th. After 300 days of cooling the activity of alpha emitters is of equal importance to that of the fission products (Fig. 3B), a result which takes ~900 days in NU fuel (Fig. 1B). The production of higher mass actinoids is strongly suppressed in irradiated ThO_2 (i.e., all plutonium and americium radionuclides provide $\leq$1% of $\tilde{E}(50)$) due to the many neutron captures required for their formation and because the production of these actinoids necessarily proceeds through uranium isotopes (^{232}U, ^{233}U and ^{235}U) with high probabilities of fission relative to capture.

Production of ^{232}U in irradiated thorium proceeds via the formation of ^{231}Pa by an (n,2n) reaction on ^{232}Th (Eqn. 5) or an (n,γ) on ^{230}Th (Eqn. 6), followed by a highly probable (σ_{th} = 200 b) subsequent capture to ^{232}U (as shown in Eqn. 7).

$$^{232}\text{Th (n, 2n) } ^{231}\text{Th} \rightarrow \text{beta} + {}^{231}\text{Pa} \quad (5)$$

$$^{230}\text{Th (n, }\gamma\text{) } ^{231}\text{Th} \rightarrow \text{beta} + {}^{231}\text{Pa} \quad (6)$$

$$^{231}\text{Pa (n, }\gamma\text{) } ^{232}\text{Pa} \rightarrow \text{beta} + {}^{232}\text{U} \quad (7)$$

The (n,2n) reaction of Eqn. 5 normally dominates the (n, γ) of Eqn. 6 because of the very high ratio of ^{232}Th to ^{230}Th in the fuel. However, the (n,γ) cross-section is large, so even small amounts of initial ^{230}Th in the fuel can dramatically increase the amount of ^{232}U produced. Sensitivity studies[§§] on the irradiation of thorium fuel have shown that the difference between 0 and 25 ppm ^{230}Th in the initial fuel has the effect of increasing the production of ^{232}U by approximately 10%. Since ^{232}U ($T½$ = 69.8 y) and its daughter ^{228}Th ($T½$ = 1.9 y), dominate the $\tilde{E}(50)$ for long cooling times (summing to ~80% of the $\tilde{E}(50)$ after 2700 days cooling, as can be seen in Table 12), the Reference Hazard for alpha emitters can be considered to have about a ±10% uncertainty at these times (assuming a possible range of 0 to 50 ppm ^{230}Th), unless the initial ^{230}Th content of the fuel is known.

The S type inhalation dose coefficient of ^{231}Pa ($e_{j,inh}(50) = 1.7\text{x}10^{-5}$ Sv Bq^{-1}) is itself quite high. However, the importance of ^{231}Pa to the committed effective dose is severely reduced by its relatively low activity in the fuel.

The presence of different radionuclides within thorium fuels, compared to NU fuels, presents significant difficulties to bioassay analysis for radiological protection purposes. A significant challenge is that both ^{228}Th and ^{232}U are released slowly from an insoluble thorium oxide matrix in the lungs. Therefore, the activity excreted in a 24-hour urine sample would be insufficient by current bioassay methods employed for NU fuels to determine an intake equivalent to a $\tilde{E}(50)$ of 1 mSv. Instead, bioassay would have to proceed using faecal analyses, where the intake estimate is highly sensitive to the collection time allocated since exposure, which can only be partially mitigated by a series of frequent sample analyses.

3.3.2 LEU component of $(LEU,Th)O_2$ fuel bundle

The 35 elements in the outer two rings of an $(LEU,Th)O_2$ fuel bundle contain only LEU (Table 3). The calculated total $\tilde{E}(50)$ at discharge for this irradiated LEU fuel (Table 13, Fig. 4B) is similar to that calculated for NU fuel (Table 4, Fig. 1B). This is because the $\tilde{E}(50)$ of both LEU and NU at discharge is dominated by short-lived fission products. The production of these products is proportional to the fission power produced by the fuel bundle which varies little (from 30 to 37 W g^{-1}) between differently fuelled bundles (Table 1 to Table 3). However, the longer irradiation time (510 days) of the LEU fuel, compared to NU fuel (187 days), in the mixed $(Th, LEU)O_2$ bundle (Table 13) increases the population of higher actinoids, and hence $\tilde{E}(50)$ at discharge compared to NU fuel (Table 4). The activity of actinoids produced in LEU fuel is approximately double that of NU fuel. Notable exceptions are 1), ^{239}Pu, whose amount changes little in irradiated LEU relative to NU because it has a high fission cross-section that results in it coming into early equilibrium with its production, and 2) ^{238}Pu, whose production increases by more than 2 times because of the creation of ^{242}Cm and its subsequent decay, a production mode which is insignificant in NU fuel.

After 2700 days cooling of irradiated LEU fuel, the fission product $\tilde{E}(50)$ is dominated by long-lived radionuclides, chiefly, ^{90}Sr and ^{137}Cs (Fig 4A, Table 14). These long-lived fission products build up continuously throughout irradiation, making LEU, with its longer irradiation time, much more toxic after a long cooling period than an equivalent mass of the NU fuel (compare Fig. 4B with Fig. 1B).

Although the relative proportions of actinoids at various cooling times are different for LEU fuel compared to NU fuel, ^{239}Pu and ^{240}Pu remain important contributors to the toxicity of LEU, contributing ~18% and ~7%, respectively of the total $\tilde{E}(50)$ at 2700 days cooling. Therefore, quantification of LEU intakes and internal dosimetry can be made, as for NU fuel, by radionuclide bioassay analysis of urine employing mass spectrometry for Pu isotope quantification. Alternatively, faecal analysis and alpha-spectrometry could be employed to achieve the required sensitivity of 1 mSv for exposure the entire LEU fuel spectrum of radionuclides.

4. RESULTS - RADIOLOGICAL PROTECTION

4.1 Overall toxicity of fuels

The toxicity of the four fuels analyzed is compared in Fig. 5 which plots the $\tilde{E}(50)$ (Sv kg^{-1}) resulting from the inhalation of irradiated fuel particulates (5 μm AMAD, $\sigma = 2.5$ μm; ICRP solubility type S) as a function of cooling time. The toxicity of the unpoisoned fuels is similar at discharge, as it is dominated by short-lived fission products whose levels are essentially dependent on fuel specific power (W g^{-1}) at discharge, a quantity which varies only by about ±10% in these simulations, as seen Table 1 and Table 3. The LVRF central element has a much lower specific power because the dysprosium significantly reduces the thermal neutron flux density and power (Table 2), and correspondingly lowers the discharge toxicity.

For all the fuels studied, the toxicity falls by an order of magnitude or more between 0 and 2700 days cooling, mainly due to the decay of the short-lived fission products and fuel beta emitters (^{239}Np in uranium and ^{233}Pa in thorium). The final level of toxicity in all the fuels is dominated by the level of the longer-lived actinoids with a smaller, but significant, contribution (10-20%) by the fission products ^{90}Sr and ^{137}Cs.

The toxicity of the LVRF central element is considerably lower than that of the other fuels, due to low power at discharge and low actinoid creation.

While similar at discharge, the toxicity of both the thorium dioxide central elements and the LEU outer elements in a (LEU,Th)O_2 bundle becomes higher than that of NU fuel after a few hundred days. This result is due to the long irradiation time of these fuels, which leads to the creation of more long-lived fission products and actinoids. By 2700 days cooling, when long-lived radionuclides dominate the toxicities of all the fuels, the toxicities of the LEU and thorium fuels are similar and are more than twice that of irradiated NU fuel.

4.2 Bioavailability and biokinetics of radionuclides

The toxicity calculations made here assume ICRP inhalation type S characteristics for all radionuclides. However, the actuality is that the fuel particle consists of a small fraction of radionuclide elements whose oxides (e.g. Am, Np, Cm) are not classified by the ICRP in Publication 68 (ICRP 1994) as type S, but as type F, M or vapours. Other examples include iodine and caesium particulates, recommended by ICRP's Publication 68 to have type F solubility. It is also conceivable that elemental iodine may be released as a vapour as a consequence of high fuel temperature. In the analysis of the toxicity of a nuclear fuel, it is presumed that radionuclides are released from the inhaled fuel particles at a rate that is determined by the type S bulk solubility of the matrix in which they are embedded and not at a rate appropriate for pure compounds of the same radionuclide. It follows that the dose calculations undertaken for some radionuclides where ICRP does not provide type S type inhalation dose coefficients cannot be verified against ICRP publications. However, in these cases, only the assumed solubility of the radionuclide in the lung was changed to S type in GenmodPC internal dosimetry code and all other dosimetric parameters (including use of M or F type gut absorption factors) and values employed for the radionuclides are as recommended by the ICRP.

Type S solubility is a worst-case assumption for the inhalation dose coefficient of radionuclides such as: ^{144}Ce, ^{228}Th, ^{232}U, ^{233}Pa, ^{238}Pu, but not of ^{239}Pu, consistent with the ICRP type S classification for uranium dioxide and thorium dioxide. In practice, it is highly likely that the in-vivo solubility/bioavailability of the fuel matrices concerned will vary not only as a function of chemical species (uranium dioxide, plutonium dioxide, thorium dioxide), but also with the fuel burnup. It is known that not only can irradiation of fuel affect its solubility (Roth and Jonsson, 2009; Nilsson and Jonsson, 2011), but also that the porosity of the oxide particles may increase – giving them a larger effective specific surface. However, although physical and chemical data are available, to date *no* biological data are available on the bioavailability of the irradiated fuels considered here. In contrast, the biokinetics/metabolism of many of the important radionuclides (e.g., ^{90}Sr, ^{137}Cs, ^{224}Ra, U-isotopes, Pu-isotopes, and ^{241}Am) is well established and has been incorporated into appropriate ICRP dosimetric models (e.g. ICRP, 1995). For some radionuclides, particularly those produced by thorium fuels (e.g., ^{233}Pa and ^{228}Th) and for cerium and curium isotopes, much less information is available on their biological behaviour, and the ICRP dosimetric models recommended for these isotopes are less firmly based.

4.3 Dosimetry code

The most commonly used internal dosimetry code for radiological protection dosimetry is IMBA, marketed by the Health Protection Agency (formerly the National Radiological Protection Board) in the UK. This code was first produced in 1997 to meet the needs of the UK nuclear industry – particularly British Nuclear Fuels Ltd. IMBA Professional Plus is available as a series of modules that together comprise a comprehensive dosimetry tool for radiological protection dosimetry. IMBA implements standard ICRP internal dosimetry calculations. The extent to which IMBA can be modified to change dosimetric parameters for calculation is limited because the code, unlike GenmodPC, does not solve the compartmental algorithms of the elemental and systemic (e.g. lung and gut models) biokinetic models, but displays retention, excretion and dose data to best-fit curves of results derived elsewhere from the implementation of the standard ICRP models. This restricts the usefulness of IMBA when non-standard radionuclide assumptions are made or if models need to be modified.

For the present report calculations were made using GenmodPC v5 (Richardson and Dunford, 1998). This code implements ICRP models, and the dosimetric data generated are verified against published ICRP dose coefficients. Because it runs the ICRP models, any parameters within the model may be changed to meet the need for non-standard dose calculations. Comparison of the GenmodPC radionuclide library with the Reference Hazard produced for the NU fuel considered in this report indicated no important omissions for periods of cooling of $\geq$100 days. For the fuels at discharge, missing radionuclides include: fission products ^{85}Kr, ^{95m}Nb and ^{144m}Pr (present in all fuels), ^{165m}Dy (present only in the LVRF center element) and thorium decay chain products: ^{220}Rn, ^{216}Po, ^{212}Po and ^{208}Tl (present mainly in irradiated thorium). These radionuclides are of minor dosimetric significance and in all cases were calculated to account for 0.2% or less of the total radionuclide activity in the fuel. Moreover, since the basic metabolic data for the elements of these missing radionuclides are presently coded within GenmodPC v5, it would be relatively trivial task to modify files to include the emission data for the missing radionuclides.

4.4 Implications for bioassay

Currently within Canada the most commonly used method to identify and quantify $\tilde{E}(50)$ following intakes of irradiated fuel is either a semi-annual or annual urine bioassay to quantify ^{239}Pu intake, followed by dose attribution for the remainder spectrum of radionuclides using a work-specific Reference Hazard. This is possible because the TIMS detection limit for ^{239}Pu (~2 μBq) is low and because the fraction of ^{239}Pu in the Reference Hazard for normal NU fuels is high for all times of cooling and for all normal fuel burnups. Without TIMS (or accelerator mass spectrometry (AMS), which has similar detection limits) the minimum $\tilde{E}(50)$ that could be attributed to intakes of NU fuels using urine bioassay analysis would be greater than 5% of the annual worker 20 mSv dose limit required by the Canadian regulator for the internal dosimetry of workers. An alternative method of analysis, alpha-spectrometry, has detection limits for ^{239}Pu that are 500 to 1000 times higher than those achieved with TIMS. While the use of urine bioassay for ^{239}Pu is adequate for NU fuel, and meets the dosimetry needs for LVRF fuel, the results of this study show that the methods currently employed need to be upgraded for both high burnup LEU and thorium fuels which may contain inadequate quantities of ^{239}Pu to make this method tenable. In the absence of TIMS analytical methods for alpha emitters in faecal samples, alpha-spectrometry may be used for dose attribution, but the monitoring period should be monthly rather than semi-annually. Thorium oxide fuels that contain no higher actinoids present a significant dosimetric challenge, as intakes that result in a committed effective dose of 1 mSv result in insufficient excretion of both ^{228}Th and ^{232}U to be adequately detected by current urinalysis methods. Exposures to thorium fuels require a bioassay regimen that includes the analysis of faecal samples that are collected monthly.

5. CONCLUSIONS

The radionuclide content of a typical CANDU NU bundle and three components of advanced HWR fuels at typical operational burnups were predicted at discharge after different periods of cooling. The three advanced fuel components were: a CANFLEX LVRF center element; thorium from the center 8 elements of a heterogeneous (LEU,Th)O_2 bundle; and LEU from the outer 35 elements of an (LEU,Th)O_2 bundle. A Reference Hazard for each of these different fuels at discharge and after 100 days, 300 days, 900 days and 2700 days of cooling has been generated. The number of radionuclides contributing to the Reference Hazards decreases rapidly due to the loss of short-lived and intermediate-lived radionuclides within the radionuclide mix calculated for each fuel. Examination of the results led to the following conclusions:

1. The toxicity of all the fuels fell rapidly with cooling as the short-lived and intermediate-lived fission products (and some fuel-derived constituents, including: ^{239}Np, ^{233}Pa and ^{242}Cm) decayed away. The contribution of fission products to the total dose from inhaling different fuels decreased rapidly with cooling such that, after longer periods of cooling, long-lived alpha emitting actinoid isotopes had the greatest contribution to inhalation dose.
2. Activated dysprosium in the center element of LVRF bundles presents a significant dosimetric hazard at discharge, but this activity becomes insignificant after a few days of decay. The presence of the strong neutron absorber Dy during irradiation suppresses both the creation of fission products and actinoids, making this fuel by far the least toxic of those studied.
3. The toxicity of higher burnup LEU fuels is greater than that of NU fuel due to the higher levels of plutonium isotopes (other than ^{239}Pu, whose removal comes into equilibrium with its production relatively early), americium and curium created by multiple neutron captures on ^{238}U.
4. The radiation toxicity of irradiated thorium was higher at discharge than CANDU NU fuel for the fuel cycle considered (which postulated a 30% higher burnup and a 20% lower specific power than that of NU fuel). In addition, the toxicity of irradiated thorium does not decline as much with cooling because of the presence of the highly toxic and slowly decaying ^{232}U, and its daughter ^{228}Th.

5. The higher calculated toxicity of high burnup uranium and thorium fuels will present significant challenges to health physics personnel and will require an increase in physical barriers between fuel and workers as well as modifications to current bioassay practices to prevent unacceptably high exposures.
6. While the bioavailability and biokinetics of uranium fuels are well understood, less is known about thorium fuels. In particular, thorium oxide fuels may be much less biosoluble than other CANDU fuels, resulting in higher inhalation (and lower ingestion) radiation doses per unit activity of intake. More work needs to be performed to understand the biology of thorium and its compounds. Similarly, ^{244}Cm contributes greatly to the dose received following the inhalation of some fuels, but its biokinetics is less well understood than that of plutonium and uranium and more work is required.
7. GenmodPC v5 contains sufficient radionuclide data to calculate intake doses for all the fuels considered (with a few minor omissions relating to radionuclides that contribute <0.2% to the total activity). The ability to undertake the same tasks with IMBA will depend upon the IMBA Professional Plus version and modules available for use at any location.
8. Urine biomonitoring with analysis for ^{239}Pu by TIMS is suitable to monitor worker intakes of the NU fuel and LVRF (at all times after discharge), but is unsuitable for the heterogeneous (LEU,Th)O_2 fuel. For the higher burnup LEU fuel, either faecal biomonitoring with analysis for ^{239}Pu by TIMS/AMS or analyses for ^{238}Pu, ^{241}Am or $^{242/244}$Cm by alpha-spectrometry will need to be employed to monitor intakes. Urine biomonitoring with analysis for ^{239}Pu by alpha-spectrometry would be generally unsuitable for high burnup LEU fuel, since the high detection limit for ^{239}Pu results in a lowest measurable $\tilde{E}(50)$ that is greater than the 1 mSv required by the Canadian regulator.
9. Standard biomonitoring for ^{239}Pu will be completely unsuitable for irradiated thorium. This material may require a monthly faecal monitoring regimen for excreted ^{232}U/^{228}Th.

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8. TABLES

Table 1. Element configuration and composition of unirradiated NU fuel.

Element Location	Number of elements	Fuel radius (cm)	Fuel material (% mass)	Fuel density (g cm^{-3})	Fuel radionuclides (% mass)
Center	1	0.6075	100 UO_2	10.593	0.71 ^{235}U, 99.29 ^{238}U
Inner	6	0.6075	100 UO_2	10.593	0.71 ^{235}U, 99.29 ^{238}U
Intermediate	12	0.6075	100 UO_2	10.593	0.71 ^{235}U, 99.29 ^{238}U
Outer	18	0.6075	100 UO_2	10.593	0.71 ^{235}U, 99.29 ^{238}U
Average Burnup		6.95 MWd kg^{-1} initial heavy elements (iHE)			
Irradiation Time		187.4 days			
Average Power		37.1 W g^{-1}			
Power at Discharge		30.7 W g^{-1}			

Table 2. Element configuration and composition of unirradiated LVRF fuel bundle.

Element Location	Number of elements	Fuel radius (cm)	Fuel material (% mass)	Fuel density (g cm^{-3})	Fuel radionuclides (% mass)
Center	1	0.6326	88.45 UO_2 11.55 Dy_2O_3	10.142	0.005 ^{234}U, 0.711 ^{235}U, 99.29 ^{238}U 2.34 ^{160}Dy, 18.93 ^{161}Dy, 25.54 ^{162}Dy, 24.94 ^{163}Dy, 28.25 ^{164}Dy
Inner	7	0.6326	100 UO_2	10.554	1^{235}U, 99 ^{238}U
Intermediate	14	0.5350	100 UO_2	10.554	1 ^{235}U, 99 ^{238}U
Outer	21	0.5350	100 UO_2	10.554	1 ^{235}U, 99 ^{238}U
Center Element Burnup	2 MWd kg^{-1} initial heavy elements (iHE)				
LEU Burnup	7.46 MWd kg^{-1} iHE				
Average Burnup	7.33 MWd kg^{-1} iHE				
Irradiation Time	200 days				
Average Power	36.7 W g^{-1}				
Power at Discharge (center element)	9.51 W g^{-1}				
Power at Discharge (remaining elements)	32.7 W g^{-1}				

Table 3. Element configuration and composition of unirradiated heterogeneous MOX (LEU,Th)O_2 fuel bundle.

Element Location	Number of elements	Fuel radius (cm)	Fuel material (% mass)	Fuel density (g cm^{-3})	Fuel radionuclides (% mass)
Center	1	0.6310	100 ThO_2	9.46	99.9975 ^{232}Th, 0.0025 ^{230}Th
Inner	7	0.6310	100 ThO_2	9.46	99.9975 ^{232}Th, 0.0025 ^{230}Th
Intermediate	14	0.5360	100 UO_2	10.593	1.7 ^{235}U, 98.3 ^{238}U
Outer	21	0.5360	100 UO_2	10.593	1.25 ^{235}U, 98.75 ^{238}U
Thorium Burnup	9.11 MWd kg^{-1} initial heavy elements (iHE)				
LEU Burnup	19.85 MWd kg^{-1} iHE				
Average Burnup	17.48 MWd kg^{-1} iHE				
Irradiation Time	510 days				
Average Specific Power	34.3 W g^{-1}				
Power at Discharge (thorium elements)	24.6 W g^{-1}				
Power at Discharge (LEU elements)	25.2 W g^{-1}				

Table 4. Natural uranium Reference Hazard (at discharge). Only radionuclides shown are those with an $\tilde{E}(50)$ *of* ≥1% of the fuel total. The 'Total' includes all radionuclides, including those with an $E(50)$ of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{89}Sr	8.68x10^{2}	0.55	1.8x10^{5}	3.6
	^{91}Y	1.10x10^{3}	0.70	2.5x10^{5}	5.0
	^{95}Zr	1.46x10^{3}	0.93	2.2x10^{5}	4.5
	^{95}Nb	1.29 x10^{3}	0.82	6.2 x10^{4}	1.2
	^{97}Zr	1.51x10^{3}	0.96	7.8x10^{4}	1.6
	^{99}Mo	1.75x10^{3}	1.11	7.1x10^{4}	1.4
	^{103}Ru	1.49x10^{3}	0.95	1.2x10^{5}	2.4
	^{106}Ru	1.94x10^{2}	0.12	2.5x10^{5}	4.9
	^{131}I	9.87x10^{2}	0.63	3.9x10^{5}	7.6
	^{132}Te	1.38x10^{3}	0.88	1.5x10^{5}	3.1
	^{133}I	1.93x10^{3}	1.23	1.5x10^{5}	2.8
	^{140}Ba	1.70x10^{3}	1.08	1.0x10^{5}	5.9
	^{140}La	1.73x10^{3}	1.10	9.8x10^{4}	2.0
	^{141}Ce	1.57x10^{3}	1.00	1.8x10^{5}	3.6
	^{143}Ce	1.40x10^{3}	0.89	5.2x10^{4}	1.0
	^{143}Pr	1.42x10^{3}	0.90	1.2x10^{5}	2.3
	^{144}Ce	5.25x10^{2}	0.33	5.6x10^{5}	11.1
Fuel beta emitters	^{241}Pu	1.99x10^{1}	0.04	6.3x10^{4}	1.2
	^{239}Np	2.65x10^{4}	49.53	1.1x10^{6}	20.1
Fuel alpha emitters	^{240}Pu	2.12x10^{-1}	16.18	6.6x10^{4}	1.3
	^{242}Cm	8.77x10^{-1}	67.03	1.3x10^{5}	2.6
Total		2.11x10^{5}		5.0x10^{6}	

Table 5. Natural uranium Reference Hazard (100 days of cooling). Only radionuclides with an *Ẽ(50)* more than 1% of the fuel total are shown. ‘Total’ includes all radionuclides, including those with an *Ẽ(50)* of <1%.

Radionuclide category	Nuclide	Activity ($Ci\ kg^{-1}$)	Activity (% of total category)	*Ẽ(50)* ($Sv\ kg^{-1}$)	*Ẽ(50)* (% of total fuel)
Fission products	^{89}Sr	$2.20x10^{2}$	5.59	$4.5x10^{4}$	3.5
	^{90}Sr	$1.62x10^{1}$	0.41	$4.6x10^{4}$	3.6
	^{91}Y	$3.39x10^{2}$	8.60	$7.6x10^{4}$	5.9
	^{95}Zr	$4.94x10^{2}$	12.53	$7.6x10^{4}$	5.9
	^{95}Nb	$8.24x10^{2}$	20.88	$4.0x10^{4}$	3.1
	^{103}Ru	$2.55x10^{2}$	6.47	$2.0x10^{4}$	1.6
	^{106}Ru	$1.61x10^{2}$	4.07	$2.0x10^{5}$	15.7
	^{137}Cs	$2.32x10^{1}$	0.59	$1.8x10^{4}$	1.4
	^{141}Ce	$1.87x10^{2}$	4.74	$2.2x10^{4}$	1.7
	^{144}Ce	$4.12x10^{2}$	10.44	$4.4x10^{5}$	33.6
Fuel beta emitters	^{241}Pu	$1.96x10^{1}$	99.97	$6.2x10^{4}$	4.8
Fuel alpha emitters	^{238}Pu	$5.32x10^{-2}$	3.73	$2.1x10^{4}$	1.6
	^{239}Pu	$1.60x10^{-1}$	11.20	$4.9x10^{4}$	3.8
	^{240}Pu	$2.12x10^{-1}$	14.85	$6.6x10^{4}$	5.1
	^{242}Cm	$5.85x10^{-1}$	41.07	$8.7x10^{4}$	6.7
Total		$3.96x10^{3}$		$1.3x10^{6}$	

Table 6. Natural uranium Reference Hazard (300 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' includes all radionuclides, including those with an $\tilde{E}(50)$ of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	1.60x10^{1}	1.27	4.6x10^{3}	6.0
	^{95}Zr	5.66x10^{1}	2.83	8.7x10^{3}	1.2
	^{106}Ru	1.11x10^{2}	0.67	1.4x10^{5}	18.6
	^{137}Cs	2.29x10^{1}	0.75	1.8x10^{4}	2.3
	^{144}Ce	2.53x10^{2}	0.23	2.7x10^{5}	35.4
Fuel beta emitters	^{241}Pu	1.91x10^{1}	99.99	6.0x10^{4}	7.9
Fuel alpha emitters	^{238}Pu	5.47x10^{-2}	7.60	2.1x10^{4}	2.8
	^{239}Pu	1.60x10^{-1}	22.21	4.9x10^{4}	6.5
	^{240}Pu	2.12x10^{-1}	29.44	6.6x10^{4}	8.7
	^{241}Am	3.01x10^{-2}	4.18	9.6x10^{3}	1.3
	^{242}Cm	2.50x10^{-1}	34.80	3.7x10^{4}	4.9
Total		1.14x10^{3}		7.6x10^{5}	

Table 7. Natural uranium Reference Hazard (900 days of cooling). Only radionuclides with an *$\tilde{E}(50)$* more than 1% of the fuel total are shown. 'Total' includes all radionuclides, including those with an *$\tilde{E}(50)$* of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	1.54×10^{1}	4.98	4.4×10^{4}	10.9
	^{106}Ru	3.61×10^{1}	11.69	4.6×10^{4}	11.5
	^{137}Cs	2.21×10^{1}	7.13	1.7×10^{4}	4.3
	^{144}Ce	5.88×10^{1}	19.03	6.2×10^{4}	15.5
	^{147}Pm	3.52×10^{1}	11.40	4.1×10^{3}	1.0
Fuel beta emitters	^{241}Pu	1.76×10^{1}	99.99	5.6×10^{4}	13.8
Fuel alpha emitters	^{238}Pu	5.51×10^{-2}	10.28	2.2×10^{4}	5.4
	^{239}Pu	1.60×10^{-1}	29.77	4.9×10^{4}	12.3
	^{240}Pu	2.12×10^{-1}	39.47	6.6×10^{4}	16.4
	^{241}Am	7.83×10^{-2}	14.60	2.5×10^{4}	6.2
Total		3.27×10^{2}		4.0×10^{5}	

Table 8. Natural uranium Reference Hazard (2700 days of cooling). Only radionuclides with an *$\tilde{E}(50)$* more than 1% of the fuel total are shown. 'Total' includes all radionuclides, including those with an *$\tilde{E}(50)$* of <1%.

Radionuclide category	Nuclide	Activity ($Ci\ kg^{-1}$)	Activity (% of total category)	$\tilde{E}(50)$ ($Sv\ kg^{-1}$)	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	$1.36x10^1$	16.66	$3.9x10^4$	12.7
	^{137}Cs	$1.97x10^1$	24.06	$1.5x10^4$	5.0
Fuel beta emitters	^{241}Pu	$1.39x10^1$	99.99	$4.4x10^4$	14.3
Fuel alpha emitters	^{238}Pu	$5.31x10^{-2}$	8.35	$2.1x10^4$	6.8
	^{239}Pu	$1.60x10^{-1}$	25.11	$4.9x10^4$	16.1
	^{240}Pu	$2.11x10^{-1}$	33.27	$6.6x10^4$	21.4
	^{241}Am	$2.01x10^{-1}$	31.65	$6.4x10^4$	20.9
	^{244}Cm	$8.88x10^{-3}$	1.40	$3.1x10^3$	1.0
Total		$9.69x10^1$		$3.1x10^5$	

Table 9. LVRF Reference Hazard (at discharge). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. ‘Total’ includes all radionuclides, including those with an $\tilde{E}(50)$ of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products and activation products	^{89}Sr	1.55x10^{2}	0.11	3.2x10^{4}	2.7
	^{91}Y	1.89x10^{2}	0.13	4.3x10^{4}	3.6
	^{95}Zr	2.18x10^{2}	0.15	3.4x10^{4}	2.9
	^{97}Zr	2.67x10^{2}	0.18	1.4x10^{4}	1.2
	^{99}Mo	3.00x10^{2}	0.21	1.2x10^{4}	1.0
	^{103}Ru	1.84x10^{2}	0.13	1.5x10^{4}	1.3
	^{131}I	1.53x10^{2}	0.10	6.0x10^{4}	5.0
	^{132}Te	2.28x10^{2}	0.16	2.5x10^{4}	2.2
	^{133}I	3.29x10^{2}	0.23	2.6x10^{4}	2.1
	^{140}Ba	2.88x10^{2}	0.20	1.7x10^{4}	4.3
	^{140}La	2.88x10^{2}	0.20	1.6x10^{4}	1.5
	^{141}Ce	2.44x10^{2}	0.17	2.8x10^{4}	2.4
	^{143}Pr	2.52x10^{2}	0.17	2.1x10^{4}	1.8
	^{165}Dy	6.44x10^{4}	44.17	2.1x10^{5}	17.8
	^{166}Ho	5.00x10^{3}	3.43	1.5x10^{5}	13.2
Fuel beta emitters	^{239}Np	5.55x10^{3}	49.71	2.1x10^{5}	18.0
Fuel alpha emitters	^{239}Pu	6.04x10^{-2}	71.05	1.9x10^{4}	1.6
Total		1.57x10^{5}		1.2x10^{6}	

Table 10. LVRF Reference Hazard (2700 days of cooling). Only radionuclides with an *Ẽ(50)* more than 1% of the fuel total are shown. ‘Total’ includes all radionuclides, including those with an *Ẽ(50)* of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	*Ẽ(50)* (Sv kg^{-1})	*Ẽ(50)* (% of total fuel)
Fission products	^{90}Sr	2.04x10^{0}	17.94	5.8x10^{3}	15.5
	^{137}Cs	2.52x10^{0}	22.19	2.0x10^{3}	5.2
Fuel beta emitters	^{241}Pu	4.29x10^{-1}	99.87	1.4x10^{3}	3.6
Fuel alpha emitters	^{238}Pu	3.95x10^{-3}	4.48	1.6x10^{3}	4.2
	^{239}Pu	6.19x10^{-2}	70.09	1.9x10^{4}	51.3
	^{240}Pu	1.57x10^{-2}	17.82	4.9x10^{3}	13.1
	^{241}Am	6.20x10^{-3}	7.03	2.0x10^{3}	5.3
Total		1.19x10^{1}		3.7x10^{4}	

Table 11. Thorium in (LEU,Th)O_2 fuel bundle Reference Hazard (at discharge). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' includes all radionuclides, including those with an $\tilde{E}(50)$ of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{89}Sr	1.10 x10^{3}	1.21	2.3x10^{5}	3.7
	^{91}Y	1.13x10^{3}	1.24	2.6x10^{5}	4.2
	^{95}Zr	1.05x10^{3}	1.15	1.6x10^{5}	2.7
	^{131}I	6.70x10^{2}	0.73	2.6x10^{5}	4.2
	^{132}Te	8.64x10^{2}	0.95	9.7x10^{4}	1.6
	^{133}I	1.11x10^{3}	1.22	8.1x10^{4}	1.3
	^{140}Ba	1.20x10^{3}	1.31	2.1x10^{5}	3.4
	^{140}La	1.21x10^{3}	1.33	7.2x10^{4}	1.2
	^{141}Ce	1.17x10^{3}	1.28	1.3x10^{5}	2.2
	^{143}Pr	1.08x10^{3}	1.18	8.8x10^{4}	1.4
	^{144}Ce	4.45x10^{2}	0.49	4.7x10^{5}	7.8
Fuel beta emitters	^{233}Pa	2.68 x10^{4}	49.63	3.1x10^{6}	51.4
Fuel alpha emitters	^{232}U	2.71x10^{-1}	49.33	2.6x10^{5}	4.3
Total		1.45x10^{5}		6.1x10^{6}	

Table 12. Thorium in (LEU,Th)O_2 fuel bundle Reference Hazard (2700 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' includes all radionuclides, including those with an $\tilde{E}(50)$ of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	2.25×10^{1}	22.46	6.4×10^{4}	9.5
	^{137}Cs	2.12×10^{1}	21.14	1.7×10^{4}	2.5
Fuel alpha emitters	^{224}Ra	2.45×10^{-1}	16.37	2.5×10^{4}	3.7
	^{228}Th	2.44×10^{-1}	16.31	2.9×10^{5}	43.3
	^{232}U	2.53×10^{-1}	16.93	2.4×10^{5}	36.1
	^{233}U	1.01×10^{-1}	6.76	2.6×10^{4}	3.9
Total		1.02×10^{2}		6.8×10^{5}	

Table 13. LEU in (LEU,Th)O_2 fuel bundle Reference Hazard (at discharge). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. ‘Total’ includes all radionuclides, including those with an $\tilde{E}(50)$ of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{89}Sr	5.65x10^{2}	0.47	1.2x10^{5}	1.9
	^{91}Y	7.91x10^{2}	0.66	1.8x10^{5}	2.9
	^{95}Zr	1.20x10^{3}	1.01	1.9x10^{5}	3.0
	^{95}Nb	1.30x10^{3}	1.01	6.3x10^{4}	1.0
	^{103}Ru	1.29x10^{3}	1.08	1.1x10^{5}	1.7
	^{106}Ru	4.17x10^{2}	0.35	5.3x10^{5}	8.6
	^{131}I	7.33x10^{2}	0.62	2.8x10^{5}	4.5
	^{132}Te	1.02x10^{3}	0.86	1.2x10^{5}	1.9
	^{133}I	1.42x10^{3}	1.19	1.0x10^{5}	1.7
	^{140}Ba	1.22x10^{3}	1.02	2.1x10^{5}	3.4
	^{140}La	1.28x10^{3}	1.08	7.6x10^{4}	1.2
	^{141}Ce	1.15x10^{3}	0.96	1.3x10^{5}	2.1
	^{143}Pr	9.95x10^{2}	0.84	8.1x10^{4}	1.3
	^{144}Ce	8.85x10^{2}	0.74	9.4x10^{5}	15.2
Fuel beta emitters	^{239}Np	1.91x10^{4}	49.24	7.3x10^{5}	11.7
	^{241}Pu	4.10x10^{1}	0.11	1.3x10^{5}	2.1
Fuel alpha emitters	^{238}Pu	2.47x10^{-1}	2.68	9.7x10^{4}	1.6
	^{240}Pu	4.17x10^{-1}	4.54	1.3x10^{5}	2.1
	^{242}Cm	8.15x10^{0}	88.61	1.2x10^{6}	19.6
	^{244}Cm	1.98x10^{-1}	2.15	6.9x10^{4}	1.1
Total		1.58x10^{5}		6.2x10^{6}	

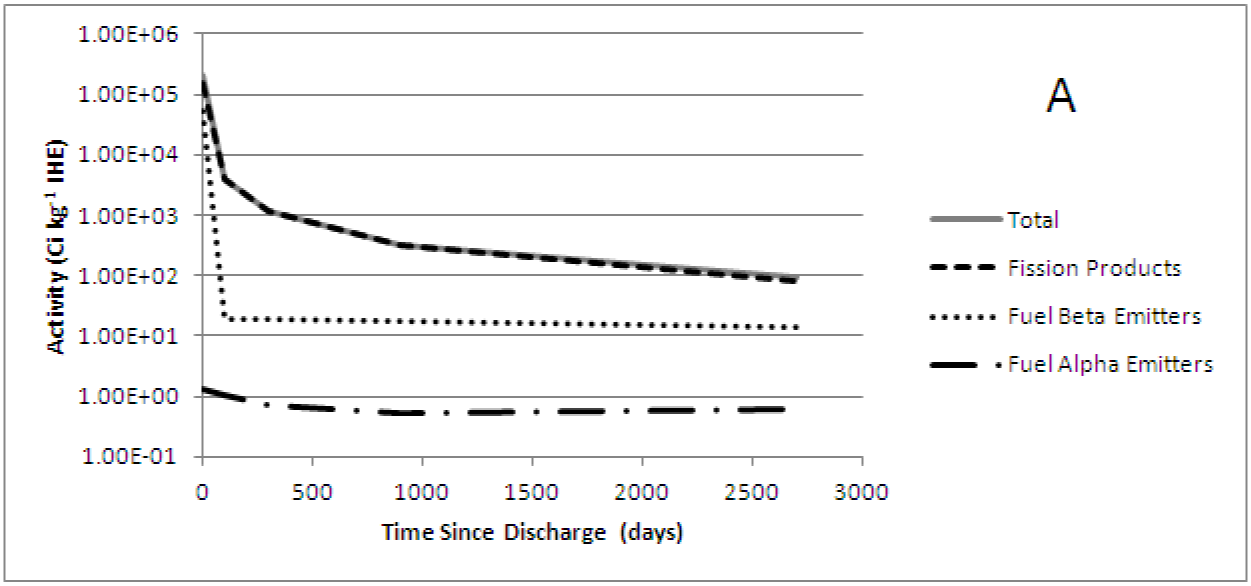
Table 14. LEU in (LEU,Th)O_2 fuel bundle Reference Hazard (2700 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' includes all radionuclides, including those with an $\tilde{E}(50)$ of <1%.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	3.38 x10^1	16.79	9.6 x10^4	13.5
	^{137}Cs	5.06 x10^1	25.08	3.9 x10^4	5.5
Fuel beta emitters	^{241}Pu	2.87 x10^1	99.98	9.0 x10^4	12.6
Fuel alpha emitters	^{238}Pu	2.77 x10^{-1}	19.27	1.1 x10^5	15.2
	^{239}Pu	1.60 x10^{-1}	11.14	5.0 x10^4	6.9
	^{240}Pu	4.17 x10^{-1}	29.01	1.3 x10^5	18.1
	^{241}Am	4.27 x10^{-1}	29.65	1.4 x10^5	19.0
	^{244}Cm	1.49 x10^{-1}	10.36	5.2 x10^4	7.3
Total		2.32x10^2		7.2x10^5	

Table 15. Inhalation Dose Coefficients to an Adult Worker for Alpha Emitters with a 5 μm AMAD. The GENMODPC coefficients were used when available.

	GENMODPC v.5.1.0		ICRP 68	
Radionuclide	Solubility Type	$e_{j,inh}(50)$ (Sv Bq^{-1})	Solubility Type	$e_{j,inh}(50)$ (Sv Bq^{-1})
^{241}Am	S	8.6x10^{-6}	M	2.7x10^{-5}
^{242m}Am	S	5.5x10^{-6}	M	2.4x10^{-5}
^{243}Am	S	8.2x10^{-6}	M	2.7x10^{-5}
^{242}Cm	S	4.0x10^{-6}	M	3.7x10^{-6}
^{243}Cm	S	1.0x10^{-5}	M	2.0x10^{-5}
^{244}Cm	S	9.5x10^{-6}	M	1.7x10^{-5}
^{237}Np	S	8.1x10^{-6}	M	1.5x10^{-5}
^{231}Pa			S	1.7x10^{-5}
^{238}Pu	S	1.1x10^{-5}	S	1.1x10^{-5}
^{239}Pu	S	8.4x10^{-6}	S	8.3x10^{-5}
^{240}Pu	S	8.4x10^{-6}	S	8.3x10^{-5}
^{242}Pu			S	7.7x10^{-6}
^{224}Ra	S	2.7x10^{-6}	M	2.4x10^{-6}
^{227}Th	S	7.6x10^{-6}	S	7.6x10^{-6}
^{228}Th	S	3.2x10^{-5}	S	2.5x10^{-5}
^{229}Th	S	4.8x10^{-5}	S	4.8x10^{-5}
^{230}Th	S	7.2x10^{-6}	S	7.2x10^{-6}
^{232}Th	S	1.2x10^{-5}	S	1.2x10^{-5}
^{232}U	S	2.6x10^{-5}	S	2.6x10^{-5}
^{233}U	S	7.0x10^{-6}	S	6.9x10^{-6}
^{234}U	S	6.8x10^{-6}	S	6.8x10^{-6}
^{235}U	S	6.1x10^{-6}	S	6.1x10^{-6}
^{236}U	S	6.3x10^{-6}	S	6.3x10^{-6}
^{238}U	S	5.7x10^{-6}	S	5.7x10^{-6}

9. FIGURES



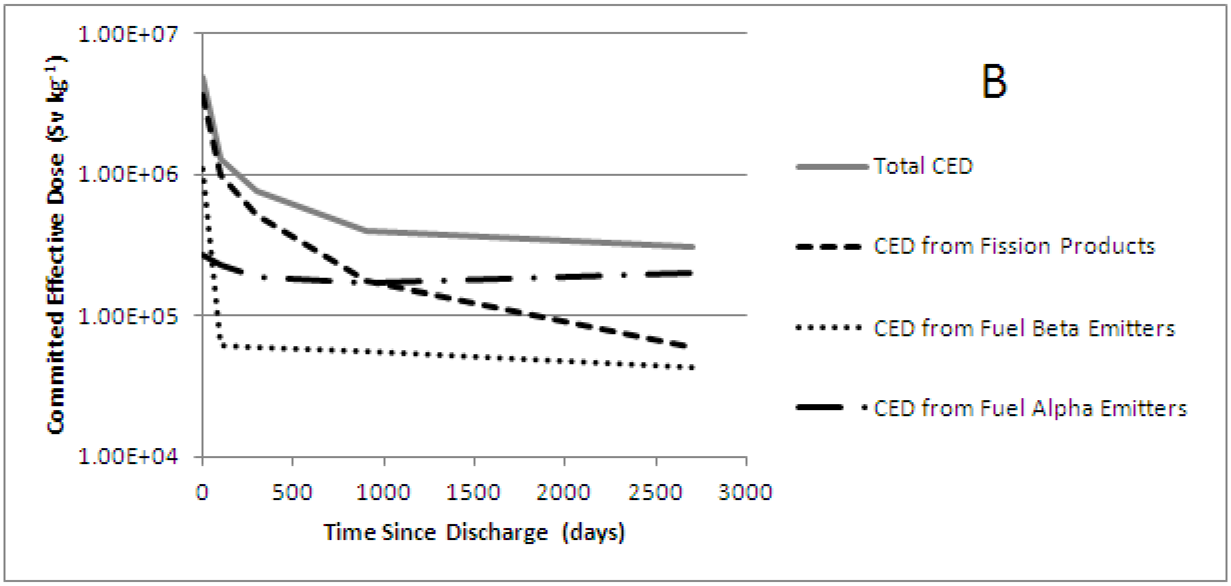


Figure 1

Average of all elements from a CANDU natural uranium bundle. The specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.

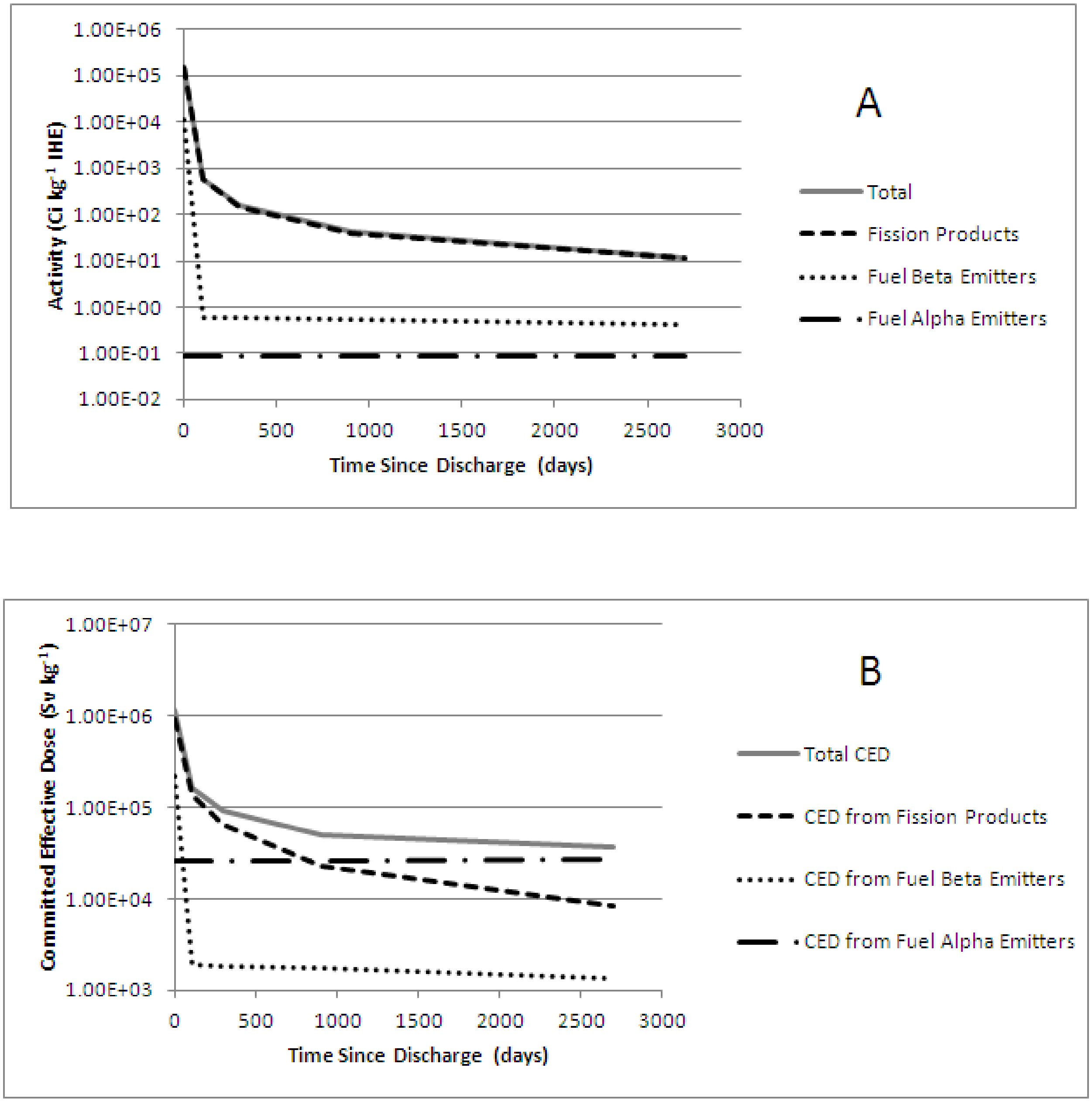


Figure 2

Center element from a CANDU low void reactivity bundle. The specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.

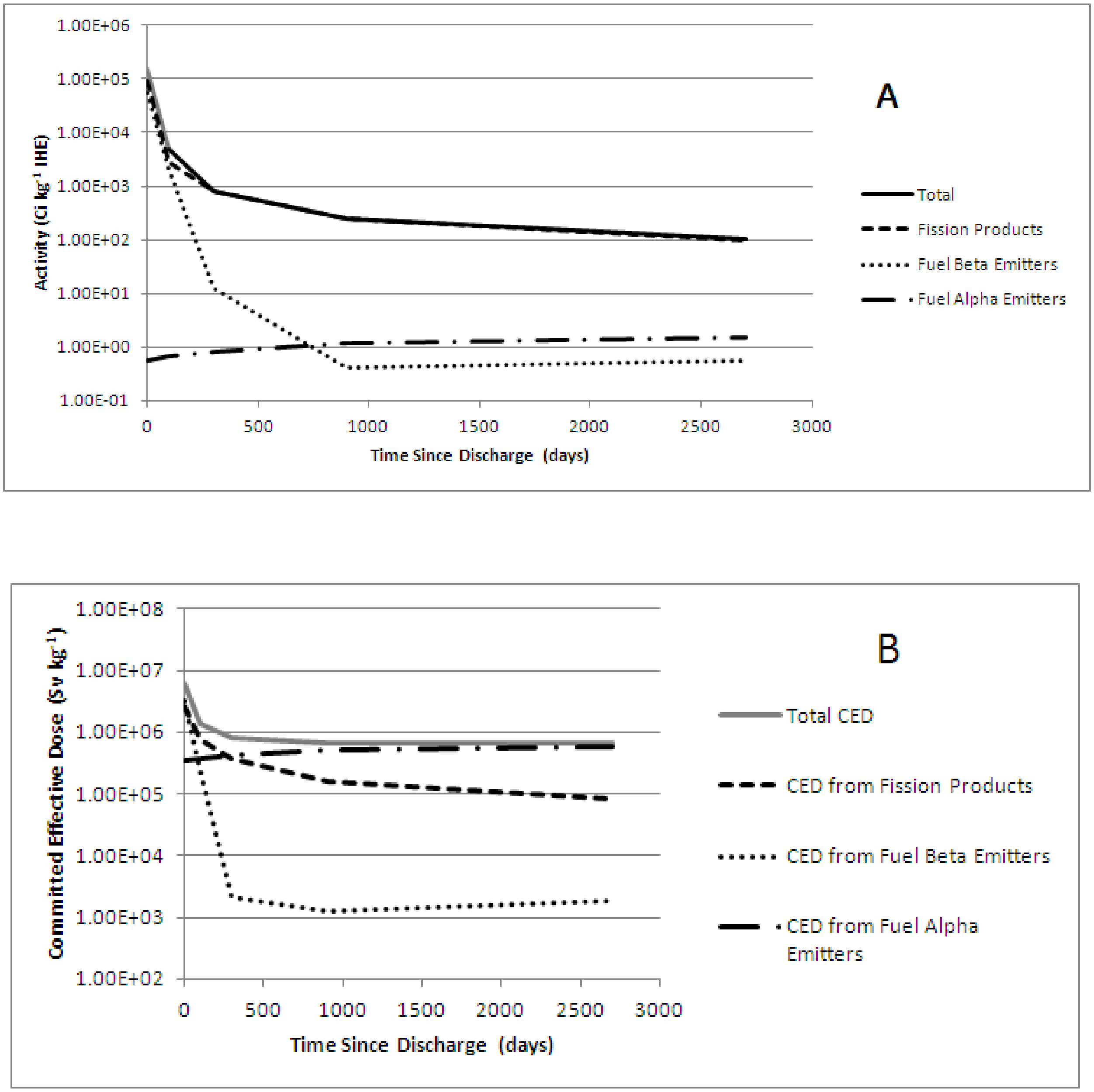


Figure 3

Center eight thorium elements from a CANFLEX (LEU,Th)O_2 bundle. The fuel specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.

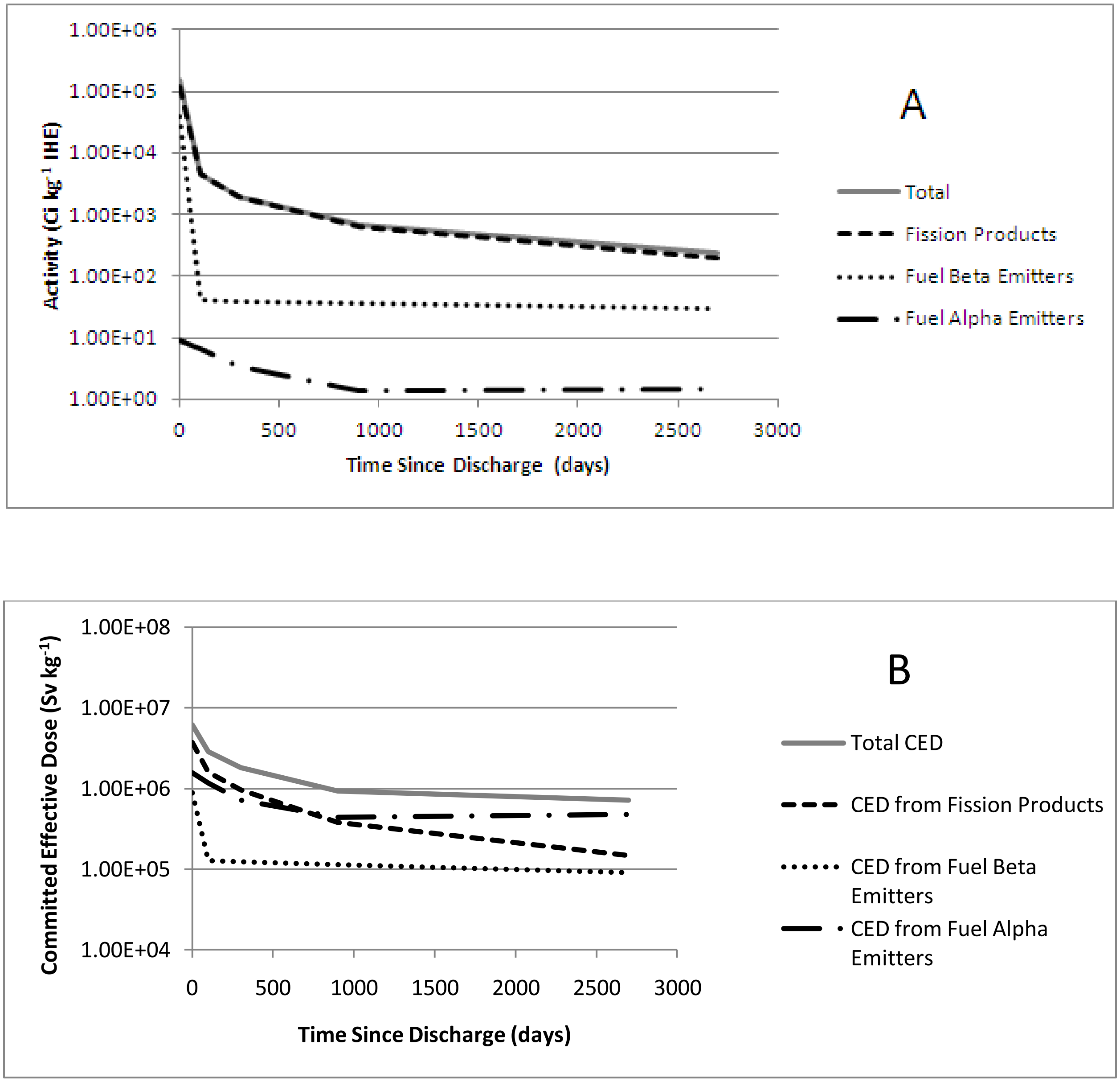


Figure 4

Outer 35 elements of an (LEU,Th)O_2 bundle. The fuel specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.

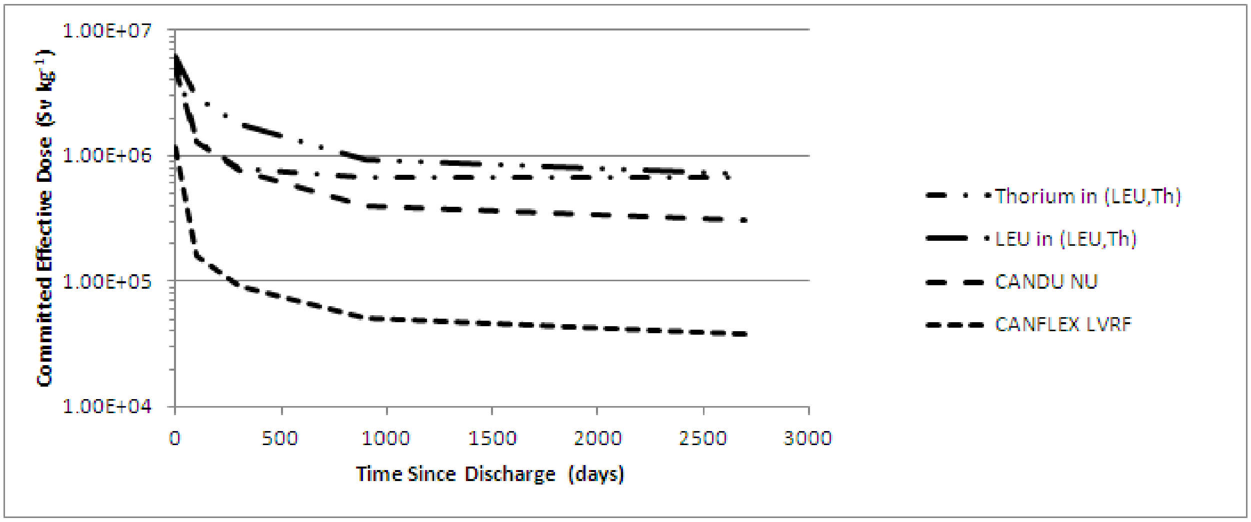


Figure 5

The calculated specific committed effective dose (Sv kg^{-1} inhalation, 5µm AMAD, ICRP Class S) i.e., $\tilde{E}(50)$ resulting from the inhalation of different CANDU fuels.

10. ENDNOTES

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[*] CANDU is a registered trademark of Atomic Energy of Canada Ltd.

[†] CANFLEX is a registered trademark Atomic Energy of Canada Ltd and the Korea Atomic Energy Research Institute.

[‡] Possible CANDU fuels include reformed, but otherwise un-reprocessed, LWR or AGR oxide fuels.

[§] Burnup is a term for the total energy produced by a nuclear fuel per unit mass. A typical unit would be MWd kg^{-1}.

[**] A kg is a convenient mass for calculations, not a realistic mass for inhalation exposure.

[††] Some radionuclides, like ^{233}U, which are created by the decay of a neutron absorbing intermediary (in this case ^{233}Pa) may be created more efficiently by a reactor history with regular downtime to allow the decay to occur, but the effect is usually small.

[‡‡] The thorium series comprises the naturally occurring decay products of ^{232}Th. This series contains isotopes of mass 228, 224 etc. and terminates in the creation of stable ^{208}Pb.

[§§] This is one conclusion from an AECL study of the expected composition of $(Pu,Th)O_2$ fuel taken to approximately 60 MWd kg^{-1} burnup in a supercritical water reactor. This work is unrestricted but has not yet appeared in the open literature.