Relative Hazard of High-Level Waste Over Long Time Periods

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1 INTRODUCTION

An issue associated with geologic disposal of nuclear waste is the time period for which containment and isolation of the waste should be of regulatory concern. With time, the radionuclides will decay, and the radiological hazard of the waste will decrease. At some time, the hazard of the nuclear waste will become comparable to the hazard of naturally occurring radioactive sources, such that the geologic disposal facility hazard becomes similar to a concentrated uranium ore body. This may be of some value in decision making even though such a concentrated uranium ore body may not actually exist in nature. A number of reports and analyses have addressed the reduction of radioactivity and radiological hazard of the waste with time (Environmental Protection Agency, 1985, 1982a, 1982b; Levi, 1980) including several by the Environmental Protection Agency (EPA) prior to issuing the since-remanded standard 40 CFR Part 191 (U.S. Environmental Protection Agency, 1989). To date, regulations have used a 10,000 yr regulatory time period. The National Academy of Sciences (NAS), as directed by the United States Congress in the Energy Policy Act of 1992, has provided EPA guidance (National Academy of Sciences, 1995) for an environmental standard for Yucca Mountain (YM), and has recommended:

> "... [the] calculation of the maximum risks of radiation releases whenever they occur as long as the geologic characteristics of the repository environment do not change significantly [underline added for emphasis]. The time scale for long-term geologic processes at Yucca Mountain is on the order of approximately one million years."

The purpose of this recommendation was to focus on the time at which future populations are at maximum risk. Because both the EPA and the Nuclear Regulatory Commission (NRC) are evaluating the time period of regulatory interest, these analyses were conducted to determine when the radioactivity and more importantly, the radiological hazard of the spent fuel become comparable to naturally occurring radioactive materials.

2 DESCRIPTION OF MODELING APPROACH

2.1 REPOSITORY AND ORE BODY

The approach used in this work is to compare the variations in total radioactivity and radiological hazard of a spent nuclear fuel repository and a hypothetical uranium ore body over a 100 million year time period. The ore body is defined to have the same amount of uranium and occupy the same volume as the repository. The hypothetical ore body contains only U-235 and U-238 and their radioactive daughters. The primary difference between the spent fuel repository and the hypothetical ore body is that repository waste has a significant man-made (through irradiation and fissioning) hazard as compared to the hazard of the ore body which is from naturally occurring nuclides. The major difference between this paper and previous work is that the most recently available data (e.g. solubilities, radionuclide inventories) and site characteristics of YM are used.

Figure 1 shows the major steps in the extraction of ore and the subsequent use of the nuclear fuel. The ore is transformed through three primary activities: mining, processing, and irradiating. Each process generates byproducts or waste. In the mining process, only the uranium is extracted from the ore body, hence the byproduct materials contain the radioactive daughters of U-235 (primarily Pa-231 and Ac-227) and U-238 (primarily U-234, Th-230, Ra-226, and Pb-210). These byproduct materials represent some radiological hazard, therefore regulations have been established for their long-term care and disposal (U.S. Environmental Protection Agency, 1995; Nuclear Regulatory Commission 1995a, 1995b). Processing includes refining and enriching the uranium to make it suitable as a reactor fuel. Processing also generates depleted uranium which possesses some radiological hazard. Interestingly, the largest single use of depleted uranium is for military kinetic-energy projectiles (Adams, 1995) which after use, presumably remain scattered about the ground surface. After a sufficiently long time, the radiological hazard of spent nuclear fuel is essentially the same as depleted uranium which, under current practices, generally remains on the target range and battlefield. After irradiation in a nuclear reactor, current regulations call for disposal of the spent fuel in a geologic repository (Nuclear Regulatory Commission, 1995c). The extraction and use of the original ore body thus generates three components of waste which eventually will be disposed of: mine waste, depleted uranium, and spent fuel.

2.2 RADIOACTIVITY AS A FUNCTION OF TIME

In Figure 2, the total radioactivity of spent fuel is shown along with the radioactivity of a number of the dominant radionuclides. At early times, the radioactivity is dominated primarily by fission products, while at later times it is dominated by transuranics (and their daughters). Strontium and cesium dominate the activity up to 100–200 yr, americium and plutonium up to 100,000 yr, and technetium and uranium beyond 100,000 yr. At times beyond 10 million years, the activity is dominated by U-235, U-238, and their daughters which are naturally occurring radionuclides. Figure 2 is in agreement with a similar figure shown in a study that was performed to support the rulemaking process for 40 CFR Part 191 in the mid 1980's [Figures A-4 and A-5 of (U.S. Environmental Protection Agency, 1985)].

In Figure 3, the inventory of the hypothetical ore body is shown. The ore body is enriched to 3.5 percent U-235 to match the preirradiation uranium content of the spent fuel. The inventory ranges over ten orders of magnitude, indicating the ore is predominantly uranium with trace amounts of daughters. The daughters are in equilibrium and their activity is 0.318 Curies per Metric Tonne of Uranium (Ci/MTU) for U-238, U-234, Th-230, Ra-226, and Pb-210, and is 0.085 Ci/MTU for U-235, Pa-231, and Ac-227. The total radioactivity is 1.85 Ci/MTU for the nuclides tracked in this work. Because U-235 and U-238 have very long half-lives, their inventory and radioactivity remain essentially constant over 100 million years.

In Figure 4, the total radioactivity of the spent fuel repository and the hypothetical ore body are compared. By one thousand years, the radioactivity of spent fuel is one percent of what it was at ten years after irradiation. At ten years from the reactor, the spent fuel radioactivity is about 182,800 Ci/MTU. The ore body radioactivity is about constant at 1.85 Ci/MTU. At approximately 10^5 yr the spent fuel radioactivity will have decreased by 99.99 percent and be within an order of magnitude of the ore body radioactivity. Beyond 10 million years, the total radioactivity it is essentially equal to the ore body.

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2.3 RADIOLOGICAL HAZARD AS A FUNCTION OF TIME

Although informative, a comparison of total radioactivity does not account for the radiological hazard of the waste, since different radionuclides have different radiotoxicities. For example, ingesting one curie of Am-241 is approximately one hundred times more hazardous than ingesting one curie of Cs-137 (U.S. Environmental Protection Agency, 1988; U.S. Department of Energy 1988). In this work, we compare the radiological hazards associated with drinking groundwater which has been contaminated by either a repository or a hypothetical ore body. In order to make such a comparison, some modeling assumptions and site specific data are employed. The model, as illustrated in Figure 5, consists of steadily percolating groundwater which flows through the repository and into the saturated zone. Some of the percolating groundwater contacts some of the waste packages. The water that contacts the waste becomes contaminated with radionuclides. For the analyses, 43 different radionuclides are considered from ten to one hundred million years after irradiation (Lozano et al., 1994).

The repository is assumed to have a footprint area of 5 km^2 (TRW Environmental Safety Systems, 1994), a 1 mm/yr spatially homogeneous percolation rate (Nuclear Regulatory Commission, 1995d; Wilson et al., 1994), a cross-sectional area of 10 m^2 and a payload of 10 MTU per waste package (TRW Environmental Safety Systems, 1994), and a package areal density of one waste package per 500 m^2 (corresponding to an 80 MTU/Acre areal mass loading). The repository and the ore body contain 63,000 MTU, which is consistent with current designs for a proposed repository at Yucca Mountain, Nevada (TRW Environmental Safety Systems, 1994). These assumptions lead to approximately 2 percent of the percolating groundwater contacting 1 percent of the waste packages. To be consistent, the same assumption is used for the ore body. The percentage of percolating groundwater that contacts the waste is the ratio of the cross-sectional area of all the waste packages compared to the total repository footprint area. The percentage of waste packages contacted is based on the ratio of the area of a single waste package to that area necessary to funnel water in order to sustain dripping. Sustained dripping is estimated to require a focusing from a 1000 m^2 area in order to capture a sufficient amount of water.

The drinking water pathway dose conversion factors (DCF_{dw}), assuming a drinking rate of two liters per day, and solubilities of the radionuclides with associated uncertainties are summarized in Table 1. The DCF_{dw}'s are based on a drinking water pathway being the sole means of exposure, and the ingestion dose conversion factors used to calculate the DCF_{dw}'s are taken from (U.S. Environmental Protection Agency, 1988; U.S. Department of Energy, 1988). The solubilities are based largely on earlier work (Nuclear Regulatory Commission 1995d; Wilson et al., 1994).

Figure 6 shows the relative radiological hazard which is the ratio of the doses received by drinking groundwater contaminated by the repository and ore body. The ratio of drinking water doses is not affected by dilution if the contaminant plumes behave the same for the repository and ore body. Sorption of radionuclides is neglected, which may affect the relative hazard. Mean values for radionuclide solubilities and bulk waste dissolution (release) rate were used to generate Figure 6. The radiological hazard is dominated by strontium and cesium up to 100-200 yr, by plutonium and americium up to 20,000 yr, and by neptunium and uranium daughter products beyond 20,000 yr. The plateaus in some of the radionuclide dose curves are due to the solubility-limited release of the radionuclide. Plutonium and americium have relatively low solubilities and hence are solubility-limited at early times. If the radionuclides are highly soluble or if the radionuclide inventory is small, then the radionuclide concentrations and the relative radiological hazard are controlled by the release rate. The maximum

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release rate that was assumed for generating Figure 6 was one part in 100,000 of the current inventory per year based on other work (Nuclear Regulatory Commission, 1995d).

By comparing Figures 2 and 6, one concludes that total radioactivity roughly correlates with radiological hazard. The radiological hazard and total radioactivity are highest during the first 1,000 yr. In general, the hazard of a radionuclide can be significantly diminished if it has a relatively low solubility in water or a low ingestion dose conversion factor. In general, alpha-emitters have higher ingestion dose conversion factors, and this is reflected in current regulatory limits. Extended containment and retardation of radionuclides can significantly reduce radiological hazard, yet are not included in this model.

Figure 7 shows the range of radiological hazard due to uncertainties in radionuclide solubilities and release rates. The range of solubilities is based on Nuclear Regulatory Commission, (1995d) and Wilson et al., (1994). In general solubilities are highly uncertain due to uncertainties in the geochemical conditions. For both the repository and ore body, 100 distinct estimates have been generated by lognormally sampling the radionuclide solubilities and release rate [the common logarithm of the release rate was assumed to be -5 with a standard error of 1/2 based on previous work, Nuclear Regulatory Commission, (1995d)]. If one draws a horizontal line on Figure 7 at a relative hazard equal to 1, it can be observed that none of the 100 realizations cross below the line prior to about 1,000 yr and half of the lines cross below prior to about 10 million years. Prior to 1,000 yr the repository is distinctly more hazardous than the ore body. Beyond 10 million years there is a negligible difference between the repository and ore body.

The relative radiological hazard is initially about four orders of magnitude greater than the ore body. The hazard diminishes most rapidly over the first few hundred to a few thousand years. Beyond about 10,000 yr, the radiological hazard diminishes less rapidly. The apparent increase in hazard at 100,000 to 500,000 yr is due to the ingrowth of radionuclides such as Th-230, Th-229, Ra-226, and Pb-210 as observed in Figure 2. By 10,000 yr the relative hazard will have decreased by 99.9 percent and be within less than an order of magnitude of the hypothetical ore body. A time period of interest for regulation of a proposed repository of 10,000 yr would, therefore, focus attention on the time period when the waste has a significant man-made hazard component which is readily discernable from a hypothetical ore body even after considering uncertainties associated only with solubilities and release rates. The findings of this study are consistent with those of many earlier studies (U.S. Environmental Protection Agency, 1982a, 1985).

3 ASSUMPTIONS AND LIMITATIONS

The assumptions and limitations are discussed throughout this work as the analysis is described, but are summarized here for convenience.

- The only radiological hazard considered is drinking contaminated groundwater.
- Groundwater percolates through a 5 km^2 repository area at a rate of 1 mm/yr such that 2 percent of the groundwater contacts 1 percent of the waste thereby becoming contaminated to the maximum extent reasonable (either solubility limited or release rate limited).

- Radionuclide solubilities and release rates (for highly soluble elements) are based on prior work, see Table 1.

- The hypothetical uranium ore body contains the same quantity of uranium as the pre-irradiation nuclear fuel, as well as the decay daughters of the uranium in equilibrium.

- The contamination of the groundwater by the ore body is the same as for the repository. The primary difference is that the spent fuel has radioactive fission products and activation products that are not present in the ore body.

4 SUMMARY AND CONCLUSIONS

The total radioactivity of a spent fuel repository decreases by 4 orders-of-magnitude within 10^5 yr after irradiation. During this time, about 99.99 percent of the gross radioactivity at 10 yr from reactor has naturally dissipated. Beyond this time, the radioactivity is comparable (within an order of magnitude) to that of a hypothetical uranium ore body. Only after 10^7 yr is the difference in gross radioactivity indistinguishable. The relative radiological hazard associated with a spent fuel repository decreases rapidly (by 3 orders-of-magnitude) within 10^4 yr. During this time, about 99.9 percent of the relative hazard has naturally dissipated. Beyond this time, the radiological hazard is comparable (within an order of magnitude) to that of a hypothetical uranium ore body. Only after 10^7 yr is the difference in radiological hazard indistinguishable.

Based on the large total radioactivities and radiological hazards during early times, it is suggested that the regulatory period of interest focus attention on times less than about 10^4 yr when the waste has a significant and distinct man-made hazard (statistically greater than the hazard of the naturally occurring component of the waste). Ultimately, the time period of regulatory interest will be decided after considering a spectrum of technical, social, ethical, and political perspectives. It is suggested that this study, as well as other earlier studies, have a valuable role in the decision making process.

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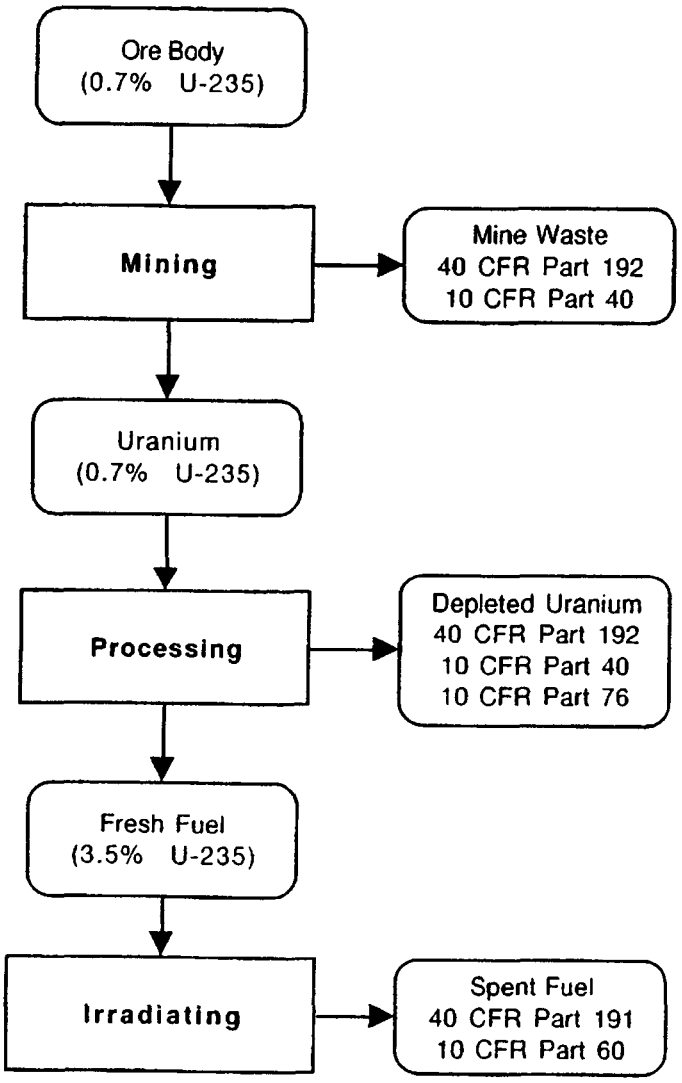

Figure 1. Major steps in extraction and use of uranium, along with major byproducts of activities and associated regulations

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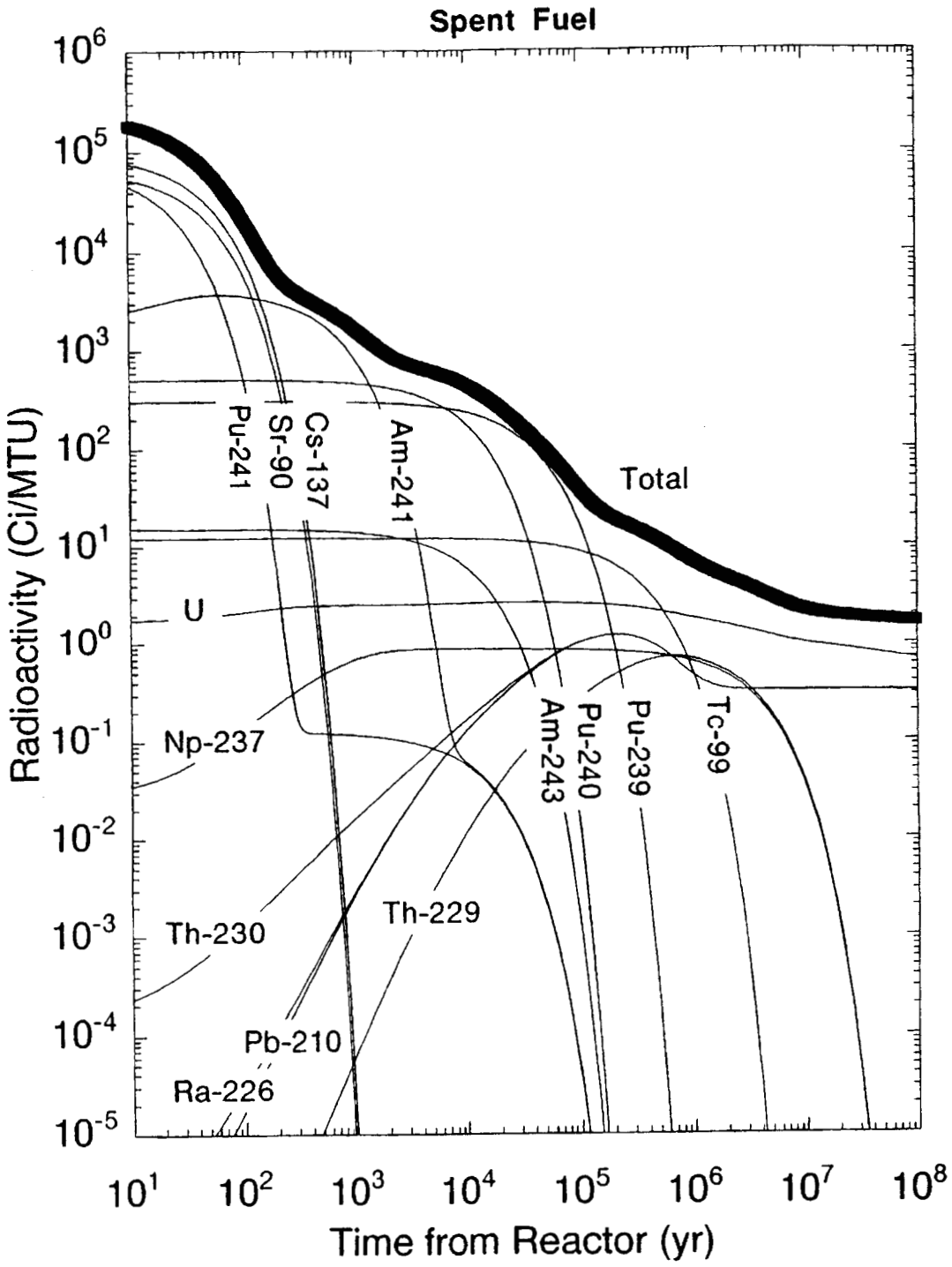

Figure 2. Radioactivity of spent nuclear fuel

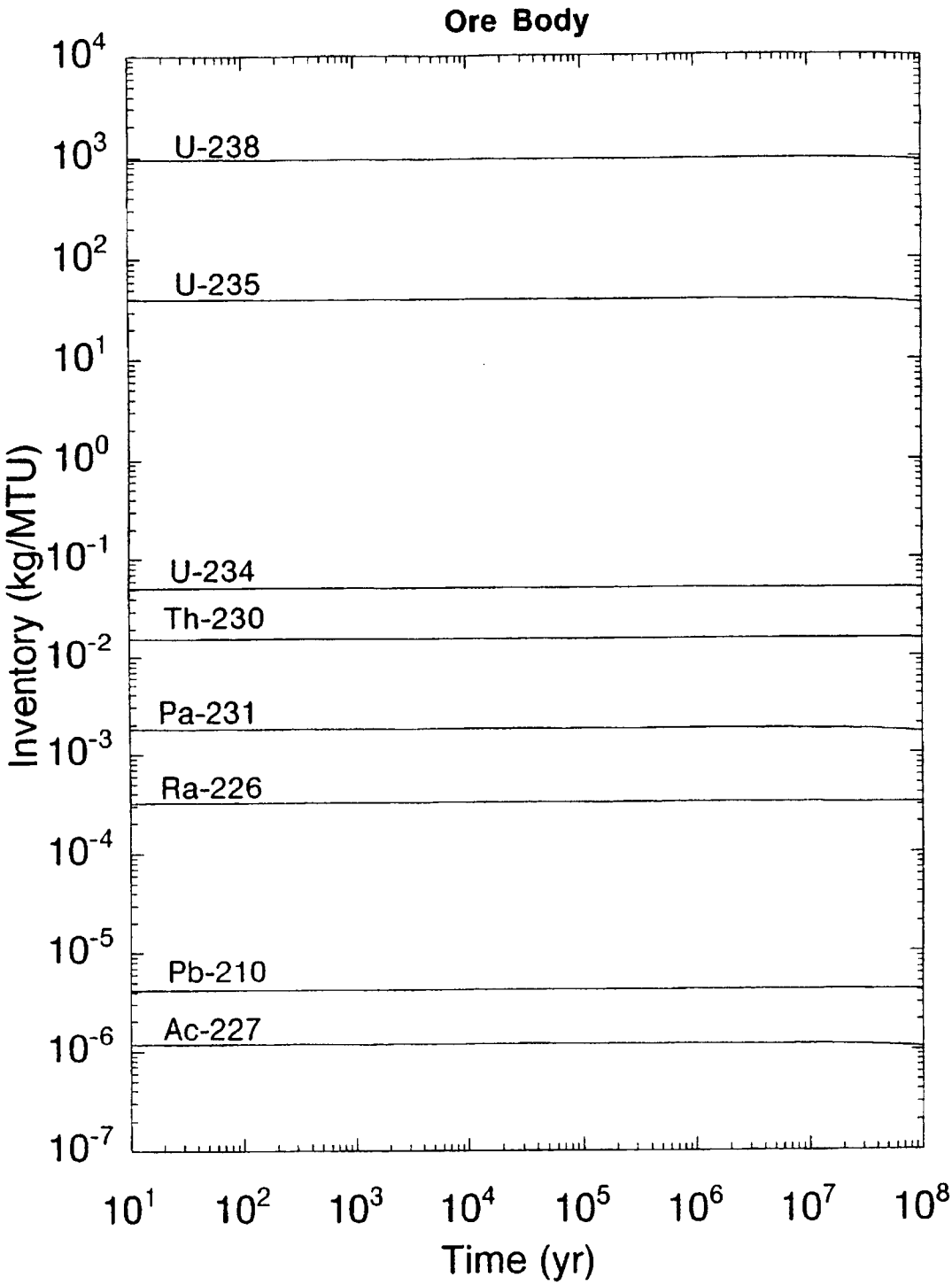

Figure 3. Radionuclide inventory of the ore body

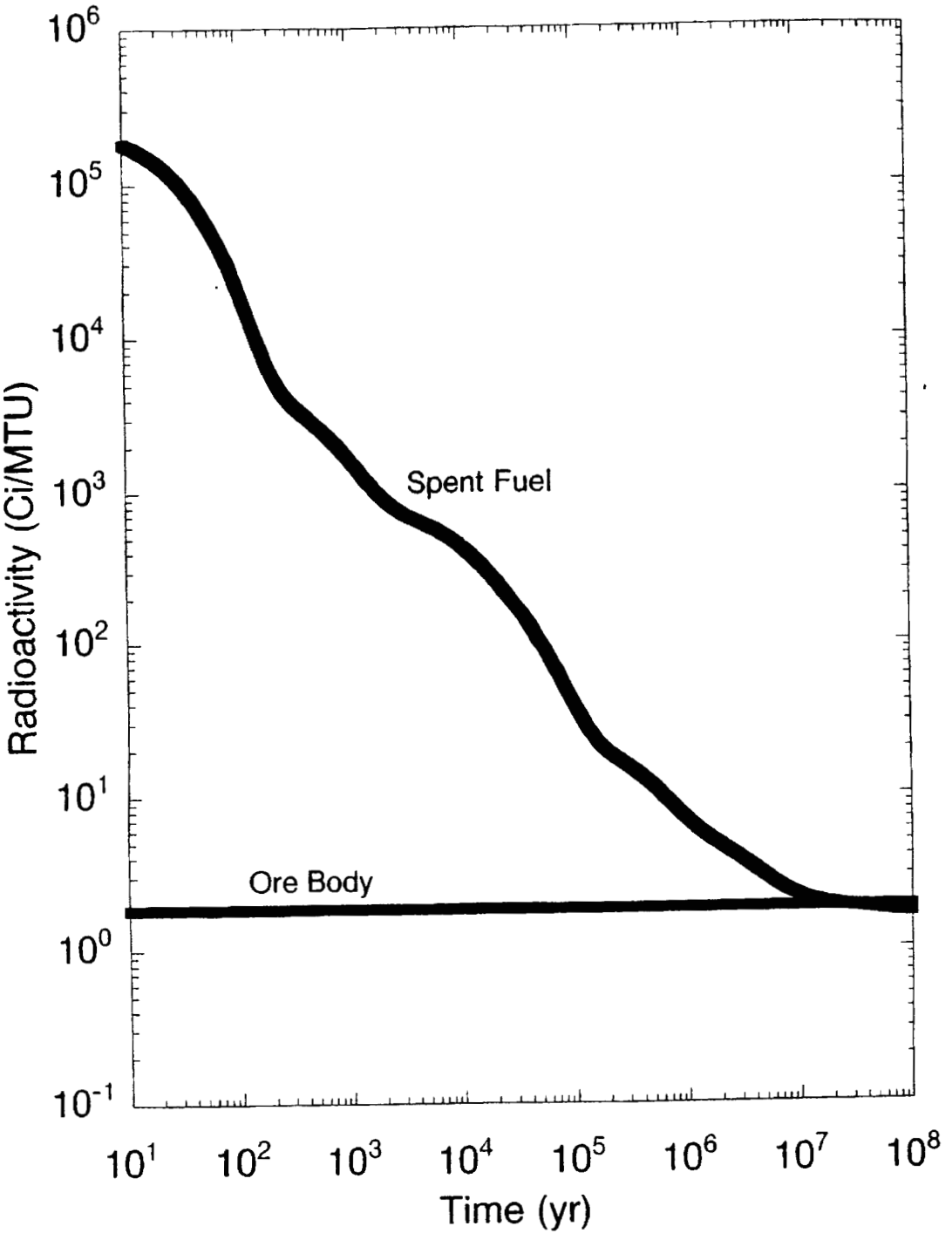

Figure 4. Comparison of radioactivity of spent fuel and hypothetical ore body

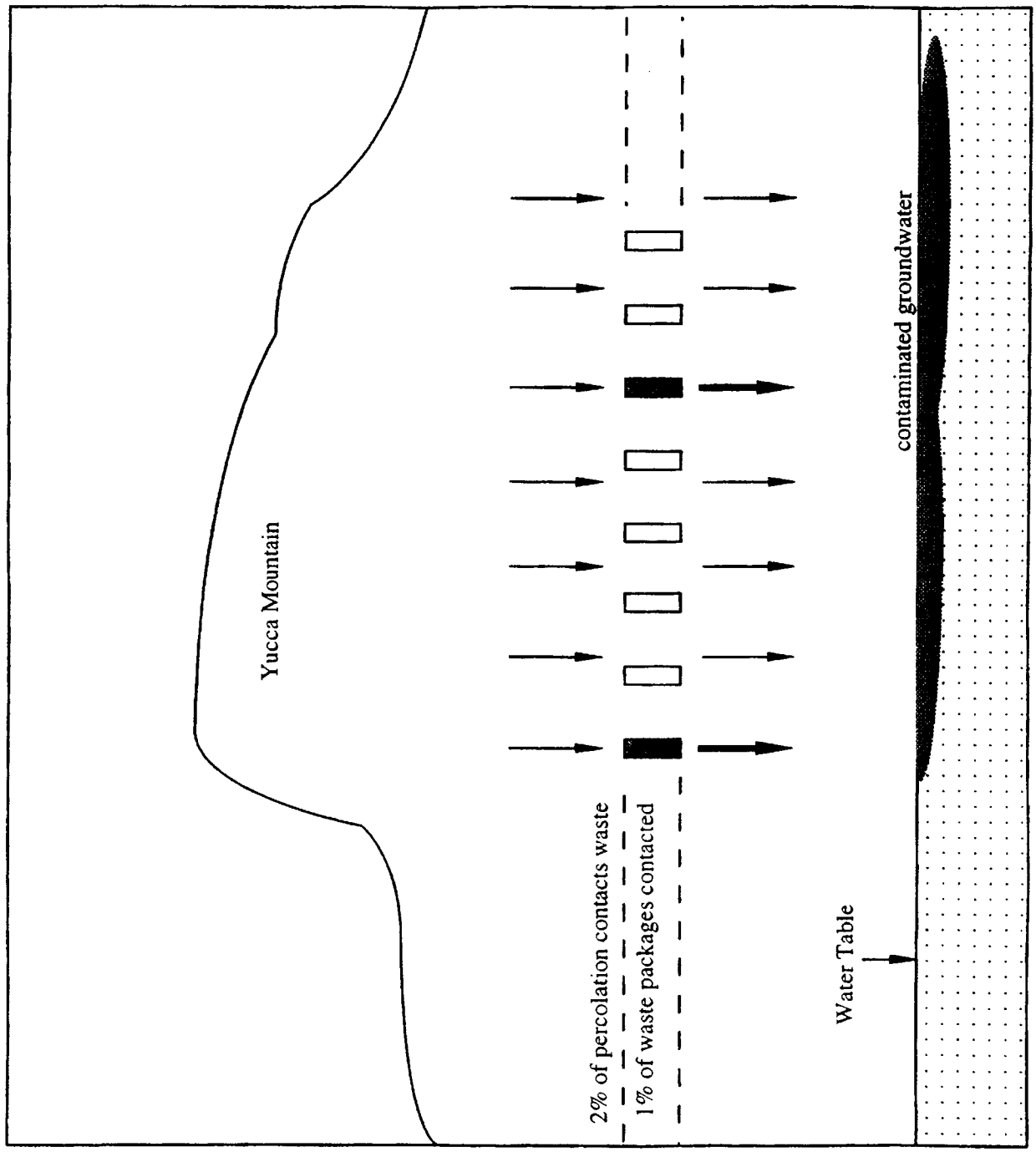

Figure 5. Drinking water dose model

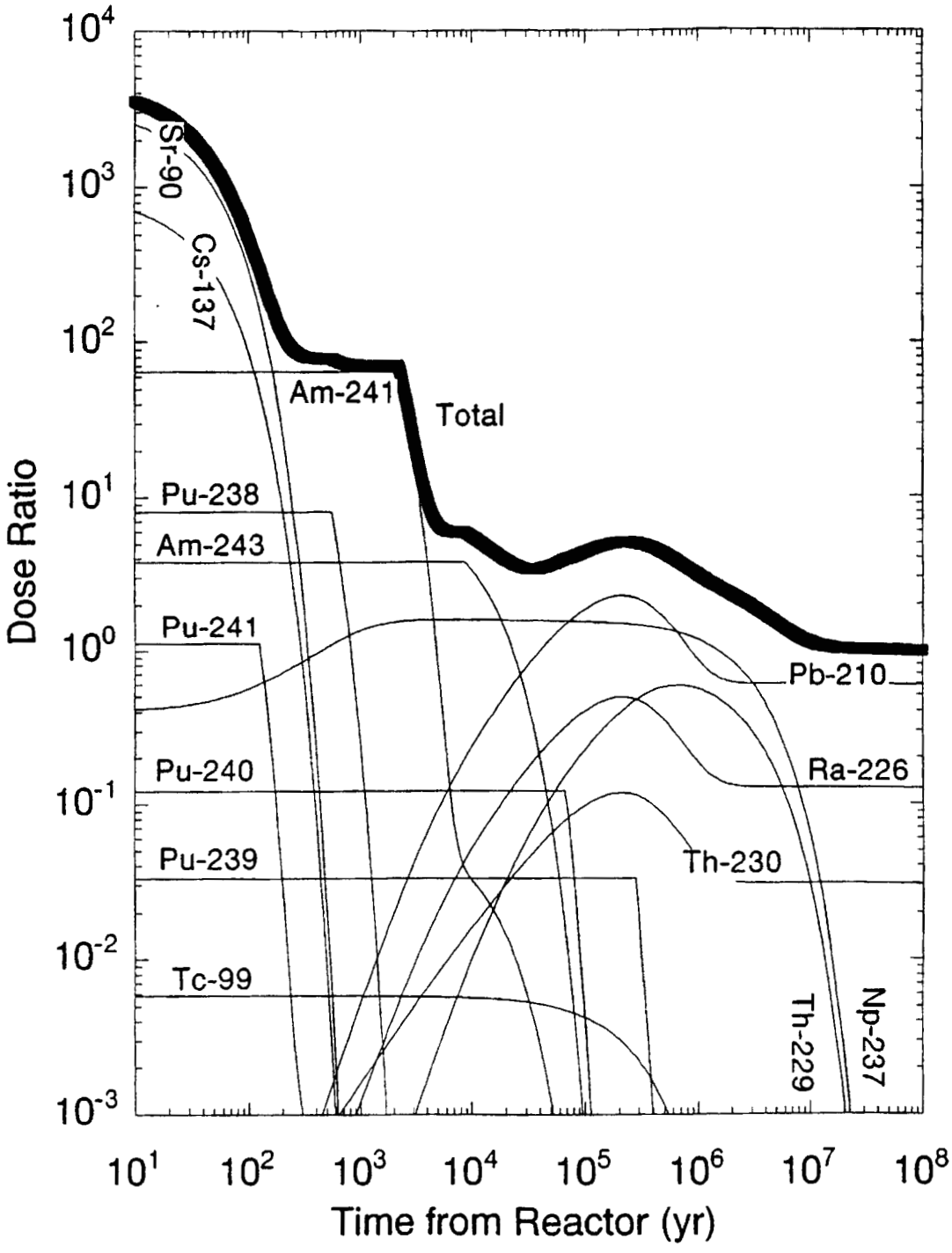

Figure 6. Relative radiological hazard of spent nuclear fuel calculated as the ratio of doses from drinking groundwater contaminated by the repository and ore body.

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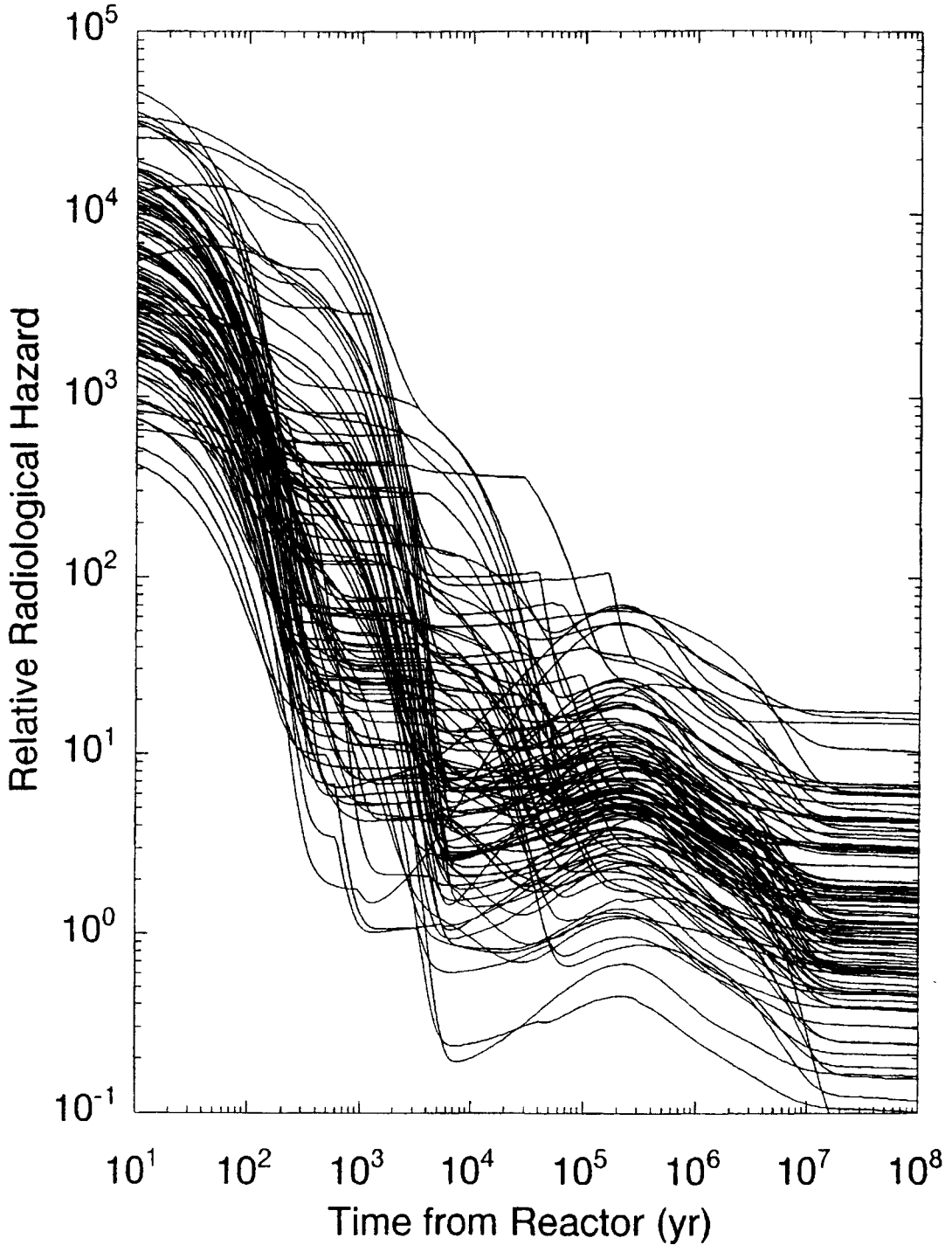

Figure 7. Comparison of hazards accounting for uncertainties in radionuclide solubilities and release rates

Table I. Solubilities and dose conversion factors for 43 radionuclides used in this work

Nuclide	Common Logarithm of Solubility [mol/L] based on NRC (1995d) and Wilson et al. (1994)	Drinking Water Dose Conversion Factors [rem/yr/Ci/m^3] based on EPA (1988) and DOE (1988)
U-238	-5.3 ± 1	1.68E+06
Cm-246	-5.3 ± 1	3.29E+07
Pu-242	-10.0 ± 2	2.99E+07
Am-242m	-8.5 ± 2	3.07E+07
Pu-238	-10.0 ± 2	2.77E+07
U-234	-5.3 ± 1	1.90E+06
Th-230	-8.0 ± 2	3.87E+06
Ra-226	-7.0 ± 2	8.03E+06
Pb-210	-6.0 ± 1	3.72E+07
Cm-243	-5.3 ± 1	2.12E+07
Am-243	-8.5 ± 2	3.29E+07
Pu-239	-10.0 ± 2	3.14E+07
U-235	-5.3 ± 1	1.83E+06
Pa-231	-7.0 ± 2	8.03E+07
Ac-227	-10.0 ± 2	1.02E+08
Cm-245	-8.5 ± 2	3.29E+07
Pu-241	-10.0 ± 2	6.28E+05
Am-241	-8.5 ± 2	3.29E+07
Np-237	-3.7 ± 1	2.85E+07
U-233	-5.3 ± 1	1.97E+06
Th-229	-8.0 ± 2	2.56E+07
Cm-244	-8.5 ± 2	1.68E+07
Pu-240	-10.0 ± 2	3.14E+07
U-236	-5.3 ± 1	1.83E+06
U-232	-5.3 ± 1	9.49E+06
Sm-151	-10.0 ± 2	2.48E+03
Cs-137	Large*	3.65E+05
Cs-135	Large	5.18E+04
I-129	Large	2.04E+06
Sn-126	-7.3 ± 2	1.24E+05
Sn-121m	-7.3 ± 2	9.49E+03
Ag-108m	-10.0 ± 2	5.48E+04
Pd-107	-6.0 ± 1	1.02E+03
Tc-99	Large	9.49E+03
Mo-93	-6.0 ± 1	9.49E+03
Nb-94	-8.0 ± 2	3.72E+04
Zr-93	-9.0 ± 2	1.17E+04
Sr-90	-3.7 ± 1	9.49E+05
Se-79	Large	6.06E+04
Ni-63	-2.7 ± 1	3.94E+03
Ni-59	-2.7 ± 1	1.46E+03
Cl-36	Large	2.19E+04
C-14	Large	1.53E+04

*Highly soluble radionuclides are release rate constrained.