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AN EVALUATION OF THE IMPORTANT RADIONUCLIDES FOR PERFORMANCE ASSESSMENT

Prepared for

Nuclear Regulatory Commission
Contract NRC-02-93-005

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May 1995

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ABSTRACT

This report describes a method and calculations to determine which radionuclides, expected to be present in spent fuel, are important to consider for performance assessments of a proposed repository at Yucca Mountain, Nevada. The method, which ranks the radionuclides according to rudimentary calculations of their ability to deliver doses to humans at the accessible environment, is performed for 27 different nuclides that other performance assessment efforts have found to be important, according to two different groundwater flow regimes. In the first regime, groundwater is assumed to flow in the rock matrix where sorption is able to retard the transport of radionuclides to the accessible environment. Any radionuclide that delivers a maximum dose of greater than 0.2 mrem/yr is considered important. In the second regime, groundwater is assumed to flow in the rock fractures where no sorption of radionuclides is assumed. Since all nuclides in this regime will reach the accessible environment at the same time after release, it is possible to make judgments of the radionuclide importance in a relative manner. All radionuclides that contributed to 99 percent of the dose at times of 2,000, 11,000, and 101,000 yr after repository closure are considered important under this regime. The radionuclides found to be important in this report are: ^{246}Cm, ^{245}Cm, ^{243}Am, ^{241}Am, ^{242}Pu, ^{240}Pu, ^{239}Pu, ^{237}Np, ^{238}U, ^{236}U, ^{234}U, ^{233}U, ^{231}Pa, ^{230}Th, ^{229}Th, ^{227}Ac, ^{226}Ra, ^{210}Pb, ^{135}Cs, ^{129}I, ^{99}Tc, ^{79}Se, and ^{14}C. This list is composed of radionuclides that are deemed important under either groundwater flow regime.

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ACKNOWLEDGMENTS

This report was prepared to document work performed by the Center for Nuclear Waste Regulatory Analyses (CNWRA) for the Nuclear Regulatory Commission (NRC) under Contract No. NRC-02-93-005. The activities reported here were performed on behalf of the NRC Office of Nuclear Material Safety and Safeguards (NMSS), Division of Waste Management (DWM). This report is an independent product of the CNWRA and does not necessarily reflect the views or regulatory position of the NRC.

The authors wish to thank R.T. Pabalan and P.A. LaPlante for their technical reviews and B. Sagar for his programmatic review.

QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: CNWRA-generated original data contained in this report meets quality assurance requirements described in the CNWRA Quality Assurance Manual. Sources for other data should be consulted for determining the level of quality for those data.

ANALYSES AND CODES: No computer codes were used in this report.

1 INTRODUCTION

Before estimating the total system performance in Iterative Performance Assessment (IPA) Phase 3, it was decided to reassess the radionuclides that should be considered in performance assessments (PAs) of the proposed high-level nuclear waste (HLW) repository at Yucca Mountain, Nevada. This reassessment takes into consideration the following:

(i) A potential shift from a release-based standard to a dose-based standard

(ii) A potential increase in the time of regulatory interest (conceivably up to 1,000,000 yr)

(iii) Recent improvement in the understanding of the release and sorption processes for a potential repository at Yucca Mountain

(iv) Insights and conclusions from recent PA reports

In quantifying which radionuclides are most important for characterizing the hazard of spent nuclear fuel disposal at Yucca Mountain, many complicated issues related to source characterization, geologic transport, and radiation dose calculation must be resolved. Arguably, the most important process to be considered in this quantification is hydrologic transport. This importance is due to the potential for sorption of radionuclides on geologic media to greatly increase (by perhaps several orders of magnitude) the time required for the nuclide to reach the accessible environment. During this increased travel time, radionuclide decay can, in some cases, lead to a reduced hazard. The increase in radionuclide travel time relative to the groundwater travel time (GWTT) is generally referred to as retardation.

One of the more challenging technical problems in predicting a potential repository's performance at Yucca Mountain is modeling water flow in the unsaturated zone below the repository. For example, it is believed that if the rock becomes fully saturated due to groundwater recharge, then water flow would be dominated by flow in fractures that permeate the rock. Conversely, if the rock matrix is not fully saturated and water is under tension, then water would tend to flow through the rock matrix. Whether water is flowing in the rock fractures or the rock matrix can have a serious effect on groundwater velocity and retardation calculations and thus, on what radionuclides are the most important contributors to radiation doses to humans at the accessible boundary of a potential repository.

The purpose of this report is to summarize rudimentary calculations of radiation dose to humans at the accessible environment in aqueous release scenarios under two groundwater flow regimes:

(i) Flow is dominated by flow in the rock matrix, where flow is slow with great opportunity for retardation of radionuclides via sorption on geologic media

(ii) Flow is dominated by flow in the rock fractures, where flow is fast and there is minimal opportunity for retardation of radionuclides via sorption on geologic media

It was determined from these simplified calculations which of the radionuclides from the spent fuel inventory are important contributors to radiation dose to humans at the accessible environment. The radionuclides considered in this study are; (i) heavy elements Cm (isotopes 245 and 246), Am (isotopes 241 and 243), Pu (isotopes 238, 239, 240 and 242), U (isotopes 233, 234, 235, 236 and 238), ^{237}Np,

^{231}Pa, Th (isotopes 229 and 230), ^{227}Ac, ^{226}Ra, and ^{210}Pb, and (ii) fission and activation products Cs (isotopes 135 and 137), ^{129}I, ^{99}Tc, ^{79}Se, ^{59}Ni, and ^{14}C. The list of considered radionuclides was based on the results of other PAs and reports (Wilson et al., 1993; Wescott et al., 1995; Eslinger et al., 1993; Andrews et al., 1994; Duguid et al., 1994). It should also be noted that the radiation dose calculations assume that the only mechanism for delivering radiation doses to humans is the drinking of contaminated water. While this approach may be nonconservative, it is acceptable for the order of magnitude type calculations presented here. Also, this approach obviates the extensive speculations about water use and human activity scenarios that are beyond the scope of this study.

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2 METHODS

2.1 INTRODUCTION

The approach of this paper is to estimate the ability of each of the radionuclides to deliver radiation doses to humans residing at the accessible environment. This estimate is made by calculating an importance index for each of the radionuclides considered in this report. The following section describes the mathematical development of the importance index.

2.2 THE IMPORTANCE INDEX (F_i)

The calculation of F_i (in mrem/yr) begins by accounting for the maximum amount of radionuclide i released from the repository per year. This amount is given by:

$$F_i = I_{\max_i} * DR_i \tag{2-1}$$

where:

$I_{\max_i}$ The maximum repository inventory, in time, for radionuclide i (in picocuries), (Lozano et al., 1994) assuming that no radionuclides leave the waste packages (conservative)

DR_i The fractional dissolution rate of radionuclide i into the water flowing through the repository (in inverse years) (Duguid et al., 1994; Kerrisk, 1985)

The values of $I_{\max}$ were calculated in Lozano et al. (1994) using the Oak Ridge Isotope GENeration Code, Version 2, ORIGEN2 for light-water reactor spent fuel. Values for DR were calculated in Kerrisk (1985) by assuming saturation of groundwater flow through the repository with radionuclides for solubility limited radionuclides and by assuming, for the nonsolubility limited radionuclides, that dissolution occurs at the bulk dissolution rate. For the special cases of Cs, I, C, and Sr, the DR values were adjusted upward to account for diffusion of the radionuclides within the bulk waste leading to higher dissolution rates. The values for DR in Duguid et al. (1994) were calculated using a procedure defined in the National Research Council, National Academy of Sciences (NAS) (1983) report. The amount of radionuclide i released from the repository is then diluted in the amount of water flowing through the repository. The importance index becomes:

$$F_i = I_{\max_i} * DR_i * \frac{1}{Vw} \tag{2-2}$$

where

V_w The volumetric flow rate of water through the repository $= 9.43 \times 10^4$ (l/yr) (Duguid et al., 1994)

The volumetric flow rate through the repository was calculated assuming a yearly recharge of 1 mm/yr at Yucca Mountain flowing uniformly through a repository area of 9.43×10^4 m^2. Next, radioactive decay of nuclide i during transport to the accessible environment is accounted for. The importance index becomes:

$$F_i = I_{\max_i} * DR_i * \frac{1}{Vw} \exp\left(\frac{-ln2}{t_{1/2_i}} R_{f_i} t_w\right) \quad (2\text{-}3)$$

where

t_w The GWTT from the repository to the accessible environment, assumed to be 1,000 yr for these calculations (assumed minimum value)

R_{f_i} The radionuclide retardation factor, that is, the ratio of radionuclide travel time to the GWTT (Duguid et al., 1994; Kerrisk, 1985; or Wilson et al., 1993)

$t_{1/2_i}$ The radionuclide half life (yr) (Duguid et al., 1994)

Retardation factors in Duguid et al. (1994) were taken largely from the NAS (1983) report and were adjusted somewhat using data from Barnard et al. (1991). Retardation factors in Kerrisk (1985) were chosen from a compilation of sorption data from Daniels et al. (1982). Retardation factors in Wilson et al. (1993) were compiled from an expert elicitation of sorption factors contained in the report. The most conservative estimate from the range of sorption factors in Wilson et al. (1993) was used to determine the R_f from that reference. The importance index must also account for dilution of the contaminated groundwater during transport. The importance index becomes:

$$F_i = I_{\max_i} * DR_i * \frac{1}{Vw} * \exp\left(\frac{-ln2}{T_{1/2_i}} R_{f_i} t_w\right) * DF \quad (2\text{-}4)$$

where

DF The dilution factor, calculated by diluting the water flowing through the repository with the yearly water usage in the area (conservative) = 3.4×10^{-5} (Duguid et al., 1994)

The dilution factor in Duguid et al. (1994) was calculated assuming that the water flowing through the repository is diluted with the yearly water usage in the Yucca Mountain area, that is, no credit is taken for dilution with existing groundwater. Finally, it is assumed that a person residing in the accessible environment procures all of his drinking water from a well that is contaminated with radionuclides from the repository. The importance index is written in its final form as:

$$F_i = I_{\max_i} * DR_i * \frac{1}{Vw} * \exp\left(\frac{-ln2}{T_{1/2_i}} R_{f_i} t_w\right) * DF * DCF_{\text{ing}_i} * L \quad (2\text{-}5)$$

where

DCF_{ing} The dose conversion factor for ingestion for radionuclide i (in mrem/picocurie) (U.S. Department of Energy, 1988)

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L The yearly ingestion rate of the contaminated water = 730 l/yr (assumes a drinking rate of two l/day)

The time at which a nuclide in the repository could deliver a maximum dose, as estimated by the importance index, must also be considered. This time is given by:

$$t_{del_i} = t_{max_i} + R_{f_i} * t_w \tag{2-6}$$

where

t_{del_i} The estimated time of delivery of the maximum dose from radionuclide i

t_{max_i} The time at which the inventory of radionuclide i reaches a maximum in the waste package

By using the best available data, if one could show that an estimate of a given radionuclide's ability to deliver a maximum dose is very small, for example, less than 0.2 mrem/yr, then one could discount that radionuclide in any dose calculations. Similarly, if one could show that a vast majority of the radiation dose at all times, for example 99 percent is comprised of contributions from several radionuclides, then one could discount the complementary set of radionuclides without introducing significant error. One would, of course, compile the 99 percent contribution by starting with the highest F_i and continuing to sum with progressively lower F_i until 99 percent of the total F_i is reached. This would lead to the smallest set of nuclides comprising the 99 percent contribution.

The choice of the 99 percent cutoff is obvious. The 0.2 mrem/yr cutoff was chosen because the authors felt that 0.2 mrem/yr is small compared to the 15 mrem/yr total dose limit given in the Code of Federal Regulations, Title 40, Part 191, for undisturbed repository performance. It is noted, however, that the method would work equally well with a lower cutoff if the reader so desires.

It should also be noted that the method presented here assumes that ingestion of drinking water is the dominant pathway for receiving dose. In actuality, many pathways can lead to exposures. Data for these pathways at Yucca Mountain is currently unavailable. If, in the future, better data become available for assessing radiation doses from radionuclides contained in well waters for Yucca Mountain, then the method should be revisited using these data.

2.3 DIFFICULTIES WITH THE METHOD FOR CERTAIN DAUGHTER PRODUCTS

Several of the considered radionuclides are daughter products in a decay chain. These radionuclide decay chains are:

(i) $^{245}Cm \rightarrow {}^{241}Am \rightarrow {}^{237}Np \rightarrow {}^{233}U \rightarrow {}^{229}Th$

(ii) $^{246}Cm \rightarrow {}^{242}Pu \rightarrow {}^{238}U \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow {}^{210}Pb$

(iii) $^{239}Pu \rightarrow {}^{235}U \rightarrow {}^{231}Pa \rightarrow {}^{227}Ac$

(iv) $^{240}Pu \rightarrow {}^{236}U$

Of these daughters, it is expected that for ^{233}U, ^{229}Th, ^{231}Pa, ^{227}Ac, ^{230}Th, ^{226}Ra, and ^{210}Pb for matrix flow, and that for ^{210}Pb and ^{227}Ac for fracture flow, parent decay in the accessible environment could be the dominant source for radiation dose. To correct the method for this effect, for these daughter products, secular equilibrium of the daughter with its parent in the environment is assumed for calculating groundwater radionuclide concentrations in the accessible environment and, hence, importance indices. This assumption may overestimate the importance of the daughter products, but maintains the conservative nature of the calculation. For these daughters, the importance index is calculated by:

$$F_{i_{\text{daughter}}} = \frac{DCF_{\text{ing}_{\text{daughter}}}}{DCF_{\text{ing}_{\text{parent}}}} F_{i_{\text{parent}}} \tag{2-7}$$

The time at which these daughters will deliver this maximum dose is given by

$$t_{\text{del}_{\text{daughter}}} = t_{\text{del}_{\text{parent}}} + t_{1/2_{\text{daughter}}} \tag{2-8}$$

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3 DETERMINATION OF THE KEY RADIONUCLIDES FOR THE CASE OF MATRIX DOMINATED FLOW

3.1 ASSUMPTIONS

The F_i values for several radionuclides were calculated assuming a GWTT for Yucca Mountain of 1,000 yr. This assumption is reasonable for two reasons; (i) the Nuclear Regulatory Commission (NRC) rule on subsystem performance (10 CFR Part 60.113) states that the GWTT for a proposed repository will be at least 1,000 yr, and (ii) the best estimates for the GWTT for Yucca Mountain for present conditions are around 1,000 yr or greater[1].

In order for the method of calculating the importance index of a radionuclide to be accurate, the pathway described by Eq. (2-5) must be the dominant pathway for providing radionuclides to the accessible environment. If it is not the dominant pathway, as it is not for the daughters listed in Section 2.3, then Eq. (2-7) is used to calculate the importance index. This alternative means of calculating F_i must be used if the parent of a given daughter is: (i) released at a relatively fast rate, (ii) not highly retarded, and (iii) long-lived. For the general case, the above three criteria are not usually met, thus Eq. (2-5) is reasonable.

Data for the radionuclide inventory, transport, and dose parameters (I_{max}, R_f, DR, DCF_{ing}) were taken from a number of sources. Generally, the most conservative values were used in the calculation of F_i. Only when there was sufficient evidence (e.g., more recent data) for using a nonconservative value for a particular constant was a nonconservative value used. In any case, only the data for the dissolution rates differed greatly from reference to reference, and for this parameter, conservative numbers were used. Parameters used in this report are listed in the Appendix.

3.2 CALCULATION OF THE IMPORTANCE INDEX

The importance indices of the considered radionuclides for matrix flow are displayed in Table 3-1. The authors recommend a 0.2 mrem/yr cutoff for the radionuclides that should be considered important for PAs at Yucca Mountain. This cutoff dose rate is recommended because of the conservative nature of the calculation and the fact that 0.2 mrem/yr is a relatively low dose rate (when compared to the 40 CFR Part 191 standard previously stated). However, should it be necessary to consider lower doses, the method would work equally well with a lower cutoff rate.

It is noted that the importance indices for the radionuclides delimited by an asterisk (*) in Table 3-1 are exaggerated because secular equilibrium with their more mobile parents in the environment is assumed. This assumption is made to maintain the conservativeness of the calculation. Also, values for retardation factors are very uncertain as well as nonconstant throughout a given media, and this uncertainty has a very significant effect on F_i. To rectify this effect, the excessively low cutoff of 0.2 mrem/yr is used to determine which nuclides are considered important.

[1]Personal conversation with G. Wittmeyer, Center for Nuclear Waste Regulatory Analyses, on February 14, 1995.

Table 3-1. A listing of the importance index (F_i) for the case of matrix flow

Radionuclide	Importance Index (F_i) in mrem/yr	Time of Deliverance of F_i (yr)
^{237}Np	7,220	1.7×10^4
^{229}Th*	6,479	N/A
^{233}U*	500	N/A
^{129}I	156	1.0×10^3
^{210}Pb*	124	2.5×10^5
^{135}Cs	42	1.5×10^5
^{14}C	42	1.0×10^3
^{226}Ra*	27	N/A
^{99}Tc	26	5.0×10^3
^{230}Th*	13	N/A
^{242}Pu	9	5.0×10^4
^{234}U	6	2.6×10^4
^{227}Ac*	5	N/A
^{79}Se	5	2.5×10^4
^{231}Pa*	4	N/A
^{236}U	1	1.3×10^5
^{238}U	0.89	2.5×10^4
^{59}Ni	0.16	5.0×10^3
^{235}U	0.10	1.3×10^5
^{239}Pu	0.03	N/A
^{245}Cm	-	N/A
^{240}Pu	-	N/A
^{246}Cm	-	N/A
^{243}Am	-	N/A
^{137}Cs	-	N/A
^{241}Am	-	N/A
^{238}Pu	-	N/A

4 DETERMINATION OF THE KEY RADIONUCLIDES FOR THE CASE OF FRACTURE DOMINATED FLOW

For the case in which groundwater flow is assumed to be predominantly in the rock fractures, the same methodology can be used to calculate the importance index. However, we very conservatively assume no sorption of radionuclides on the rock surfaces in the fractures. For this case, three points should be noted: (i) the radionuclide retardation factor used for the importance index calculation is set equal to one, (ii) the radionuclide inventory used for the importance index calculation is no longer the maximum inventory, but is the inventory at 1,000 yr before the time of interest after repository closure, and (iii) since all of the released radionuclides will be arriving at the accessible environment at the same time, it is possible to interpret the importance index in a relative manner. The importance indices, as calculated for this case, are shown in Tables 4-1, 4-2, and 4-3. (Again, asterisks indicate those nuclides with exaggerated indices due to the parent equilibrium assumption.) Since the relative importance of the considered radionuclides will vary with time, the importance indices are calculated at times of 2,000, 11,000, and 101,000 yr after repository closure. The results of these calculations can be shown most clearly through the use of pie charts. These pie charts, generated from Tables 4-1, 4-2, and 4-3, are shown in Figures 4-1, 4-2, and 4-3.

The authors recommend a 99 percent cutoff for the radionuclides that should be considered important for PAs at Yucca Mountain. This percentage is used as a cutoff because little error would be introduced by neglecting the radionuclides that comprise the remaining 1 percent of the radiation dose. However, should it be deemed that this cutoff is too low, the method would work equally well with a higher cutoff point.

Interestingly, if one could estimate the fraction of a radionuclide that is being transported in colloidal form, then the methods described in this section could work equally well for estimating the relative importance of the radionuclides considering the effects of colloidal transport. Since, to the knowledge of the authors, these estimates do not exist at this time, no attempt was made to specifically consider colloidal transport.

Table 4-1. A listing of the importance index (F_i) for the case of fracture flow at 2,000 yr after repository closure

Radionuclide	F_i in mrem/yr
^{241}Am	9,219
^{237}Np	7,255
^{240}Pu	7,197
^{239}Pu	4,665
^{245}Cm	933
^{243}Am	572
^{246}Cm	181
^{129}I	156 - 99% cutoff point
^{135}Cs	43
^{14}C	42
^{99}Tc	27
^{242}Pu	23
^{210}Pb*	19
^{230}Th	14
^{227}Ac*	13 - 99.9% cutoff point
^{231}Pa	10
^{234}U	7
^{79}Se	6
^{226}Ra	4
^{238}U	0.9
^{236}U	0.8
^{229}Th	0.4
^{59}Ni	0.16
^{235}U	0.05
^{233}U	0.01
^{238}Pu	0.005
^{137}Cs	-
	TOTAL = 30,388

Table 4-2. A listing of the importance index (F_l) for the case of fracture flow at 11,000 yr after repository closure

Radionuclide	F_l in mrem/yr
^{237}Np	7,255
^{239}Pu	4,665
^{240}Pu	2,056
^{210}Pb*	615
^{245}Cm	444
^{243}Am	381
^{227}Ac*	179
^{129}I	156
^{231}Pa	140
^{230}Th	137
^{226}Ra	133
^{229}Th	83 - 99% cutoff point
^{135}Cs	43
^{99}Tc	27
^{242}Pu	23
^{246}Cm	18
^{14}C	14 - 99.9% cutoff point
^{234}U	7
^{79}Se	6
^{236}U	1
^{238}U	0.9
^{241}Am	0.7
^{233}U	0.18
^{59}Ni	0.16
^{235}U	0.05
^{137}Cs	-
^{238}Pu	-
	TOTAL = 16,362

Table 4-3. A listing of the importance index (F_i) for the case of fracture flow at 101,000 yr after repository closure

Radionuclide	F_i in mrem/yr
^{237}Np	7,255
^{210}Pb*	6,154
^{229}Th	2,074
^{226}Ra	1,327
^{227}Ac*	765
^{230}Th	684
^{231}Pa	601
^{239}Pu	311
^{129}I	156
^{135}Cs	43 - 99% cutoff point
^{99}Tc	27
^{242}Pu	23 - 99.9% cutoff point
^{234}U	7
^{233}U	2
^{79}Se	2
^{236}U	1
^{238}U	0.9
^{245}Cm	0.16
^{240}Pu	0.15
^{235}U	0.10
^{59}Ni	0.08
^{243}Am	0.004
^{14}C	-
^{238}Pu	-
^{137}Cs	-
^{246}Cm	-
^{241}Am	-
	TOTAL = 19,433

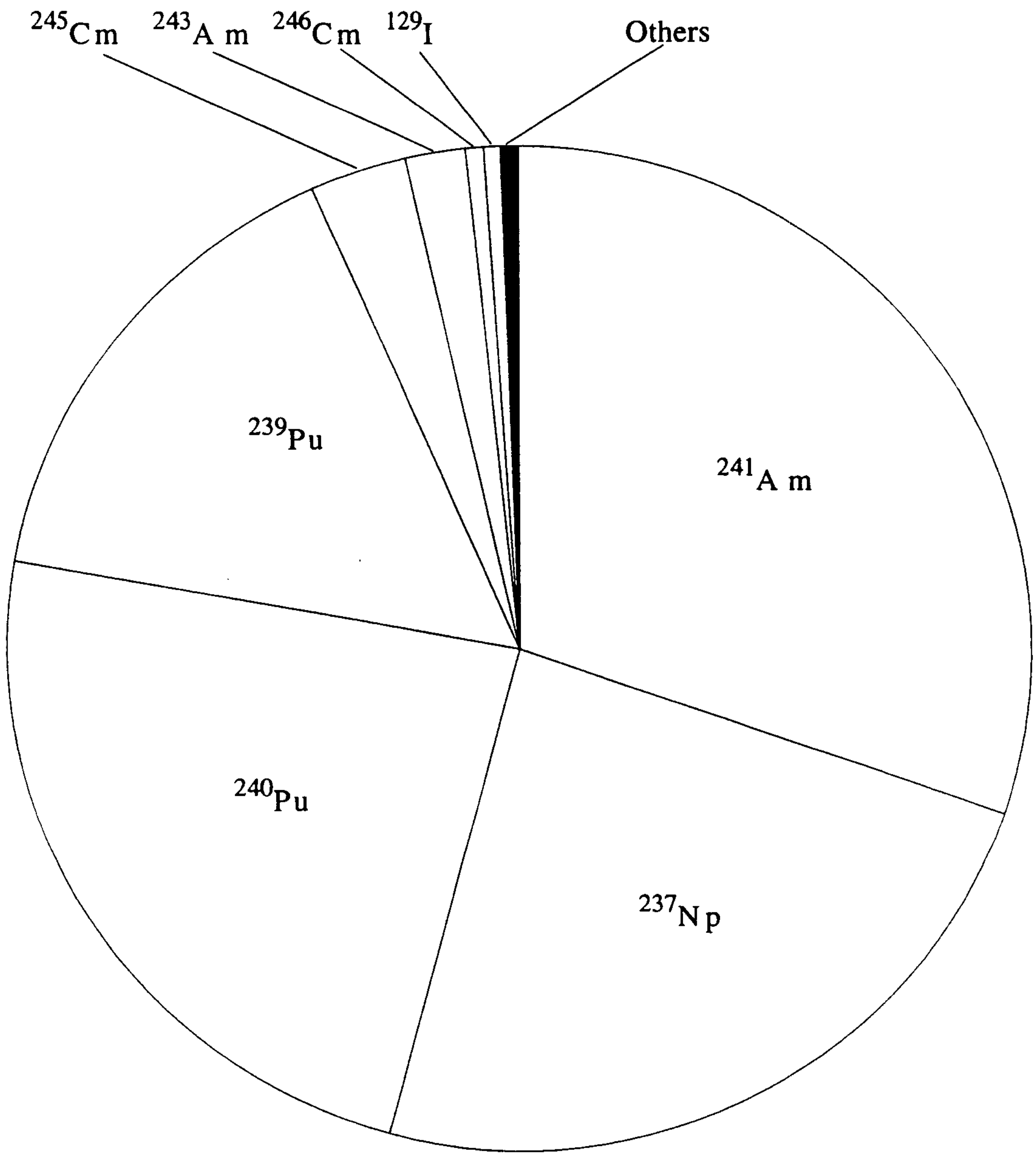

Figure 4-1. Relative contributions of radionuclides to total importance index for the fracture flow case at 2,000 yr after repository closure

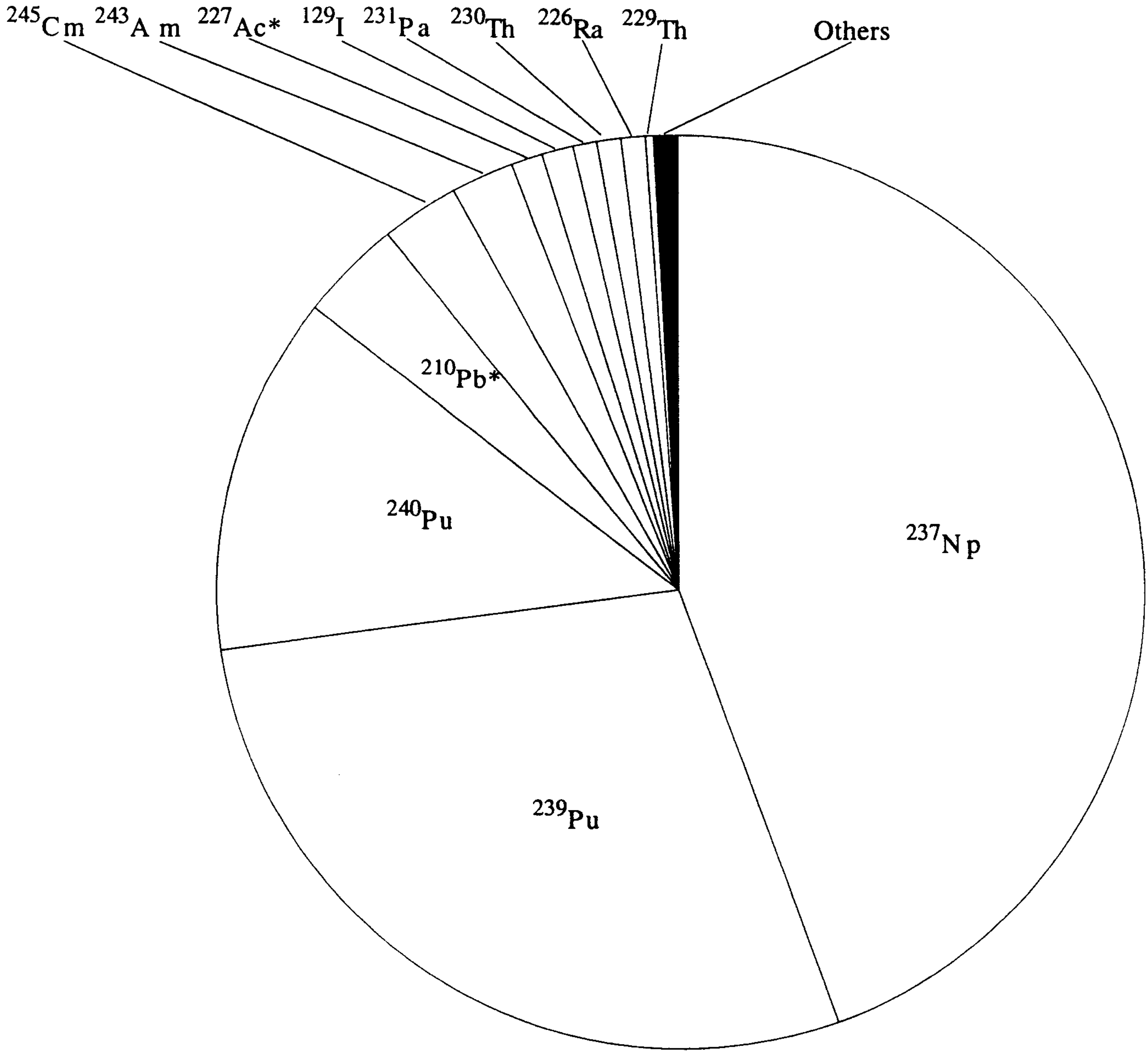

Figure 4-2. Relative contributions of radionuclides to total importance index for the fracture flow case at 11,000 yr after repository closure

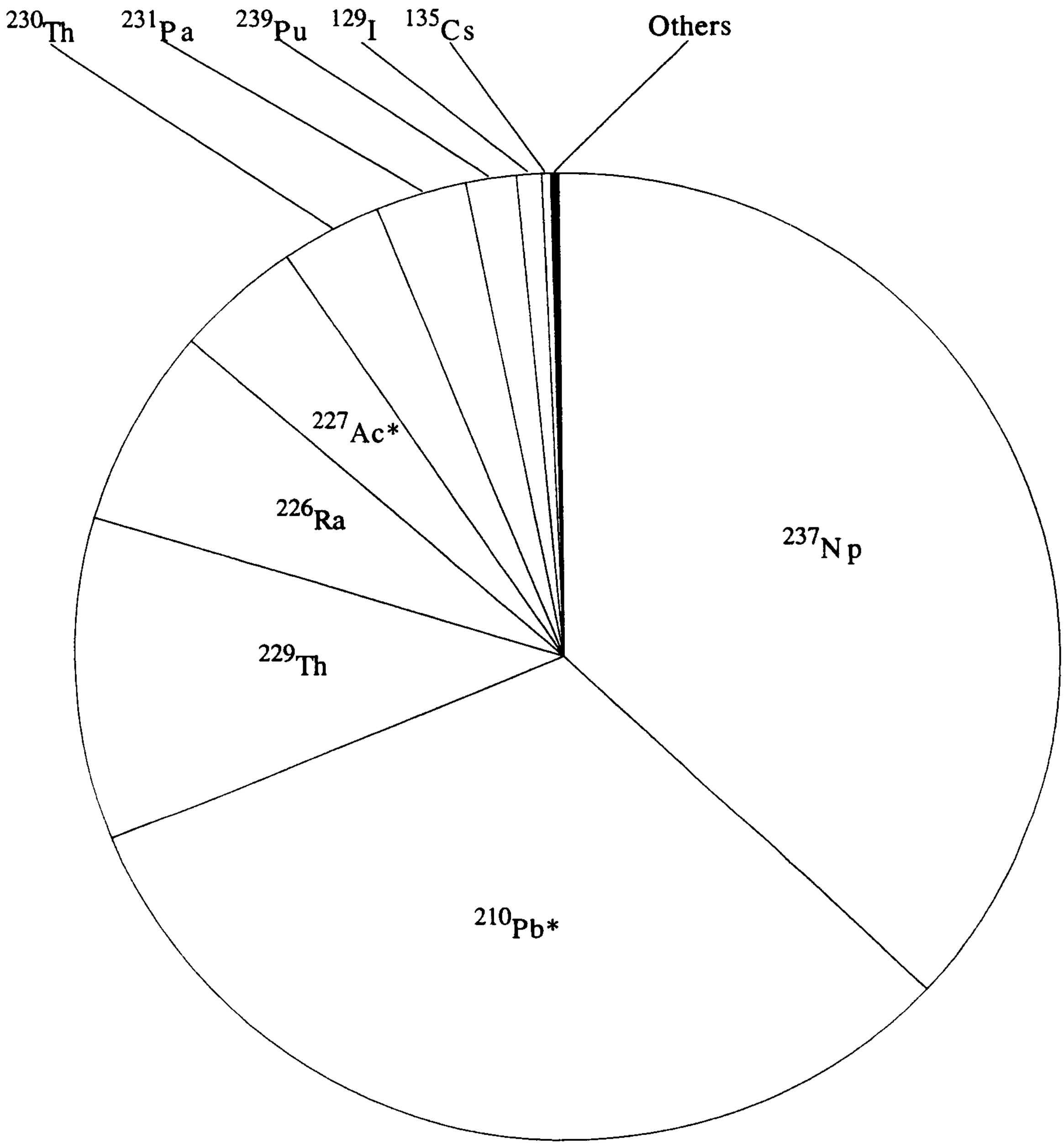

Figure 4-3. Relative contributions of radionuclides to total importance index for the fracture flow case at 101,000 yr after repository closure

5 CONCLUSIONS

5.1 DETERMINATION OF THE IMPORTANT RADIONUCLIDES

In conclusion, any radionuclide that is deemed important in either Table 3-1 or Figures 4-1, 4-2, or 4-3 should be considered important for PAs of a potential repository located at Yucca Mountain. Thus, we conclude that the following radionuclides are important: ^{246}Cm, ^{245}Cm, ^{243}Am, ^{241}Am, ^{242}Pu, ^{240}Pu, ^{239}Pu, ^{237}Np, ^{238}U, ^{236}U, ^{234}U, ^{233}U, ^{231}Pa, ^{230}Th, ^{229}Th, ^{227}Ac, ^{226}Ra, ^{210}Pb, ^{135}Cs, ^{129}I, ^{99}Tc, ^{79}Se, and ^{14}C. This list uses a cutoff of 0.2 mrem/yr in Table 3-1 and a cutoff of 99 percent in Figures 4-1, 4-2, and 4-3.

The authors emphasize that many of the parameters used here have, of course, large uncertainties. Due to these uncertainties and the simplified treatment adopted here, the importance index should not be considered a definitive estimate of environmental dose effects. The utility of this exercise lies chiefly in determining which radionuclides should be included in Total-System Performance Assessments (TSPAs).

This report is consistent with the conclusions of two earlier products of the Center for Nuclear Waste Regulatory Analyses (CNWRA) (1994a,b), which discussed important radionuclides. Whereas the present letter is general in its treatment, those reports were focused on confirming the importance of uranium isotopes in HLW research. The importance index in effect combines radionuclide characteristics that were treated less quantitatively (e.g., solubility, sorption) or less directly (e.g., dose conversion) in the earlier letters. It is notable that, in the present study, uranium isotopes remain on the list of important radionuclides.

5.2 SIGNIFICANT DIFFERENCES WITH NUCLEAR REGULATORY COMMISSION ITERATIVE PERFORMANCE ASSESSMENT PHASE 2

In comparing the above list of important radionuclides with the list of radionuclides considered in NRC IPA Phase 2, there are several differences:

(i) ^{137}Cs and ^{59}Ni were not found to be important. In no case was it shown that these radionuclides were significant contributors of radiation dose to humans.

(ii) ^{236}U was found to be important. This radionuclide is capable of contributing enough dose to deem it important for the case of groundwater flow predominantly through the rock matrix.

(iii) ^{233}U, ^{229}Th, ^{231}Pa, and ^{227}Ac were found to be important. These radionuclides are daughters of a more mobile parent (Np-237 for ^{233}U and ^{229}Th, and ^{235}U for ^{231}Pa and ^{227}Ac) and thus have the potential to deliver a significant dose through parent decays in the accessible environment. Retaining these four radionuclides is analogous to retaining ^{226}Ra and ^{210}Pb for PA calculations.

(iv) ^{242}Pu was found to be important. Due to the long half-life of this radionuclide, it was shown that it could be a significant contributor of radiation dose to humans.

(v) ^{94}Nb was not found to be important. This radionuclide was shown to be insignificant by other PAs and reports; therefore, it was not considered in this study.

Table 5-1 shows the results of the above comparison in tabular form and lists those radionuclides considered in aqueous release calculations in earlier U.S. Department of Energy (DOE) TSPAs.

The differences between the conclusions of this study and the nuclides considered in IPA Phase 2 are due mainly to the comprehensive, quantitative, and conservative nature of the radionuclides considered in this study. In essence, a "mini" performance assessment was performed here to determine how the nuclide list used in IPA Phase 2 should be updated for IPA Phase 3. Due to the conservative nature of these calculations, it is possible that radionuclides shown here to be important may, upon more refined analysis, be shown to be insignificant dose contributors.

Table 5-1. A comparison of the results of this study with other Total-System Performance Assessments. An "X" indicates that radionuclide was considered in transport calculations.

Radionuclide	This Study	IPA Phase 2	SNL TSPA-1993	PNL TSPA
^{246}Cm	X	X		
^{245}Cm	X	X		
^{243}Am	X	X		X
^{241}Am	X	X		
^{242}Pu	X			
^{240}Pu	X	X		
^{239}Pu	X	X	X	X
^{238}Pu				
^{237}Np	X	X	X	X
^{238}U	X	X		
^{236}U	X			
^{235}U				
^{234}U	X	X	X	X
^{233}U	X			
^{231}Pa	X		X	
^{230}Th	X	X		
^{229}Th	X			
^{227}Ac	X			
^{226}Ra	X	X		
^{210}Pb	X	X		
^{137}Cs		X		
^{135}Cs	X	X		X
^{129}I	X	X	X	X
^{126}Sn				X
^{99}Tc	X	X	X	X
^{94}Nb		X		
^{79}Se	X	X	X	X
^{59}Ni		X		
^{14}C	X	X	X	X

6 RECOMMENDATIONS FOR FURTHER RESEARCH

Due to the limited scope of this project, there are several recommendations that the authors wish to make for further investigations. First, the values of the importance index are sensitive to the data that are used as input to calculate them. Therefore, a sensitivity analysis of the method with respect to the input data would be informative.

Second, assuming no retardation of radionuclides in the fracture flow case is, in the opinion of these authors, overly conservative. A more realistic approximation could be to consider that the fractures have a sorption coefficient, and hence retardation factor, that is reduced from that of the surrounding rock matrix by a factor that is the ratio of the fracture surface area to the surface area of the pore volume in the rock matrix for a given volume of rock. Of course, this approach does not consider possible mineralogical differences, nor effects such as competition of radionuclides for available absorber sites, multilayer adsorption layers, or sorption kinetics that would become more important for the reduced number of adsorber sites available in the case of fracture flow.

Third, current CNWRA efforts are underway that are using site specific information about Yucca Mountain to determine doses on a per picocurie/liter radionuclide concentration in the groundwater that are based on a larger suite of groundwater related pathways. These constants, when determined, could be used with the method developed in this paper to more accurately determine values of F_i. Essentially, the constants would substitute for DCF_{ing}*L in Eq. (2-5).

Finally, this ranking should be considered an ongoing exercise, to be revisited as new data, regulatory standards, models, and design concepts are introduced.

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APPENDIX

DATA USED FOR CALCULATING THE IMPORTANCE INDEX

Table A-1: Radionuclide inventory data used to calculate the importance indices (Lozano et al., 1994)

Radionuclide	I_{max} (pCi)	I [10^3 yr] (pCi)	I [10^4 yr] (pCi)	I [10^5 yr] (pCi)
^{14}C	8.6×10^{16}	8.6×10^{16}	2.8×10^{16}	7.0×10^{11}
^{59}Ni	1.6×10^{17}	1.6×10^{17}	1.6×10^{17}	7.0×10^{16}
^{79}Se	2.8×10^{16}	2.8×10^{16}	2.8×10^{16}	7.0×10^{15}
^{99}Tc	7.7×10^{17}	7.7×10^{17}	7.7×10^{17}	7.7×10^{17}
^{129}I	2.1×10^{15}	2.1×10^{15}	2.1×10^{15}	2.1×10^{15}
^{135}Cs	2.3×10^{16}	2.3×10^{16}	2.3×10^{16}	2.3×10^{16}
^{137}Cs	5.6×10^{20}	7.0×10^{11}	0.0	0.0
^{210}Pb	7.0×10^{16}	2.1×10^{14}	7.0×10^{15}	7.0×10^{16}
^{226}Ra	7.0×10^{16}	2.0×10^{16}	7.0×10^{15}	7.0×10^{16}
^{227}Ac	2.1×10^{15}	2.8×10^{13}	3.5×10^{14}	2.1×10^{15}
^{229}Th	7.0×10^{16}	7.0×10^{12}	1.4×10^{15}	3.5×10^{16}
^{230}Th	7.0×10^{16}	1.4×10^{15}	1.4×10^{16}	7.0×10^{16}
^{231}Pa	2.1×10^{15}	3.5×10^{13}	4.9×10^{14}	2.1×10^{15}
^{237}Np	7.0×10^{16}	7.0×10^{16}	7.0×10^{16}	7.0×10^{16}
^{233}U	7.0×10^{16}	2.1×10^{14}	3.5×10^{15}	3.5×10^{16}
^{234}U	1.4×10^{17}	1.4×10^{17}	1.4×10^{17}	1.4×10^{17}
^{235}U	2.1×10^{15}	1.0×10^{15}	1.0×10^{15}	2.1×10^{15}
^{236}U	2.8×10^{16}	1.6×10^{16}	2.4×10^{16}	2.8×10^{16}
^{238}U	2.1×10^{16}	2.1×10^{16}	2.1×10^{16}	2.1×10^{16}
^{238}Pu	1.4×10^{20}	7.0×10^{16}	0.0	0.0
^{239}Pu	2.1×10^{19}	2.1×10^{19}	2.1×10^{19}	1.4×10^{18}
^{240}Pu	3.5×10^{19}	3.5×10^{19}	1.0×10^{19}	7.0×10^{14}
^{242}Pu	1.1×10^{17}	1.1×10^{17}	1.1×10^{17}	1.1×10^{17}
^{241}Am	2.8×10^{20}	7.0×10^{19}	5.6×10^{15}	0.0
^{243}Am	1.1×10^{18}	1.1×10^{18}	7.0×10^{17}	7.0×10^{12}
^{245}Cm	8.4×10^{15}	8.4×10^{15}	4.0×10^{15}	1.4×10^{12}
^{246}Cm	1.8×10^{15}	1.8×10^{15}	1.8×10^{14}	0.0

Table A-2: Radionuclide data used to calculate the importance indices (most conservative values used) (Duguid et al., 1994; Kerrisk, 1985; U.S. Department of Energy, 1988; Wilson et al., 1993)

Radionuclide	DR (yr^{-1})	$t_{1/2}$ (yr)	R_f	DCF_{ing}	t_{max} (yr)
^{14}C	1.0×10^{-3}	5,730	1	2.1×10^{-6}	0.0
^{59}Ni	2.0×10^{-5}	8.0×10^{4}	5	2.0×10^{-7}	0.0
^{79}Se	1.0×10^{-4}	6.5×10^{4}	25	8.3×10^{-6}	0.0
^{99}Tc	1.0×10^{-4}	2.1×10^{5}	5	1.3×10^{-6}	0.0
^{129}I	1.0×10^{-3}	1.7×10^{7}	1	2.8×10^{-4}	0.0
^{135}Cs	1.0×10^{-3}	3.0×10^{6}	150	7.1×10^{-6}	0.0
^{137}Cs	1.0×10^{-3}	30	150	5.0×10^{-5}	0.0
^{210}Pb	1.0×10^{-4}	22	50	5.1×10^{-3}	2.0×10^{5}
^{226}Ra	1.0×10^{-4}	1,600	500	1.1×10^{-3}	2.0×10^{5}
^{227}Ac	1.0×10^{-4}	22	1,500	1.4×10^{-2}	1.0×10^{5}
^{229}Th	7.0×10^{-5}	7,340	500	3.5×10^{-3}	5.0×10^{5}
^{230}Th	7.0×10^{-5}	7.7×10^{4}	500	5.3×10^{-4}	2.0×10^{5}
^{231}Pa	1.0×10^{-4}	3.3×10^{4}	1,500	1.1×10^{-2}	3.0×10^{5}
^{237}Np	1.0×10^{-4}	2.1×10^{6}	16	3.9×10^{-3}	2,000
^{233}U	7.0×10^{-7}	1.6×10^{5}	25	2.7×10^{-4}	5.0×10^{5}
^{234}U	7.0×10^{-7}	2.4×10^{5}	25	2.6×10^{-4}	500
^{235}U	7.0×10^{-7}	7.0×10^{8}	25	2.5×10^{-4}	1.0×10^{5}
^{236}U	7.0×10^{-7}	2.3×10^{7}	25	2.7×10^{-4}	1.0×10^{5}
^{238}U	7.0×10^{-7}	4.5×10^{9}	25	2.3×10^{-4}	0.0
^{238}Pu	2.0×10^{-7}	88	500	3.8×10^{-3}	0.0
^{239}Pu	2.0×10^{-7}	2.4×10^{4}	500	4.3×10^{-3}	0.0
^{240}Pu	2.0×10^{-7}	6,540	500	4.3×10^{-3}	0.0
^{242}Pu	2.0×10^{-7}	3.8×10^{5}	500	4.1×10^{-3}	0.0
^{241}Am	5.0×10^{-7}	458	1,000	4.5×10^{-3}	100
^{243}Am	5.0×10^{-7}	7,380	1,000	4.5×10^{-3}	0.0
^{245}Cm	1.0×10^{-4}	9,300	500	4.5×10^{-3}	0.0
^{246}Cm	1.0×10^{-4}	4,730	500	4.5×10^{-3}	0.0

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