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Geochemical Behavior of Long-Lived Radioactive Wastes

Ferruccio Gera

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GEOCHEMICAL BEHAVIOR OF LONG-LIVED RADIOACTIVE WASTES

Ferruccio Gera

JULY 1975

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION



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GEOCHEMICAL BEHAVIOR OF LONG-LIVED RADIOACTIVE WASTES

Ferruccio Gera

ABSTRACT

The hazard potential associated with the heavy elements present in high-level radioactive waste decreases greatly in the first few tens of thousands of years of decay; however, further reduction in the hazard potential becomes extremely slow after about 100,000 years. In the time period between 100,000 and 5 million years the hazard potential of high-level waste is reduced by a factor of between 10 and 20. Current evidence seems to indicate that if radioactive waste containment were to fail after a period of 100,000 years or more, some environmental contamination would result; however, the contamination levels would be low. The radiological risk would not be significantly different from that now existing in various localities as a result of the accumulation of natural radioactive elements.

With the partial exception of radium, which is concentrated in the fruit of specific perennial plants, the long-lived alpha-emitters are characterized by very low biologic availability in terrestrial ecosystems. The biologic availability may be somewhat higher in aquatic ecosystems due to the significant reconcentration factors in particular organisms. Despite the fact that many of these elements are frequently discussed as a group (e.g., the actinides or the transuranics), significant differences of environmental behavior exist among them. Inhalation hazards are controlled primarily by the efficiency of resuspension mechanisms. Soil erosion by wind is more effective for particles between 50 and 500 μm . On the other hand, the pulmonary deposition of particles larger than 10 μm is practically negligible.

Natural high-background areas and the piles of uranium mill tailings are instances of unusual accumulations of long-lived alpha-emitters in surface materials. Data concerning the levels of activity in foods grown on radioactive soils seem to confirm the low biologic availability of the natural radioactive elements. Surveys of uranium mill tailings indicate little dispersal of the radioactive elements into the environment; even though untreated tailings piles would appear to be particularly vulnerable to resuspension of dust particles by wind.

1. INTRODUCTION

The duration of required containment for radioactive wastes is a function of the level of activity considered to be the threshold below which dispersal into the environment can be tolerated. For the long-lived alpha-emitters contained in wastes generated by fuel reprocessing and fabrication, it is not presently possible to specify the required containment time to meet this criterion. However, it is clear that these wastes will have to be contained for time periods well in excess of the recorded history of mankind. Obviously, no manmade structure can be guaranteed to last for such long periods of time. In fact, "storage" of waste in engineered facilities is necessarily a temporary measure and although useful in overall waste management strategies should not be confused with "disposal" of radioactive wastes.

Deep geologic formations with appropriate properties can provide reliable containment for geologic time periods; however, there is obviously a progressive increase of the probability of containment failure with time even for the most favorable geologic environments. In general, it seems possible to estimate the maximum rate of geologic processes capable of breaching the waste containment, and to design a disposal facility in such a manner that it will withstand at least a few hundred thousand years of geologic change. It is known, however, that the longest-lived nuclides in the waste will present some radiological hazard for millions of years, and it is not possible to absolutely guarantee that waste will be contained for such long periods of time.

Erosion of the overburden and interaction with ground water are two possible mechanisms that, given enough time, have the potential for causing dispersal of activity into the environment. This does not mean that environmental contamination will necessarily occur, but only that the possibility of containment failure at some future time cannot be excluded; hence, it is interesting to evaluate the radiological consequences of such an event.

This report attempts to put the long-term hazard potential of high-level wastes into proper perspective by following the changes in the isotopic composition of the waste up to 10 million years after generation. It also includes an extensive review of the available information on the

geological and environmental behavior of the long-lived nuclides. The sequence of possible events that could lead to environmental dispersion of the activity contained in radioactive waste buried deep in the earth has been discussed in a previous report.¹ The conclusion was reached that the most likely mechanism leading to transfer of activity to the biosphere is failure of isolation from ground water. It can be stated generally that the assessment of the hazard resulting from such a failure of geologic containment requires a knowledge of the mechanisms and rates of transfer of the various nuclides from the solid to the liquid phase, their behavior in ground waters and, upon reaching the surface, their behavior in surface waters, food chains, and the atmosphere.

Undoubtedly, our knowledge of the various links and transfer mechanisms is less than perfect, but much can be deduced by the interpretation of available information. The geochemistry of natural radioactive elements provides an insight into the underground mobility of some nuclides. The areas where unusually high concentrations of long-lived alpha-emitters are present in surface materials, either naturally (high-background areas) or as a result of human action (uranium mill tailings), offer an opportunity for studying the environmental behavior of several nuclides of interest and the relationship between their concentration in various environmental areas and human exposure.

2. HEAVY NUCLIDES AND LONG-TERM HAZARD POTENTIAL

Staff members of the Oak Ridge National Laboratory have discussed the composition and distribution of long-lived alpha-emitters in high-level waste in other reports.^{1,2} More complete analyses of the variation of composition of high-level waste as a function of decay time have also been published.^{3,4}

The computer program ORIGEN has been used to calculate the activities of the radionuclides contained in high-level waste after various decay times.⁵ The activities of long-lived fission products present in thousand-year-old high-level solid waste are shown in Table 1. The critical long-lived fission product is ⁹⁹Tc; the amounts of ⁹⁹Tc shown in Table 1 correspond to 1.1 and 1.4 limits of annual intake by ingestion (LAI_{ing}), and 9.5 and 12.1 limits of annual intake by inhalation (LAI_{inh}) per cubic centimeter of waste.

Table 1. Activity of Long-Lived Fission Products in High-Level Solid Wastes from LWR and LMFBR Fuels

Nuclide	Half-life	Ci/m^3 After 10^3 Years of Decay	
		LWR	LMFBR
^{79}Se	6.5×10^4 y	3.94	6.43
$^{93}\text{Zr}_{\downarrow}$	1.5×10^6 y	18.90	17.60
$^{93\text{m}}\text{Nb}^b$	13.6 y	18.90	17.60
^{99}Tc	2.12×10^5 y	143.00	182.00
^{107}Pd	7×10^6 y	1.10	2.88
$^{126}\text{Sn}_{\downarrow}$	10^5 y	5.42	15.20
$^{126\text{m}}\text{Sb}_{\downarrow}^b$	19 m	5.42	15.20
$^{126}\text{Sb}^b$	12.5 d	5.37	15.10
$^{129}\text{I}^c$	1.7×10^7 y	0.000522	0.00104
^{135}Cs	3×10^6 y	2.86	14.00
^{151}Sm	87 y	4.36	21.30

^a1 m³ of high-level solid waste produced by reprocessing 10 metric tons (MT) of fuel.

^bShort-lived nuclide below the arrow produced by decay of parent above.

^c0.1% of ^{129}I originally in fuel assumed to remain in high-level solid waste.

The number of LAI contained in a unit volume of waste is used in this report as an indication of the relative hazard potential of the waste. The LAI is preferred to other possible values related to the radiotoxicity of the nuclides [e.g., radiation concentration guides (RCG), maximum permissible body burdens (MPBB)] because it is considered by the author to be the most satisfactory expression of equivalent risk^{1,6} since the intake of a "limit" of any nuclide results in equivalent 50-year dose commitments.

The hazard associated with the long-lived fission products, which is relatively minor, would become comparable to the hazard of the heavy radionuclides only after extremely long decay times (i.e., greater than 10 million years). Iodine is fairly volatile, and therefore most ¹²⁹I becomes separated from the waste stream during fuel reprocessing and waste solidification; only 0.1% of the original ¹²⁹I is assumed to remain in the high-level solid waste. Fission products will not be discussed further in this report.

Table 2 shows the relevant properties of the heavy nuclides present in high-level solid waste, but does not include nuclides with a half-life shorter than a month or those present in the waste with concentrations less than 10^{-21} Ci/m³.

Tables 3 and 4 show the radioactivity of heavy nuclides in high-level solid waste produced by the reprocessing of light water reactor (LWR) and liquid metal fast breeder reactor (LMFBR) fuels, respectively, after decay times of up to 10 million years.

In order to provide an indication of the relative hazard potential associated with the heavy nuclides, the numbers of LAI_{ing} and LAI_{inh} per cubic centimeter of waste for each nuclide have been calculated and are shown in Tables 5 through 8. The LAI values used to calculate the tables are given in ref. 7, with the exceptions of the limits for ²²⁹Th, ²³⁶Pu, and ²⁵⁰Cm that were not listed. These LAI have been estimated on the basis of the radiotoxicity of other isotopes of the same elements. The number of LAI contained in the waste is a satisfactory indication of the relative hazard potential if it is assumed that the waste could be ingested directly or inhaled and that the risk refers to only one generation of humans.

Table 2. Properties of Heavy Radionuclides with Half-Lives Longer Than 30 Days

Nuclide	Half-Life	Mode of Decay	Lowest Limits of Annual Intake for Members of the Public ^a (μCi) and Relative Critical Organ	
			Ingestion	Inhalation
²¹⁰ Pb	21 y	β ⁻	9.6 · 10 ⁻² (Sol. - Total body)	3.1 · 10 ⁻² (Sol. - Kidney)
²¹⁰ Po	138 d	α	5.8 · 10 ⁻¹ (Sol. - Spleen)	5.0 · 10 ⁻² (Insol. - Lung)
²²⁶ Ra	1,604 y	α	9.6 · 10 ⁻³ (Sol. - Bone)	7.1 · 10 ⁻³ (Sol. - Bone)
²²⁸ Ra	6.7 y	β ⁻	2.2 · 10 ⁻² (Sol. - Bone)	9.5 · 10 ⁻³ (Insol. - Lung)
²²⁷ Ac	21.5 y	β ⁻ , α(1 · 10 ⁻²)	1.5 · 10 ⁻¹ (Sol. - Bone)	5.8 · 10 ⁻⁴ (Sol. - Bone)
²²⁸ Th	1.91 y	α	5.8 (Sol. - Bone)	1.5 · 10 ⁻³ (Insol. - Lung)
²²⁹ Th ^b	7,300 y	α	1.4 ^b (Sol. - Bone)	5.6 · 10 ⁻⁴ ^b (Sol. - Bone)
²³⁰ Th	80,000 y	α, SF (< 5 · 10 ⁻¹⁶)	1.4 (Sol. - Bone)	5.6 · 10 ⁻⁴ (Sol. - Bone)
²³² Th	1.41 · 10 ¹⁰ y	α, SF (< 1.4 · 10 ⁻¹¹)	1.2 (Sol. - Bone)	4.8 · 10 ⁻⁴ (Sol. - Bone)
²³¹ Pa	32,500 y	α	7.0 · 10 ⁻¹ (Sol. - Bone)	2.8 · 10 ⁻⁴ (Sol. - Bone)
²³² U	72 y	α, SF (9 · 10 ⁻¹³)	6.7 · 10 ⁻¹ (Sol. - Bone)	6.9 · 10 ⁻³ (Insol. - Lung)
²³³ U	1.62 · 10 ⁵ y	α, SF (1.3 · 10 ⁻¹²)	3.4 (Sol. - Bone)	3.0 · 10 ⁻² (Insol. - Lung)
²³⁴ U	2.47 · 10 ⁵ y	α, SF (1.2 · 10 ⁻¹¹)	3.4 (Sol. - Bone)	3.0 · 10 ⁻² (Insol. - Lung)
²³⁵ U	7.1 · 10 ⁸ y	α, SF (2 · 10 ⁻⁹)	3.0 (Sol. - Kidney)	3.2 · 10 ⁻² (Insol. - Lung)
²³⁶ U	2.39 · 10 ⁷ y	α, SF (1 · 10 ⁻⁹)	3.6 (Sol. - Bone)	3.1 · 10 ⁻² (Insol. - Lung)
²³⁸ U	4.51 · 10 ⁹ y	α, SF (5 · 10 ⁻⁷)	4.7 · 10 ⁻¹ (Sol. - Kidney)	1.8 · 10 ⁻² (Sol. - Kidney)
²³⁷ Np	2.13 · 10 ⁶ y	α, SF (< 2 · 10 ⁻¹²)	2.5 (Sol. - Bone)	1.0 · 10 ⁻³ (Sol. - Bone)
²³⁶ Pu ^b	2.85 y	α, SF (8.0 · 10 ⁻¹⁰)	4.0 ^b (Sol. - Bone)	4.8 · 10 ⁻⁴ ^b (Sol. - Bone)
²³⁸ Pu	89 y	α, SF (1.8 · 10 ⁻⁹)	4.0 (Sol. - Bone)	4.8 · 10 ⁻⁴ (Sol. - Bone)
²³⁹ Pu	24,400 y	α, SF (4.4 · 10 ⁻¹²)	3.6 (Sol. - Bone)	4.3 · 10 ⁻⁴ (Sol. - Bone)
²⁴⁰ Pu	6,760 y	α, SF (5 · 10 ⁻⁸)	3.6 (Sol. - Bone)	4.3 · 10 ⁻⁴ (Sol. - Bone)
²⁴¹ Pu	14.6 y	β ⁻ , α(2.3 · 10 ⁻⁵)	1.8 · 10 ² (Sol. - Bone)	2.3 · 10 ⁻² (Sol. - Bone)
²⁴² Pu	3.79 · 10 ⁵ y	α, SF (5.0 · 10 ⁻⁶)	3.8 (Sol. - Bone)	4.5 · 10 ⁻⁴ (Sol. - Bone)
²⁴⁴ Pu	8.29 · 10 ⁷ y	α, SF (1.2 · 10 ⁻³)	3.4 (Sol. - Bone)	4.1 · 10 ⁻⁴ (Sol. - Bone)
²⁴¹ Am	433 y	α, SF (1.5 · 10 ⁻¹²)	3.0 (Sol. - Kidney)	1.5 · 10 ⁻³ (Sol. - Bone & Kidney)
^{242m} Am	151 y	IT, α(4.3 · 10 ⁻³), SF (1.6 · 10 ⁻¹⁰)	3.5 (Sol. - Bone)	1.4 · 10 ⁻³ (Sol. - Bone)
²⁴³ Am	7,660 y	α, SF (2.3 · 10 ⁻¹⁰)	3.5 (Sol. - Bone)	1.4 · 10 ⁻³ (Sol. - Bone)
²⁴² Cm	163 d	α, SF (6.0 · 10 ⁻⁸)	1.9 · 10 (Sol. - GI)	3.0 · 10 ⁻² (Sol. - Liver)
²⁴³ Cm	32 y	α, EC (3.0 · 10 ⁻³)	4.1 (Sol. - Bone)	1.6 · 10 ⁻³ (Sol. - Bone)
²⁴⁴ Cm	18.2 y	α, SF (1.3 · 10 ⁻⁶)	5.7 (Sol. - Bone)	2.3 · 10 ⁻³ (Sol. - Bone)
²⁴⁵ Cm	8,260 y	α	2.8 (Sol. - Bone)	1.2 · 10 ⁻³ (Sol. - Bone)
²⁴⁶ Cm	4,715 y	α, SF (3.0 · 10 ⁻⁴)	2.9 (Sol. - Bone)	1.2 · 10 ⁻³ (Sol. - Bone)
²⁴⁷ Cm	1.64 · 10 ⁷ y	α	2.9 (Sol. - Bone)	1.2 · 10 ⁻³ (Sol. - Bone)
²⁴⁸ Cm	3.52 · 10 ⁵ y	α, SF (1 · 10 ⁻¹)	3.5 · 10 ⁻¹ (Sol. - Bone)	1.5 · 10 ⁻⁴ (Sol. - Bone)
²⁵⁰ Cm ^b	17,440 y	SF	3.5 · 10 ⁻² ^b (Sol. - Bone)	1.5 · 10 ⁻⁵ ^b (Sol. - Bone)
²⁴⁹ Bk	314 d	β ⁻ , α(1.4 · 10 ⁻⁵), SF (5 · 10 ⁻¹⁰)	4.7 · 10 ² (Sol. - GI)	2.3 · 10 ⁻¹ (Sol. - Bone)
²⁴⁹ Cf	352 y	α, SF (6.0 · 10 ⁻⁹)	3.3 (Sol. - Bone)	3.9 · 10 ⁻⁴ (Sol. - Bone)
²⁵⁰ Cf	13.1 y	α, SF (8.0 · 10 ⁻⁴)	1.0 · 10 (Sol. - Bone)	1.2 · 10 ⁻³ (Sol. - Bone)
²⁵¹ Cf	900 y	α	3.4 (Sol. - Bone)	4.2 · 10 ⁻⁴ (Sol. - Bone)
²⁵² Cf	2.65 y	α, SF (3.2 · 10 ⁻²)	5.8 (Sol. - Bone)	1.6 · 10 ⁻³ (Sol. - Bone)

^aTable IIA from "Basic Safety Standards for Radiation Protection," Safety Series No. 9, International Atomic Energy Agency, Vienna (1967).⁷

^bNo limit of annual intake (LAI) is listed for these nuclides; the values used have been estimated on the basis of the LAI's of other nuclides of the same element.

SF = Spontaneous fission; IT = Isomeric transition; EC = Orbital electron capture; GI = Gastro-intestinal tract.

Table 3. Radioactivity of Heavy Nuclides in LWR High-Level Solid Waste^a as a Function of Decay Time

Nuclide	(Ci/m ³) After Decay Times (years) of:									
	10	10 ²	10 ³	10 ⁴	5 · 10 ⁴	10 ⁵	5 · 10 ⁵	10 ⁶	5 · 10 ⁶	10 ⁷
²¹⁰ Pb	1.95-7 ^b	7.52-6	6.94-4	2.82-2	1.46-1	2.23-1	1.63-1	5.39-2	1.57-2	1.57-2
²¹⁰ Po	1.95-7	7.52-6	6.94-4	2.82-2	1.46-1	2.23-1	1.63-1	5.39-2	1.57-2	1.57-2
²²⁶ Ra	1.10-6	1.11-5	6.94-4	2.82-2	1.46-1	2.23-1	1.63-1	5.39-2	1.57-2	1.57-2
²²⁸ Ra	2.21-10	3.89-10	1.08-9	1.20-8	8.51-8	1.82-7	9.49-7	1.90-6	9.00-6	1.68-5
²²⁷ Ac	7.71-5	2.38-4	2.60-4	3.91-4	1.33-3	2.42-3	3.37-3	3.37-3	3.35-3	3.34-3
²²⁸ Th	1.41-3	4.01-4	7.03-8	1.20-8	8.51-8	1.82-7	9.49-7	1.90-6	9.00-6	1.68-5
²²⁹ Th	4.22-7	7.30-6	6.95-4	5.56-2	5.79-1	1.18	3.00	2.91	7.46-1	1.48-1
²³⁰ Th	2.11-4	3.36-4	3.72-3	3.62-2	1.45-1	2.21-1	1.63-1	5.38-2	1.57-2	1.57-2
²³² Th	3.25-10	3.89-10	1.08-9	1.20-8	8.51-8	1.82-7	9.49-7	1.90-6	9.00-6	1.68-5
²³¹ Pa	2.47-4	2.48-4	2.60-4	3.91-4	1.33-3	2.41-3	3.37-3	3.37-3	3.35-3	3.34-3
²³² U	8.72-4	3.91-4	6.74-8	0	0	0	0	0	0	0
²³³ U	1.48-4	1.47-3	1.53-2	1.57-1	7.19-1	1.29	2.99	2.89	7.46-1	1.48-1
²³⁴ U	6.48-2	2.38-1	4.41-1	4.32-1	3.88-1	3.39-1	1.21-1	4.16-2	1.57-2	1.57-2
²³⁵ U	8.56-4	8.57-4	8.73-4	1.17-3	2.56-3	3.17-3	3.37-3	3.37-3	3.35-3	3.34-3
²³⁶ U	1.44-2	1.46-2	1.68-2	3.04-2	3.92-2	3.93-2	3.88-2	3.82-2	3.41-2	2.95-2
²³⁸ U	1.57-2	1.57-2	1.57-2	1.57-2	1.57-2	1.57-2	1.57-2	1.57-2	1.57-2	1.57-2
²³⁷ Np	3.41	3.45	3.68	3.75	3.71	3.65	3.21	2.73	7.46-1	1.48-1
²³⁶ Pu	1.39-3	4.31-13	0	0	0	0	0	0	0	0
²³⁸ Pu	9.29+2	4.90	1.23	2.83-18	0	0	0	0	0	0
²³⁹ Pu	1.62+1	1.67+1	2.06+1	4.11+1	2.31+1	5.78	6.96-5	2.42-6	2.04-6	1.65-6
²⁴⁰ Pu	4.43+1	8.61+1	7.98+1	3.17+1	5.24-1	3.11-3	1.93-8	2.64-8	2.97-8	2.85-8
²⁴¹ Pu	3.20+3	4.80+1	3.15	1.48	5.16-2	7.79-4	2.10-18	0	0	0
²⁴² Pu	6.93-2	7.15-2	7.65-2	8.07-2	7.68-2	7.01-2	3.37-2	1.35-2	8.98-6	9.59-10
²⁴⁴ Pu	6.07-13	6.07-12	6.07-11	6.01-10	2.89-9	5.51-9	1.93-8	2.64-8	2.97-8	2.85-8
²⁴¹ Am	1.63+3	1.50+3	3.59+2	1.48	5.16-2	7.79-4	2.21-18	0	0	0
^{242m} Am	8.74+1	5.80+1	9.56-1	1.43-18	0	0	0	0	0	0
²⁴³ Am	8.82+2	1.80+2	1.66+2	7.34+1	1.96	2.11-2	2.47-6	2.42-6	2.04-6	1.65-6
²⁴² Cm	7.17+1	4.75+1	7.84-1	1.17-18	0	0	0	0	0	0
²⁴³ Cm	2.96+1	4.21	1.44	0	0	0	0	0	0	0
²⁴⁴ Cm	1.64+4	5.22+2	6.38-13	7.82-13	3.76-12	7.16-12	2.51-11	3.43-11	3.86-11	3.70-11
²⁴⁵ Cm	3.41	3.39	3.14	1.48	5.15-2	7.78-4	2.09-18	0	0	0
²⁴⁶ Cm	6.83-1	6.74-1	5.90-1	1.57-1	4.36-4	2.78-7	2.81-21	0	0	0
²⁴⁷ Cm	2.52-6	2.52-6	2.52-6	2.52-6	2.52-6	2.51-6	2.47-6	2.42-6	2.04-6	1.65-6
²⁴⁸ Cm	7.91-6	7.91-6	7.90-6	7.76-6	7.17-6	6.50-6	2.96-6	1.10-6	4.19-10	2.22-14
²⁵⁰ Cm	9.23-13	9.19-13	8.87-13	6.20-13	1.26-13	1.72-14	2.06-21	0	0	0
²⁴⁹ Bk	7.93-6	0	0	0	0	0	0	0	0	0
²⁴⁹ Cf	9.70-5	8.12-5	1.38-5	2.77-13	0	0	0	0	0	0
²⁵⁰ Cf	2.17-4	1.84-6	8.87-13	6.20-13	1.26-13	1.72-14	2.06-21	0	0	0
²⁵¹ Cf	2.72-6	2.54-6	1.27-6	1.24-9	0	0	0	0	0	0
²⁵² Cf	3.27-5	1.88-15	0	0	0	0	0	0	0	0

^a 1 m³ of solid waste produced by reprocessing 10 MT of fuel. Specific power = 30 MW/MT; burnup = 33,000 MWD/MT; flux = 2.92 · 10¹³ n/cm² s. Fuel reprocessing 160 days after discharge; 0.5% of U and Pu, and 100% of other heavy elements in waste.

^b Read as 1.95 x 10⁻⁷.

Table 4. Radioactivity of Heavy Nuclides in LMFBR High-Level Solid Waste^a as a Function of Decay Time

Nuclide	(Ci/m ³) After Decay Times (years) of:									
	10	10 ²	10 ³	10 ⁴	5 · 10 ⁴	10 ⁵	5 · 10 ⁵	10 ⁶	5 · 10 ⁶	10 ⁷
210 _{Pb}	1.01-8 ^b	3.22-6	2.52-3	1.21-1	6.27-1	9.58-1	6.69-1	1.84-1	1.44-2	1.44-2
210 _{Po}	1.01-8	3.22-6	2.52-3	1.21-1	6.27-1	9.58-1	6.69-1	1.84-1	1.44-2	1.44-2
226 _{Ra}	6.71-8	6.30-6	2.52-3	1.21-1	6.27-1	9.58-1	6.69-1	1.84-1	1.44-2	1.44-2
228 _{Ra}	4.78-12	1.09-11	2.33-10	1.59-8	1.70-7	3.80-7	2.05-6	4.12-6	1.95-5	3.66-5
227 _{Ac}	8.04-7	2.70-6	7.36-6	2.36-4	3.86-3	8.47-3	1.26-2	1.26-2	1.25-2	1.25-2
228 _{Th}	1.22-2	3.29-3	5.68-7	1.59-8	1.70-7	3.80-7	2.05-6	4.12-6	1.96-5	3.66-5
229 _{Th}	1.53-6	4.91-6	6.37-4	8.68-2	9.48-1	1.94	4.95	4.80	1.23	2.44-1
230 _{Th}	1.77-5	3.95-4	1.46-2	1.55-1	6.25-1	9.50-1	6.69-1	1.84-1	1.44-2	1.44-2
232 _{Th}	7.06-12	1.09-11	2.33-10	1.59-8	1.70-7	3.80-7	2.05-6	4.12-6	1.96-5	3.66-5
231 _{Pa}	2.62-6	2.89-6	7.36-6	2.36-4	3.86-3	8.46-3	1.26-2	1.26-2	1.25-2	1.25-2
232 _U	7.16-3	3.20-3	5.53-7	0	0	0	0	0	0	0
233 _U	6.42-5	7.72-4	1.64-2	2.50-1	1.18	2.12	4.93	4.77	1.23	2.44-1
234 _U	1.08-1	8.01-1	1.90	1.86	1.67	1.45	4.82-1	1.29-1	1.44-2	1.44-2
235 _U	1.34-4	1.51-4	3.25-4	2.29-3	8.97-3	1.17-2	1.26-2	1.26-2	1.25-2	1.25-2
236 _U	4.84-4	1.25-3	8.70-3	5.51-2	8.52-2	8.55-2	8.46-2	8.33-2	7.42-2	6.42-2
238 _U	1.43-2	1.43-2	1.43-2	1.43-2	1.43-2	1.43-2	1.44-2	1.44-2	1.44-2	1.44-2
237 _{Np}	1.52	2.15	5.25	6.20	6.12	6.02	5.29	4.50	1.23	2.44-1
236 _{Pu}	1.13-2	3.52-12	0	0	0	0	0	0	0	0
238 _{Pu}	3.52+3	2.10+3	1.16+1	3.50-17	0	0	0	0	0	0
239 _{Pu}	1.93+2	1.94+2	2.02+2	2.32+2	1.04+2	2.57+1	2.98-4	1.58-7	1.33-7	1.08-7
240 _{Pu}	2.75+2	2.98+2	2.72+2	1.08+2	1.79	1.06-2	1.53-8	1.53-8	1.49-8	1.43-8
241 _{Pu}	1.84+4	2.57+2	8.18-1	3.85-1	1.34-2	2.03-4	5.45-19	0	0	0
242 _{Pu}	7.21-1	7.47-1	7.96-1	7.85-1	7.29-1	6.66-1	3.20-1	1.28-1	8.53-5	9.10-9
244 _{Pu}	1.50-8	1.50-8	1.50-8	1.50-8	1.51-8	1.51-8	1.53-8	1.53-8	1.49-8	1.43-8
241 _{Am}	2.26+4	2.01+4	4.76+3	3.87-1	1.34-2	2.03-4	5.75-19	0	0	0
242 _{mAm}	1.08+3	7.16+2	1.18+1	1.76-17	0	0	0	0	0	0
243 _{Am}	5.41+2	5.36+2	4.94+2	2.19+2	5.83	6.28-2	1.61-7	1.58-7	1.33-7	1.08-7
242 _{Cm}	8.85+2	5.87+2	9.68	1.45-17	0	0	0	0	0	0
243 _{Cm}	3.65+2	5.19+1	1.77-7	0	0	0	0	0	0	0
244 _{Cm}	1.01+4	3.21+2	1.99-11	1.96-11	1.96-11	1.96-11	1.98-11	1.99-11	1.93-11	1.85-11
245 _{Cm}	8.88-1	8.81-1	8.17-1	3.84-1	1.34-2	2.02-4	5.44-19	0	0	0
246 _{Cm}	4.01-2	3.95-2	3.46-2	9.21-3	2.56-5	1.63-8	5.75-22	0	0	0
247 _{Cm}	1.65-7	1.65-7	1.65-7	1.64-7	1.64-7	1.64-7	1.61-7	1.58-7	1.33-7	1.08-7
248 _{Cm}	1.19-7	1.19-7	1.19-7	1.17-7	1.08-7	9.79-8	4.45-8	1.66-8	6.31-12	3.34-16
250 _{Cm}	1.89-13	1.89-13	1.82-13	1.27-13	2.58-14	3.52-15	4.23-22	0	0	0
249 _{Bk}	1.18-7	0	0	0	0	0	0	0	0	0
249 _{Cf}	1.30-6	1.09-6	1.85-7	3.71-15	0	0	0	0	0	0
250 _{Cf}	2.85-7	2.42-9	1.82-13	1.27-13	2.58-14	3.52-15	4.23-22	0	0	0
251 _{Cf}	7.90-11	7.37-11	3.68-11	3.60-14	0	0	0	0	0	0
252 _{Cf}	1.80-11	1.04-21	0	0	0	0	0	0	0	0

^a 1 m³ of solid waste produced by reprocessing 10 MT of fuel. Specific power = 49 MW/MT; burnup = 37,000 MWD/MT; flux = 2.49 · 10¹⁵ n/cm² s. Fuel is a mixture of core and blankets. Fuel reprocessing 90 days after discharge; 0.5% of U and Pu, and 100% of other heavy elements in waste.

^b Read as 1.01 × 10⁻⁸.

Table 5. Number of Limits of Annual Intake by Ingestion of Heavy Nuclides in 1 cm³ of LWR High-Level Solid Waste as a Function of Decay Time

Nuclide	LAI _{ing} /cm ³ After Decay Times (years) of:									
	10	10 ²	10 ³	10 ⁴	5 · 10 ⁴	10 ⁵	5 · 10 ⁵	10 ⁶	5 · 10 ⁶	10 ⁷
²¹⁰ Pb	2.03-6 ^a	7.83-5	7.23-3	2.94-1	1.52	2.32	1.70	5.61-1	1.63-1	1.63-1
²¹⁰ Po	3.36-7	1.30-5	1.20-3	4.86-2	2.52-1	3.84-1	2.81-1	9.29-2	2.71-2	2.71-2
²²⁶ Ra	1.14-4	1.16-3	7.23-2	2.94	<u>1.52+1</u>	<u>2.32+1</u>	<u>1.70+1</u>	<u>5.61</u>	<u>1.63</u>	<u>1.63</u>
²²⁸ Ra	1.00-8	1.77-8	4.91-8	5.45-7	3.87-6	8.27-6	4.31-5	8.64-5	4.09-4	7.64-4
²²⁷ Ac	5.14-4	1.50-3	1.73-3	2.61-3	8.87-3	1.61-2	2.25-2	2.25-2	2.23-2	2.23-2
²²⁸ Th	2.43-4	6.91-5	1.21-8	2.07-9	1.47-8	3.14-8	1.64-7	3.28-7	1.55-6	2.90-6
²²⁹ Th	3.01-7	5.21-6	4.96-4	3.97-2	4.13-1	8.43-1	2.14	2.08	5.33-1	1.06-1
²³⁰ Th	1.51-4	2.40-4	2.66-3	2.59-2	1.04-1	1.58-1	1.16-1	3.84-2	1.12-2	1.12-2
²³² Th	2.71-10	3.24-10	9.00-10	1.00-8	7.09-8	1.52-7	7.91-7	1.58-6	7.50-6	1.40-5
²³¹ Pa	3.53-4	3.54-4	3.71-4	5.59-4	1.90-3	3.44-3	4.81-3	4.81-3	4.79-3	4.77-3
²³² U	1.30-3	5.83-4	1.01-7	0	0	0	0	0	0	0
²³³ U	4.35-5	4.32-4	4.50-3	4.62-2	2.11-1	3.79-1	8.79-1	8.50-1	2.19-1	4.35-2
²³⁴ U	1.90-2	7.00-2	1.30-1	1.27-1	1.14-1	9.97-2	3.56-2	1.22-2	4.62-3	4.62-3
²³⁵ U	2.85-4	2.86-4	2.91-4	3.90-4	8.53-4	1.06-3	1.12-3	1.12-3	1.12-3	1.11-3
²³⁶ U	4.00-3	4.05-3	4.67-3	8.44-3	1.09-2	1.09-2	1.08-2	1.06-2	9.47-3	8.19-3
²³⁸ U	3.34-2	3.34-2	3.34-2	3.34-2	3.34-2	3.34-2	3.34-2	3.34-2	3.34-2	3.34-2
²³⁷ Np	1.36	1.38	1.47	1.50	1.48	1.46	1.28	1.09	2.98-1	5.92-2
²³⁶ Pu	3.47-4	1.08-13	0	0	0	0	0	0	0	0
²³⁸ Pu	2.32+2	1.22+2	3.07-1	7.07-19	0	0	0	0	0	0
²³⁹ Pu	4.50	4.64	5.72	1.14+1	6.42	1.60	1.93-5	6.72-7	5.67-7	4.58-7
²⁴⁰ Pu	1.23+1	2.39+1	2.22+1	8.80	1.45-1	8.64-4	5.36-9	7.33-9	8.25-9	7.92-9
²⁴¹ Pu	1.78+1	2.67-1	1.75-2	8.22-3	2.87-4	4.33-6	1.17-20	0	0	0
²⁴² Pu	1.82-2	1.88-2	2.01-2	2.12-2	2.02-2	1.84-2	8.87-3	3.55-3	2.36-6	2.52-10
²⁴⁴ Pu	1.78-13	1.78-12	1.78-11	1.77-10	8.50-10	1.62-9	5.68-9	7.76-9	8.73-9	8.38-9
²⁴¹ Am	5.43+2	<u>5.00+2</u>	<u>1.20+2</u>	4.93-1	1.72-2	2.60-4	7.37-19	0	0	0
^{242m} Am	2.50+1	1.66+1	2.73-1	4.09-19	0	0	0	0	0	0
²⁴³ Am	5.20+1	5.14+1	4.74+1	<u>2.10+1</u>	5.60-1	6.03-3	7.06-7	6.91-7	5.83-7	4.71-7
²⁴² Cm	3.77	2.50	4.13-2	6.16-20	0	0	0	0	0	0
²⁴³ Cm	7.22	1.03	3.51-9	0	0	0	0	0	0	0
²⁴⁴ Cm	<u>2.88+3</u>	9.16+1	1.12-13	1.37-13	6.60-13	1.26-12	4.40-12	6.02-12	6.77-12	6.49-12
²⁴⁵ Cm	1.22	1.21	1.12	5.29-1	1.84-2	2.78-4	7.46-19	0	0	0
²⁴⁶ Cm	2.35-1	2.32-1	2.03-1	5.41-2	1.50-4	9.59-8	9.69-22	0	0	0
²⁴⁷ Cm	8.69-7	8.69-7	8.69-7	8.69-7	8.69-7	8.65-7	8.52-7	8.34-7	7.03-7	5.69-7
²⁴⁸ Cm	2.26-5	2.26-5	2.26-5	2.22-5	2.05-5	1.86-5	8.46-6	3.14-6	1.20-9	6.34-14
²⁵⁰ Cm	2.64-11	2.63-11	2.53-11	1.77-11	3.60-12	4.91-13	5.89-20	0	0	0
²⁴⁹ Bk	1.69-8	0	0	0	0	0	0	0	0	0
²⁴⁹ Cf	2.94-5	2.46-5	4.18-6	8.39-14	0	0	0	0	0	0
²⁵⁰ Cf	2.17-5	1.84-7	8.87-14	6.20-14	1.26-14	1.72-15	2.06-22	0	0	0
²⁵¹ Cf	8.00-7	7.47-7	3.73-7	3.65-10	0	0	0	0	0	0
²⁵² Cf	5.64-6	3.24-16	0	0	0	0	0	0	0	0
TOTAL	~3,800	~820	~200	~48	~27	~31	~24	~11	~3	~2

Each column has an underlined value, indicating the critical heavy nuclide at that particular time.

^aRead as 2.03 x 10⁻⁶.

Table 6. Number of Limits of Annual Intake by Inhalation of Heavy Nuclides in 1 cm³ of LWR High-Level Solid Waste as a Function of Decay Time

Nuclide	LAI _{inh} /cm ³ After Decay Times (years) of:									
	<u>10</u>	<u>10²</u>	<u>10³</u>	<u>10⁴</u>	<u>5 · 10⁴</u>	<u>10⁵</u>	<u>5 · 10⁵</u>	<u>10⁶</u>	<u>5 · 10⁶</u>	<u>10⁷</u>
²¹⁰ Pb	6.29-6 ^a	2.43-4	2.24-2	9.10-1	4.71	7.19	5.26	1.74	5.06-1	5.06-1
²¹⁰ Po	3.90-6	1.50-4	1.39-2	5.64-1	2.92	4.46	3.26	1.08	3.14-1	3.14-1
²²⁶ Ra	1.55-4	1.56-3	9.77-2	3.97	2.06+1	3.14+1	2.30+1	7.59	2.21	2.21
²²⁸ Ra	2.33-8	4.09-8	1.14-7	1.26-6	8.96-6	1.92-5	9.99-5	2.00-4	9.47-4	1.77-3
²²⁷ Ac	1.33-1	4.10-1	4.48-1	6.74-1	2.29	4.17	5.81	5.81	5.78	5.76
²²⁸ Th	9.40-1	2.67-1	4.69-5	8.00-6	5.67-5	1.21-4	6.33-4	1.27-3	6.00-3	1.12-2
²²⁹ Th	7.54-4	1.30-2	1.24	9.93+1	1.03+3	2.11+3	<u>5.36+3</u>	<u>5.20+3</u>	<u>1.33+3</u>	<u>2.64+2</u>
²³⁰ Th	3.77-1	6.00-1	6.64	6.46+1	2.59+2	3.95+2	2.91+2	9.61+1	2.80+1	2.80+1
²³² Th	6.77-7	8.10-7	2.25-6	2.50-5	1.77-4	3.79-4	1.98-3	3.96-3	1.87-2	3.50-2
²³¹ Pa	8.82-1	8.86-1	9.28-1	1.40	4.75	8.61	1.20+1	1.20+1	1.20+1	1.19+1
²³² U	1.26-1	5.67-2	9.77-6	0	0	0	0	0	0	0
²³³ U	4.93-3	4.90-2	5.10-1	5.23	2.40+1	4.30+1	9.97+1	9.63+1	2.49+1	4.93
²³⁴ U	2.16	7.93	1.47+1	1.44+1	1.29+1	1.13+1	4.03	1.39	5.23-1	5.23-1
²³⁵ U	2.67-2	2.68-2	2.73-2	3.66-2	8.00-2	9.91-2	1.05-1	1.05-1	1.05-1	1.04-1
²³⁶ U	4.64-1	4.71-1	5.42-1	9.81-1	1.26	1.27	1.25	1.23	1.10	9.52-1
²³⁸ U	8.72-1	8.72-1	8.72-1	8.72-1	8.72-1	8.72-1	8.72-1	8.72-1	8.72-1	8.72-1
²³⁷ Np	3.41+3	3.45+3	3.68+3	3.75+3	3.71+3	3.65+3	3.21+3	2.73+3	7.46+2	1.48+2
²³⁶ Pu	2.90	8.98-10	0	0	0	0	0	0	0	0
²³⁸ Pu	1.93+6	<u>1.02+6</u>	2.56+3	5.90-15	0	0	0	0	0	0
²³⁹ Pu	3.77+4	3.88+4	4.79+4	<u>9.56+4</u>	<u>5.37+4</u>	<u>1.34+4</u>	1.62-1	5.63-3	4.74-3	3.84-3
²⁴⁰ Pu	1.03+5	2.00+5	1.86+5	7.37+4	1.22+3	7.23	4.49-5	6.14-5	6.91-5	6.63-5
²⁴¹ Pu	1.39+5	2.09+3	1.37+2	6.43+1	2.24	3.39-2	9.13-17	0	0	0
²⁴² Pu	1.54+2	1.59+2	1.70+2	1.79+2	1.71+2	1.56+2	7.49+1	3.00+1	2.00-2	2.13-6
²⁴⁴ Pu	1.48-9	1.48-8	1.48-7	1.47-6	7.05-6	1.34-5	4.71-5	6.44-5	7.24-5	6.95-5
²⁴¹ Am	1.09+6	1.00+6	<u>2.39+5</u>	9.87+2	3.44+1	5.19-1	1.47-15	0	0	0
^{242m} Am	6.24+4	4.14+4	6.83+2	1.02-15	0	0	0	0	0	0
²⁴³ Am	1.30+5	1.29+5	1.19+5	5.24+4	1.40+3	1.51+1	1.76-3	1.73-3	1.46-3	1.18-3
²⁴² Cm	2.39+3	1.58+3	2.61+1	3.90-17	0	0	0	0	0	0
²⁴³ Cm	1.85+4	2.63+3	9.00-6	0	0	0	0	0	0	0
²⁴⁴ Cm	<u>7.13+6</u>	2.27+5	2.77-10	3.40-10	1.63-9	3.11-9	1.09-8	1.49-8	1.68-8	1.61-8
²⁴⁵ Cm	2.84+3	2.82+3	2.62+3	1.23+3	4.29+1	6.48-1	1.74-15	0	0	0
²⁴⁶ Cm	5.69+2	5.62+2	4.92+2	1.31+2	3.63-1	2.32-4	2.34-18	0	0	0
²⁴⁷ Cm	2.10-3	2.10-3	2.10-3	2.10-3	2.10-3	2.09-3	2.06-3	2.02-3	1.70-3	1.37-3
²⁴⁸ Cm	5.27-2	5.27-2	5.27-2	5.17-2	4.78-2	4.33-2	1.97-2	7.33-3	2.79-6	1.48-10
²⁵⁰ Cm	6.15-8	6.13-8	5.91-8	4.13-8	8.40-9	1.15-9	1.37-16	0	0	0
²⁴⁹ Bk	3.45-5	0	0	0	0	0	0	0	0	0
²⁴⁹ Cf	2.49-1	2.08-1	3.54-2	7.10-10	0	0	0	0	0	0
²⁵⁰ Cf	1.81-1	1.53-3	7.39-10	5.17-10	1.05-10	1.43-11	1.72-18	0	0	0
²⁵¹ Cf	6.48-3	6.05-3	3.02-3	2.95-6	0	0	0	0	0	0
²⁵² Cf	2.04-2	1.17-12	0	0	0	0	0	0	0	0
Total	~10,700,000	~2,700,000	~600,000	~230,000	~60,000	~20,000	~9,100	~8,200	~2,200	~500

Each column has an underlined value, indicating the critical heavy nuclide at that particular time.

^aRead as 6.29 X 10⁻⁶.

Table 7. Number of Limits of Annual Intake by Ingestion of Heavy Nuclides in 1 cm³ of LMFBR High-Level Solid Waste as a Function of Decay Time

Nuclide	LAI _{ing} /cm ³ After Decay Times (years) of:									
	10	10 ²	10 ³	10 ⁴	5 · 10 ⁴	10 ⁵	5 · 10 ⁵	10 ⁶	5 · 10 ⁶	10 ⁷
²¹⁰ Pb	1.05-7 ^a	3.35-5	2.62-2	1.26	6.53	9.98	6.97	1.92	1.50-1	1.50-1
²¹⁰ Po	1.74-8	5.55-6	4.34-3	2.09-1	1.08	1.65	1.15	3.17-1	2.48-2	2.48-2
²²⁶ Ra	6.99-6	6.56-4	2.62-1	1.26+1	<u>6.53+1</u>	<u>9.98+1</u>	<u>6.97+1</u>	<u>1.92+1</u>	<u>1.50</u>	<u>1.50</u>
²²⁸ Ra	2.17-10	4.95-10	1.06-8	7.23-7	7.73-6	1.73-5	9.32-5	1.87-4	8.86-4	1.66-3
²²⁷ Ac	5.36-6	1.80-5	4.91-5	1.57-3	2.57-2	5.65-2	8.40-2	8.40-2	8.33-2	8.33-2
²²⁸ Th	2.10-3	5.67-4	9.79-8	2.74-9	2.93-8	6.55-8	3.53-7	7.10-7	3.38-6	6.31-6
²²⁹ Th	1.09-6	3.51-6	4.55-4	6.20-2	6.77-1	1.39	3.54	3.43	8.79-1	1.74-1
²³⁰ Th	1.26-5	2.82-4	1.04-2	1.11-1	4.46-1	6.78-1	4.78-1	1.31-1	1.03-2	1.03-2
²³² Th	5.88-12	9.08-12	1.94-10	1.32-8	1.42-7	3.17-7	1.71-6	3.43-6	1.63-5	3.05-5
²³¹ Pa	3.74-6	4.13-6	1.05-5	3.37-4	5.51-3	1.21-2	1.80-2	1.80-2	1.79-2	1.79-2
²³² U	1.07-2	4.78-3	8.25-7	0	0	0	0	0	0	0
²³³ U	1.89-5	2.27-4	4.82-3	7.35-2	3.47-1	6.23-1	1.45	1.40	3.62-1	7.18-2
²³⁴ U	3.18-2	2.36-1	5.59-1	5.47-1	4.91-1	4.26-1	1.42-1	3.79-2	4.23-3	4.23-3
²³⁵ U	4.47-5	5.03-5	1.08-4	7.63-4	2.99-3	3.90-3	4.20-3	4.20-3	4.17-3	4.17-3
²³⁶ U	1.34-4	3.47-4	2.42-3	1.53-2	2.37-2	2.37-2	2.35-2	2.31-2	2.06-2	1.78-2
²³⁸ U	3.04-2	3.04-2	3.04-2	3.04-2	3.04-2	3.04-2	3.06-2	3.06-2	3.06-2	3.06-2
²³⁷ Np	6.08-1	8.60-1	2.10	2.48	2.45	2.41	2.12	1.80	4.92-1	9.76-2
²³⁶ Pu	2.82-3	8.80-13	0	0	0	0	0	0	0	0
²³⁸ Pu	8.80+2	5.25+2	2.90	8.75-18	0	0	0	0	0	0
²³⁹ Pu	5.36+1	5.39+1	5.61+1	<u>6.44+1</u>	2.89+1	7.14	8.28-5	4.39-8	3.69-8	3.00-8
²⁴⁰ Pu	7.64+1	8.28+1	7.55+1	3.00+1	4.97-1	2.94-3	4.25-9	4.25-9	4.14-9	3.97-9
²⁴¹ Pu	1.02+2	1.43	4.54-3	2.14-3	7.44-5	1.13-6	3.03-21	0	0	0
²⁴² Pu	1.90-1	1.97-1	2.09-1	2.07-1	1.92-1	1.75-1	8.42-2	3.37-2	2.24-5	2.39-9
²⁴⁴ Pu	4.41-9	4.41-9	4.41-9	4.41-9	4.44-9	4.44-9	4.50-9	4.50-9	4.38-9	4.21-9
²⁴¹ Am	<u>7.53+3</u>	<u>6.70+3</u>	<u>1.59+3</u>	1.29-1	4.47-3	6.77-5	1.92-19	0	0	0
^{242m} Am	3.09+2	2.05+2	3.37	5.03-18	0	0	0	0	0	0
²⁴³ Am	1.55+2	1.53+2	1.41+2	6.26+1	1.67	1.79-2	4.60-8	4.51-8	3.80-8	3.09-8
²⁴² Cm	4.66+1	3.09+1	5.09-1	7.63-19	0	0	0	0	0	0
²⁴³ Cm	8.90+1	1.27+1	4.32-8	0	0	0	0	0	0	0
²⁴⁴ Cm	1.77+3	5.63+1	3.49-12	3.44-12	3.44-12	3.44-12	3.47-12	3.49-12	3.39-12	3.25-12
²⁴⁵ Cm	3.17-1	3.15-1	2.92-1	1.37-1	4.79-3	7.21-5	1.94-19	0	0	0
²⁴⁶ Cm	1.38-2	1.36-2	1.19-2	3.18-3	8.83-6	5.62-9	1.98-22	0	0	0
²⁴⁷ Cm	5.69-8	5.69-8	5.69-8	5.65-8	5.65-8	5.65-8	5.55-8	5.45-8	4.59-8	3.72-8
²⁴⁸ Cm	3.40-7	3.40-7	3.40-7	3.34-7	3.09-7	2.80-7	1.27-7	4.74-8	1.80-11	9.54-16
²⁵⁰ Cm	5.40-12	5.40-12	5.20-12	3.63-12	7.37-13	1.01-13	1.21-20	0	0	0
²⁴⁹ Bk	2.51-10	0	0	0	0	0	0	0	0	0
²⁴⁹ Cf	3.94-7	3.30-7	5.61-8	1.12-15	0	0	0	0	0	0
²⁵⁰ Cf	2.85-8	2.42-10	1.82-14	1.27-14	2.58-15	3.52-16	4.23-23	0	0	0
²⁵¹ Cf	2.32-11	2.17-11	1.08-11	1.06-14	0	0	0	0	0	0
²⁵² Cf	3.10-12	1.79-22	0	0	0	0	0	0	0	0
Total	~11,000	~7,800	~1,900	~175	~110	~125	~86	~30	~4	~2

Each column has an underlined value, indicating the critical heavy nuclide at that particular time.

^aRead as 1.05 x 10⁻⁷.

Table 8. Number of Limits of Annual Intake by Inhalation of Heavy Nuclides in 1 cm^3 of LMFBR High-Level Solid Waste as a Function of Decay Time

Nuclide	LAI _{inh} /cm ³ After Decay Times (years) of:									
	<u>10</u>	<u>10²</u>	<u>10³</u>	<u>10⁴</u>	<u>5 · 10⁴</u>	<u>10⁵</u>	<u>5 · 10⁵</u>	<u>10⁶</u>	<u>5 · 10⁶</u>	<u>10⁷</u>
²¹⁰ Pb	3.26-7 ^a	1.04-4	8.13-2	3.90	2.02+1	3.09+1	2.16+1	5.93	4.64-1	4.64-1
²¹⁰ Po	2.02-7	6.44-5	5.04-2	2.42	1.25+1	1.92+1	1.34+1	3.68	2.88-1	2.88-1
²²⁶ Ra	9.45-6	8.87-4	3.55-1	1.70+1	8.83+1	1.35+2	9.42+1	2.59+1	2.03	2.03
²²⁸ Ra	5.03-10	1.15-9	2.45-8	1.67-6	1.79-5	4.00-5	2.16-4	4.34-4	2.05-3	3.85-3
²²⁷ Ac	1.39-3	4.65-3	1.27-2	4.07-1	6.65	1.46+1	2.17+1	2.17+1	2.15+1	2.15+1
²²⁸ Th	8.13	2.19	3.79-4	1.06-5	1.13-4	2.53-4	1.37-3	2.75-3	1.31-2	2.44-2
²²⁹ Th	2.73-3	8.77-3	1.14	1.55+2	1.69+3	3.46+3	<u>8.84+3</u>	<u>8.57+3</u>	<u>2.20+3</u>	<u>4.36+2</u>
²³⁰ Th	3.16-2	7.05-1	2.61+1	2.77+2	1.12+3	1.70+3	1.19+3	3.29+2	2.57+1	2.57+1
²³² Th	1.47-8	2.27-8	4.85-7	3.31-5	3.54-4	7.92-4	4.27-3	8.58-3	4.08-2	7.62-2
²³¹ Pa	9.36-3	1.03-2	2.63-2	8.43-1	1.38+1	3.02+1	4.50+1	4.50+1	4.46+1	4.46+1
²³² U	1.04	4.64-1	8.01-5	0	0	0	0	0	0	0
²³³ U	2.14-3	2.57-2	5.47-1	8.33	3.93+1	7.07+1	1.64+2	1.59+2	4.10+1	8.13
²³⁴ U	3.60	2.67+1	6.33+1	6.20+1	5.57+1	4.83+1	1.61+1	4.30	4.80-1	4.80-1
²³⁵ U	4.19-3	4.72-3	1.02-2	7.16-2	2.80-1	3.66-1	3.94-1	3.94-1	3.91-1	3.91-1
²³⁶ U	1.56-2	4.03-2	2.81-1	1.78	2.75	2.76	2.73	2.69	2.39	2.07
²³⁸ U	7.94-1	7.94-1	7.94-1	7.94-1	7.94-1	7.94-1	8.00-1	8.00-1	8.00-1	8.00-1
²³⁷ Np	1.52+3	2.15+3	5.25+3	6.20+3	6.12+3	6.02+3	5.29+3	4.50+3	1.23+3	2.44+2
²³⁶ Pu	2.35+1	7.33-9	0	0	0	0	0	0	0	0
²³⁸ Pu	7.33+6	4.37+6	2.42+4	7.29-14	0	0	0	0	0	0
²³⁹ Pu	4.49+5	4.51+5	4.70+5	<u>5.39+5</u>	<u>2.42+5</u>	<u>5.98+4</u>	6.93-1	3.67-4	3.09-4	2.51-4
²⁴⁰ Pu	6.39+5	6.93+5	6.32+5	2.51+5	4.16+3	2.46+1	3.56-5	3.56-5	3.46-5	3.33-5
²⁴¹ Pu	8.00+5	1.12+4	3.56+1	1.67+1	5.83-1	8.83-3	2.37-17	0	0	0
²⁴² Pu	1.60+3	1.66+3	1.77+3	1.74+3	1.62+3	1.48+3	7.11+2	2.84+2	1.90-1	2.02-5
²⁴⁴ Pu	3.66-5	3.66-5	3.66-5	3.66-5	3.68-5	3.68-5	3.73-5	3.73-5	3.63-5	3.49-5
²⁴¹ Am	<u>1.51+7</u>	<u>1.34+7</u>	<u>3.17+6</u>	2.58+2	8.93	1.35-1	3.83-16	0	0	0
^{242m} Am	7.71+5	5.11+5	8.43+3	1.26-14	0	0	0	0	0	0
²⁴³ Am	3.86+5	3.83+5	3.53+5	1.56+5	4.16+3	4.49+1	1.15-4	1.13-4	9.50-5	7.71-5
²⁴² Cm	2.95+4	1.96+4	3.23+2	4.83-16	0	0	0	0	0	0
²⁴³ Cm	2.28+5	3.24+4	1.11-4	0	0	0	0	0	0	0
²⁴⁴ Cm	4.39+6	1.39+5	8.65-9	8.52-9	8.52-9	8.52-9	8.61-9	8.65-9	8.39-9	8.04-9
²⁴⁵ Cm	7.40+2	7.34+2	6.81+2	3.20+2	1.12+1	1.68-1	4.53-16	0	0	0
²⁴⁶ Cm	3.34+1	3.29+1	2.88+1	7.67	2.13-2	1.36-5	4.79-19	0	0	0
²⁴⁷ Cm	1.37-4	1.37-4	1.37-4	1.37-4	1.37-4	1.37-4	1.34-4	1.32-4	1.11-4	9.00-5
²⁴⁸ Cm	7.93-4	7.93-4	7.93-4	7.80-4	7.20-4	6.53-4	2.97-4	1.11-4	4.21-8	2.23-12
²⁵⁰ Cm	1.26-8	1.26-8	1.21-8	8.47-9	1.72-9	2.35-10	2.82-17	0	0	0
²⁴⁹ Bk	5.13-7	0	0	0	0	0	0	0	0	0
²⁴⁹ Cf	3.33-3	2.79-3	4.74-4	9.51-12	0	0	0	0	0	0
²⁵⁰ Cf	2.37-4	2.02-6	1.52-10	1.06-10	2.15-11	2.93-12	3.52-19	0	0	0
²⁵¹ Cf	1.88-7	1.75-7	8.76-8	8.57-11	0	0	0	0	0	0
²⁵² Cf	1.12-8	6.50-19	0	0	0	0	0	0	0	0
Total	~30,000,000	~20,000,000	~4,700,000	~960,000	~270,000	~73,000	~17,000	~14,000	~3,600	~800

Each column has an underlined value, indicating the critical heavy nuclide at that particular time.

^aRead as 3.26×10^{-7} .

Considering that direct ingestion or inhalation of radioactive solid waste seems a rather far-fetched assumption, and that the hazard potential should express the global hazard beginning at the time of containment failure until decay of the radioactive nuclides, a different expression of the waste hazard would be more meaningful. Procedures to calculate hazard indexes of radioactive substances have been proposed by several authors.^{6,8-11} Unfortunately, either these indexes are applicable only to particular conditions or their calculation requires data about the modes of exposure and the fraction of activity to which man is actually exposed. These data are usually difficult to obtain even for specific environmental situations. In the case of high-level waste for which containment failure might take place at some unspecified future time, meaningful assumptions about the nature of the environment, its use by man, and possible pathways are practically impossible.

In addition to the general difficulties, not much is known about the environmental behavior of many heavy elements, especially the transuranics. Therefore, in this report, the number of LAI will be considered as a first approximation of the hazard potential associated with the waste after various decay times. The totals shown in Tables 5-8 are somewhat arbitrary; in fact, intakes can be totaled only if the relative dose commitments are referred to the same critical organ. Most of the radionuclides in the tables are bone-seekers; therefore, it can be assumed that bone is the critical organ, and that the totals are more or less legitimate.

Observation of the tables reveals some very interesting facts. After about 100,000 years of decay, the reduction of the hazard potential becomes an extremely slow process; in the time interval between 100,000 and 5 million years there is a 10- to 20-fold reduction in the number of LAI. Considering ingestion as the mode of exposure, the main contribution to the long-term hazard ($>10^5$ years) is due to natural radioactive elements of the uranium series, with ^{226}Ra as the critical nuclide; the neptunium series is next in importance. Observation of the inhalation hazard indicates a completely different situation; ^{239}Pu is still the critical nuclide after 100,000 years, but a few tens of thousands of years later, the main hazard becomes associated with the neptunium series in which ^{229}Th is the critical nuclide. The very long-term inhalation hazard ($>10^6$ years) is almost completely due to the neptunium series.

Another important conclusion drawn from the tables is that the increase in hazard potential of the waste in the time interval between 100,000 and 1 million years that has been mentioned in the literature^{4,12} is mostly an artifact. Some daughter nuclides which reach equilibrium with their parents in that time interval are not listed in the Code of Federal Regulations (CFR); therefore, the potential hazard was not calculated from the appropriate radiation concentration guides (RCG), but from the default value that the CFR prescribes for unlisted nuclides.⁴

The variation in potential hazard from 100,000 to 10 million years is so limited and slow that it is virtually impossible to make a rational case for any specific length of required containment falling within that time interval. Either containment failure can be considered acceptable after a period on the order of 100,000 years, or containment must be assured for periods of time exceeding at least 5 million years. The second alternative is clearly impracticable since totally reliable geologic predictions of the detail required over such long time frames are beyond present capability. This results in two options: (1) proving that containment failure sometime after 100,000 years would result in tolerable levels of environmental contamination; or (2) treating the waste to remove the nuclides responsible for the long-term hazard and eliminating them by some other method.

Bell and Dillon³ have compared the long-term hazard of radioactive waste with the hazard associated with naturally occurring radioactive elements. Typical concentrations of U_3O_8 in uranium deposits are on the order of 0.1 to 0.3% (U_3O_8 is 85% uranium); higher concentrations are not common, and values of 1% uranium can be considered exceptional. Obviously, the uranium concentration in uraniferous minerals is much higher (e.g., pitchblende, a mixture of uranium oxides, is about 60% uranium). Typical concentrations of thorium deposits are somewhat similar; for example, the monazite sands of Brazil can contain as much as 10 to 20% monazite, which is 4 to 6% thorium oxide and 0.15 to 0.25% uranium oxide.¹³

Following the same procedure used for the waste, and taking 1% as the concentration of uranium or thorium, the potential hazard associated with 1 cm³ of uranium ore (assuming a density of 3 g/cm³) is 1.2 LAI_{ing} and 23 LAI_{inh}; the hazard is 0.15 LAI_{ing} and 9.3 LAI_{inh} for 1 cm³ of

thorium ore. The potential hazard of 1 cm^3 of pitchblende (density ~ 9) is $220 \text{ LAI}_{\text{ing}}$ and $4200 \text{ LAI}_{\text{inh}}$; this is comparable to LWR and LMFBR waste aged from less than a thousand to a few thousand years respectively, as far as ingestion is concerned, but waste is still more hazardous than pitchblende after a million years of decay if intake by inhalation is assumed. Ten-million-year-old high-level waste is more hazardous than geologic materials containing 1% uranium or thorium. The difference is quite small for ingestion, but the hazard potential of waste is still 20 to 30 times greater in case of inhalation. This raises the question of the probability of inhalation as a significant mode of exposure and of its contribution to the total hazard potential.

Available experience concerning human exposure to widely dispersed radionuclides indicates that ingestion is usually the most important mode of internal exposure. In the case of fallout plutonium, inhalation is critical despite the fact that the intake by ingestion is two to four times greater than the intake by inhalation.¹⁴ This is due to the very low absorption from the gastrointestinal tract that determines a $\text{LAI}_{\text{inh}}/\text{LAI}_{\text{ing}}$ ratio equal to about 10^{-4} . Inhalation is obviously the critical mode of exposure for fallout plutonium that was originally released into the atmosphere in finely divided form; however, it will be much less important for plutonium or other actinides originally present in solid waste and slowly dispersed into the environment by geologic processes.

As far as natural background radiation is concerned, the highest dose is usually received through external irradiation. This is due to the great geochemical stability of most radioactive compounds and low biologic availability of most natural radioactive elements.

The comparison of the hazard potentials associated with radioactive wastes and natural radionuclides is a potentially useful method for putting the long-term aspects of the waste disposal problem into proper perspective. If inhalation is really an insignificant potential contribution to exposure by nuclides originally buried deep in the earth's crust, the hazard potential associated with high-level waste after only a few thousand years of radioactive decay is then comparable to the hazard potential of uranium minerals.

It can be seen that a comparison based only on the number of LAI contained in waste and ores or minerals neglects some important aspects of the problem. In fact, the actual hazard is greatly dependent on a number of factors, such as transfer of the radionuclides from the solid to the liquid phase, dispersal and accumulation mechanisms, biologic availability, reconcentration in edible organisms, etc.

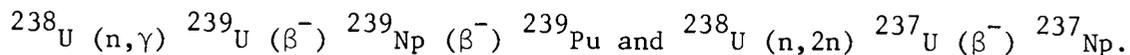
The following sections of the report review the available data concerned with the behavior of natural alpha-emitters and transuranium elements in geologic systems and in terrestrial and aquatic ecosystems. Resuspension mechanisms are discussed briefly in order to obtain some feeling for the probability that a significant fraction of the activity would become airborne. Hopefully, this discussion will lead to a better understanding of the similarities and differences between natural radioactive materials and radioactive wastes, and to a more realistic assessment of the possible environmental impact of the nuclear fission industry.

3. GEOCHEMICAL DATA

The long-lived parents of the three naturally occurring radioactive decay series are ^{232}Th , ^{235}U , and ^{238}U . A fourth series, the neptunium series, with mass numbers defined by the general formula $(4n + 1)$, is not known in nature, and is shown with the other three in the Appendix.*

Uranium and thorium, as minor constituents of the lithosphere, are present in all geologic materials. Table 9 shows the average content of uranium and thorium in various rock types.

* ^{237}Np and ^{239}Pu are formed in nature when atoms of ^{238}U capture neutrons which can be produced by the spontaneous fission of uranium, cosmic radiation, and interaction between alpha particles and light nuclei. The reactions are:



However, the concentrations, especially for ^{237}Np , are so low that these nuclides can be measured only with great effort.

Table 9. Concentration of Uranium and Thorium
in Various Rocks^a

Type of Rock	Concentration (ppm)			Reference No. ^b
	U	Th	Th/U	
IGNEOUS ROCKS				
<u>Acid Rocks</u> (mostly granites)				
North America	4	13	3.2	16
World	3.5	18	5.1	17
<u>Intermediate Rocks</u>				
North America	2.6	10	3.8	16
World	1.8	7	3.9	17
<u>Basic Rocks</u>				
Basalts	0.8	5	6.2	18
Basalts	0.8	3	3.7	17
Average for all igneous rocks	3	10	3.3	
SEDIMENTARY ROCKS				
Placers enriched in uranium- and thorium-bearing heavy minerals	2	60	30	19
Sandstones	0.5-1	2-5		
Shales, average	3.7	12	3.2	20
Grey-green shales, USA	3.2	13.1	4.1	20
Bentonites, USA	5.0	24	4.8	20
Bauxites	9.3	53.2	5.7	20
Residual clays	1.8	13	7.2	20
Shales, Russian platform	4.1	11	2.6	21
Bituminous and carbonaceous shales	50-80 up to ~1250 ^c			
Limestones	1.3	1.1	0.9	18
Limestones, USA	2.2	1.1	0.5	20
Limestones, Russian platform	2.1	2.4	1.1	21
Phosphate rocks	30-100 up to ~650			
Lignites and coals	1-200 up to ~10,000 ^d			

^a Modification of table from Antoni Polanski, Geochemistry of Isotopes, TT61-31327 (English Transl.), published by the Scientific Publications Foreign Cooperation Center, Warsaw, Poland, 1965.

^b Refer to references in text.

^c Shales of St. Hippolyte, France.

^d Kolm, Cambrian coal, Sweden.

Uranium and thorium usually begin their geochemical cycles together. They are closely associated in the same accessory minerals of igneous rocks such as zircon, sphene, allanite, monazite, or xenotime; however, the differences in geochemical behavior (particularly in the postmagmatic phases, and during weathering, transport, and sedimentation) are fairly significant.

From our point of view, the greatest interest lies in the behavior of uranium and thorium during the weathering of rocks and minerals. Most uranium- and thorium-containing minerals dispersed in igneous rocks are extremely resistant to chemical weathering.¹⁵ Several minerals of pegmatitic and hydrothermal deposits, particularly those rich in thorium (such as thorite and thorianite), are similarly characterized by very low solubility. Most of the dispersal of the radioactive elements contained in these minerals is thus achieved through the physical processes of erosion and transport by running water. Weathering, transport, and sedimentation can lead to the accumulation of uranium- and thorium-bearing minerals and, in favorable circumstances, placer deposits can be formed. Famous examples are the monazite sands found on the Brazilian coast between Cabo Frio and Recife, and on the Travancore coast in India.

Many minerals containing radioactive elements, particularly the uranium minerals of hydrothermal deposits, eventually undergo chemical weathering. Secondary products are formed and can be found in the oxidation zones associated with uranium deposits, especially uranite and pitchblende. During weathering under oxidizing conditions, a fraction of uranium goes into solution as stable uranyl complexes, causing partial separation of the two elements.²² Chemical weathering of geologic materials thus results in a relative enrichment of thorium in the residual fraction.

Once the two elements reach a sedimentation basin they will eventually precipitate to the bottom. Thorium, having reached the sedimentation basin primarily by association with suspended particles, is rapidly removed by sedimentation; its residence time* in sea water has been estimated

*The residence time, τ , of elements or radionuclides in environmental compartments can be defined as the average time spent in the compartment before removal. It is expressed as: $\tau = A/(dA/dt)$, where A is the total amount of the element in the compartment, and dA/dt is the amount removed per unit of time. A necessary assumption is that complete mixing of the element takes place in a time which is short when compared with τ .

at about 300 to 350 years. Uranium, on the other hand, is characterized by a residence time in sea water on the order of 500,000 years.²³⁻²⁵ Sackett et al.²⁶ have estimated a residence time of uranium in the world oceans of 200,000 years. Precipitation of uranium involves chemical and biological processes which are especially active in reducing environments where the soluble hexavalent form becomes unstable and is reduced to tetravalent uranium. Consequently, liquid hydrocarbons and bituminous sediments such as dark shales are usually rich in uranium.

Some Tertiary and Cretaceous lignites are also uraniferous. The most commonly accepted explanation for this is that uranium was adsorbed on the surfaces of lignite particles from uranium-rich circulating ground waters, the source of the uranium being located in nearby bodies of igneous rocks. A direct relationship has been observed between the uranium content in peat and in mosses living in the same environment, indicating that, at least in specific cases, the uranium accumulation could have taken place in the living plants.²⁷ Some coal deposits are significantly enriched in uranium; a famous example is kolm, a Cambrian coal from Sweden, which can contain more than 1% uranium. The alum-shales in which kolm is contained have an average uranium content between 50 and 100 ppm, but the uranium concentration is about 200 ppm in proximity to the kolm seams.²⁸

Phosphate rocks of marine origin can also be markedly enriched in uranium, up to about 650 ppm in some cases. It is believed that the uranium is mainly syngenetic, although limited redistribution during diagenesis is quite possible. Uranium in phosphorites is dispersed in the phosphate phase and apparently has been enriched by coprecipitation.²⁹ At least two mechanisms seem to be responsible for the presence of uranium in phosphate minerals: (1) isomorphous replacement of U^{4+} for Ca^{2+} in apatite, and (2) adsorption of uranium on the surfaces of the minute phosphate crystals and the associated organic matter. According to Bliskovskiy and Smirnov,³⁰ the weathering of phosphorites can result in a further increase in the uranium concentration.

In summary, the geochemistry of uranium and thorium is quite similar in the magmatic phase; however, differences develop in postmagmatic stages, becoming even more significant when the rocks and minerals are exposed to exogenous processes. Consequently, high values of the Th/U ratio are

observed in continental sediments, such as residual clays and bauxites. In general, any elution process, particularly if under oxidizing conditions, will lead to enrichment of thorium relative to uranium. Low values of the Th/U ratio are found in sediments accumulated by chemical processes (e.g., carbonates and evaporites). Finally, extremely low values of the Th/U ratio are found in certain sediments such as dark shales, and in some coals and phosphatic rocks.

The short-lived daughters of uranium and thorium are usually found associated with their parents in radioactive equilibrium. In specific circumstances some separation can take place, the net result is a relative geochemical individualization, especially for nuclides with fairly long half-lives or great environmental mobility. Obvious examples are ^{226}Ra and radon.

The interaction between ground water and uranium- and thorium-rich rocks is of particular interest, since this is similar to the hypothetical first step of radionuclide mobilization after waste containment failure. Radium-226 is the most hazardous nuclide, for exposure by ingestion, both in high-level radioactive waste aged more than 50,000 years and in the natural radioactive series.

In radioactive equilibrium, the Ra/U ratio is 3.6×10^{-7} ; the specific activity of ^{226}Ra is ~ 1 Ci/g. Radium is an alkaline-earth element, being located in the second group of the periodic system of elements, and barium is its closest geochemical analogue. Radium occurs in nature in a very dispersed state and no minerals are known.

The chemistry of ground waters is always controlled by the chemical composition of the enclosing rocks, and the content of radioactive substances is no exception. The enrichment of natural waters with radioactive elements, particularly uranium and radium, is dependent on the following processes:

- (1) primary transfer from the rock into the water;
- (2) stability in solution;
- (3) separation of the element from solution.

3.1 Mobilization of Uranium and Radium

Uranium and thorium in rocks are usually bound in the crystal lattice of minerals; therefore, their transfer to ground water is controlled by the solubility of the minerals, which is influenced by water chemistry, amount of free oxygen, temperature, etc.³¹ Radioactive decay transforms uranium and thorium atoms into atoms of different elements that might not be able to occupy the same position in the crystal lattice. Radium, in particular, can be leached out of the minerals in which it was generated, and thus accumulated in the interstitial water. Of course, the transfer of radium from the solid to the liquid phase is greatly dependent on the size of the rock particles since leaching is a surface-related process.

On the other hand, if the grain size is in the clay range, radium can be transferred efficiently to the surface of the particles, but release from the solid phase is impeded by the low permeability and by surface attraction phenomena.

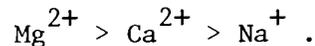
Starik and Lazarev³² have studied the effect of crushing monazite on the extraction of radium, thorium, and uranium. Monazite is practically insoluble, and leaching of the radioelements occurs by extraction from the pores and surfaces of the mineral grains. The extraction of radium and uranium increased with the degree of crushing up to about 3%, for an average particle size <0.07 mm. On the other hand, the extraction of thorium was not changed by crushing, despite a more than 100-fold increase of the specific surface.

In relatively permeable rocks, the transfer of radium from the rock grains to the mobile interstitial water can be imagined as divided into the following steps:

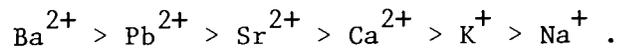
- (1) Radioactive decay, through recoil and the creation of lattice instability, moves radium atoms to the surface of rock particles.
- (2) Radium is leached out of the particle surfaces into the film of adsorbed water surrounding the rock grains.
- (3) Radium diffuses through the adsorbed water and reaches mobile water.

The transfer from the particle surfaces to the adsorbed water is controlled by an adsorption equilibrium, and the transfer to mobile water is dependent on a diffusion equilibrium. Due to the slowness of diffusion processes, a long contact time between ground water and formation, realized in conditions of stagnant or slow-moving waters, promotes their enrichment in radium. Stagnant waters are usually highly mineralized. High salinity further enhances the enrichment in radium, caused by the competition and exchange between the ions in solution and the ions adsorbed on the rock particle surfaces.^{33,34}

The nature of the ions present in ground water is also important. The dissolution of uranium is appreciably influenced by cationic composition of the water only in carbonate rocks. On the basis of their effect on the dissolution of uranium, cations can be arranged in this order:



The effect of cationic composition of the water on leaching of radium is much stronger. In general, the greater the chemical affinity of an element to radium, the greater its effect on leaching. The sequence is:



The anionic composition of the water significantly affects the solubility of uranium, but has little effect on radium. Bicarbonate ions are the most efficient since they cause the formation of readily soluble uranyl complexes of the type $[\text{UO}_2(\text{HCO}_3)_3]^{-}$.

Additional factors that are important in the transfer of radioactive elements into ground water are: (1) water temperature, (2) pH, (3) gas content, (4) electrochemical properties.

The transfer of uranium and radium into solution is directly proportional to the water temperature.

The solubility of uranium and radium increases at low and high values of pH. Therefore, acidic and alkaline waters are usually enriched in radioactive elements.

The gases with most effect on the migration of radioactive elements are O_2 and CO_2 . Free oxygen in ground water decreases with depth down to an "oxygen surface" that represents the lower boundary of the presence of free oxygen. Above the oxygen surface is the domain of oxidation processes, whereas reduction processes are prevalent at greater depth. The depth of the oxygen surface can vary from almost at the surface to more than 1 km.³¹ Free oxygen produces a marked effect on the solubility of uranium.

Carbon dioxide increases the transfer of uranium into solution by the formation of soluble bicarbonates of the very stable uranyl ion. The enrichment with radium is also affected.

The redox potential (Eh) of ground water is an index of the intensity of oxidation processes, and is related to several of the preceding factors. Above the oxygen surface the value of Eh is directly proportional to the content of free oxygen, while for the deeper zone it is inversely proportional to the content of hydrogen sulfide. Obviously, the chemical weathering of uranium-bearing minerals and the enrichment of uranium in solution takes place to a much greater extent in waters characterized by high values of Eh. Radium, on the other hand, is affected much less by the redox potential and shows greater stability in solution in waters of low Eh.³⁵

3.2 Removal of Uranium and Radium from Ground Water

Once uranium and radium have been transferred into solution they migrate with the water. The extent of migration is dependent upon their stability in solution, and on the intensity of various processes leading to their removal from solution. Any variation of the factors discussed above as affecting the mobilization of uranium and radium can induce precipitation. Decrease of temperature, loss of gas, neutralization of acidic or alkaline waters, or any other change in the chemistry of the water might cause separation of uranium and/or radium from solution.

The principal processes responsible for precipitation of uranium are:

- (1) Hydrolysis and coagulation of uranium hydroxides (mostly in clays).

- (2) Adsorption on the surface of clay particles and organic matter, responsible for the enrichment of uranium in clays, bituminous shales, peat, coal, etc.
- (3) Breakdown of complex ions with formation of less soluble compounds. Among the various carbonates, coprecipitation with siderite is most efficient in the removal of uranium from solution.
- (4) Reduction of uranium from the hexavalent to the tetravalent oxidation state.
- (5) Formation of insoluble salts - vanadates, phosphates, arsenates, carbonates, and silicates.

As already mentioned, no radium minerals are known in nature; however, radium will follow its geochemical analogues. Therefore, precipitation of barium compounds from ground water will result in the removal of radium from solution. Radium follows calcium and magnesium to a lesser extent. Significant enrichment of radium also takes place in precipitates of iron and manganese, probably due to the colloidal nature of these compounds and the adsorption of radium on the surface of colloids. Adsorption and ion exchange are responsible for the enrichment of radium in peat, coal, and clays. Table 10 shows the concentration of radium in the waters of some Russian mineral springs and the enrichment in the spring deposits. Obviously, in favorable circumstances, significant accumulation and re-concentration of radium can take place.

3.3 Abundance of Natural Alpha-Emitters in the Hydrosphere

The complexity of the geochemical behavior of uranium and radium indicates that the concentrations of these elements in natural waters will vary within wide limits. In general, the content of radioactive elements in surface waters is lower than in ground waters due to the dilution by meteoric waters. A relationship exists between the climate and the uranium content in ground waters, with arid regions being characterized by significantly higher levels of uranium.³⁵ In specific closed basins in arid areas, the concentration of uranium can be as high

Table 10. Content of Radium in Water and Deposits of Some Russian Mineral Springs^a

Spring	Nature of Deposit	Radium in Water (pg/liter)	Radium in Deposit (pg/g)	<u>Radium in Deposit</u> Radium in Water
Malakovka	Ferruginous precipitate	7,000	153,000	22,000
Yamkun	Travertine	1,000	14,000	14,000
Zheleznovodsk	Travertine	21,000	4,600	220
	Ferruginous precipitate	15,000	60,000	4,000
Pyatigorsk	Travertine	300	1,800	6,000
	Manganic precipitate	300	410,000	1,350,000
Obershlem	Clayey precipitate	6,000	2,830,000	471,000
Vishnevogorsk	Peat	100	2,000	20,000
Baku	Ferruginous precipitate	90,000	7,800	87
Ukhta	Ferruginous precipitate	748,000	400,000	530

^aModification of table from A. N. Tokarev and A. V. Shcherbakov, Radiohydrogeology, Moscow (1956) AEC-tr-4100 (1960).

as $\sim 4 \times 10^{-2}$ g/liter.³¹ The average concentrations of uranium and radium in sea water are 3 μ g/liter and 0.1 pg/liter respectively. The volume of the oceans is 1.37×10^9 km³; therefore, it can be calculated that about 1.5×10^8 g of ²²⁶Ra and 4.5×10^{15} g of uranium are present in sea water. Table 11 shows the ranges and mean concentrations of radium and uranium in surface and various ground waters. All values of the Ra/U ratio higher than 3.6×10^{-7} indicate an enrichment of radium relative to uranium, while the opposite is true for values lower than equilibrium. In ground waters below the oxidation zone, the equilibrium is usually displaced toward radium.

A deficiency of radium exists in the oceans and, to a lesser extent, in lakes. This is undoubtedly related to the very low solubility and removal by sedimentation of ²³⁰Th, which is the immediate precursor of ²²⁶Ra in the radioactive series (see the Appendix). This interpretation is supported by the excess of radium relative to uranium that has been reported for recent deep-sea sediments.^{36,37} When the concentrations of ²³⁰Th in sea water and deep-sea sediments are analyzed, it is found that the deficiency of this nuclide in water is much more marked than for radium; conversely, there is an excess of ²³⁰Th with respect to ²²⁶Ra in the sediments. This situation can be explained when it is understood that ²³⁰Th, with a half-life of 80,000 years, separates from sea water and accumulates in the sediments, where its decay produces ²²⁶Ra, part of which is leached out of the sediments and returned to the water.^{38,39}

It has been observed that disequilibrium usually exists between ²³⁸U and ²³⁴U.^{24,40-42} In sea water the ²³⁴U/²³⁸U activity ratio is 1.14, indicating an excess of ²³⁴U of 14%. This is due to the fact that ²³⁴U is removed from weathering rocks more readily than ²³⁸U. Electrons are lost during the decay from ²³⁸U to ²³⁴U and change to the more soluble +6 state is facilitated.²⁴

Natural waters contain significantly less thorium than uranium. In sea water, which represents about 98% of the hydrosphere, the average thorium concentration has been estimated as falling between 2×10^{-8} and 5×10^{-8} g/liter.^{25,35} The U/Th ratio is thus about 100, or a variation of about 300 with respect to the relative abundance of the two elements in the lithosphere. On the other hand, the Th/U ratio in sea bottom

Table 11. Content of Uranium and Radium in Natural Waters^a

Type of Waters	Natural Conditions	Radium (g/liter)			Uranium (g/liter)			$\frac{Ra}{U}$
		min.	max.	mean	min.	max.	mean	
Surface Waters	Oceans and seas	8.0×10^{-14}	4.5×10^{-11}	1.0×10^{-13}	3.6×10^{-8}	5.0×10^{-6}	3.0×10^{-6}	3×10^{-8}
	Lakes	1.0×10^{-13}	8.0×10^{-12}	1.0×10^{-12}	2.0×10^{-7}	4.0×10^{-2}	8.0×10^{-6}	1×10^{-7}
	Rivers	1.0×10^{-13}	4.0×10^{-12}	2.0×10^{-13}	2.0×10^{-8}	5.0×10^{-5}	6.0×10^{-7}	3×10^{-7}
Waters of Sedimentary Rocks	Zone of intensive water circulation	1.0×10^{-13}	6.0×10^{-12}	2.0×10^{-12}	2.0×10^{-7}	8.0×10^{-6}	5.0×10^{-6}	5×10^{-7}
	Zone of highly impeded water circulation	1.0×10^{-11}	1.0×10^{-8}	3.0×10^{-10}	2.0×10^{-8}	6.0×10^{-6}	2.0×10^{-7}	1×10^{-3}
Waters of Magmatic Acid Rocks	Zone of intensive water circulation (waters of the weathering shell)	1.0×10^{-12}	7.0×10^{-12}	2.0×10^{-12}	2.0×10^{-7}	3.0×10^{-5}	7.0×10^{-6}	1×10^{-6}
	Zone of impeded water circulation (waters of deep tectonic fissures)	2.0×10^{-12}	9.0×10^{-12}	4.0×10^{-12}	2.0×10^{-7}	8.0×10^{-6}	4.0×10^{-6}	2×10^{-6}
Waters of Uranium Deposits	Zone of intensive water circulation (waters of the oxidation zone)	8.0×10^{-12}	2.0×10^{-9}	8.0×10^{-11}	5.0×10^{-5}	9.0×10^{-2}	6.0×10^{-4}	1×10^{-7}
	Zone of impeded water circulation (waters of the reduction zone)	1.0×10^{-11}	8.0×10^{-10}	6.0×10^{-11}	2.0×10^{-6}	3.0×10^{-5}	8.0×10^{-6}	1×10^{-5}

^aModification of table from A. N. Tokarev and A. V. Shcherbakov, Radiohydrogeology, Moscow (1956) AEC-tr-4100 (1960).

sediments is roughly similar to the average value in continental rocks.^{22,43} This would indicate that, overall, the two elements are transferred to the oceans with similar efficiency. However, while the geochemistry of uranium is based mainly on aqueous solutions, the geochemical cycle of thorium is almost completely independent of aqueous solutions.

Sackett et al.²⁶ have recently attempted a world balance of the geochemical cycle of uranium, and have found that the input of uranium to the ocean is higher than the amount removed by deposition by at least a factor of 3. This discrepancy could be explained by the presently high input of uranium to the ocean, or by an underestimate of uranium removal. The actions of man, particularly in the use of phosphate fertilizers and the enhanced leaching of soils caused by world-wide cultivation, are probably responsible for the observed imbalance.

A short-lived nuclide in the ^{235}U series, ^{227}Th , is present in sea water at a concentration of only a few percent of the equilibrium value. This indicates that one of the longer-lived precursors, most likely ^{231}Pa (half-life, 32,500 years), is efficiently removed from sea water.³⁷

Of all natural radioactive elements present in natural waters, radium is usually the most significant from the radiological point of view.^{44,45} The limit of ^{226}Ra in drinking water supplies has been fixed by the U. S. Public Health Service at 3 pCi/liter.⁴⁶ Many natural waters, not only highly mineralized brines of sedimentary rocks and waters of specific mineral springs, but also waters of aquifers used extensively for drinking water supplies, contain radium in excess of this limit.

High levels of radium in ground waters are known in several areas of the United States.⁴⁷⁻⁴⁹ Three important examples of radium-rich aquifers are (1) limestones, dolomites, and sandstones of Cambrian and Ordovician age in the upper Mississippi Valley (Illinois, Iowa, Wisconsin, Michigan, and Indiana); (2) limestones and sandstones of Ordovician age in Kansas, Oklahoma, and Missouri; and (3) the Cheyenne sandstone member of the Purgatoire Formation of Early Cretaceous age in southeastern Colorado and northeastern New Mexico. Scott⁴⁷ says that in the first two aquifers most of the uranium is probably associated with shale beds and lenses that are fairly numerous throughout the formation, whereas it is likely that uraniumiferous minerals are uniformly dispersed in the Cheyenne sandstone.

A correlation exists between radium concentration and salinity; fresh ground waters contain ^{226}Ra at concentrations ranging from fractions of a picocurie per liter to a few tens of picocuries per liter, while brines (dissolved solids >3000 ppm) can contain as much as several hundreds of picocuries per liter.

Samuels⁵⁰ estimates that in the upper Mississippi Valley at least one million people are exposed to drinking waters containing more than 3 picocuries of ^{226}Ra per liter, and about 50,000 people have more than 10 picocuries per liter in their water supplies. This obviously affects the radium intake in the exposed populations. The average daily intake of ^{226}Ra for the U. S. population is probably in the range 1 to 2 pCi,⁵¹⁻⁵³ of which about 90% is from food and 10% is from drinking water.⁵⁴ In the high radium areas, the daily intake can be significantly higher. Samuels⁵⁰ estimates that the contributions to the intake from food and water are probably reversed, with about 90% due to ingestion of water.

3.4 Transuranium Elements

As already mentioned, minute amounts of neptunium and plutonium are formed in nature by the interaction of neutrons with ^{238}U nuclei. Unfortunately, very little is known about the geochemistry of these elements and their long-term mobility through geologic systems.

Several authors have reported the occurrence of ^{239}Pu in uranium ores with α , which is defined as the $^{239}\text{Pu}/^{238}\text{U}$ ratio, on the order of 10^{-11} to 10^{-12} .⁵⁵⁻⁵⁷ These values are in fairly good agreement with the α obtained theoretically, assuming complete utilization of available neutrons.

Cherdyntsev et al.⁵⁸ have measured the ^{239}Pu content in some volcanic waters and found values up to 10^{-13} g/liter with an α as high as 10^{-7} . This is orders of magnitude higher than the α observed in uranium minerals. Excluding the possibility of contamination by artificial plutonium, Cherdyntsev et al. propose an additional natural source of ^{239}Pu in the form of an unknown osmium-type transuranium element with probable atomic number 108.^{58,59} Among the plutonium-bearing minerals associated with the

plutonium-rich waters are sphene, molybdenite, magnetite, and particularly apatite and zircon. The $^{239}\text{Pu}/^{238}\text{U}$ ratio in these minerals is of the same order of magnitude as in the water, indicating that, in this case, no fractionation between the two elements takes place during transfer from one phase to the other.

Additional instances of high plutonium levels have been reported more recently for rocks and minerals produced by recent volcanic activity, and for the associated waters.⁶⁰⁻⁶² For example, unusually high plutonium concentrations have been observed in limonite and water samples from the fumarole field of the Mutnov volcano in Kamchatka, and in the manganese-iron suspensions of an Indonesian submarine volcano. Tables 12 and 13 show the plutonium content in some recent volcanic products as reported by Cherdyntsev et al.⁶⁰ The difference in Pu/U ratios between the limonite and the water in the Kamchatka fumarole area seems to indicate that plutonium is removed from solution somewhat more actively than uranium, at least in this particular environment.

Table 12. Plutonium Content in Waters of Volcanic Regions^a

Samples	^{239}Pu (10^{-12} g/liter)	U (10^{-6} g/liter)	Pu/U (10^{-8})
Fumarole area, Kamchatka	3.1 ± 0.3	2.4	130 ± 10
<u>Caucasus</u>			
High altitude springs, Dzhulisu, Elbrus	0.018 ± 0.006	0.43	4.1 ± 1.5
Hot springs, Nakalakavi	0.030 ± 0.008	0.37	8 ± 2
Sulfate spring, Abano	2.0 ± 0.4	16	13 ± 2
Carbonate spring, Khasut	0.029 ± 0.009	0.16	18 ± 5.5

^aTable taken from V. V. Cherdyntsev, V. L. Zverev, V. M. Kuptsov, and G. T. Kislitsina, "Plutonium-239 in Nature," *Geochem. Int.* 5. 355-61 (1968).

Table 13. Plutonium Content in Recent Volcanic Rocks and Minerals^a

Sample	²³⁹ Pu (10 ⁻¹⁴ g/g)	U (10 ⁻⁶ g/g)	Pu/U (10 ⁻⁸)
<u>1. Minerals from fumarole areas</u>			
Limonite, Kamchatka	740 ± 60	0.3	1700 ± 200
Same, another sample	470 ± 30	0.3	1600 ± 100
" " "	177 ± 5	0.12	1440 ± 70
" " "	125 ± 5	0.75	170 ± 10
Same, iron-enriched rock	40 ± 1	0.03	1300 ± 100
Manganese-iron suspension, submarine volcano, Indonesia	740 ± 20	56	12.5 ± 0.4
<u>2. Silicic volcanic rocks, Caucasus</u>			
Rhyolite, Mt. Kuyun-Dag	4.7 ± 0.8	0.95	4.9 ± 0.9
Same, banded rock	13 ± 2	0.31	42 ± 7
" tuff	7.4 ± 2.4	0.82	9.1 ± 3.0
" volcanic bomb	39 ± 8	0.90	43 ± 9
Trachyliparite, Mt. Zmeyka	<2	17	<0.1
Pumice, Mt. Bogutlu	11.8 ± 0.8	0.38	31 ± 2
" Dzhvlar	1.4 ± 0.3	0.14	10 ± 2.5
Tuff, village of Fontan	5.6 ± 1.2	0.47	12 ± 2.5
" Mt. Elbrus	4.1 ± 1.0	0.60	6.8 ± 1.6
Andesite-dacite, Erman plateau	2.1 ± 0.5	0.44	4.0 ± 1.3
Same, flow I, Chkher ravine, Mt. Kazbek	1.5 ± 0.6	0.55	2.8 ± 1.1
Same, flow III	0.44 ± 0.14	0.06	7.3 ± 2.4
Tuff at base of flows, same location	<1	0.19	<5
Holocene andesite-dacite flow, Tkasheti	6.7 ± 2.0	0.12	56 ± 17
Andesite-dacite, Galgat cone	1.4 ± 0.4	0.07	20 ± 5
Same	3.8 ± 1.0	0.14	27 ± 7
Tuff, same location	8.0 ± 0.4	0.41	19.5 ± 1.7
Volcanic bomb, large cone, Galgat	12.0 ± 0.75	0.075	160 ± 10
Same, small cone, Galgat	26.2 ± 0.6	0.145	181 ± 4
<u>3. Intermediate volcanic rocks, Caucasus</u>			
Andesite, Kel'sk plateau	4.6 ± 0.8	0.32	14 ± 2
Same, Tsitelakhati R., Kel'sk plateau	5.8 ± 0.3	0.32	18 ± 1
Same, Mt. Kabardzhino	4.7 ± 0.8	0.66	7.1 ± 1.3
Ash, Shatskiy dome	2.9 ± 0.7	0.29	10 ± 2.5
Tuff, Bakuriani	1.6 ± 0.4	0.33	4.8 ± 1.2
Tuff-pumice, Mt. Malyy Abul	2.6 ± 0.8	0.56	4.6 ± 1.4
Andesite, Aragats volcano	<0.035	0.18	<0.2
<u>4. Mafic volcanic rocks, Caucasus</u>			
Basalt, Aragats volcano	2.6 ± 0.5	0.30	8.5 ± 1.7
Basalt, Dzhermuk volcano, flow I	1.3 ± 0.3	0.24	5.4 ± 1.2
Same, " " last flow (V)	2.4 ± 0.4	0.15	16.0 ± 2.7
Basaltic andesite, village of Tambovka	5.2 ± 0.8	0.40	13 ± 2
Cone material, Dzhermuk	<0.25	1.3	<0.2
<u>5. Volcanic rocks, Italy</u>			
Pumice, Pompeii	17 ± 6	37	0.47 ± 0.16
Trachyte	30 ± 15	50	0.6 ± 0.3
Piperno	---	---	3.2 ± 1.5
<u>6. Spring deposits</u>			
Carbonate spring, Abano, upper reaches of Terek River	21 ± 3	0.14	150 ± 22
Ferruginous travertine, Khasaut, Narzan valley	5.5 ± 1.1	0.60	9.2 ± 1.9
Carbonates, Kaler, Armenia	2 ± 1	2.2	9.0 ± 4.4
Carbonates, Monakh Bay	<0.12	0.15	<0.9
<u>7. Marine Mn - Fe nodules</u>			
Pacific ocean, sp. No. 844	60 ± 5	---	---
Same, sp. No. 842	36 ± 6	6.5	5.5 ± 1.0
Same, sp. No. 851	<16	4.25	(1.9 ± 1.0)
Gulf of Finland, sp. No. 850	20 ± 7	6.1	3.2 ± 1.0
Manganese ore, Caucasus	---	---	<0.2
<u>8. Accessory minerals of monzonite, Caucasus</u>			
Apatite	22 ± 7	20.2	1.0 ± 0.3
Sphene	17 ± 7	31	0.56 ± 0.25
Magnetite from placer	5 ± 2	5	1.0 ± 0.4
Hornblende	5.0 ± 1.3	0.18	28 ± 4

^aTable taken from V. V. Cherdyn'tsev, V. L. Zverev, V. M. Kuptsov, and G. T. Kisiitsina, "Plutonium-239 in Nature," *Geochem. Int.* 5, 355-61 (1968).

In general, these data suggest a considerable geochemical mobility of plutonium. Unfortunately, no determinations of the chemical state of plutonium in the enriched waters exist. The available information is too scanty and too restricted to volcanic environments to provide a reliable indication of the general behavior of plutonium in geologic systems.

Andelman and Rozzell⁶³ have reviewed the behavior of plutonium in the water environment and found that plutonium hydrolyzes in the pH range of natural waters. The hydrolysis products exhibit colloidal behavior and are easily adsorbed on the particle surfaces. As with uranium, the solubility of plutonium is enhanced by the presence of bicarbonate ions, with which it forms stable carbonate complexes. Silver⁶⁴ has discussed the difficulties of forecasting the behavior of plutonium in the aqueous environment due to the various possible oxidation states of the element and the great chemical variability of natural waters.

The underground migration of plutonium colloids through geologic formations would be restricted by filtration and surface adsorption, rather than by ion exchange. Controlling factors would be the size of colloid particles and the dimensions of interstices through which water circulates. Christenson and Thomas⁶⁵ have studied the distribution of plutonium underneath one of the Los Alamos waste seepage pits, and have observed migration through at least 8.5 m of tuff. The data indicate that movement took place essentially along fissures in the tuff beds.

Study of the distribution of plutonium in soil profiles has shown some penetration of the element.⁶⁶⁻⁶⁹ The downward migration of plutonium is obviously a function of rainfall and the physicochemical characteristics of both the plutonium particles and the soil. In all reported cases, most of the plutonium was contained in the first 10 cm of soil, and the amount that penetrated below 30 cm was always less than 5% of the total. The actual mechanism of infiltration is not clear; at the Nevada Test Site, it is believed to be due mainly to mechanical movement of high-density particles along fissures formed by alternate freezing and thawing, and alternate wetting and drying.⁷⁰ At Alamogordo, the evidence seems to indicate that very little vertical migration of plutonium occurred during the period 1948-1956.⁷¹ The mechanism of infiltration in humid areas

could be significantly different and should be controlled by the percolation of water. Biologic processes could also contribute to the downward migration of plutonium.

Almost no data are available concerning the behavior of neptunium, americium and curium in geologic materials and ground waters. Data about americium and curium in soils seem to indicate a behavior not too different from plutonium, as all these elements, including neptunium, at typical soil pH, are known to form insoluble hydroxocolloids; however, neptunium exists primarily in the +5 oxidation state, whereas plutonium is +4 and americium and curium are +3.⁷²⁻⁷⁴

If Cherdyntsev's explanation for the high plutonium concentrations in some recent volcanic products is correct, measurable amounts of ^{243}Am and ^{247}Cm - relatively long-lived nuclides in the decay series of the unknown transuranium element - must exist in some geologic environments. If radioactive equilibrium were to exist in some plutonium-rich minerals, the $^{243}\text{Am}/^{239}\text{Pu}$ and $^{247}\text{Cm}/^{239}\text{Pu}$ ratios would be unity on the basis of activity, and 0.3 and 670, respectively, by weight. Data on the geochemical behavior of these elements might thus become available through the analysis of appropriate geologic materials.

The global contamination of surface waters with fallout plutonium would offer an opportunity for the study of the behavior of this element in the water environment. Unfortunately, little has been learned in this area, and we are still limited to few studies of plutonium distribution in the oceans.⁷⁵⁻⁸⁰ Noshkin⁸⁰ has clearly summarized the situation: "We know very little about the behavior of plutonium, relatively nothing of the behavior of the other transuranics in the ocean, and far less of the behavior and fate of the elements in fresh water environments."

The total amount of plutonium dispersed into the environment can be estimated on the basis of known environmental levels, or from the magnitude of total atmospheric nuclear tests. The following values of ^{239}Pu activities are inclusive of ^{240}Pu , which cannot be distinguished from ^{239}Pu by alpha spectrometry because of the similarity of energy of the alpha particles.

Stewart et al.⁸¹ have estimated the total deposition of ^{239}Pu in the United Kingdom to the end of 1955 as 0.4 mCi/km^2 . The total

deposition of fallout plutonium in Tokyo for the period 1958-1969 has been estimated as ~ 1 mCi/km².⁸² Hardy and Krey⁶⁸ measured the accumulated ²³⁹Pu in six soil profiles in the New York area, and found an average value of 2.3 ± 0.3 mCi/km². De Bortoli and Gaglione⁶⁶ have analyzed 11 soil profiles collected in the area of Ispra, Italy, in 1966 and 1967, and reported a mean ²³⁹Pu deposition of 2.1 ± 0.7 mCi/km². Noshkin⁸⁰ reports that in Buzzards Bay, Massachusetts, the total deposition of ²³⁹Pu has been estimated at 2.3 ± 0.2 mCi/km². Aarkrog⁸³ estimates that up to 1968, the total ²³⁹Pu deposition in Thule was ~ 0.3 mCi/km².

Hardy et al.⁶⁹ have reported the results of world-wide deposition analyses for ²³⁸Pu, ²³⁹Pu, and ⁹⁰Sr. The heaviest fallout deposition has taken place in the middle latitudes of the Northern Hemisphere, with much lower values for the polar and equatorial regions. From the average deposition per 10° latitude bands, Hardy et al.^{69,84} estimate the global inventory of ²³⁹Pu at the end of 1970 as 326 ± 36 kCi.

Alternatively, if we consider the total production of atmospheric ⁹⁰Sr through 1972 to be a little more than 21 MCi, and the average activity ratio between ²³⁹Pu and ⁹⁰Sr as being 0.023 and 0.028, we obtain a total production of fallout ²³⁹Pu ranging from 500 to 600 kCi.⁸⁵⁻⁸⁸ The actual inventory of man-made ²³⁹Pu in the environment is very likely to fall in the range of 300 to 600 kCi (from 5000 to 10,000 kg), which is sufficiently accurate for the present discussion.

The measurements of ²³⁹Pu concentrations in ocean waters have shown that the upper layers are significantly deficient in the nuclide. In fact, the ²³⁹Pu/⁹⁰Sr and ²³⁹Pu/¹³⁷Cs ratios are quite low, indicating fixation of plutonium on sinking particles. It is not known if the removal of plutonium from surface waters is due mainly to adsorption on clay minerals or to uptake by microorganisms.⁷⁸⁻⁸⁰ Noshkin and Bowen⁸⁰ indicate that plutonium residence time in ocean water is so short that biologic processes can only account for part of the removal. On the other hand, it is known that a number of marine species have fairly high plutonium concentration factors. It thus seems logical that sedimentation of plutonium could be due to a combination of biological and physicochemical processes.

Wahlgren and Nelson⁸⁹ have studied the behavior of fallout plutonium in Lake Michigan, finding an average concentration of ²³⁹Pu in the water

of 0.7×10^{-3} pCi/liter. If all deposited plutonium had remained in the water phase, the concentration would have been 0.028 pCi/liter. The resulting half-time for removal from Lake Michigan is 1.0 ± 0.3 years.

The half-time of residence of ^{239}Pu in the upper layers of the North Pacific has been estimated as about 3.5 years.⁹⁰ Few measurements have been made of the concentration of plutonium in sea bottom sediments.^{79,80,91} Cores from Buzzards Bay, Massachusetts, the North and South Atlantic, the Mediterranean Sea, and the Irish Sea show that practically all plutonium is contained in the first 20 cm of sediments. In most cases, the rate of sedimentation is too low to account for the observed distribution; therefore, it must be concluded that secondary migration through the sediments has taken place. The penetration of plutonium and other fallout nuclides in the sedimentary profile can be explained by diffusion, biological processes, or a combination of the two.

Aarkrog⁸³ has studied the fate of plutonium in the Thule environment, following the detonation of the chemical explosive in the triggering mechanism of four nuclear weapons. Sediments from Bylot Sound, 1 km from the point of impact, contained as much as 16 pCi/g (dry weight) of ^{239}Pu . Bivalves 15 km away had plutonium concentrations two orders of magnitude above background, indicating significant dispersion through the marine environment.

Sediments in proximity of the Bikini Atoll, where several underwater test explosions have been carried out, contained as much as 8 pCi/g (dry weight) of ^{239}Pu .⁹² As could be predicted, plutonium levels were higher in association with fine-grained sediments.

4. ENVIRONMENTAL BEHAVIOR

The distribution of the elements in the lithosphere and hydrosphere is controlled by their geochemical behavior. However, the final transfer from the environment to human beings is mainly dependent on mobility and concentration in biologic systems. In certain cases, inhalation and direct ingestion of water can be the critical mechanisms of uptake from the environment, but the internal exposure of human populations is generally a function of the content of radionuclides in the diet.

Natural radioactive elements are dispersed in all parts of the environment and, therefore, are and have always been present in edible organisms and human beings. Several studies have been made of the content of natural radioactive elements in various foods and in the total diet.^{51-54,93-96} The only important sources of internal radiation in the uranium and thorium series are radium and radon and its decay products. Table 14 shows a recent estimate of the internal radiation doses delivered to the average inhabitant of the United States by natural radionuclides.⁹⁷

Radon is a noble gas and is characterized by greater environmental mobility than the other natural radioactive elements. It causes widespread exposure of the population through inhalation. The environmental behavior and the biologic availability of the radon's daughters, primarily ^{210}Pb and ^{210}Po , are greatly dependent on the wide atmospheric distribution of radon, and on natural fallout as the mechanism of entry into the food chains. Therefore, their study would be of little use in understanding the behavior of nuclides without a rare-gas precursor. For this reason, radon and its daughters will not be considered further in the present discussion.

As already mentioned, the average intake of ^{226}Ra resulting from a typical U. S. diet has been estimated as 1 to 2 pCi/day. In Germany and the United Kingdom, the values are 3 and 1.2 pCi/day, respectively.^{54,98} The average dietary intake of ^{226}Ra has been estimated as 1.4 pCi/day in the region of Varese, Italy.⁹⁵

The average dietary intake of uranium in three large U. S. cities has been estimated as about 1.3 $\mu\text{g}/\text{day}$.⁹³ Hamilton⁹⁶ has calculated the average intake in the United Kingdom as about 1 $\mu\text{g}/\text{day}$. It has been reported that in one area of the Soviet Union the average daily intake of uranium reaches 30 μg .⁹⁹

The average intake of ^{239}Pu in nine U. S. cities was estimated by Michelson et al.⁵¹ as 0.1 pCi/day. Magno et al.¹⁴ have calculated an average dietary intake of ^{239}Pu in the United States of 0.007 pCi/day.

In order to put these dietary intakes into perspective, it may be useful to compare them with the following limits of intake by ingestion: ~ 26 pCi/day for ^{226}Ra ; ~ 3800 $\mu\text{g}/\text{day}$ for uranium; and $\sim 10,000$ pCi/day for

Table 14. Estimated Average Annual Internal Radiation^a Doses from Natural Radioactivity in the United States^a

Radionuclide ^b	Annual Doses (mrem/person)		
	Whole Body	Endosteal Cells (Bone)	Bone Marrow
³ H	0.004	0.004	0.004
¹⁴ C	1.0	1.6	1.6
⁴⁰ K	17	8	15
⁸⁷ Rb	0.6	0.4	0.6
²¹⁰ Po	3.0	21	3.0
²²² Rn	3.0	3.0	3.0
²²⁶ Ra	-	6.1	0.3
²²⁸ Ra	-	7	0.3
TOTAL	25	47	24

^aTable from A. W. Klement, Jr., C. R. Miller, R. P. Minx, and B. Shleien, Estimates of Ionizing Radiation Doses in the United States 1960-2000, U.S.E.P.A., ORP/CSD 72-1 (1972).

^bOther natural radionuclides would contribute to the exposure, but are such small fractions that they would not affect the totals within the accuracy of these estimates. As an example, doses from ³H are shown here.

^{239}Pu . These values are LAI_{ing} for members of the public divided by 365. Obviously, only radium contributes significantly to the average exposure of the human population.

No data are available about the concentration of the other transuranium elements in the human diet.

4.1 Terrestrial Ecosystems

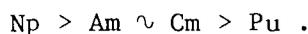
Radium, being an alkaline earth, has a behavior in biologic systems roughly similar to calcium, strontium, and barium; however, significant quantitative differences do exist, particularly with the lighter members of the group. Radium is adsorbed on soil particles to a considerably larger extent than calcium and is generally characterized by significantly lower biologic availability. Once radium has been taken up by plants, limited transfer takes place from the roots to the aerial parts, but the situation can be different in certain perennial species. For example, concentrations of ^{226}Ra as high as 2730 pCi/kg have been reported in Brazil nuts.¹⁰⁰ This plant is also known to concentrate barium and, to a lesser extent, lead. The high radium concentration in Brazil nuts is found in plants growing on soils with normal levels of radium.

Table 15 shows typical levels of ^{226}Ra in soil in various geographical regions. Assuming 1.4 as the average density of dry soil, and 1 pCi/g as the average activity concentration of ^{226}Ra , one can calculate that the ^{226}Ra content per square kilometer of land area to a depth of 30 cm would be 0.42 Ci.

The uptake of plutonium by plants has been studied experimentally.¹⁰¹⁻¹⁰³ The uptake is somewhat dependent on the plant species and on the type of soil, but the values are always very low; usually the leaf/soil concentration ratios are about 10^{-4} .

Cline¹⁰³ has observed an uptake of americium by beans and barley 20 to 30 times greater than the uptake of plutonium. Thomas and Jacobs⁷³ observed practically no uptake of curium by a common forage grass, tall fescue, but bean plants did show measurable curium concentrations in the aerial portion. Price⁷⁴ reported that the uptake by tumbleweed and

cheatgrass from Hanford soil occurred in the following order:



An interesting conclusion derived from these data is that transuranium elements are characterized by significant differences in biological behavior in soil-plant systems.

Table 15. Typical Concentrations of ^{226}Ra in Soil
Excluding High-Background Areas^a

Region	Soil (dry weight basis) (pCi/g)	Reference ^b
British Guiana	0.3 to 1.6	104
Czechoslovakia	0.1 to 3.8	105
Germany	0.15 to 1.3	106
Ireland	1.3 to 2.9	105
Italy (zone of Varese)	0.72 ± 0.11	95
United Kingdom	0.08 to 1.5	105,106
U.S.A.	0.8 to 2.8	105
U.S.S.R.	0.1 to 1.3	105

^aModification of a table from R. Scott Russell and K. A. Smith, "Naturally Occurring Radioactive Substances: The Uranium and Thorium Series," in Radioactivity and Human Diet, R. Scott Russell (ed.), Pergamon, Oxford, 1966.

^bRefers to references in text.

There is some indication that the uptake of plutonium by plants may increase with successive croppings and the passage of time. Possible explanations for the increase in plutonium uptake are either the increased contact between plutonium particles and roots with the development of the root system, or the increased complexing of plutonium due to organic matter resulting from decay of root tissue.¹⁰⁷⁻¹⁰⁹ Price¹¹⁰ has observed that when plutonium is added to the soil in the form of organic acid complexes, such as oxalate or citrate, it is taken up by plants more readily than when added as a dilute nitric acid solution.

Table 16 shows the ^{226}Ra and ^{228}Th content in various food items as reported by Turner et al.¹⁰⁰

Table 16. Activity of ^{226}Ra and ^{228}Th in Various Foods^a

Foodstuff	^{226}Ra (pCi/kg)	^{228}Th (pCi/kg)	$^{228}\text{Th}/^{226}\text{Ra}$	No. of Samples
Brazil nuts	2730 ^b	1200 ^b	0.4	4
Buttered Brazil nuts	520	170	0.3	1
Brazil nuts, chocolate	240	70	0.3	1
Marzipan Brazil nuts	140	100	0.7	1
Cereal 1 (U.K.)	68	61	0.9	1
Cereal 2 (U.K.)	62 ^b	46 ^b	0.7	3
Cockles	57 ^b	19 ^b	0.3	2
Tea (China)	51	51	1.0	1
Tea (India)	48	40	0.8	1
Tea (Paraguay)	43	40	0.9	1
Tea (Brazil)	40	36	0.9	1
Cereal 3 (U.K.)	25	22	0.9	1
Peanuts	18	9	0.5	1
Mussels	18.2	6.1	0.3	1
Peanut butter	9	17	2.0	1
Sardines (tinned)	6	2.4	0.4	1
Walnuts	3	3	1.0	1
Soya	nm ^c	5.2	-	1
Cereal 13 (U.K.)	nm ^c	5.0	-	1
Strawberry jam	2.4	1.9	0.8	1
Salami	2	2	1.0	1
Calf liver	1	2.5	2.5	1
Sausages	2 ^b	1.7 ^b	0.9	2
Cheese 2	1.5	1.5	1.0	1
Plaice	1.5	1.5	1.0	1
Egg	2	nm ^c	-	1
Pears (tinned)	1.1	0.6	0.5	1
Bacon	nm ^c	1.4	-	1
Tomato soup (tinned)	nm ^c	0.8	-	1

^aData from R. C. Turner, J. M. Radley, and W. V. Mayneord, "The Naturally Occurring α -Ray Activity of Foods," Health Phys. 1, 268-75 (1958).

^bMaximum value.

^cnm = nonmeasurable.

Comparison of the contents of ^{226}Ra and ^{239}Pu in the first 30 cm of soil with their respective dietary intakes seems to indicate that, at the present, the transfer from soil to man is achieved with roughly similar efficiencies. The values used are shown in Table 17. Obviously, calculations of this kind must be interpreted with the greatest caution since significant differences exist between the two elements. The primary potential sources of error are the different mechanisms of entry into the biosphere, the difference of distribution in the soil profile, and the fact that environmental plutonium is likely to be in a transient phase.

Table 17. Relationship Between Dietary Intake and Concentration in Soil of Radium and Plutonium

	Dietary Intake (pCi/day)	Content in Top 30 cm of Soil (mCi/km ²)	Intake/Soil Content ^a
^{226}Ra	2	420	1
^{239}Pu	0.007	2	0.74

^aTaken equal to 1 for radium.

Both radium and plutonium are bone-seekers; thus, the greatest fraction of activity absorbed by animals ends up in nonedible parts of the body, thereby reducing the transfer to man through flesh foods.

Ng et al.¹¹¹ have compiled a large mass of data relative to the transfer of radionuclides through the food chains. Table 18 shows some of the factors relative to the heavy elements. It must be observed that these data are meant to provide maximum possible doses; therefore, where no information was available, very conservative estimates have been tabulated. For example, the data relative to transplutonium elements are all default values. The concentration of plutonium in soil is based on the concentration of uranium, assuming a Pu/U ratio equal to 10^{-13} ; however, it is obvious that most of the plutonium in the top layer of soil is

Table 18. Factors Relevant to the Transfer of Heavy Elements in Terrestrial Foodchains^a

Element	Concentrations, C. (ppm) and Concentration Factors, C.F. in:						Fraction Secreted in Milk ^b
	Soil	Plants		Meat			
	C.	C.	C.F.	C.	C.F. (Soil)	C.F. (Plants)	
Pb	1.0×10	6.8×10^{-1}	6.8×10^{-2}	1.0×10^{-2}	1.0×10^{-3}	1.5×10^{-2}	6.2×10^{-4}
Po	1.0×10^{-11}	1.0×10^{-11}	1.0	1.0×10^{-9}	1.0×10^2	1.0×10^2	3.0×10^{-3}
Ra	8.0×10^{-7}	2.5×10^{-10}	8.3×10^{-4}	4.3×10^{-10}	5.4×10^{-4}	1.7	1.5×10^{-2}
Ac	1.0×10^{-10}	2.5×10^{-13}	2.5×10^{-3}	1.0×10^{-8}	1.0×10^2	4.0×10^4	5.0×10^{-6}
Th	6.0	2.5×10^{-2}	4.2×10^{-3}	6.0×10^2	1.0×10^2	2.4×10^4	5.0×10^{-6}
Pa	1.0×10^{-7}	2.5×10^{-10}	2.5×10^{-3}	1.0×10^{-5}	1.0×10^2	4.0×10^4	5.0×10^{-6}
U	1.0	2.5×10^{-3}	2.5×10^{-3}	5.0×10^{-3}	5.0×10^{-3}	2.0	5.0×10^{-4}
Np	1.0×10^{-12}	2.5×10^{-15}	2.5×10^{-3}	1.0×10^{-10}	1.0×10^2	4.0×10^4	5.0×10^{-6}
Pu	1.0×10^{-13}	2.5×10^{-17}	2.5×10^{-4}	1.0×10^{-11}	1.0×10^2	4.0×10^5	1.5×10^{-6}
Am	1.0×10^{-30}	2.5×10^{-34}	2.5×10^{-4}	1.0×10^{-28}	1.0×10^2	4.0×10^5	5.0×10^{-6}
Cm	1.0×10^{-30}	2.5×10^{-33}	2.5×10^{-3}	1.0×10^{-28}	1.0×10^2	4.0×10^4	5.0×10^{-6}
Bk	1.0×10^{-30}	2.5×10^{-33}	2.5×10^{-3}	1.0×10^{-28}	1.0×10^2	4.0×10^4	1.5×10^{-6}
Cf	1.0×10^{-30}	2.5×10^{-33}	2.5×10^{-3}	1.0×10^{-28}	1.0×10^2	4.0×10^4	1.5×10^{-6}

^aData from Y. C. Ng, C. A. Burton, S. E. Thompson, R. K. Tandy, H. K. Kretner and M. W. Pratt, Prediction of the Maximum Dosage to Man from the Fallout of Nuclear Devices. IV. Handbook for Estimating the Maximum Internal Dose from Radionuclides Released to the Biosphere, UCRL-50163, Part IV (1968).

^bFraction of daily intake secreted in 1 liter of cow milk.

accumulated by fallout. Assuming 2 mCi of ^{239}Pu per square kilometer uniformly distributed in the top 30 cm of soil, one obtains a concentration of 7.8×10^{-8} ppm, which is almost six orders of magnitude higher than the value in Table 18.

The grass-cow-milk pathway is more important for radium than for the other heavy elements. This is due to the greater absorption of radium through the gastrointestinal tract and the relative similarity of behavior between radium and calcium. It has been reported that the ratio of radium to calcium in milk is about 0.02 to 0.03 of that in the diet of the cow. The corresponding ratios for strontium and barium are 0.1 and 0.05, respectively.¹⁰⁴

The transfer of some transuranium elements from plasma to milk in the sheep, following a single intravenous administration, has been studied by McClellan et al.¹¹² The milk/plasma concentration ratios fall in the range 2 to 5 for americium and curium, and in the range 0.02 to 0.15 for uranium, neptunium, and plutonium; these values can be compared with the mean milk/plasma ratio for calcium of 35. The obvious indication of these data is that the transuranium elements show significant variability in their behavior in animals.

In conclusion, it can be said that the potential hazard associated with the heavy radionuclides in terrestrial environments is somewhat reduced by several factors: (1) the strong adsorption on soil particles; (2) the low solubility of most compounds; (3) the low uptake by plants (with few exceptions); and (4) the marked discrimination factors against transfer of these elements from one trophic level to the other.¹¹³

As a whole, radium should be characterized by a higher biologic availability than the actinides. The fairly high biologic availability of plutonium that was indicated in Table 17 is probably caused by the fact that an unknown fraction of the plutonium in the diet enters the foodstuff through atmospheric deposition.

The actinides are undoubtedly characterized by a certain similarity of behavior; however, on closer examination, important differences are revealed. Hence the assumption that the whole group is characterized by the same environmental behavior would lead to significant errors.

4.2 Aquatic Ecosystems

The contribution of aquatic foods chains to the total diet is quite variable. In the United States, aquatic organisms constitute a minor fraction of the diet. In other countries, such as Japan, the population relies on aquatic foods to a much greater extent. The dependence on sea foods can be exceptional for certain groups of people; for example, it is known that some fishermen eat as much as 1 kg of fish daily while at sea.

In general, aquatic organisms can concentrate trace elements with great efficiency. Many examples are known of environmental contaminants dispersed in aquatic ecosystems that in the higher trophic levels reach toxic concentrations. Aquatic foods are likely to be critical contributors to the intake of radionuclides whenever they represent a significant fraction of the diet.

Table 19 shows the concentrations (C.) of heavy elements in water and aquatic organisms and the relative concentration factors (C.F.).¹¹⁴ As in Table 18, many of the data in Table 19 are default values and should be taken as very tentative guesses.

The C.F. of thorium in marine fish that is reported in Table 19 is three orders of magnitude higher than for the other actinides. Why such a large difference in behavior should exist is not clear.

Actual measurements of the concentrations of heavy elements in aquatic organisms are rather scarce, but a few examples are mentioned below. The data obtained from the measurement of ^{239}Np in the Columbia River indicate significant association of the nuclide with suspended particles. While ^{239}Np has been found in the lower aquatic forms, significant amounts have not been detected in the higher trophic levels.¹¹⁵ The C.F. of ^{239}Np in green algae, sponge, and insect larvae in the Columbia River have been determined as ~ 300 , ~ 40 , and ~ 16 , respectively.¹¹⁶ The lack of ^{239}Np in fish cannot, however, be interpreted as proof of no uptake since a system of mobile organisms in flowing water that has variable concentrations, such as fish in the Columbia River, would be very unlikely to even approach equilibrium, at least for a short-lived nuclide such as ^{239}Np ($T_{1/2} = 2.3$ days).

Table 19. Concentrations of Heavy Elements in Various Compartments of Aquatic Ecosystems^a

Element	Concentrations, C. (ppm) and Concentration Factors, C.F. in: ^b													
	Seawater	Marine Plants		Marine Invertebrates		Marine Fish		Freshwater	Freshwater Plants		Freshwater Invertebrates		Freshwater Fish	
	C.	C.	C.F.	C.	C.F.	C.	C.F.	C.	C.	C.F.	C.	C.F.	C.	C.F.
Pb	1.00-3 ^c	1.00	1000(5000)	1.00	1000	3.00-1	300	5.00-3	1.00	200	5.00-1	100	1.50	300(100)
Po	5.00-15	1.00-11	2000(2000)	1.00-10	20,000(5000)	1.00-11	2000(300)	2.00-14	4.10-11	2000	4.00-10	20,000	1.00-12	50(500)
Ra	1.00-10	1.00-8	100	1.00-8	100	5.00-9	50	4.00-10	1.00-6	2500	1.00-7	250	2.00-8	50
Ac	4.00-15	2.00-11	5000	4.00-12	1000	1.00-13	25	1.37-15	7.00-12	5000	1.37-12	1000	3.40-14	25
Th	1.00-5	3.00-2	3000	2.00-2	2000	1.00-1	10,000	2.00-5	3.00-2	1500	1.00-2	500	6.00-4	30
Pa	5.00-12	3.00-11	6	5.00-11	10	5.00-11	10	1.77-12	2.00-9	1130	2.00-10	113	2.00-11	11
U	3.00-3	2.00-1	67	3.00-2	10	3.00-2	10	1.00-3	1.00	1000(0.5)	1.00-1	100(60)	1.00-2	10(2)
Np	5.00-15	3.00-14	6	5.00-14	10	5.00-14	10	2.00-15	2.00-12	1000(300)	8.00-13	400	2.00-14	10
Pu	2.00-11	7.00-9	350(1000)	2.00-9	100(200)	7.00-11	3.5(3)	1.00-10	3.50-8	350	1.00-8	100	3.50-10	3.5
Am	1.00-30	5.00-27	5000	1.00-27	1000	2.50-29	25	3.00-31	1.50-27	5000	3.00-28	1000	7.50-30	25
Cm	1.00-30	5.00-27	5000	1.00-27	1000	2.50-29	25	3.00-31	1.50-27	5000	3.00-28	1000	7.50-30	25
Bk	1.00-30	5.00-27	5000	1.00-27	1000	2.50-29	25	3.00-31	1.50-27	5000	3.00-28	1000	7.50-30	25
Cf	1.00-30	5.00-27	5000	1.00-27	1000	2.50-29	25	3.00-31	1.50-27	5000	3.00-28	1000	7.50-30	25

^aData from S. E. Thompson, C. A. Burton, D. J. Quinn, and Y. C. Ng, Concentration Factors of Chemical Elements in Edible Aquatic Organisms, UCRL-50564, Rev. 1 (1972).

^bValues of concentration factors are usually derived; values in parenthesis are measured.

^cRead as 1.00×10^{-3} .

The literature gives some C.F. values for radium higher than those reported in Table 19. For example, De Bortoli and Gaglione⁹⁵ have measured a C.F. as high as 228 in fish from Lake Maggiore, Italy. In the Animas River, Colorado and New Mexico, C.F. in algae and aquatic insects were as high as 1000.¹¹⁷

The available information on C.F. of plutonium in marine invertebrates and algae has been recently reviewed by Noshkin.⁷⁹ Table 20 shows the data, and proves that several marine organisms are characterized by a significant affinity for plutonium, particularly North Atlantic Sargassum, for which a mean C.F. of 21,000 has been measured. Hodge et al.⁹⁰ have measured concentrations of ²³⁹Pu in the liver of albacore tuna of the North Pacific of about 0.1 pCi/kg (wet weight).

On the basis of average contents in marine fish, Aten¹¹⁸ has estimated the following C.F. from sea water: radium = 15, uranium \leq 20, and plutonium = 13. Assuming 0.1 pCi/liter as the average concentration of ²²⁶Ra in sea water, the radium concentrations in fish reported by Muth et al.⁵⁴ would indicate concentration factors of 28, 40, and 63 in the flesh of green herrings, cod, and haddock, respectively. Turner's¹⁰⁰ data in Table 15 would give a C.F. of 60 in sardines (with bone) and 15 in plaice. The ²²⁶Ra concentrations measured in edible bivalves were significantly higher at 57 pCi/kg in cockles and 18.2 pCi/kg in mussels. The corresponding C.F. would be 570 and 182. The reliability of these calculated C.F. is quite low due to the possible variations of radium concentration in sea water and to the limited number of samples; however, the order of magnitude seems to be fairly well established.

The heavy radionuclides would thus appear to be characterized by significant potential mobility through aquatic food chains. The residence time in the water phase is different for the various elements under consideration, with radium and uranium having greater stability in solution in sea water than thorium and the transuranics. The biologic availability could be relatively high even for elements with short residence time in the liquid phase if removal is due to biological processes, or if recycling from bottom sediments takes place.

Table 20. Concentration Factors of ^{239}Pu in Marine Invertebrates and Algae^a

Organism	Tissue	Collection Date	Location	Mean Value	Range	Reference
Blue Mussel	Body	1970	Cape Cod	300 (7) ^b	250-350	118
Mussel	Body	1964	California coast	260 (2)	230-290	75
Brown Mussel	Body	1970	Cape Cod	340 (1)		119
Blue Mussel	Shell	1970	Cape Cod	490 (3)		119
Soft Shell Clam	Body	1970	Cape Cod	440 (1)		119
Oyster	Body	1970	Cape Cod	130 (2)	100-160	119
Scallop	Adductor	1970	Cape Cod	24 (2)	10-37	119
	Body	1970	Cape Cod	520 (3)	410-690	119
	Shell	1970	Cape Cod	600 (1)		119
Whelk	Body	1970	Cape Cod	140 (1)		119
	Shell	1970	Cape Cod	300 (1)		119
Moonshell	Body	1970	Cape Cod	660 (1)		119
	Shell	1970	Cape Cod	690 (1)		119
Starfish	Body	1970	Cape Cod	1020 (1)		119
Brittle Star	Body	1970	Cape Cod	760 (1)		119
Marine Worm	Body	1970	Cape Cod	4100 (1)		119
Sponge	Body	1970	Cape Cod	2100 (1)		119
Bivalves (mixed)	Body	1968	Danish inner waters	8235 (?) ^c		83
Crustacea	Body	1968	Danish inner waters	1765 (5)	1060-4471 ^c	83
Zooplankton		1961	Atlantic	2300 (1)		120
		1964	California coast	2590 (1)		75
Salpa		1958	Atlantic	1567 (3)	900-2400	120
Algae		1964	Pacific coast	1020 (4)	660-1570	75
		1971	Pacific coast	890 (19)	260-3500	120
		1970-1971	Cape Cod	620 (6)	100-1600	119
Sargassum Sp.		1965-1971	Atlantic	21000 (6)	3000-100,000	119
		1971	Pacific coast	390 (2)	325-450	119

^aData from V. E. Noshkin, "Ecological Aspects of Plutonium Dissemination in Aquatic Environments," Health Phys. 22, 537-49 (1972).

^bNumber of samples in parenthesis.

^cComputed using an average open ocean seawater value of 0.00085 pCi/kg.

4.3 Resuspension

Tables 5-8 show clearly that the hazard potential associated with the actinides present in high-level solid waste is several orders of magnitude higher if the intake is assumed to occur through inhalation. One of the main factors to be considered in any environmental hazard evaluation is the probability of the activity being inhaled; or, more precisely, the fraction of activity that would become airborne if dispersal of the radionuclides into the environment were to occur.

The natural radioactivity in air is due mainly to radon and its daughters. This would seem to confirm that resuspension of natural radioactive elements is a fairly inefficient mechanism when compared to the loss of radon from the lithosphere.

Suspended particles in the atmosphere can have a cosmic or terrestrial origin, but the greatest fraction is derived from the earth. The terrestrial aerosols are generated by (1) the action of the wind on land and water, (2) biologic processes, (3) volcanic activity, (4) forest and brush fires, (5) the action of man, and (6) the emission of radon. Briefly, the prime factors contributing to the formation of atmospheric aerosols are:

1. Suspension of marine salts; aerosols of marine origin are characterized by a chemical composition related to the dissolved solids in sea water and by particles of variable size. The main resuspension mechanisms from the surface of large bodies of water are wave breaking and the bursting of small gas bubbles.^{121,122} The aerosols formed by this last process have a chemical composition related to the surface film from which they originate. Generally, the surface film does not differ significantly from the composition of the body of water; however, certain pollutants (e.g., pesticides, detergents, oil) can be greatly concentrated in the surface film and preferentially transferred to the atmosphere.
2. Wind erosion of soil materials; most eroded matter is in the form of large particles (>20 μm).

3. Automobile traffic, home heating, industrial activities, fires; combustion processes produce particles of small size and can be the most important source of local atmospheric particulates.
4. Volcanic activity; particles of variable size are produced. Strong volcanic eruptions have been proposed as a significant source of stratospheric aerosols,¹²³ but they are intermittent; on a global basis, they should not contribute more than the continuous volcanic emanations.

Hidy and Brock¹²⁴ have reviewed the global sources of tropospheric aerosols, classifying them as primary and secondary. Primary particles are those found in the atmosphere in the same form in which they were produced (e.g., dust, smoke, sea spray, etc.). Secondary aerosols are those produced in the atmosphere by chemical reactions, for example, the particles produced by reactions involving hydrocarbons, and sulfur and nitrogen compounds emanated by vegetation or by the decay of organic matter. The production of secondary aerosols has been reviewed by Robinson and Robbins.¹²⁵ Table 21 summarizes the most important sources of atmospheric aerosols and estimates their relative contributions to the total.

The size of the particles controls their fate in the atmosphere; if they are larger than 10 μm , removal by sedimentation is quite rapid, whereas particles smaller than 1 μm can remain airborne for relatively long times. The smaller particles can reach high altitudes, and will eventually achieve worldwide distribution if they enter the stratosphere.

Bagnold,¹²⁶ Chepil and Woodruff,¹²⁷ and the Committee on Sedimentation of the American Society of Civil Engineers¹²⁸ have reviewed the physics of erosion and transport of soil particles by the wind. Chepil¹²⁹ had defined the relative erodibility of soil particles as a function of their size, as shown in Table 22.

Sand, silt, and clay particles have diameter ranges of 1000 to 50 μm , 50 to 2 μm , and less than 2 μm , respectively. Therefore, grains of sand and silt size are much more subject to erosion and transport by wind than are clay particles, which are generally aggregated to form large clods or adhere to larger grains.¹³⁷ A dislodged particle moves downwind by

Table 21. Important Sources of Atmospheric Aerosols and Worldwide Production^a

Source	Production rate (tons/day)	Max. % of total	Reference ^b
A. Natural			
1. Primary			
Sea spray	3×10^6 ^c	28	130,131 121,132
Wind erosion	2×10^4 to 10^6	9.3	133
Cosmic dust	50 to 550	0.005	134
Volcanic dust	10^4	0.09	124
Forest fires	4×10^5	3.8	124
2. Secondary			
Vegetation (hydrocarbons-terpenes)	5×10^5 to 3×10^6	28	135
Sulfur cycle ($\text{H}_2\text{S} \rightarrow \text{SO}_4^{2-}$)	10^5 to 10^6	9.3	136
Nitrogen cycle ($\text{NO}_x \rightarrow \text{NO}_3^-$)	10^6	9.3	136
(ammonia)	7×10^5	6.5	136
Volcanoes (volatiles, SO_2 , H_2S)	10^3	0.009	124
Subtotal: Natural	10.1×10^6	94	
B. Anthropogenic			
1. Primary			
Combustion and industrial Dust rise by cultivation	1×10^5 to 3×10^5 10^2 to 10^3	2.8 0.009	124 124
2. Secondary			
Hydrocarbons	7×10^3	0.065	136
Sulfates	3×10^5	2.8	136
Nitrates	6×10^4	0.56	136
Ammonia	3×10^3	0.028	136
Subtotal: Anthropogenic	6.7×10^5	6	
TOTAL: All sources	10.8×10^6	100	

^aData from G. M. Hidy and J. R. Brock, "An Assessment of the Global Sources of Tropospheric Aerosols," Second Int. Clean Air Congress, Washington, D.C., December 6-11, 1970, H. M. Englund and W. T. Beery, Eds., pp. 1088-97, Academic, New York, 1971.

^bRefers to reference numbers in text.

^cAbout 90% of this is redeposited on the sea.

Table 22. Relative Erodibility of Soil Particles According to Size^a

Particles Diameter (μm)	Relative Erodibility
$d < 20$	Nonerodible except at wind speeds in excess of 80 km/hr at 15 cm above ground
$20 \leq d < 50$	Difficultly erodible
$50 \leq d < 500$	Highly erodible
$500 \leq d < 1000$	Difficultly erodible
$1000 \leq d$	Nonerodible except at wind speeds in excess of 80 km/hr at 15 cm above ground

^aTable from J. W. Healy and J. J. Fuquay, "Wind Pickup of Radioactive Particles from the Ground," Proc. U. N. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva, 1958, 18, 291-95 (1958).

suspension, surface creep, or saltation. When saltating particles fall back to the ground, the impact can disaggregate clay-size particles and cause their resuspension.¹³⁸ Table 23 shows that small particles, once they become suspended, can be carried to great distances and remain in the atmosphere for long periods of time.

Table 23. Maximum Theoretical Values for Time of Flight, Range, and Height of Rise of Particles in the Atmosphere for a Wind of 55 km/hr^a

Particles Diameter (μm)	Velocity of Fall (cm/sec)	Time of Flight	Range	Height
1	0.00824	8 to 80 years	4×10^6 to 40×10^6 km	6 to 60 km
10	0.824	7 to 70 hr	400 to 4000 km	60 to 600 m
100	82.4	3 to 30 sec	40 to 400 m	0.6 to 6 m

^aModification of a table from Committee on Sedimentation, "Sediment Transportation Mechanics: Wind Erosion and Transportation," J. Hydraulics Div. Am. Soc. Civil Eng. 91, 267-87 (1965).

Wind erosion is a complex geologic process controlled by many factors, such as wind intensity, nature of the ground, surface roughness, status of aggregation and moisture content of the soil, vegetation, etc. An example of a region characterized by fairly intense wind erosion is the Great Plains. Most erosion takes place during the brief and violent duststorms that are frequent in the area. Hagen and Woodruff¹³⁹ have reported that long-term mean concentrations of particulate matter in air at Great Plains locations range between 10 and 140 $\mu\text{g}/\text{m}^3$. Much higher values are observed during duststorms. For example, in March 1963, a concentration of 843 $\mu\text{g}/\text{m}^3$ was recorded in Ward County, North Dakota. From the relation between dust concentration and visibility, the median dust concentration during duststorms in the 1950's was 4.85 mg/m^3 , whereas in the driest area of the southern Great Plains the average concentrations during duststorms have

been estimated at 10 to 15 mg/m³. These are very high levels of dust concentration; however, the mass median diameter (MMD) of suspended particles was about 50 μm, and deposition was quite rapid when the wind subsided.

Gillette et al.¹⁴⁰ have measured the size distribution of aerosols originating from farmland, and have found a strong similarity to the size distribution of the particles in the soil. They also concluded that soil dust was the dominant aerosol source in the rural area where their observations were conducted, but that most of the suspended matter arose far from the study area. Apparently, under normal meteorological conditions, the erosion of local soil represents a relatively minor contribution to the total atmospheric aerosols in the area.^{124,133}

Lee¹⁴¹ has reported on the size distribution of particulate matter in several urban environments. Since most of the dust suspended in city air is due to human activity, these data cannot be extrapolated to conditions of natural resuspension. Table 24 shows the size distribution of particulate matter in air in six U. S. cities, at three U. K. sites, and in Ankara, Turkey. The Eskdalemuir Observatory in southern Scotland is the only site with relatively unpolluted air. With the exception of Ankara, the average MMD of suspended particles is significantly less than 1 μm. An additional interesting conclusion is that the average size of the dust particles is directly proportional to the total amount of suspended matter; aerosols in unpolluted areas are thus generally characterized by a much smaller average MMD. Lee has also observed that significant growth of aerosol size takes place during periods of temperature inversion, and that aggregation primarily affects the smaller particles.

The size distribution of atmospheric aerosols has important health implications, since the pulmonary deposition of suspended particles is inversely proportional to their size. The Task Group on Lung Dynamics of ICRP¹⁴² has indicated that pulmonary deposition of particles larger than 10 μm is practically negligible. The pulmonary deposition increases from about 20% to 70 to 80% of inhaled aerosol as the MMD decreases from 1 to 0.01 μm. Aerosol particles smaller than 0.01 μm are very unstable due to aggregation processes and are not considered important.

Table 24. Size Distribution of Particulate Matter Suspended in Air^a

Locality	No. of Samples	Average Concentration ($\mu\text{g}/\text{m}^3$)	Average MMD ^b (μm)	Average Geometric S.D. ^c	Average Mass of Particulate (%)	
					$\leq 1 \mu\text{m}$	$\leq 2 \mu\text{m}$
<u>United States^d</u>						
Chicago	21	86.5	0.76	8.18	55	68
Cincinnati	18	74.3	0.70	5.49	59	74
Denver	21	59.7	0.40	10.50	65	75
Philadelphia	20	58.5	0.47	5.65	67	80
St. Louis	22	73.1	0.83	6.80	54	68
Washington, D.C.	23	56.3	0.46	5.22	68	81
<u>United Kingdom^e</u>						
London	28	95.8	0.45	8.24	65	76
Kew Observatory	33	62.7	0.36	5.87	72	84
Eskdalemuir Observatory	35	41.3	0.14	10.63	82	89
<u>Turkey^f</u>						
Ankara	10	115.9	1.79	5.20	37	53

^aData from R. E. Lee, Jr., "The Size of Suspended Particulate Matter in Air," Science 178, 567-75 (1972).

^bMMD = mass median diameter.

^cS.D. = standard deviation; the geometric standard deviation, which is a measure of the particle dispersion, is estimated from the ratio of the 84th percentile to the 50th percentile.

^dSamples collected over a 1-year period, January to December, 1970.

^eSamples collected over a 4-month period, February to May, 1970.

^fSamples collected over a 10-day period, April 26 to May 5, 1971.

Hamilton¹⁴³ has reported some measurements of uranium concentrations in air at Sutton, United Kingdom, in the North and South Atlantic Ocean, and in Antarctica. The observed mean concentrations of uranium in air are shown in Table 25.

Table 25. Uranium Concentrations in Air^a

Locality	Uranium (10^{-12} g/m ³)
Sutton, United Kingdom	62 ± 78
N. Atlantic Ocean	4.1 ± 1.2
S. Atlantic Ocean	2.2 ± 0.5 ^b
Antarctic Pack Ice	1.2 ± 0.3 ^c
Antarctic Base Roi Baudouin	3 ± 1 ^d

^aData from E. I. Hamilton, "The Concentration of Uranium in Air from Contrasted Natural Environments," Health Phys. 19, 511-20 (1970).

^bMean for 5 samples; no uranium found in 4 samples.

^cMean for 3 samples; no uranium found in 15 samples.

^dMean for 18 samples; no uranium found in 2 samples.

The surface lithology of the Sutton area is characterized by chalk overlain by a fairly thin layer of soil. The average uranium content of the chalk and of the soil is 0.47 ± 0.13 and 1.12 ± 0.41 ppm, respectively, which is significantly less than in most geologic materials. However, the correlation of the uranium concentrations in soil and air is uncertain for an urban environment such as Sutton due to the addition of significant amounts of artificial particulates. The average concentration of dust in the air at Sutton was 59 ± 32 $\mu\text{g}/\text{m}^3$, with 1.03 ± 1.01 ppm of uranium. Examination of the dust particles has revealed the presence of carbon debris, chalk particles, organic structures, brick debris, iron oxides,

and occasionally, spherules of silicate glass, which are listed in the order of abundance. It is interesting that the uranium concentration in atmospheric dust is about the same as in the local soil. If this coincidence were observed in open country it would seem to prove that no discrimination against uranium occurs during resuspension; in Sutton, however, this is probably due to the low uranium content of British coal (1.41 ± 0.53 ppm of uranium).

Thorium measurements in air in seven nonurban sites in the United Kingdom have been reported by Peirson et al.¹⁴⁴ At Wraymires, which is located in the vicinity of Lake Windermere in the Lake District, England, the average particulate concentration in air in 1971 was about $25 \mu\text{g}/\text{m}^3$, with a thorium mean concentration of $160 \times 10^{-12} \text{ g}/\text{m}^3$, or 6.4 ppm, which is about the average thorium concentration in soil.^{105,145} Data for the other six sites are reported for only the first quarter of 1972, during which thorium concentrations ranged between $<37 \times 10^{-12}$ and $160 \times 10^{-12} \text{ g}/\text{m}^3$ of air. Resuspension of soil dust is the source of airborne thorium in these relatively undisturbed areas; unfortunately no data were reported on the thorium content of the soil in the Lake Windermere area.

The thorium concentrations in air and in suspended aerosols in the San Francisco Bay area are shown in Table 26.¹⁴⁶ The analysis of the data shows thorium belonging to the soil-derived group of elements. The concentration of thorium in suspended matter is lower than in the British study by a factor ranging from 2 to 6; this could be due to the dilution of soil-derived particles with anthropogenic aerosols or with particles of marine origin. The mean concentrations of thorium in sea water and in sea salt are 5×10^{-5} ppm, and 1.5×10^{-3} ppm, respectively.^{121,147} Consequently, aerosols of marine origin are practically thorium-free.

Rahn and Winchester¹⁴⁸ have studied the chemical composition of the atmospheric aerosol in various North American continental regions. The soil-derived particles were of fairly large size and of refractory nature. The elements found in the soil-derived large particles show aerosol/soil concentration ratios nearly equal to those of iron, an element abundant in soil. The spread of the particle size distribution in the soil-derived fracture was less than for the aerosol particles of different origin.

Table 26. Thorium Concentrations in Air in the San Francisco Bay Area^a

Locality	Total Particulates ($\mu\text{g}/\text{m}^3$)	Th in Air ($10^{-12} \text{ g}/\text{m}^3$)	Th in Particulates (ppm)
Pittsburg	104	320 (40) ^b	3.1
Richmond	62	140 (30)	2.3
San Rafael	43	100 (30)	2.3
San Francisco	59	60 (20)	1.0
Redwood City	58	120 (30)	2.1
San Jose	80	170 (30)	2.1
Fremont	120	360 (40)	3.0
Livermore	96	260 (40)	2.7

^aData from W. John, R. Kaifer, K. Rahn, and J. J. Wesolowski, "Trace Element Concentrations in Aerosols from the San Francisco Bay Area," *Atmos. Environ.* 7, 107-18 (1973).

^bAnalytical errors are given in parentheses.

The concentration of soil-derived particles tended to be lower in winter than in summer, probably caused by the greater dryness of the land in the warm season, and the snow cover in the winter. On closer observation, a correlation with local wind velocities was also noticeable for the soil-derived particles. Table 27 shows some thorium concentrations in air at U. S. and Canadian locations.

On the basis of this admittedly scant evidence, it can be tentatively assumed that no significant fractionation takes place during wind erosion and dust resuspension under natural conditions. The result is that the concentration of radium and actinides in the soil-derived fraction of atmospheric aerosol would be of the same order of magnitude as in the exposed surface material. This conclusion would also appear to be supported by Chepil's observation that density is not a critical factor in the resuspension of particles smaller than about $50 \mu\text{m}$.¹⁴⁹

Table 27. Thorium Concentrations in Air in Continental Areas of North America^a

Locality	Date	Thorium in Air (10^{-12} g/m ³)	Aerosol/soil (Normalized Enrichments ^b)
Markstown Park, E. Chicago, Ind.	Feb. 1970	270	--
Central Fire Station, E. Chicago, Ind.	Feb. 1970	230	--
Ann Arbor, Mich.	Feb. 1970	97	--
Mackinac Island, Mich.	Feb. 1970	29	--
Niles, Mich.	Summer 1970	110	0.72
Mackinac Island, Mich.	Summer 1970	18	0.45
Algonquin, Ont.	Summer 1970	56	1.2
Riding Mountain, Man.	Summer 1970	58	1.4
Prince Albert, Sask.	Summer 1970	40	1.4
Jasper, Alta.	Summer 1970	36	1.0
Twin Gorges, N. W. Territories	Summer 1970	52	4.6
Mackinac Island, Mich.	Winter 1970	28	--
Algonquin, Ont.	Winter 1970	7.1	--
Twin Gorges, N. W. Territories	Winter 1970	30	--
Northwest Ind., ^c Industrialized	Summer 1969	570	--
Northwest Ind., ^c Semirural	Summer 1969	350	--

^aTable from K. A. Rahn and J. W. Winchester, Sources of Trace Elements in Aerosols - An Approach to Clean Air, COO-1705-9 (1971).

^bThe normalized enrichment factor is the aerosol/soil concentration ratio for the element under consideration divided by the aerosol/soil concentration ratio for iron.

^cThe Northwest Indiana area is the Hammond - East Chicago - Gary - Whiting metropolitan complex, one of the nation's most heavily industrialized areas.

In the case of surfaces contaminated by the deposition of radioactive particles, the situation is completely different (at least in the initial period), and the evaluation of the potential for resuspension could not be based on geochemical considerations. Healy¹⁵⁰ has clearly summarized the situation, describing the two limiting conditions of plutonium freshly deposited at the surface and uniformly mixed with the soil. In the first case, the resuspension will be a function of the amount of plutonium per unit area. In the second case, the amount resuspended is a function of the soil dust picked up from the surface; hence the concentration in the soil would seem to be the controlling factor.

Langham¹⁵¹ has reviewed the problem of plutonium resuspension, and has concluded that the resuspension factor (R_f) is dependent on so many interrelated ill-defined variables that it is currently impossible to produce an analytical evaluation of exposure by resuspension. Some of the factors affecting resuspension are the nature of the contaminated surface (soil type, vegetation, etc.), micrometeorology (wind velocity, turbulence, rainfall, etc.), and the amount of physical activity (action of animals, traffic, plowing, etc.). In addition, the fraction of activity available for resuspension changes with time as a function of many factors, such as the status of aggregation of soil particles, water content, migration along the soil profile, etc.

Some values of resuspension factors for plutonium, expressed as

$$R_f \approx \frac{\text{Air Concentration } (\mu\text{g}/\text{m}^3)}{\text{Surface Deposition } (\mu\text{g}/\text{m}^2)}$$

have been measured by Langham. A R_f of $7 \times 10^{-5}/\text{m}$ was found to apply to disturbed Nevada desert conditions; equilibrium calculations applicable to dusty rural areas gave a value of $7 \times 10^{-6}/\text{m}$. A R_f of $7 \times 10^{-7}/\text{m}$ is reported for undisturbed desert conditions. The corresponding surface contamination values considered tolerable for lifetime occupancy with normal activity are 0.7, 7.0, and 70 $\mu\text{Ci}/\text{m}^2$, respectively. Langham¹⁵¹ stated that 10^{-6} can be considered as a reasonable average value of the R_f to use in the assessment of the inhalation hazard associated with occupancy of a plutonium-contaminated area. However, the variability of

this factor is extreme since values ranging from about 10^{-2} to 10^{-11} have been reported.

Jordan¹⁵² has discussed the Palomares accident of January 17, 1966, in which a refueling mishap involved a B-52 bomber armed with nuclear weapons. The triggering devices of two thermonuclear weapons detonated upon impact with the ground, and uranium and plutonium were dispersed as finely divided oxide particles over an area of about 500 hectares. The radii of the areas with plutonium contamination in excess of $32 \mu\text{Ci}/\text{m}^2$ were 80 m in one case and 65 m in the other. The cleanup procedure consisted of scraping and removing the top layer of soil where the contamination exceeded $32 \mu\text{Ci}/\text{m}^2$. All areas with contamination levels between 32 and $0.32 \mu\text{Ci}/\text{m}^2$ were plowed to a depth of 40 cm. The plowing obviously had the effect of increasing the immediate resuspension and decreasing the long-term hazard by dilution and by accelerating migration through the soil profile.

Subsequent monitoring performed by the Spanish authorities failed to show either dangerous air concentrations or significant plutonium uptakes on the part of the local inhabitants;¹⁵³ however, resuspension of plutonium was measurable at all four sampling stations. Plutonium concentrations in air that exceeded the concentration limits of soluble compounds for members of the public were recorded on only two occasions. Both cases were at a station located in an area contaminated at levels between 3.2 and $0.32 \mu\text{Ci}/\text{m}^2$ but surrounded by farmland which suffered higher levels of contamination. The highest plutonium concentrations in air were always observed in coincidence with wind velocities higher than 35 km/hr. The average concentrations of plutonium in air at the station with the highest levels were $1.2 \times 10^{-15} \mu\text{Ci}/\text{cm}^3$ in 1966 and $1.2 \times 10^{-14} \mu\text{Ci}/\text{cm}^3$ in 1967. Assuming an average surface contamination in the surrounding area of about $3.2 \mu\text{Ci}/\text{m}^2$, an R_f on the order of 10^{-9}m^{-1} can be calculated. Caution is required before using R_f values determined for small contaminated areas, since it is known that the greatest fraction of suspended matter is usually derived from sources removed from the immediate vicinity; therefore, local resuspension factors could be greatly underestimated.

Additional data on the resuspension of plutonium particles are provided by the study of areas contaminated in less dramatic fashion (e.g., the vicinity of the Rocky Flats plutonium processing plant, and the

already mentioned Nevada Test Site). Volchok¹⁵⁴ has reported that plutonium air concentrations near Rocky Flats are more than an order of magnitude greater than at Denver, New York, and Ispra. In addition, the variations are not related to the seasonal trend of worldwide fallout, but are a function of local wind intensities. On the basis of the plutonium concentration in Rocky Flats soil reported by Krey and Hardy,⁶⁷ an R_f of about $10^{-9}/m$ has been calculated by Volchok.¹⁵⁴

The concepts of surface contamination and resuspension factors are probably meaningless in a discussion of long-term hazards associated with high-level wastes, since the most likely mechanism of release of activity from the disposal formation involves leaching and transport by ground water. In any event, the transfer of long-lived nuclides from the perfect containment, which prevails as long as the disposal formation is not breached to the condition of biologic availability discussed here, will be accomplished by slow geologic processes. This will inevitably result in significant environmental dispersion of the radionuclides, and in thorough mixing with geologic materials.

In order to evaluate an upper limit of the inhalation hazard, it can be assumed that no dispersion takes place, and that the waste becomes exposed at the surface of an area equal to the original repository - for example, 7.8 km^2 . After 100,000 years of decay the number of LAI by inhalation per cubic centimeter of waste would be 20,000 and 73,000, respectively, for LWR and LMFBR waste. If the waste were originally a glass with a density of 2.7 g/cm^3 , the number of LAI per gram would be 7400 and 27,000, respectively, and twice that much if the original solid was a calcine. Assuming a mean particulate concentration in air over the exposed waste of $100 \text{ } \mu\text{g/m}^3$, and a standard breathing rate of $7000 \text{ m}^3/\text{year}$, the amount of dust inhaled annually by an individual living at the site on a permanent basis would be 0.7 g. It is also necessary to assume that during disinterment the ratio between the areas of the waste and the total repository on a horizontal section would be roughly preserved at 1:2000.

With this set of fairly extravagant assumptions, it can be calculated that the intake by inhalation could fall in the range 4 to 50 LAI. Considering the conservatism of the various assumptions, namely: (1) continuous occupancy of the site; (2) no dilution of the suspended particles

by unpolluted materials; (3) no dispersion of the waste during transfer to the surface, and (4) no allowance for the large average size of soil-derived particles, it can be intuitively concluded that if the waste containment were to fail sometime in the neighborhood of 100,000 years after disposal, the inhalation hazard would be relatively limited. In case of exhumation of the waste with little environmental dispersion, the inhalation hazard would be higher but restricted to a small area; if extensive dispersion into the environment were to take place, as would probably be the case, the inhalation hazard would be reduced to very low levels. It should be emphasized that the considerations concerning resuspension and inhalation hazards are necessarily quite vague because of many uncertainties relative to waste disinterment and resuspension mechanisms.

4.4 High-Background Areas

High-background areas provide natural laboratories for the study of equilibrium conditions resulting from the existence of higher-than-normal environmental levels of long-lived alpha-emitters. Many localities are characterized by unusually high levels of background radiation. In most cases, the available information is limited to the levels of external radiation; few data exist concerning the levels of activity in the food-stuffs. It would be useful, however, to analyze the actual concentrations of radium, thorium, and uranium in various environmental compartments, and the relative exposure of the people living in the area.

The already mentioned high-background areas in India and Brazil are famous and have been studied extensively. Two types of high-background regions have been investigated in Brazil. The first is the area of monazite sands on the Atlantic Coast, including the town of Guarapari and the fishing village of Meaípe. The second is an area of alkaline intrusive rocks in the state of Minas Gerais, where the town of Araxá and the village of Tapira are located.

The monazite sands are typical placer deposits caused by the high density and physicochemical resistance of monazite, and by the interaction

of river and ocean currents. The source of monazite is in the Precambrian gneiss outcropping in the mountain range that parallels the coast. The monazite sand is usually black due to the association of monazite and ilmenite and is, therefore, easily recognizable. Levels of external radiation on the spots of black sand can be as high as 2 mR/hr.^{13,155} Small quantities of food are produced on the monazite sand deposits, but their contribution to the total diet of the local population is very limited. The levels of activity in food grown in the area are somewhat higher than normal, although the total intake of alpha emitters is not significantly high.^{44,156,157} This is due both to the dilution of locally produced foods with imported foodstuffs and to the low biologic availability of the radionuclides contained in monazite.

In the Araxá-Tapira area the mineralization consists mainly of apatite; the radioactive nuclides are mostly contained in the mineral pyrochlore, which contains as much as 60% niobium oxide, 1.9% thorium oxide, 1.3% uranium oxide, rare earths, titanium, zirconium, and vanadium. Weathering of the magmatic rock has produced a fairly radioactive soil; this is characterized by a fertility directly proportional to the content of apatite and, therefore, to the level of radioactivity. The main activities in the area are cattle raising and the manufacture of cheese; however, some farming also takes place, particularly on patches of soil naturally fertilized by the presence of apatite.¹⁵⁵ The levels of ^{226}Ra , ^{228}Ra , and ^{228}Th in crops grown in the Araxá-Tapira area were as much as 100 times higher than normal.¹⁵⁷ Manioc and its flour (the staple foods in the area), potatoes, citrus fruits, and some leafy vegetables contained the highest levels of activity; concentrations of ^{228}Ra and ^{226}Ra reached 2720 pCi/kg and 81 pCi/kg (fresh weight), respectively.¹⁵⁷

Up to about 75% of the total intake was from root and tuber vegetables (yam, taro, manioc, potatoes) with green vegetables, beans, and fruits being responsible for the balance. Despite the fact that some Brazilian mineral waters have a very high content of radium - for example, a sample of water from Caxambu, Minas Gerais, contained 131 pCi of ^{228}Ra and 79 pCi of ^{226}Ra per liter - in the Araxá-Tapira area, the contribution of water to the intake has been estimated as negligible.¹⁵⁵

The highest estimated daily intakes in the region were 240 pCi of ^{228}Ra and 40 pCi of ^{226}Ra . No actual measurements of radium in the soil or in the people living in the area have been reported. Penna Franca et al.¹⁵⁷ have estimated the highest body burdens in members of the high intake group as 3360 pCi and 560 pCi of ^{228}Ra and ^{226}Ra , respectively.

Several localities on the Kerala coast in southwest India are characterized by high values of background radioactivity. Most of the activity is due to the thorium series, since the radioactive deposits are monazite-bearing sands.¹⁵⁸ Mistry et al.^{159,160} have measured the levels of radioactivity in various foods, and some of these results are shown in Table 28. The dietary intake of ^{228}Ra has been estimated as 162 pCi/day, of which roots and tubers and flesh foods account for a very large fraction; the intake through ingestion of water is practically negligible.

The ^{228}Ra intake in the Kerala monazite area is significantly higher than in the Guarapari region of Brazil, reflecting the greater dependence of the Indian population on local foodstuffs. Unfortunately, insufficient data are available to answer the question concerning possible differences in the biologic availability of the radioactive elements.

Gopal-Ayengar et al.¹⁶¹ have reported on a dosimetric survey in the high-radiation region of the Kerala coast. Of the 70,000 individuals living in the study area, about 16,600 are probably receiving a dose higher than 500 mR/year. Of the population sample actually provided with dosimeters, 8.8% was exposed to doses exceeding 1 R/year and 1.1% to doses higher than 2 R/year. The analysis of the demographic data of the exposed population generally indicated no statistically significant variations between different-exposure-level groups with respect to fertility index, sex ratio among offspring, infant mortality rate, pregnancy terminations, multiple births, and gross abnormalities. However, the lowest fertility index and the highest infant mortality were recorded for a group of couples exposed to doses in excess of 2 R/year; the total loss of offspring in this group was significantly higher than in those receiving lower radiation doses.

This review of the available information on some high-background areas appears to confirm the low biologic availability of the long-lived natural alpha-emitters. As could be predicted, radium is the critical

Table 28. Gross Alpha and ^{228}Ra Levels
in Various Foods Collected on the Kerala Coast^a

Foodstuff	No. of Samples	Weight Factor ^b	Gross Alpha on Dry Weight Basis (pCi/kg)			^{228}Ra on Dry Weight Basis (pCi/kg)		
			Min.	Max.	Mean	Min.	Max.	Mean
Rice	6	0.87	20	130	80	<10	<10	<10
Tapioca	16	0.40	<10	1020	420	<10	920	290
Okra	6	0.11	880	3190	1780	<10	1990	1020
Coconut	6	0.63	<10	60	20	30	160	90
Plantain	12	0.39	<10	1770	340	<10	1130	250
Fish	8	0.20	1260	11,160	5810	<10	5980	2850
Milk	4	0.20	<10	<10	<10	<10	3060	1530
Chili powder	8	0.89	<10	400	140	120	2710	850
Water ^c	6		<10	800	350	<10	<10	<10

^aData from K. B. Mistry, K. G. Bharathan, and A. R. Gopal-Ayengar, "Radioactivity in the Diet of Population of the Kerala Coast Including Monazite Bearing High Radiation Areas," Health Phys. 19, 535-42 (1970).

^bWeight factor = $\frac{\text{dry weight}}{\text{fresh weight}}$.

^cValues in pCi/liter.

element and is more available than uranium and thorium. Unfortunately, the relevance of these data to the hypothetical dispersal of waste nuclides into the environment is somewhat reduced by the lack of reported concentration measurements in soils.

4.5 Uranium Mill Tailings

The operation of uranium mills results in the accumulation of large volumes of solid wastes that offer interesting examples of surface materials containing unusual amounts of long-lived alpha-emitters. In addition to the solid wastes, uranium mills produce liquid effluents that contain significant amounts of ^{226}Ra .

Of all the radium contained in the ore, only a small fraction is mobilized during the separation of uranium. It has been estimated that from less than 1% to about 2% of the radium - according to the leaching process used - becomes dissolved in the liquid effluents; most of the radium remains in the ore tailings which are usually dumped in settling ponds or in large piles.¹⁶²⁻¹⁶⁴ The tailings piles do not constitute an effective containment, however, since leaching and wind erosion continue over the years and disperse the activity into the environment.

The levels of environmental contamination caused by the presently inactive uranium and vanadium mill located at Durango, Colorado, have been studied extensively. Before 1959, this mill discharged untreated effluents to the Animas River. Surveys performed by the Public Health Service in 1958 and 1959 revealed significant levels of environmental contamination and led to the introduction of waste treatment.^{117,165}

In the summer of 1958, river water below the mill averaged 12.6 pCi of radium per liter in proximity of the plant and 2.9 pCi/liter 100 km downstream; radium levels upstream from the plant were between 0.3 and 0.6 pCi/liter. River mud had radium concentrations up to 600 pCi/g (dry weight) compared with a background level of about 1.6 pCi/g, and a concentration in the tailings of about 900 pCi/g. Fish above the uranium mill had an average radium concentration of 0.5 pCi/g (ash) compared to 14.4 pCi/g below the mill. Radium concentrations in algae ranged from 2.8 pCi/g (ash) upstream to 530 pCi/g downstream in the summer of 1958,

and up to 880 pCi/g 3 km below the mill in the spring of 1959. Aquatic insects appeared to contain amounts of radium similar to the algae.¹⁶⁴ The resulting C.F. were 500 to 1000 for algae and aquatic insects, 3 to 4 for fish flesh, and about 100 for fish skeletons.

The Animas River represents the water supply for the populations of Aztec and Farmington in New Mexico (6,000 and 22,000 people, respectively). In addition, approximately 2,000 farmers in the area use the raw river water for both domestic purposes and the irrigation of about 26,000 acres of farmland.

The daily intakes of ^{226}Ra were estimated from the data collected in the 1958 and 1959 surveys as follows: (1) for the farmers using raw river water - 21.2 pCi, of which 21% was ingested with food and 79% with water; (2) for the inhabitants of Aztec - 12.4 pCi, of which 64% was ingested with the water; and (3) for the inhabitants of Farmington - 10.2 pCi, of which the fraction due to ingestion of water had decreased to 56%.¹⁶⁶

In summary, the above-mentioned surveys have shown that a sizable group of people was exposed to concentrations of ^{226}Ra in water and diet in excess of the recommended limits. This was a result of the operation of a relatively small uranium mill, and of the release to the Animas River of about 0.5 mCi of ^{226}Ra per day in dissolved form, plus about 30 mCi per day associated with suspended solids.

Samples of water, sediments, and biota of the Animas River collected during the period 1960-1963 (after introduction of waste treatment at the mill) showed significantly lower levels of contamination. The highest mean concentrations were observed in algae and insects, with values of about 12 pCi/g (ash). The C.F. calculated for algae fell in the range between 1000 and 2000. The maximum radium concentrations in water were about 0.5 pCi/liter, not significantly different from background.¹⁶⁷

Shearer and Lee¹⁶⁸ have investigated the factors that influence the leaching of ^{226}Ra from uranium mill tailings and river sediments. They found that the ratio between the volume of leachant and the weight of solids was the most significant factor. The composition of the water had very little effect on the extraction of radium; however, the presence of barium significantly promoted leaching.

Havlik et al.^{169,170} have studied the effects of pH and chemical composition of water on the leaching of radium from uranium mill tailings and river sediments. In all experiments, more radium was leached at low pH values; the amount of liberated radium was lowest at pH 9, but the overall effect of pH was rather limited. On the other hand, the chemical composition of the leaching solution had a significant influence; for example, 95 to 100% of the radium contained in river sediments was released in 1 N solutions of NaCl and KCl. Leaching from uranium-bearing rocks was lower, but still significant; for example, a 1 N KCl solution extracted 31% of the radium in 15 min from one ore type, and 22% in 5 hr from a different ore.

Snelling and Shearer¹⁷¹ have reported the results of the survey of the tailings pile of the El Paso Natural Gas Company, at Tuba City, Arizona, which was performed in May 1967. The mill had a capacity of 300 tons of ore per day and was in operation between 1957 and 1966. The tailings piles contained about 800,000 tons of material with an estimated ²²⁶Ra content of 980 pCi/g, for a total of more than 700 Ci of ²²⁶Ra. Since the survey, the pile has been regraded, fenced, and chemically stabilized against wind erosion. Airborne particulates were sampled at ten stations located on or near the tailings area and at one "background" locality. The sampling lasted 4 days, and strong winds (estimated at 25 to 50 km/hr) were prevalent during the daylight hours of the last three days.

The average air concentrations of ²²⁶Ra at the ten stations ranged from about 3 to 1000 times the background. The highest average concentration, 5.6 pCi/m³, was observed at a station located about 50 m east of the pile, in the predominant downwind direction. This value is 2.8 times the concentration limit of ²²⁶Ra for members of the public (2 pCi/m³). A second station had an average ²²⁶Ra concentration of 1.8 pCi/m³, while all the others showed significantly lower values. The critical nuclide of the uranium series, ²³⁰Th, was not measured in the instance of inhalation; however, the estimated maximum level of ²³⁰Th was about ten times higher than the concentration limit of 0.08 pCi/m³. Radon concentrations were slightly above the limit of 3.0 pCi/liter at only two stations. It must be noted, however, that the strong wind, while increasing the resuspension of particulate matter, caused more efficient dispersion of the radon.

Water samples taken at various localities gave the results shown in Table 29. While no water contamination was detected outside the plant site, it is likely that some ground water contamination could be detected if samples were taken closer to the tailings pile.

Another tailings pile was surveyed by Snelling¹⁷² in May 1968; located at Monument Valley, Arizona, it was produced by the mill of the Foote Mineral Company. The mill was designed to process low-grade uranium ore and is presently inactive. The original tailings pile was reprocessed for recovery of uranium and was moved to a new location. The present tailings pile covers an area of about 10 hectares and contains about 1.5×10^6 tons of material, with an estimated ^{226}Ra content of ~ 90 pCi/g, for a total of ~ 120 Ci of ^{226}Ra .

Four sampling stations for airborne particulates were operated throughout a ten-day period that was characterized by light winds during the first six days (0 to 10 km/hr), and somewhat stronger winds during the last four days (15 to 30 km/hr). The samples were analyzed for ^{226}Ra , ^{230}Th , and natural uranium, in addition to gross alpha activity. The observed values are shown in Table 30.

Three samples of tailings material were analyzed for ^{226}Ra , ^{230}Th , and natural uranium with the following results: 59 pCi/g, 46 pCi/g, and 33 $\mu\text{g/g}$, respectively. The low concentrations in the tailings material are reflected in the low levels of activity in suspended particulates. The mean ^{230}Th concentration at the station on the tailings pile is about 50% of the concentration limit. Ground-water samples obtained in the vicinity of the tailings area failed to show any contamination.

A survey of the A-Z Minerals Corporation's tailings pile located at Mexican Hat, Utah, was also performed by Snelling in May 1968.¹⁷³ The mill has been inactive since 1965. The tailings area covers about 14 hectares and contains about 2.2×10^6 tons of tailings material. The estimated content of ^{226}Ra is about 1000 pCi/g, for a total ^{226}Ra content in the pile of about 2000 Ci.

Suspended particulate matter was sampled at nine stations on and around the tailings area for a period of 11 days. The wind was light during the first seven days (0 to 10 km/hr); however, winds on the order of 15 to 30 km/hr were encountered during the daylight hours of the last four

Table 29. Radioactivity in Water,
Tuba City Uranium Mill, May 1967^a

Location	Radioactivity Concentration (pCi/liter)		
	Gross Alpha	Gross Beta	²²⁶ Ra
Tailings raffinate pond	5,300	89,700	1,200
Sewage lagoon overflow	160	200	87
Shallow ground water, 3 km southeast of tailings	0.8	2	0.15
Shallow ground water, 1.5 km south of tailings	0.6	3	0.59
Moenkopi wash, 1.5 km upstream from plant	0.8	11	0.28
Moenkopi wash, 2.5 km downstream from plant	0.4	3	0.28
Deep ground water, 0.4 km north of tailings (background)	1.4	7	0.22

^aData from R. N. Snelling and S. D. Shearer, Jr., "Environmental Survey of Uranium Mill Tailings Pile, Tuba City, Arizona," Radiol. Health Data 10, 475-87 (1969).

Table 30. Average Long-Lived Radioactivity in Air,
Monument Valley Uranium Mill, May 1968^a

Location	Gross Alpha	²²⁶ Ra	²³⁰ Th	Natural Uranium
	(pCi/m ³)	(pCi/m ³)	(pCi/m ³)	(10 ⁻¹² g/m ³)
On tailings pile	0.103	0.023	0.045	62,000
70 m downwind from pile	0.070	0.005	0.007	12,000
Near original tailings pile	0.016	0.001	0.006	14,000
Housing area	0.103	0.006	0.011	40,000

^aData from R. N. Snelling, "Environmental Survey of Uranium Mill Tailings Pile, Monument Valley, Arizona," Radiol. Health Data 11, 511-17 (1970).

days. The samples were analyzed for ^{226}Ra , ^{230}Th , and natural uranium; the corresponding maximum observed concentrations were: 0.007 pCi/m^3 , 0.014 pCi/m^3 and $10,000 \text{ pg/m}^3$.

The actual analysis of tailings material gave the following average concentrations per gram: 370 pCi of ^{226}Ra , 1960 pCi of ^{230}Th , and 150 μg of uranium. The radium value is less than the 1000 pCi/g estimated on the basis of the composition of the mill feed material. The radioactive equilibrium between ^{226}Ra and ^{230}Th that probably existed in the feed material has obviously been disrupted.

Dividing the average concentrations in tailings material by the average air concentrations for the four stations either on or in the immediate proximity of the pile, and taking the ratio for ^{226}Ra equal to 1, the values of 2.5 and 0.3 are obtained for ^{230}Th and uranium, respectively. These values do not differ widely; in fact, they seem to indicate the similarity of resuspension of the three elements, rather than any significant difference.

The observed air concentrations at the Mexican Hat pile were significantly lower than at the Tuba City pile, which has a similar radium concentration in the tailings material. This is probably because the Mexican Hat pile was quite moist and displayed a clay-like consistency during the survey. Ground-water samples were obtained at two springs located 300 m and 1000 m from the tailings area and were analyzed for ^{226}Ra and uranium. The observed concentrations, shown in Table 31, indicate that the uranium content was significantly elevated, while radium had not reached the spring at 1000 m and was only a few times above background 300 m from the tailings. Obviously, uranium was mobilized more actively than radium in this oxidizing environment.

It can be assumed that after failure of containment, either through the action of ground water or the erosion of the overburden, if the long-lived alpha-emitters contained in radioactive waste were to reach the surface, they would constitute a potential hazard somewhat similar to the large piles of uranium mill tailings. The total amount of ^{226}Ra that would build up in 100,000 years in the high-level waste accumulated in the U. S. up to the year 2000 - possibly located in a single repository -

would be about 3500 to 4000 Ci. This would not be significantly different from the amount accumulated in the ore tailings of a large uranium mill. For example, the mill of the Kerr-McGee Corporation, located in Grants, New Mexico, with a nominal capacity of 7000 tons of ore per day, would accumulate tailings with about 1200 Ci of ^{226}Ra annually, provided equilibrium existed in the uranium decay series (assuming operations at full capacity for 300 days per year and an ore grade of 0.2% U_3O_8).¹⁷⁴ It would thus take only three years of operation to produce a pile of tailings containing the same amount of radium that would exist in the hypothetical repository after 100,000 years of decay.

Table 31. ^{226}Ra and Uranium in Ground Water, Mexican Hat Uranium Mill, May 1968^a

Location	Number of Samples	^{226}Ra (pCi/liter)	Natural Uranium ($\mu\text{g/liter}$)
Spring, 300 m from tailings	3	1.5	1,690
Spring, 1000 m from tailings	1	0.2	1,160
Monument Valley (background)	5	0.5	2

^aData from R. N. Snelling, "Environmental Survey of Uranium Mill Tailings Pile, Mexican Hat, Utah," Radiol. Health Data 12, 17-28 (1971).

As mentioned previously, a typical radium content in tailings at U. S. mills is about 1000 pCi/g. While this is an average value, there is a significant variation as a function of the grain size in the tailings fractions. For example, Tsivoglou and O'Connell^{163,175} reported that the slimes fraction of the tailings, which represents only a third of the total waste, contains 70 to 80% of the radium. Typical radium concentrations in the slimes are several thousand picocuries per gram (dry weight), while the sands contain only a few hundred picocuries per gram.

In 100,000-year-old high-level waste the ^{226}Ra content would be about 78,000 pCi/g, or about 10 to 20 times the concentration in tailings slimes. Even after 10 million years the ^{226}Ra content of waste would be about 5000 pCi/g.

Table 32 shows a comparison of the hazard potential associated with the 100,000-year-old waste contained in a hypothetical repository and that for a large pile of uranium mill tailings. The assumptions are that (1) the repository contained all the high-level waste produced by the U. S. nuclear power industry up to the year 2000; and (2) that the tailings pile contained about 20 million tons of material, equivalent to 10 years of operation of a mill the size of the Kerr-McGee Corporation plant in Grants, New Mexico.

Table 32. Comparison of Hazard Potentials Associated with a 100,000-Year-Old High-Level Waste Repository and a Large Uranium Mill Tailings Pile

	Amount of Material	Number of LAI_{ing}	Number of LAI_{inh}
Waste repository	11,000 m ³ LWR waste +5,900 m ³ LMFBR waste	1.1 X 10 ¹²	6.5 X 10 ¹⁴
Tailings pile	2 X 10 ⁷ tons	1.3 X 10 ¹²	2.5 X 10 ¹³

After 100,000 years of decay, the hazard potential by ingestion associated with the long-lived alpha activity contained in the hypothetical repository would be of the same order of magnitude as that associated with uranium tailings piles currently existing in several localities; the inhalation hazard potential of the waste would, of course, be somewhat higher. In addition, the concentration of ^{226}Ra in the high-level waste would be considerably higher than in the tailings. However, the waste, that in the repository was originally buried at a depth of at least several hundred meters and dispersed through a significant volume of geologic materials, could not be disinterred by slow geologic processes without substantial dilution.

5. SUMMARY AND CONCLUSIONS

The change of the hazard potential associated with the heavy nuclides present in high-level solid waste is an exceedingly slow process, particularly after about 100,000 years of decay. In the period from 100,000 to 5 million years, the hazard potential of high-level waste is decreased only by a factor of between 10 and 20. The long-term hazard is due primarily to the uranium series, assuming the intake occurs by ingestion; however, in the case of inhalation, the neptunium series provides the principal contribution after a decay time of about 150,000 years. The variation in hazard potential from 100,000 to ten million years is so limited and so slow that it is impractical to specify an exact length of time for required containment integrity within that time interval.

The available data indicate that if radioactive waste containment were to fail 100,000 to 200,000 years after disposal, some environmental contamination could occur; however, the contamination levels would be low, and the radiological risk would not be significantly different from that currently existing in various localities. This conclusion is based on a comparison of the hazard potential of large piles of uranium mill tailings with that of high-level waste in a repository after 100,000 years of decay.

On a per unit volume basis high-level waste is more hazardous than uranium ores, even after 10 million years of decay. On the other hand, the hazard potential of pitchblende equals that of high-level waste after a decay time varying between less than a thousand to a few thousand years, if the intake is by ingestion. In the case of inhalation, 1-million-year-old waste continues to be more hazardous than pitchblende.

Although very little is known about the geochemical behavior of the transuranium elements, the geochemistry of the natural alpha-emitters, particularly radium, thorium, and uranium provide an insight into the probable behavior of some waste nuclides in geologic systems. Various geologic processes leading to activity release from a disposal formation can be postulated. The mechanisms that have the greatest probability of occurrence are: (1) invasion of the disposal zone by ground water, followed by leaching and transport to the surface; (2) disinterment of the waste through erosion of the overburden; and (3) some combination of the two.

Obviously, the nature of the formation containing the waste would have a significant influence on the most probable release mechanism. For example, if the waste were contained in salt, ground water, which may eventually reach the disposal zone, would remove the various materials according to their solubility. If this occurs it is probable that some separation of the waste from the surrounding rock-salt would take place. The physicochemical nature of the waste at the time of leaching cannot be known for two reasons: (1) because the nature of the solid product that will be placed in the repository is still undefined; and (2) because it is possible that significant changes would take place in the many thousands of years following disposal. Regardless of the condition of the waste, salt would be dissolved more easily and the waste would be left behind in the less soluble materials originally contained in the salt formation. On the other hand, the prolonged contact between brine and waste would undoubtedly cause some leaching and subsequent migration of activity. It is practically impossible to speculate on the destiny of the waste and the ratio between the fraction of activity that would be transported by the saline ground water and that left associated with the insoluble impurities. From the observation of natural radioactive elements it can be assumed that the order of relative mobility would be: Ra and U > Th; where the other actinides would fit in the sequence is not clear, but they would probably be in some intermediate position.

If containment failure were to occur after a decay period of 100,000 years or more, neptunium and plutonium would be the only transuranics present in significant amounts. Regardless of the details of the release mechanism it is certain that activity would be transferred to the surface very slowly, and would undergo significant dispersion and dilution in the process. The exposure of human populations would be dependent on the biologic availability of the nuclides, the dietary habits, and the fraction of activity that would be resuspended. With the partial exception of radium, which is concentrated in the fruits of particular perennial plants, the other heavy nuclides are characterized by a very low biologic availability in terrestrial ecosystems. This is due to the low solubility of most compounds, the strong adsorption on soil particles, the very low uptake from the soil, the generally moderate transfer from the roots to

the aerial part of the plant, the very low absorption from the gastrointestinal tract of mammals, and the preferential accumulation in the skeleton.

As far as the fate of the heavy nuclides in aquatic ecosystems is concerned, it is known that significant differences exist. For example, the residence time in the liquid phase is quite long for radium and uranium, and short for thorium and plutonium. The biologic availability is generally higher in aquatic environments because of the significant concentration factors in many organisms. As examples, concentration factors of plutonium as high as 2×10^4 in North Atlantic Sargassum and between 10^3 and 10^4 in some edible bivalves have been reported.

The inhalation hazard potential of many heavy nuclides is very great due to the extremely low limits of intake by inhalation. The observation of airborne particulate matter shows that dust concentrations in air, at least in the inhalable size range, are usually well below $100 \mu\text{g}/\text{m}^3$. In fairly unpolluted environments far from the seashore, the composition of the suspended dust reflects the composition of the soil, and resuspension apparently causes no separation between elements. Another important observation is that dust of local origin is always diluted by particles coming from more remote sources. All these factors seem to contribute to the reduction of the inhalation hazard. This agrees with what has been observed for the natural long-lived alpha-emitters for which the critical intake mechanism is usually by ingestion.

Study of the high-background areas of India and Brazil confirms the low biologic availability of the natural radioactive elements contained in monazite. No obvious radiological consequences for the exposed population have been observed so far.

Large piles of uranium mill tailings exist in many localities. In several cases, the tailings have not been stabilized in any way; the largest piles can contain several thousand curies of ^{226}Ra . They represent a hazard potential comparable to a radioactive waste repository after a decay time between 100,000 and 200,000 years. Environmental surveys around the tailings show that dispersal of activity and contamination of the landscape are actually taking place, but the levels of activity are usually low. No evidence has been found of leaching and migration

of radium with the ground water at any significant distance from the tailings. Air concentrations of ^{226}Ra and ^{230}Th higher than the suggested limits for members of the public have been reported in one case, but only in the immediate proximity of the tailings.

6. ACKNOWLEDGMENT

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APPENDIX

RADIOACTIVE DECAY SERIES^a

^aFrom:

Radiological Health Handbook
U. S. Department of Health, Education, and Welfare
Public Health Service, 1970

Thorium Series (4n)*

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{232}_{90}\text{Th}$	Thorium	1.41×10^{10} y	3.95 (24%) 4.01 (76%)	---	---
$^{228}_{88}\text{Ra}$	Mesothorium I	6.7y	---	0.055 (100%)	---
$^{228}_{89}\text{Ac}$	Mesothorium II	6.13h	---	1.18 (35%) 1.75 (12%) 2.09 (12%)	0.34c‡ (15%) 0.908 (25%) 0.96c (20%)
$^{228}_{90}\text{Th}$	Radiothorium	1.910y	5.34 (28%) 5.43 (71%)	---	0.084 (1.6%) 0.214 (0.3%)
$^{224}_{88}\text{Ra}$	Thorium X	3.64d	5.45 (6%) 5.68 (94%)	---	0.241 (3.7%)
$^{220}_{86}\text{Rn}$	Emanation Thoron (Tn)	55s	6.29 (100%)	---	0.55 (0.07%)
$^{216}_{84}\text{Po}$	Thorium A	0.15s	6.78 (100%)	---	---
$^{212}_{82}\text{Pb}$	Thorium B	10.64h	---	0.346 (81%) 0.586 (14%)	0.239 (47%) 0.300 (3.2%)
$^{212}_{83}\text{Bi}$	Thorium C	60.6m	6.05 (25%) 6.09 (10%)	1.55 (5%) 2.26 (55%)	0.040 (2%) 0.727 (7%) 1.620 (1.8%)
$^{212}_{84}\text{Po}$	Thorium C'	304ns	8.78 (100%)	---	---
$^{208}_{81}\text{Tl}$	Thorium C''	3.10m	---	1.28 (25%) 1.52 (21%) 1.80 (50%)	0.511 (23%) 0.583 (86%) 0.860 (12%)
$^{208}_{82}\text{Pb}$	Thorium D	Stable	---	---	2.614 (100%) ---

*This expression describes the mass number of any member in this series, where n is an integer.
Example: $^{232}_{90}\text{Th}$ (4n).....4(58) = 232

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.

‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

Data taken from: Lederer, C. M., Hollander, J. M., and Perlman, I., Table of Isotopes (6th ed.; New York: John Wiley & Sons, Inc., 1967) and Hogan, O. H., Zigman, P. E., and Mackin, J. L., Beta Spectra (USNRDL-TR-802 [Washington, D.C.: U.S. Atomic Energy Commission, 1964]).

Neptunium Series (4n + 1)*

Nuclide	Element name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{241}_{94}\text{Pu}$ 	Plutonium	13.2y	4.85 (0.0003%) 4.90 (0.0019%)	0.021 (~100%)	0.145 (.00016%)
	Americium	458y	5.44 (13%) 5.49 (85%)	---	0.060 (36%) 0.101c‡ 0.04%
$^{237}_{92}\text{U}$ 	Uranium	6.75d	---	0.248 (96%)	0.060 (36%) 0.208 (23%)
	Neptunium	2.14×10^6 y	4.65c (12%) 4.78c (75%)	---	0.030 (14%) 0.086 (14%) 0.145 (1%)
$^{233}_{91}\text{Pa}$ 	Protactinium	27.0d	---	0.145 (37%) 0.257 (58%) 0.568 (5%)	0.31c (44%)
	Uranium	1.62×10^5 y	4.78 (15%) 4.82 (83%)	---	0.042 (?) 0.097 (?)
$^{229}_{90}\text{Th}$ 	Thorium	7340y	4.84 (58%) 4.90 (11%) 5.05 (7%)	---	0.137c (~3%) 0.20c (~10%)
	Radium	14.8d	---	0.32 (100%)	0.040 (33%)
$^{225}_{89}\text{Ac}$ 	Actinium	10.0d	5.73c (10%) 5.79 (28%) 5.83 (54%)	---	0.099 (?) 0.150 (?) 0.187 (?)
	Francium	4.8m	6.12 (15%) 6.34 (82%)	---	0.218 (14%)
$^{217}_{85}\text{At}$ 	Astatine	0.032s	7.07 (~100%)	---	---
	Bismuth	47m	5.87 (~2.2%)	1.39 (~97.8%)	0.437 (?)
$^{213}_{83}\text{Bi}$ 	Polonium	4.2μs	8.38 (~100%)	---	---
	Thallium	2.2m	---	1.99 (100%)	0.12 (50%) 0.45 (100%) 1.56 (100%)
$^{209}_{82}\text{Pb}$ 	Lead	3.30h	---	0.637 (100%)	---
	Bismuth	Stable ($>2 \times 10^{18}$ y)	---	---	---

*This expression describes the mass number of any member in this series, where n is an integer.

Example: $^{229}_{90}\text{Th}$ (4n + 1).....4(57) + 1 = 229

The (4n + 1) series is included here for completion. It is not found as a naturally-occurring series.

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.

‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

Data taken from: Table of Isotopes and USNRDL-TR-802.

Uranium Series (4n + 2)*

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{238}_{92}\text{U}$	Uranium I	$4.51 \times 10^9 \text{ y}$	4.15 (25%) 4.20 (75%)	---	---
$^{234}_{90}\text{Th}$	Uranium X ₁	24.1d	---	0.103 (21%) 0.193 (79%)	0.063c‡ (3.5%) 0.093c (4%)
$^{234}_{91}\text{Pa}^m$	Uranium X ₂	1.17m	---	2.29 (98%)	0.765 (0.30%) 1.001 (0.60%)
$^{234}_{91}\text{Pa}$	Uranium Z	6.75h	---	0.53 (66%) 1.13 (13%)	0.100 (50%) 0.70 (24%) 0.90 (70%)
$^{234}_{92}\text{U}$	Uranium II	$2.47 \times 10^5 \text{ y}$	4.72 (28%) 4.77 (72%)	---	0.053 (0.2%)
$^{230}_{90}\text{Th}$	Ionium	$8.0 \times 10^4 \text{ y}$	4.62 (24%) 4.68 (76%)	---	0.068 (0.6%) 0.142 (0.07%)
$^{226}_{88}\text{Ra}$	Radium	1602y	4.60 (6%) 4.78 (95%)	---	0.186 (4%)
$^{222}_{86}\text{Rn}$	Emanation Radon (Rn)	3.823d	5.49 (100%)	---	0.510 (0.07%)
$^{218}_{84}\text{Po}$	Radium A	3.05m	6.00 (~100%)	0.33 (~0.019%)	---
$^{214}_{82}\text{Pb}$	Radium B	26.8m	---	0.65 (50%) 0.71 (40%) 0.98 (6%)	0.295 (19%) 0.352 (36%)
$^{218}_{85}\text{At}$	Astatine	~2s	6.65 (6%) 6.70 (94%)	? (~0.1%)	---
$^{214}_{83}\text{Bi}$	Radium C	19.7m	5.45 (0.012%) 5.51 (0.008%)	1.0 (23%) 1.51 (40%) 3.26 (19%)	0.609 (47%) 1.120 (17%) 1.764 (17%)
$^{214}_{84}\text{Po}$	Radium C'	164μs	7.69 (100%)	---	0.799 (0.014%)
$^{210}_{81}\text{Tl}$	Radium C''	1.3m	---	1.3 (25%) 1.9 (56%) 2.3 (19%)	0.296 (80%) 0.795 (100%) 1.31 (21%)
$^{210}_{82}\text{Pb}$	Radium D	21y	3.72 (0.00002%)	0.016 (85%) 0.061 (15%)	0.047 (4%)
$^{210}_{83}\text{Bi}$	Radium E	5.01d	4.65 (.00007%) 4.69 (.00005%)	1.161 (~100%)	---
$^{210}_{84}\text{Po}$	Radium F	138.4d	5.305 (100%)	---	0.803 (0.0011%)
$^{206}_{81}\text{Tl}$	Radium E''	4.19m	---	1.571 (100%)	---
$^{206}_{82}\text{Pb}$	Radium G	Stable	---	---	---

*This expression describes the mass number of any member in this series, where n is an integer.

Example: $^{206}_{82}\text{Pb}$ (4n + 2).....4(51) + 2 = 206

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.

‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

Data taken from: Table of Isotopes and USNRDL-TR-802.

Actinium Series (4n + 3)*

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{235}_{92}\text{U}$	Actinouranium	$7.1 \times 10^8 \text{y}$	4.37 (18%) 4.40 (57%) 4.58c‡ (8%)	---	0.143 (11%) 0.185 (54%) 0.204 (5%)
$^{231}_{90}\text{Th}$	Uranium Y	25.5h	---	0.140 (45%) 0.220 (15%) 0.305 (40%)	0.026 (2%) 0.084c (10%)
$^{231}_{91}\text{Pa}$	Protoactinium	$3.25 \times 10^4 \text{y}$	4.95 (22%) 5.01 (24%) 5.02 (23%)	---	0.027 (6%) 0.29c (6%)
$^{227}_{89}\text{Ac}$	Actinium	21.6y	4.86c (0.18%) 4.95c (1.2%)	0.043 (~99%)	0.070 (0.08%)
$^{227}_{90}\text{Th}$ (98.6%)	Radioactinium	18.2d	5.76 (21%) 5.98 (24%) 6.04 (23%)	---	0.050 (8%) 0.237c (15%) 0.31c (8%)
$^{223}_{87}\text{Fr}$ (1.4%)	Actinium K	22m	5.44 (~0.005%)	1.15 (~100%)	0.050 (40%) 0.080 (13%) 0.234 (4%)
$^{223}_{88}\text{Ra}$	Actinium X	11.43d	5.61 (26%) 5.71 (54%) 5.75 (9%)	---	0.149c (10%) 0.270 (10%) 0.33c (6%)
$^{219}_{86}\text{Rn}$	Emanation Actin. (An)	4.0s	6.42 (8%) 6.55 (11%) 6.82 (81%)	---	0.272 (9%) 0.401 (5%)
$^{215}_{84}\text{Po}$	Actinium A	1.78ms	7.38 (~100%)	0.74 (~0.00023%)	---
$^{211}_{82}\text{Pb}$ (~100%)	Actinium B	36.1m	---	0.29 (1.4%) 0.56 (9.4%) 1.39 (87.5%)	0.405 (3.4%) 0.427 (1.8%) 0.832 (3.4%)
$^{215}_{85}\text{At}$ (.00023%)	Astatine	~0.1ms	8.01 (~100%)	---	---
$^{211}_{83}\text{Bi}$	Actinium C	2.15m	6.28 (16%) 6.62 (84%)	0.60 (0.2%)	0.351 (14%)
$^{211}_{84}\text{Po}$ (0.28%)	Actinium C'	0.52s	7.45 (99%)	---	0.570 (0.5%) 0.90 (0.5%)
$^{207}_{81}\text{Tl}$ (99.7%)	Actinium C''	4.79m	---	1.44 (99.8%)	0.897 (0.16%)
$^{207}_{82}\text{Pb}$	Actinium D	Stable	---	---	---

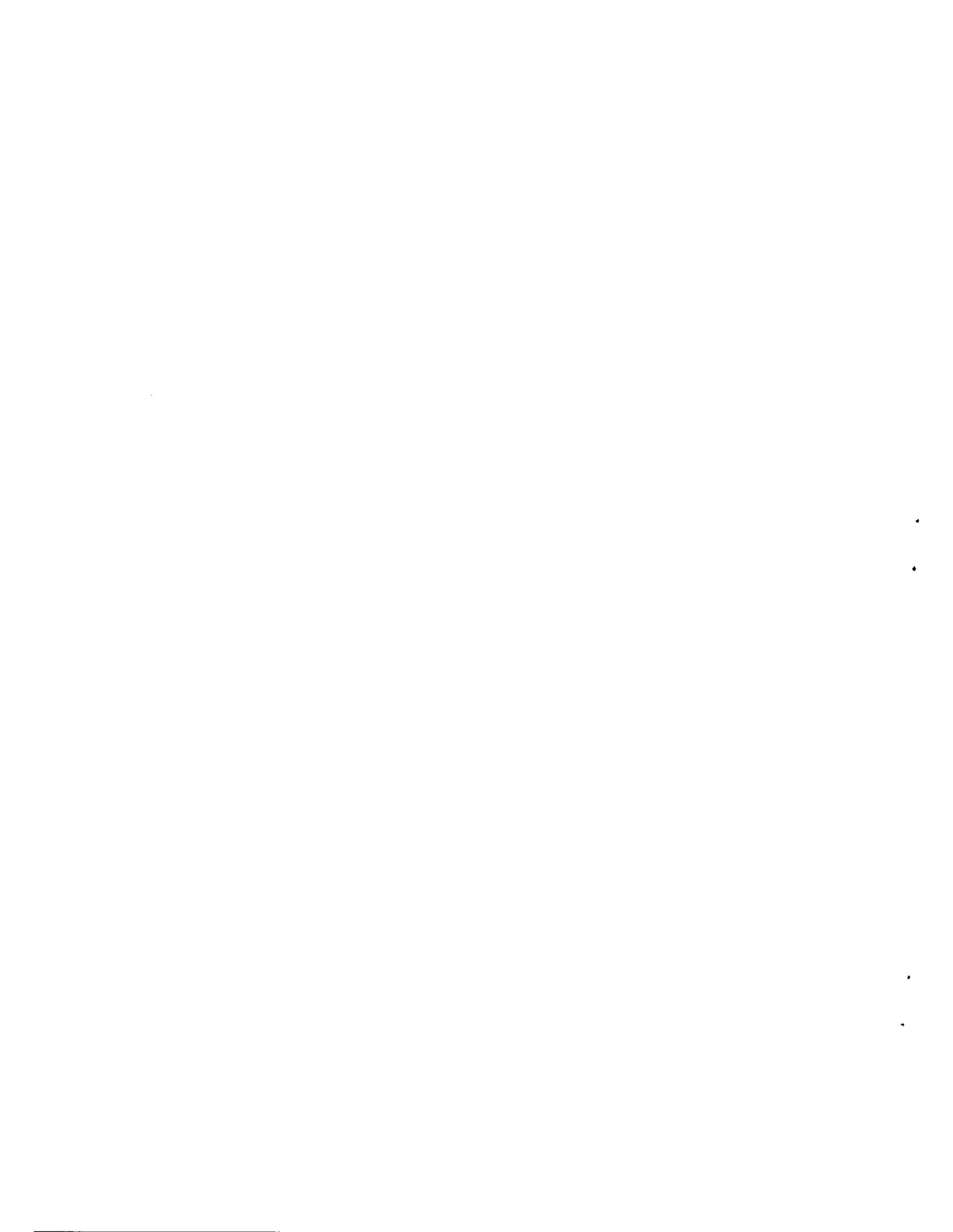
*This expression describes the mass number of any member in this series, where n is an integer.

Example: $^{207}_{82}\text{Pb}$ (4n + 3).....4(51) + 3 = 207

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.

‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

Data taken from: Table of Isotopes and USNRDL-TR-802.



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