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A review of plutonium environmental data with a
bibliography for use in risk assessments

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15 Jun 1983

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A REVIEW OF PLUTONIUM ENVIRONMENTAL DATA
WITH A BIBLIOGRAPHY FOR USE IN RISK ASSESSMENTS

Prepared for

U.S. Department of Energy
Office of Special Nuclear Projects

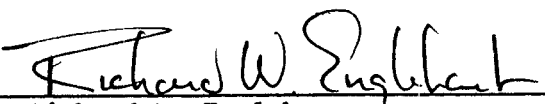
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1.0 INTRODUCTION

Plutonium fueled radioisotopic heat sources find application in a spectrum of space, terrestrial, and undersea applications to generate electrical power by thermoelectric or dynamic-cycle conversion. The principal radioisotope used for this purpose is plutonium (Pu)-238 in the form of plutonium dioxide. Such systems under postulated accident conditions could release radioactivity into the environment resulting in risks to the general population in the form of radiological doses and associated health effects.

Radioactivity could be released at high and low altitudes, on the land surface, or underwater. The released radioactivity could then be dispersed into various environmental media (air, soil, water) and interact with people through various exposure pathways leading to inhalation, ingestion, and external radiological doses and associated health effects. Possible scenarios for the release of activity, its dispersion in the environment, and subsequent population exposure addressed in this report are shown in the flow diagram presented in Figure 1.⁽¹⁾

The evaluation of the radiological impact of postulated scenarios involving releases of activity into the environment includes the following steps:

1. Identification of postulated accident release modes, including the probability of release and the release location.
2. Source term definition, including the activity of each radionuclide released and the corresponding chemical form and particle size distribution.
3. Analysis of the environmental behavior of the released radioactivity to determine the concentrations in environmental media (air, soil, and water) as a function of time.
4. Analysis of the interaction between the environmental concentrations and man, leading to ingestion, inhalation, and external doses through each environmental exposure pathway.

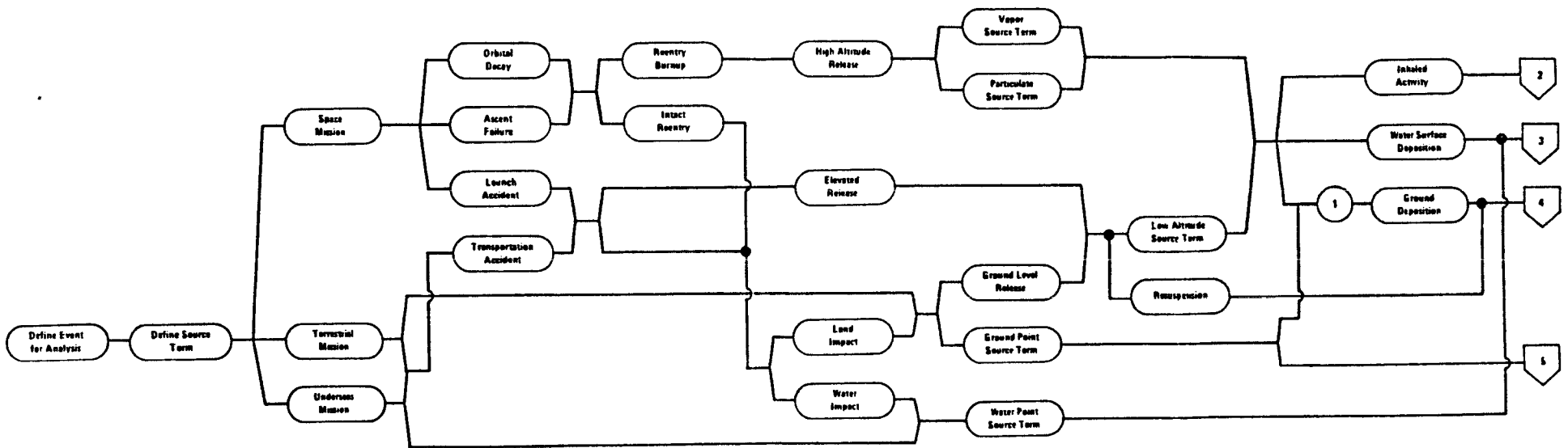


Figure 1.- Potential Plutonium Environmental Exposure Pathways

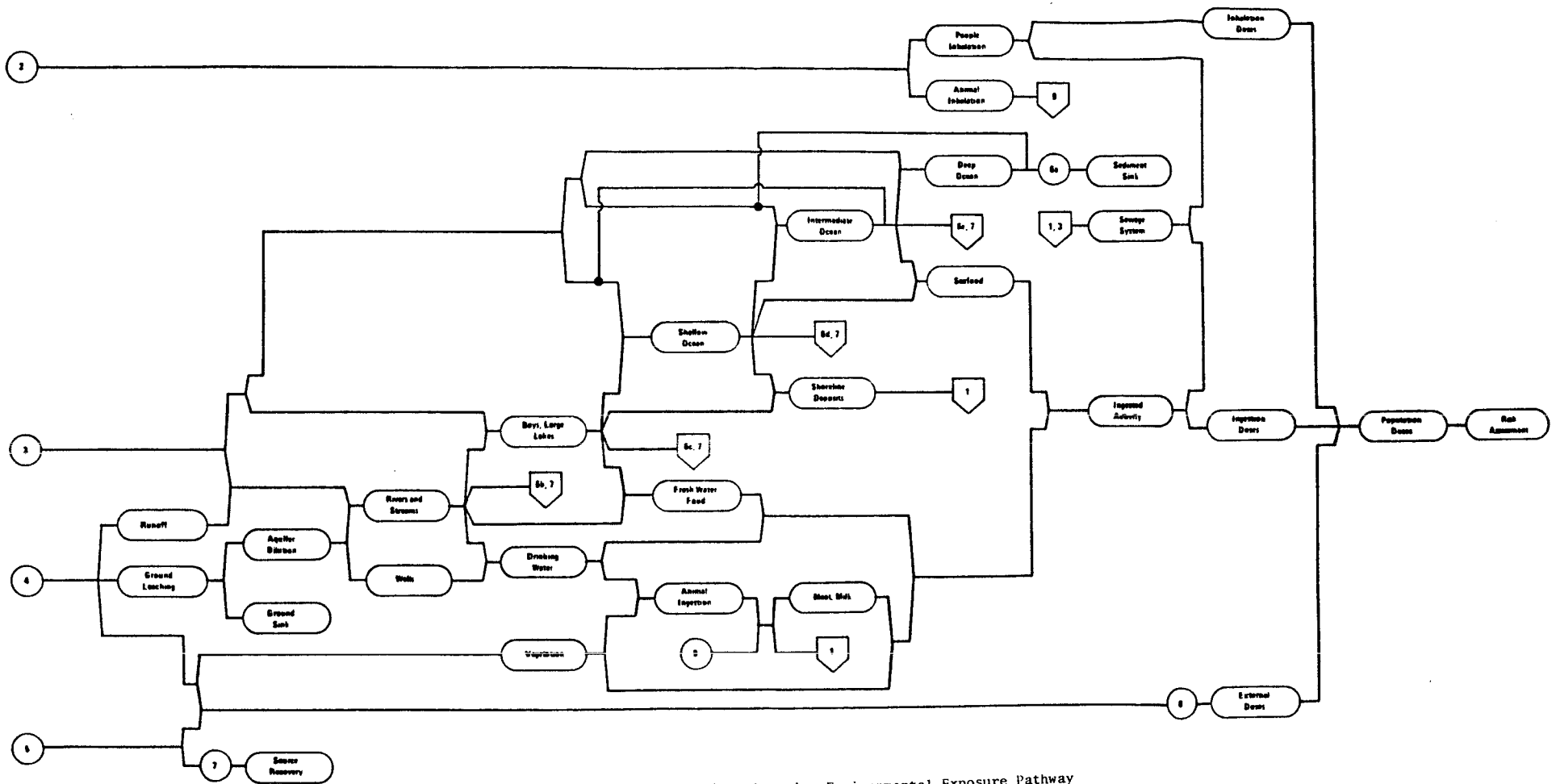


Figure 1.- (Continued) Potential Plutonium Environmental Exposure Pathway

5. The evaluation of the radiological impact on man in terms of the population doses received and the resulting health effects.

Radiological assessment models have been developed to perform the above analyses over both short time periods (up to one year) and long time periods (thousands of years) following postulated releases of plutonium.^(1,2,3) These models have been documented in the Overall Safety Manual for use in safety risk assessments of space missions utilizing plutonium-fueled electric power systems.⁽²⁾ However, the results calculated by such models depend on representative input values of a spectrum of environmental transfer parameters that characterize the behavior of plutonium in the environment.

The selection of appropriate environmental transfer parameters to be used in a given analysis will depend on the accident scenarios to be modeled and the terrestrial and aquatic environmental conditions to be encountered. The objective of this report is to summarize available plutonium environmental data to assist in this parameter value selection process. In order to provide a basis for the data presented, the availability of plutonium environmental data and the status of supporting research programs are summarized in Chapter 2. Appendix A summarizes plutonium environmental data by transport mechanism and/or environment with supporting references. Appendix B presents a bibliography on plutonium in the environment that can be used to gather more detailed information on a given parameter.

This work has been performed for the U.S. Department of Energy under Contract No. DE-AC01-80ET-32029, Task Assignment 82-8. This report is an update of an earlier NUS report that presented only a bibliography without any data summary.⁽⁴⁾

2.0 AVAILABILITY OF PLUTONIUM ENVIRONMENTAL DATA

Much research has been conducted over the past ten years on the behavior of plutonium in the environment. This research has been prompted by environmental and safety studies designed 1) to evaluate the effects of fallout following nuclear weapons testing in the atmosphere, 2) in support the Liquid Metal Fast Breeder Reactor (LMFBR) program, 3) in support of plutonium production and handling facilities, and 4) in support of plutonium-fueled nuclear systems used in space, terrestrial, and underseas applications.

Principal organizations involved in research on plutonium in the environment include the following:

- o Argonne National Laboratory (ANL)
- o Pacific Northwest Laboratory (PNL)
- o Lawrence Livermore National Laboratory (LLL)
- o Los Alamos National Laboratory (LANL)
- o Oak Ridge National Laboratory (ORNL)
- o Savannah River Ecology Laboratory (SREL)
- o U.S. DOE, Environmental Measurement Laboratory (EML)
- o U.S. DOE, Nevada Applied Ecology Group (NAEG)
- o U.S. DOE, Office of Special Nuclear Projects (OSNP)
- o U.S. DOE, Office of Health and Environmental Research (OHER)
- o U.S. Environmental Protection Agency (EPA)
- o Universities.

A primary focus point of plutonium environmental research is the Ecological Sciences Division of the DOE Office of Health and Environmental Research (OHER) and its coordinated research efforts in support of the LMFBR program. These research areas, summarized in Figure 2, cover not only plutonium but all the transuranics. Principal OHER sponsored research is conducted by PNL, ANL, SREL, ORNL, and LANL. OHER has sponsored annual workshops over the past several years that deal with transuranics in the environment with special emphasis on plutonium.⁽⁵⁻⁹⁾ Due to the tremendous amount of plutonium, environmental data collected to date, OHER sponsored the compilation and summary of this work, resulting in the publication of Transuranic Elements in the Environment.⁽¹⁰⁾

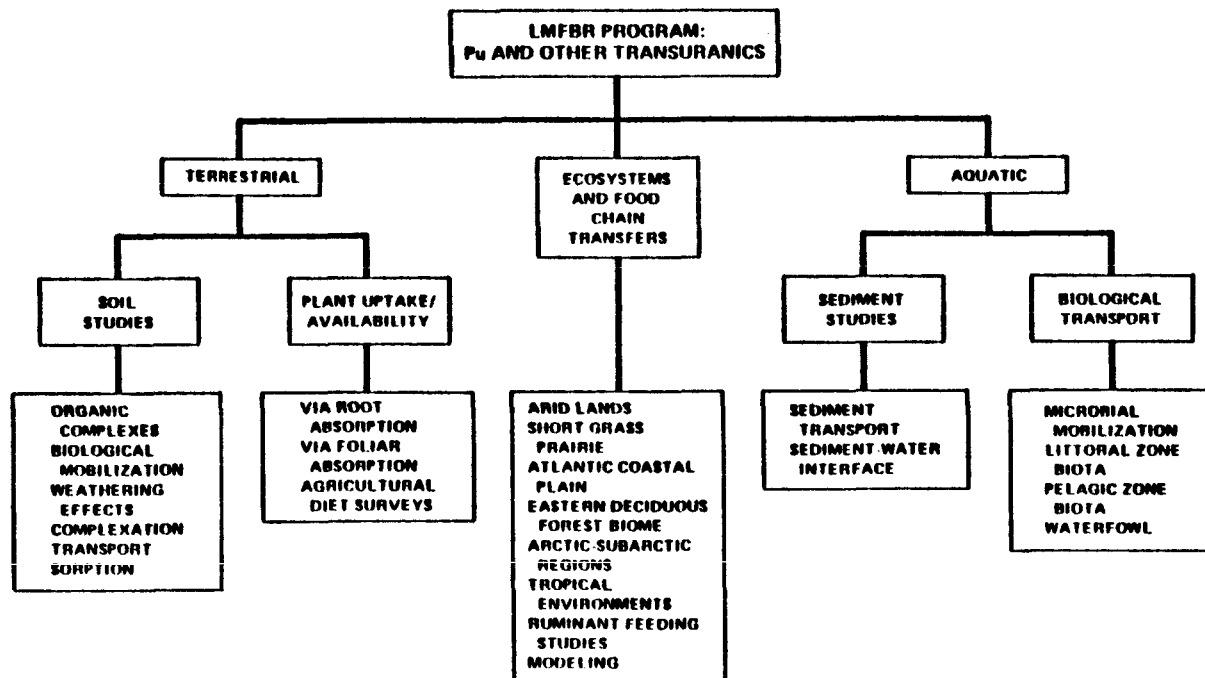


Figure 2.- Plutonium Environmental Programs Sponsored by DOE's Office of Health and Environmental Research

The document is an important source of data for use in estimating environmental transfer parameters.

Research related to plutonium in the environment resulting from nuclear weapons testing is being conducted by EML, NAEG, and ANL.

Environmental research related to plutonium fueled space nuclear systems, sponsored by the DOE Office of Special Nuclear Projects, is being conducted by LANL and the Naval Ocean Systems Center. This research work is designed to evaluate the interaction of Pu-238 oxide fuel with terrestrial and aquatic environments.

A summary of the research programs being conducted by the organizations identified related to the behavior of plutonium in the environment is presented in a separate report prepared by NUS as part of this task.⁽¹¹⁾

In reviewing the literature and the current research programs being carried out by the above organizations on plutonium environmental behavior, it is evident that a tremendous amount of data is available for use in safety risk assessments of plutonium fueled space nuclear systems. These data have been reviewed and summarized in Appendix A according to the following categories:

- o General Behavior
- o Source Terms
- o Terrestrial Ecosystems
 - General
 - Soils
 - Plant Uptake
 - Animal Uptake
 - Leaching and Runoff
 - Deposition and Resuspension
- o Aquatic Ecosystems
 - General
 - Fresh Water
 - Marine

- o Atmospheric Dispersion
- o Dose Assessment and Modeling.

The data are keyed to the appropriate models and exposure pathways described in the Overall Safety Manual.⁽²⁾ References supporting the cited data are presented in a comprehensive bibliography in Appendix B, with the references grouped in the same categories identified above as the data.

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Appendix B

Bibliography on Behavior of Plutonium in the Environment

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Appendix A

Plutonium Environmental Data Summary

Summary of Plutonium Environmental Data
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Data Category	Parameter	Applicability	Value	Data Reference	
<u>Terrestrial Ecosystems</u>	Airborne Source Terms (Release rate per gram of fuel)	6 mm particles on surface		B.2(23)	
		Humid Conditions		Analysis of LANL data by NUS	
		Pre-rain	2x10 ⁻⁸ nCi/s	"	
		Rain	0.9 nCi/per rain	"	
		Post-rain	5x10 ⁻⁹ nCi/s	"	
		Arid Conditions	2.10 ⁻⁷ nCi/s	"	
		6 mm particles on surface			
		Humid Conditions			
		Pre-rain	4x10 ⁻⁸ nCi/s	"	
		Rain	0	"	
		Post-rain	3x10 ⁻⁹ nCi/s	"	
		Leaching/Runoff Source (Release rate per gram of fuel)	6 mm particles on surface		"
		Humid Conditions	1.3x10 ⁻⁵ nCi/s	"	
	Arid Conditions	2.8x10 ⁻⁷ nCi/s	"		
	6 mm particles on surface				
	Humid Conditions	4.4x10 ⁻⁵ nCi/s	"		
	Arid Conditions	2.5x10 ⁻⁶ nCi/s	"		
Inventory Ratio ^a	Soils	0.998 to 0.986	B.1(25)		
	Vegetation	2x10 ⁻² to 3x10 ⁻⁷	"		
	Litter	3x10 ⁻² to 2x10 ⁻⁴	"		
	Animals	7x10 ⁻³ to 6x10 ⁻¹¹	"		
	Concentration Ratio ^b (Field grown plants)	Crops and Vegetables (ORNL)		B.4.3(2)	
Fruit		10 ⁻⁵ to 10 ⁻⁴	"		
Foliage		10 ⁻⁴ to 10 ⁻³	"		
Underground Crops		10 ⁻⁴ to 10 ⁻²	"		
Crops and Vegetables (SRP)					
Wheat					
Grain		10 ⁻³	"		
Foliage		10 ⁻² to 10 ⁻¹	"		
Soybean					
Grain		10 ⁻⁴	"		
Foliage	10 ⁻²	"			

a. The proportion of the total plutonium in the ecosystem to that found in each major compartment
b. Ratio of concentration in plant to concentration in soil

Summary of Plutonium Environmental Data
Page 2 of 4

Data Category	Parameter	Applicability	Value	Data Reference	
<u>Terrestrial Ecosystems</u> (Continued)		Corn			
		Grain	10 ⁻⁴	B.4.3(2)	
		Leaves	10 ⁻²	"	
		Crops and Vegetables (Cape Cod)			
		Corn and Cobs	10 ⁻²	"	
		Potatoes	10 ⁻²	"	
		Peas (shelled)	10 ⁻³	"	
		Native Vegetation (SRP)	10 ⁻² to 10 ⁰	"	
		Native Vegetation (Enewetak)	10 ⁻⁴ to 10 ⁻¹	"	
		Coconut	10 ⁻³	"	
		Native Vegetation (Rocky Flats)	10 ⁻⁴ to 10 ⁻³	"	
		Native Vegetation (NTS)	10 ⁻² to 10 ⁰	"	
		Native Vegetation (ORNL)	10 ⁻³	"	
		Concentration Ratio (Total Ecosystem)	ORNL Floodplain Ecosystem		B.4(6)
			Tree Wood	5x10 ⁻⁵	"
			Tree Root	6x10 ⁻²	"
			Tree Leaf	5x10 ⁻⁵	"
			Litter	1x10 ⁻¹	"
			Soil Fauna	2x10 ⁻²	"
			Ground Vegetation	8x10 ⁻⁴	"
			Consumer	6x10 ⁻¹¹	"
			Los Alamos Mortandad Canyon		
			Tree Wood	1x10 ⁻³	"
		Tree Leaf	1x10 ⁻³	"	
		Litter	6x10 ⁻¹	"	
		Grass	1x10 ⁰	"	
		Forb	8x10 ⁻²	"	
		Consumer	5x10 ⁻³	"	
	Concentration Ratio (Animals)	General	10 ⁻¹ to 10 ⁻⁴	B.1(25)	
		Floodplain Ecosystem (ORNL)	6x10 ⁻⁴	B.4(6)	
		Canyon Ecosystem (LANL)	5x10 ⁻³	"	
		Small Mammals (Rocky Flats)	7.8x10 ⁻³	B.4.1(25)	
	Weathering Rate	General	10% per year		

Summary of Plutonium Environmental Data
Page 3 of 4

Data Category	Parameter	Applicability	Value	Data Reference	
<u>Terrestrial Ecosystems</u> (Continued)	Watershed Erosion Rate	Savannah River	0.005% per year	B.1(25)	
		Miami River	0.05% per year	"	
		Hudson River	0.05% per year	"	
	Soil Leaching Rate	General	0.1 cm/yr through top 1 cm of soil	B.7(12)	
			0.5 cm/yr below top 1 cm of soil		
	Resuspension Factor	Nevada Test Site (Generally applied)	$10^{-4}e^{-0.15} t+10^{-9},m^{-1}$	B.4.6(9)	
			Sandy Grass	10^{-8} to 5×10^{-5}	B.4.6(28)
			Sandy Debris	2×10^{-9} to 10^{-5}	
			Rocky Flats, Co	10^{-9} to 10^{-5}	
			Palmares, Spain	2×10^{-9} to 8×10^{-6}	
New York City			5×10^{-9} to 2×10^{-6}		
Hanford			10^{-10} to 6×10^{-7}		
United Kingdom	2×10^{-9}				
<u>Aquatic Ecosystems</u>	Dissolution Rate	Grog Fuel		B.2(23)	
		10°C Fresh Water	1440 nCi/m ² -s	"	
		35°C Fresh Water	340 nCi/m ² -s	"	
		10°C Sea Water	60 nCi/m ² -s	"	
		35°C Sea Water	17 nCi/m ² -s	"	
		PPO Fuel			
		10° Fresh Water	90 nCi/m ² -s	"	
		10° Sea Water	1.2 nCi/m ² -s	"	
	K _d ^a	General	10^5	B.1(25)	
		Pu all Oxidized	10^3	"	
		Pu all Reduced	10^6	"	
		pH Decreases	K _d Decreases	"	
		Amount of Organic Carbon Increases	K _d Decreases	"	
		Amount of Sulfates Increases	K _d Decreases	B.1(25)	
		Amount of Carbonates Increases	K _d Decreases	"	
		Enewetak (marine)	1.8×10^5	B.5.3(36)	
		Enewetak (groundwater)	2.5×10^5	"	
Bikini (marine)	2.2×10^5	"			
Windscale	5×10^4	"			

A-3

a. Ratio of concentration in sediment to concentration in water

Summary of Plutonium Environmental Data
Page 4 of 4

Data Category	Parameter	Applicability	Value	Data Reference	
<u>Aquatic Ecosystems</u> (Continued)		Humbolt Bay, CA	8x10 ⁴	"	
		Mediterranean	5x10 ⁴	"	
	Seafood Density	Florida Coast	8.6x10 ⁻⁹ g/cm ³ -yr	B.7(45)	
	Concentration Ratio	Plankton		5000	B.5.1(4)
		Benthic Algae and Macrophytes		5000	"
		Benthic Invertebrates		1000	"
		Fish			
		Bottom Feeders		250	"
	Plankton Feeders		25	"	
	Piscivorous		5	"	
Particle Sink Rates	Pacific Ocean		50-400 m/yr	B.1(24)	
	North American Fresh Water Lakes		10-100 m/yr	B.1(24)	
<u>High Altitude Vapor Release</u>	Residence Half-Times	Mesosphere (45 km)	5 years	B.7(10)	
		Stratosphere (10-45 km)	0.75-2 years	"	
		Troposphere (0-10 km)	5-15 days	"	

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B.5

AQUATIC ECOSYSTEMS

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Data Category	Parameter	Applicability	Value	Data Reference	
<u>Terrestrial Ecosystems</u>	Airborne Source Terms (Release rate per gram of fuel)	6 mm particles on surface		B.2(23)	
		Humid Conditions		Analysis of LANL data by NUS	
		Pre-rain	2x10 ⁻⁸ nCi/s	"	
		Rain	0.9 nCi/per rain	"	
		Post-rain	5x10 ⁻⁹ nCi/s	"	
		Arid Conditions	2.10 ⁻⁷ nCi/s	"	
			6 mm particles on surface		
			Humid Conditions		
			Pre-rain	4x10 ⁻⁸ nCi/s	"
			Rain	0	"
			Post-rain	3x10 ⁻⁹ nCi/s	"
	Leaching/Runoff Source (Release rate per gram of fuel)	6 mm particles on surface	Humid Conditions	1.3x10 ⁻⁵ nCi/s	"
			Arid Conditions	2.8x10 ⁻⁷ nCi/s	"
		6 mm particles on surface	Humid Conditions	4.4x10 ⁻⁵ nCi/s	"
Arid Conditions			2.5x10 ⁻⁶ nCi/s	"	
Inventory Ratio ^a		Soils	0.998 to 0.986	B.1(25)	
		Vegetation	2x10 ⁻² to 3x10 ⁻⁷	"	
	Litter	3x10 ⁻² to 2x10 ⁻⁴	"		
	Animals	7x10 ⁻³ to 6x10 ⁻¹¹	"		
Concentration Ratio ^b (Field grown plants)	Crops and Vegetables (ORNL)	Fruit	10 ⁻⁵ to 10 ⁻⁴	B.4.3(2)	
		Foliage	10 ⁻⁴ to 10 ⁻³	"	
		Underground Crops	10 ⁻⁴ to 10 ⁻²	"	
	Crops and Vegetables (SRP)	Wheat			
		Grain	10 ⁻³	"	
		Foliage	10 ⁻² to 10 ⁻¹	"	
		Soybean			
		Grain	10 ⁻⁴	"	
		Foliage	10 ⁻²	"	

a. The proportion of the total plutonium in the ecosystem to that found in each major compartment
b. Ratio of concentration in plant to concentration in soil

Summary of Plutonium Environmental Data
Page 2 of 4

Data Category	Parameter	Applicability	Value	Data Reference	
<u>Terrestrial Ecosystems</u> (Continued)		Corn			
		Grain	10 ⁻⁴	B.4.3(2)	
		Leaves	10 ⁻²	"	
		Crops and Vegetables (Cape Cod)			
		Corn and Cobs	10 ⁻²	"	
		Potatoes	10 ⁻²	"	
		Peas (shelled)	10 ⁻³	"	
		Native Vegetation (SRP)	10 ⁻² to 10 ⁰	"	
		Native Vegetation (Enewetak)	10 ⁻⁴ to 10 ⁻¹	"	
		Coconut	10 ⁻³	"	
		Native Vegetation (Rocky Flats)	10 ⁻⁴ to 10 ⁻³	"	
		Native Vegetation (NTS)	10 ⁻² to 10 ⁰	"	
		Native Vegetation (ORNL)	10 ⁻³	"	
		Concentration Ratio (Total Ecosystem)	ORNL Floodplain Ecosystem		B.4(6)
			Tree Wood	5x10 ⁻⁵	"
			Tree Root	6x10 ⁻²	"
			Tree Leaf	5x10 ⁻⁵	"
			Litter	1x10 ⁻¹	"
			Soil Fauna	2x10 ⁻²	"
			Ground Vegetation	8x10 ⁻⁴	"
			Consumer	6x10 ⁻¹¹	"
			Los Alamos Mortandad Canyon		
			Tree Wood	1x10 ⁻³	"
		Tree Leaf	1x10 ⁻³	"	
		Litter	6x10 ⁻¹	"	
		Grass	1x10 ⁰	"	
		Forb	8x10 ⁻²	"	
		Consumer	5x10 ⁻³	"	
	Concentration Ratio (Animals)	General	10 ⁻¹ to 10 ⁻⁴	B.1(25)	
		Floodplain Ecosystem (ORNL)	6x10 ⁻⁴	B.4(6)	
		Canyon Ecosystem (LANL)	5x10 ⁻³	"	
		Small Mammals (Rocky Flats)	7.8x10 ⁻³	B.4.1(25)	
	Weathering Rate	General	10% per year		

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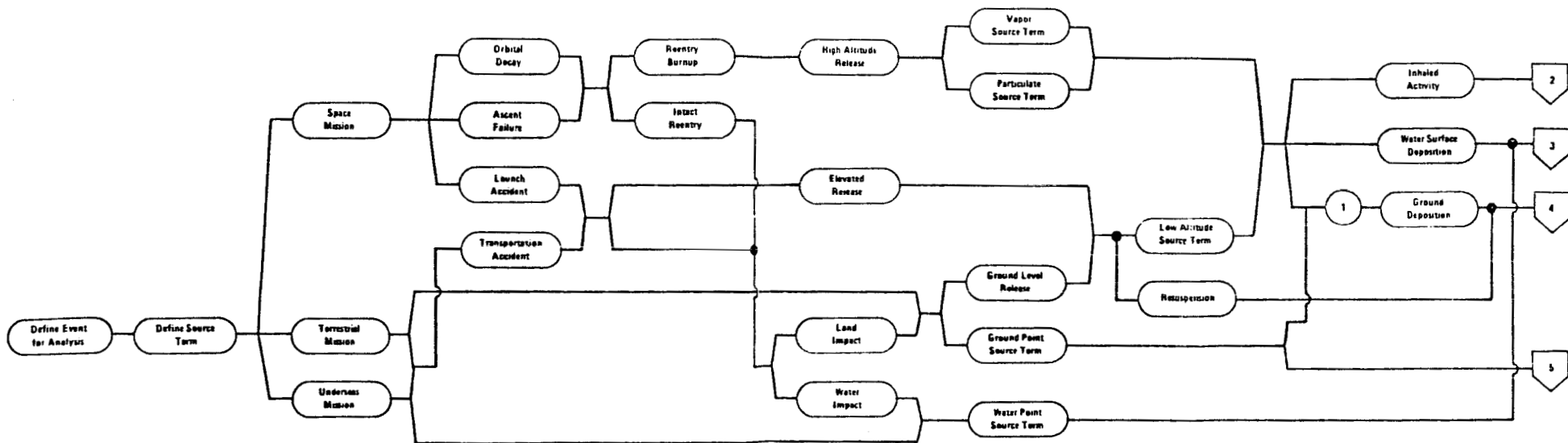


Figure 1.- Potential Plutonium Environmental Exposure Pathways

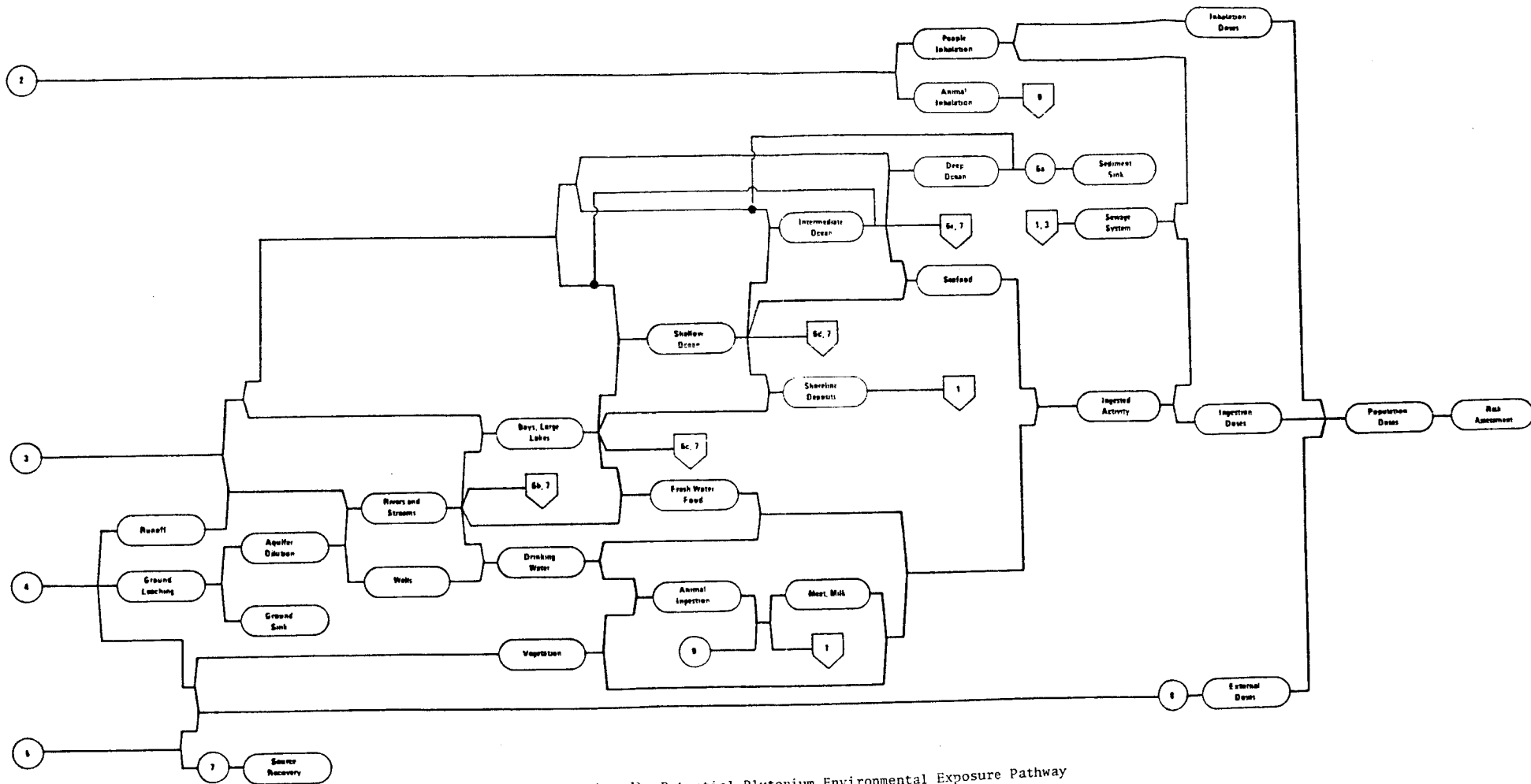


Figure 1.- (Continued) Potential Plutonium Environmental Exposure Pathway

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Summary of Plutonium Environmental Data
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Data Category	Parameter	Applicability	Value	Data Reference	
<u>Terrestrial Ecosystems</u> (Continued)	Watershed Erosion Rate	Savannah River	0.005% per year	B.1(25)	
		Miami River	0.05% per year	"	
		Hudson River	0.05% per year	"	
	Soil Leaching Rate	General		0.1 cm/yr through top 1 cm of soil	B.7(12)
				0.5 cm/yr below top 1 cm of soil	
	Resuspension Factor	Nevada Test Site (Generally applied)		$10^{-4}e^{-0.15 t+10^{-9},m^{-1}}$	B.4.6(9)
			Sandy Grass	10^{-8} to 5×10^{-5}	B.4.6(28)
			Sandy Debris	2×10^{-9} to 10^{-5}	
			Rocky Flats, Co	10^{-9} to 10^{-5}	
			Palmares, Spain	2×10^{-9} to 8×10^{-6}	
New York City			5×10^{-9} to 2×10^{-6}		
Hanford	10^{-10} to 6×10^{-7}				
United Kingdom	2×10^{-9}				
A-3 <u>Aquatic Ecosystems</u>	Dissolution Rate	Grog Fuel		B.2(23)	
		10°C Fresh Water	1440 nCi/m ² -s	"	
		35°C Fresh Water	340 nCi/m ² -s	"	
		10°C Sea Water	60 nCi/m ² -s	"	
		35°C Sea Water	17 nCi/m ² -s	"	
		PPO Fuel			
	10° Fresh Water	90 nCi/m ² -s	"		
	10° Sea Water	1.2 nCi/m ² -s	"		
	K _d ^a	General		10 ⁵	B.1(25)
		Pu all Oxidized		10 ³	"
		Pu all Reduced		10 ⁶	"
		pH Decreases		K _d Decreases	"
		Amount of Organic Carbon Increases		K _d Decreases	"
		Amount of Sulfates Increases		K _d Decreases	B.1(25)
		Amount of Carbonates Increases		K _d Decreases	"
Enewetak (marine)			1.8x10 ⁵	B.5.3(36)	
Enewetak (groundwater)			2.5x10 ⁵	"	
Bikini (marine)		2.2x10 ⁵	"		
Windscale		5x10 ⁴	"		

a. Ratio of concentration in sediment to concentration in water

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Summary of Plutonium Environmental Data
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Data Category	Parameter	Applicability	Value	Data Reference	
<u>Aquatic Ecosystems</u> (Continued)		Humbolt Bay, CA	8x10 ⁴	"	
		Mediterranean	5x10 ⁴	"	
	Seafood Density	Florida Coast	8.6x10 ⁻⁹ g/cm ³ -yr	B.7(45)	
	Concentration Ratio	Plankton		5000	B.5.1(4)
		Benthic Algae and Macrophytes		5000	"
		Benthic Invertebrates		1000	"
		Fish			
		Bottom Feeders		250	"
		Plankton Feeders		25	"
		Piscivorous		5	"
Particle Sink Rates	Pacific Ocean		50-400 m/yr	B.1(24)	
	North American Fresh Water Lakes		10-100 m/yr	B.1(24)	
<u>High Altitude Vapor Release</u>	Residence Half-Times	Mesosphere (45 km)	5 years	B.7(10)	
		Stratosphere (10-45 km)	0.75-2 years	"	
		Troposphere (0-10 km)	5-15 days	"	

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