

May 1975

Actinides in the Environment and Their Uptake by Man

Edward A. Martell

ATMOSPHERIC QUALITY AND MODIFICATION DIVISION
NATIONAL CENTER FOR ATMOSPHERIC RESEARCH
BOULDER, COLORADO

PREFACE

The preparation of this report was carried out in the summer of 1973 as a contribution to a study entitled "Public Health Implications of a Plutonium Energy Economy," sponsored by the Energy Policy Project of the Ford Foundation. Because a contribution on the important biomedical aspects of this study was never completed, the Energy Policy Project did not undertake to publish a final report on this study.

At the suggestion of the Energy Policy Project this portion of the study is being made available for distribution as an NCAR Technical Note. Another portion of the study, a report entitled *Radioactivity in Plutonium, Americium and Curium in Nuclear Reactor Fuel*, is available from its author, Professor Thomas H. Pigford, Department of Nuclear Engineering, University of California at Berkeley, 94720.

Readers interested in recent consideration of cancer risks of plutonium and the actinides are directed to several recent publications (Geesaman, 1968; Tamplin and Cochran, 1974; Bair et al., 1974; Martell, 1975) and to the Proceedings of the U.S. Environmental Protection Agency Hearings on Plutonium Standards, December 1974 and January 1975.

Edward A. Martell
March 1975

CONTENTS

Preface	iii
SECTION I. Actinides in the Environment	
Introduction	1
A. Properties of Pu Effluents	1
B. Isotopic Composition of Effluent Pu	3
C. Global Distribution of Pu	8
D. The Size of PuO ₂ Particles	11
E. Meteorological Transport	15
F. Resuspension of Deposited PuO ₂	16
G. Weathering in Soils	18
H. Plutonium in Aquatic Systems	20
SECTION II. Actinide Uptake by Man	
Introduction	23
A. Plutonium Inhalation	23
B. Uptake of PuO ₂ from Surface Soils and Urban Dust	28
C. Plutonium Uptake by Plants	33
D. Actinides in Food Chains	37
References	41

SECTION I

Actinides in the Environment

Introduction

An adequate assessment of the public health consequences of environmental releases of plutonium (Pu) and other actinides requires an understanding of their redistribution in the environment on all time scales ranging up to thousands of years. Present knowledge is fragmentary and based on less than three decades of experience with Pu fallout from nuclear tests and with Pu contamination from Pu waste handling and Pu weapons accidents. The environmental transport of Pu is governed initially by the physical and chemical properties of liquid and airborne effluents and subsequently by the modification of these properties in terrestrial and marine environments. Public exposure to Pu will result primarily from the inhalation of particulate plutonium dioxide (PuO_2) and, to a lesser extent, from Pu uptake via the food chain. An assessment of inhalation exposure is particularly dependent on the size distribution of PuO_2 in airborne effluents, the air reentrainment of deposited PuO_2 by natural processes, and the recycling of deposited PuO_2 by human and vehicular activity in rural and urban environments. Uptake of actinides via the food chain will result from a complex array of environmental processes including deposition of airborne actinides on plant surfaces, plant uptake from soils, ingestion and translocation of actinides by grazing animals and their consequent distribution in meat and dairy products, and actinide distribution in terrestrial waters and marine environments and their uptake by fish and other marine organisms. Present knowledge and some of the more pertinent available literature bearing on these processes are reviewed here.

A. Properties of Pu Effluents

The extensive literature of the chemistry and metallurgy of Pu is reviewed and summarized in the *Plutonium Handbook* (Wick, 1967). In the case of Pu we are primarily concerned with the products of corrosion and combustion of Pu and its alloys and Pu with uranium (U) in mixed oxide and carbide fuels.

Plutonium is a chemically reactive member of the actinide elements and the metal reacts rapidly with water vapor to form the extremely toxic olive-green powder, PuO_2 . Experience has shown that the presence of oxygen (O_2) in dry air inhibits Pu oxidation and that the most practical atmosphere for handling and storing Pu is freely circulating dry air. The extensive corrosion of both Pu and U when stored in dry inert gases of very low oxygen content remains unexplained. The relatively unpredictable pyrophoric behavior of metallic Pu and of several of its binary and ternary alloys which have been proposed for reactor use pose serious safety problems. Pyrophoric products form on the surface of Pu and many of its alloys during prolonged storage in closed containers, and these products are subject to spontaneous ignition when the container is opened. Similarly, pinhole corrosion of cladded or coated Pu fuel elements will lead to the bulging and blistering of their surfaces, due to hydride and hydroxide formation. The combustion of Pu in air results invariably in the production of finely divided PuO_2 particles (see below). Plutonium metal also reacts with common gases at elevated temperature: with nitrogen or ammonia to form nitrides, with hydrogen to form hydrides, with CO to form carbides, and with CO_2 to form carbides and oxides, etc.

The aqueous solution chemistry of Pu (Katz and Seaborg, 1957; Wick, 1967) is highly complex, due to (a) the multiplicity of oxidation states which may coexist in solution (III, IV, V, and VI), (b) the complex reaction kinetics governing equilibria between oxidation states, (c) the remarkable tendency of Pu (IV) to form complexes, and (d) the influence of alpha radiolysis effects. In low-level Pu liquid waste solutions discharged to the environment one is usually concerned with Pu concentrations $\leq 1.5 \times 10^{-8}$ g per liter and with the two lower oxidation states Pu (III) and Pu (IV) (Polzer, 1971). Thus it is the complexing action and the ion exchange properties of these two oxidation states in dilute solution that govern its subsequent redistribution in surface waters and sediments. The marked complexing and self-chelating action of Pu (IV) gives rise to the formation of large molecular clusters or "unfilterable" colloids under most conditions. Because of its high ionic potential Pu is readily adsorbed on cation exchange resins. On the other hand,

because of its readiness to form anionic complexes Pu also is highly adsorbed on anion exchange resins. Because of these properties Pu released in liquid effluents is highly adsorbed on soils and sediments locally near the point of release and percolates through soils exceedingly slowly under the influence of ground water transport. One possible consequence is illustrated by experience at Hanford where Pu waste solutions have, for more than 22 years, been pumped directly into floorless trenches and allowed to seep down into the soil (U.S. AEC, 1972). Experience has shown that the Pu is highly adsorbed on soils and is, for the most part, retained within a few feet of the point of release. Trench Z-9, used between 1955 and 1962, contains an estimated 100 kg of Pu irregularly distributed in the upper 30 cm of the soil floor. In AEC Report WASH-1520 (1972) it is stated that "Due to the quantity of plutonium contained in the soil of Z-9 it is possible to conceive of conditions which could result in a nuclear chain reaction." The AEC is proceeding to excavate and package the Pu-contaminated soil and plans to recover the Pu--the first Pu mining operation in history. It is suggested that similar problems may develop because of Pu accumulation in the sediments of open holding ponds which receive Pu liquid wastes at the Rocky Flats Plutonium Plant in the western environs of Denver.

B. Isotopic Composition of Effluent Pu

The isotopic composition of the Pu contaminant has considerable bearing on the specific alpha disintegration rate and its change with time, and thus on the alpha activity and dose delivered per Pu particle of given size. For weapons grade Pu processed at Rocky Flats, Owen (1968) reports an isotopic composition as follows: $< 1\%$ ^{238}Pu , $\sim 93\%$ ^{239}Pu , $\sim 6\%$ ^{240}Pu , $< 1\%$ ^{241}Pu , and $< 0.1\%$ ^{242}Pu . The ^{241}Pu , a β^- emitter of 14-year half-life, forms ^{241}Am (americium) which emits 5.5 MeV α particles with a half-life of 460 years.

For Pu used in the Light Water Reactor (LWR) and the Liquid Metal Fast Breeder Reactor (LMFBR), the isotopic composition varies substantially during the course of reactor operation and during various periods

of cooling of the actinide mixture after reactor discharge (Pigford, 1974). The important alpha emitters in the reactor contaminant mixture are ^{236}Pu , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , ^{242}Cm (curium), and ^{244}Cm with decay properties shown in Table 1. Isotopic composition and activities of actinides in reactor fuels have been computed and typical isotopic compositions of Pu, Am, and Cm in the discharge fuel of water and breeder reactors are summarized in Tables 2 and 3. It should be noted that the shorter lived Pu isotopes all increase in abundance relative to ^{239}Pu such that the specific alpha disintegration rate of the separated Pu isotope mixture for discharge fuel would be about 7.15, 7.3, and 3.7 times that of pure ^{239}Pu for U fueled water reactors, U-Pu fueled water reactors, and fast breeders, respectively. Prior to the time of reactor discharge and for more than a year thereafter, ^{242}Cm is the principal alpha emitter among the actinides in the LMFBR fuel.

The presence of ^{242}Cm , ^{244}Cm , and ^{241}Am in the spent reactor fuel mixture gives rise to penetrating neutron and gamma radiation, with most of the neutrons originating from the spontaneous fission of ^{242}Cm and ^{244}Cm (Table 1) and with the 60 keV gamma ray of ^{241}Am as the dominant source of gamma radiation. Thus, even after separation from fission products and neutron activation products, substantial shielding is necessary during the processing, shipping, and handling of the separated actinides.

The high abundance of ^{241}Pu in the discharge fuel mixtures (Table 3) gives rise to a very substantial growth of ^{241}Am in the separated Pu fraction, resulting in a total ^{241}Am activity which is comparable to the total Pu alpha activity in 70 to 80 years when the decay of the parent ^{241}Pu is essentially complete. The presence of the 460-year half-life ^{241}Am and of the 18-year half-life ^{244}Cm complicates the environmental problem. These two actinides will accumulate in the environment both by accidental release of the discharge fuel mixture, or by the direct release of separated Am and Cm. In addition, ^{241}Am will become prominent in aged Pu releases to the environment. Differences in the chemical properties of Pu, Am, and Cm will bring about their fractionation or separation by the action of physical, chemical,

Table 1

Decay Properties of Principal Alpha-Emitting Contaminants

Isotope	Half-life	α Energies (MeV)	Spontaneous fission half-life (years)
^{236}Pu	2.85 years	5.769(69%) 5.722(31%)	-----
^{238}Pu	86.4 years	5.495(72%) 5.452(28%)	4.3×10^{10}
^{239}Pu	24,360 years	5.147(72.5%) 5.134(16.8%) 5.096(10.7%)	5.5×10^{15}
^{240}Pu	6,580 years	5.162(76%) 5.118(24%)	1.22×10^{11}
^{241}Am	460 years	5.477(85%) 5.435(13%)	-----
^{242}Cm	163 days	6.110(73.7%) 6.066(26.3%)	7.2×10^6
^{244}Cm	18.1 years	5.801(76.7%) 5.759(23.3%)	1.4×10^7

Table 2

Curies of Plutonium, Americium, and Curium Radionuclides
Reprocessed Yearly, for a 1000 MWe Power Plant (Pigford, 1974)

Isotope	Uranium fueled water reactor (curies/Year)	Uranium-Plutonium fueled water reactor (curies/Year)	Fast breeder reactor (curies/Year)
^{236}Pu	0.85×10^4	0.402×10^5	0.9
^{238}Pu	7.57×10^4	5.04×10^5	2.68×10^5
^{239}Pu	8.89×10^3	0.300×10^5	0.808×10^5
^{240}Pu	1.288×10^4	1.036×10^5	1.001×10^5
^{241}Pu	3.10×10^6	3.05×10^7	1.342×10^7
^{242}Pu	37	795	292
^{241}Am	6.25×10^3	1.046×10^5	3.712×10^4
$^{242\text{m}}\text{Am}$	1.10×10^2	2.65×10^3	1.869×10^3
^{242}Am	1.10×10^2	2.65×10^3	1.869×10^3
^{243}Am	4.74×10^2	7.98×10^3	1.074×10^3
^{242}Cm	3.13×10^5	2.92×10^6	1.096×10^6
^{243}Cm	1.09×10^2	8.60×10^2	8.31×10^2
^{244}Cm	6.78×10^4	7.36×10^5	2.65×10^4
Total Pu alpha	1.06×10^5	6.78×10^5	4.49×10^5
Total Am alpha	6.83×10^3	1.13×10^5	3.82×10^4
Total Cm alpha	3.80×10^5	3.65×10^6	1.12×10^6

Table 3

Isotopic Composition of Plutonium in Processed Discharge Fuel
(Pigford, 1974)

Isotope	Uranium-fueled water reactor (wt. %)	Uranium-plutonium fueled water reactor (wt. %)	Fast-breeder reactor (wt. %)
^{236}Pu	0.007	0.005	10^{-6}
^{238}Pu	1.8	1.9	0.77
^{239}Pu	59.3	34.2	66.9
^{240}Pu	24.0	31.4	22.4
^{241}Pu	11.1	18.5	6.1
^{242}Pu	<u>3.8</u>	<u>13.9</u>	<u>3.8</u>
	100.0	100.0	100.0

and biological processes in the environment. Americium has an electronic configuration which makes it the heavy homolog of the rare earth element europium (Eu), borne out by the fact that Am forms the divalent oxidation state more readily than other actinides (Edelstein et al., 1967). Compared to its neighbors Pu and Cm, Am is intermediate in its chemical properties, with pronounced differences in the reactions of its oxides and in its ion exchange properties. The ^{241}Am and $^{239,240}\text{Pu}$ contamination in Eniwetok island soils and lagoon sediments are highly fractionated, with $^{241}\text{Am}/^{239,240}\text{Pu}$ activity ratios ranging from 0.25 to 3.5 (U.S. AEC, 1973). The environmental distribution of actinides also is reviewed by Price (1973). Because the total ^{241}Am alpha activity will be comparable to that of the combined Pu isotopes for centuries following environmental contamination by LMFBR or LWR Pu recycle fuels, it is possible that ^{241}Am may be as serious as Pu in its biological and public health effects (see below).

C. Global Distribution of Pu

Plutonium is essentially unknown in nature except for the occurrence of ^{239}Pu as a minor trace constituent (~ 1 part in 10^{14}) in pitchblende (Seaborg and Perlman, 1948). Beginning with the first nuclear explosion, the Trinity Shot near Almagordo, New Mexico, in July 1945, ^{239}Pu and to a lesser extent, ^{238}Pu and ^{240}Pu have been distributed widely over the whole earth. The cumulative fallout distribution and inventory of ^{238}Pu , ^{239}Pu , and ^{240}Pu up to the end of 1970 have been reported by Hardy et al. (1973) and are summarized in Tables 4a and 4b. The accumulation of ^{239}Pu and ^{240}Pu is the combined result of atmospheric nuclear tests by the United States, the U.S.S.R., the United Kingdom, France, and China and does not include that deposited locally within the test areas, including large amounts trapped underground from underground nuclear tests by the United States and the U.S.S.R. Nearly two-thirds of the total ^{238}Pu fallout is the result of the reentry burnup of one satellite payload which included a nuclear generator (SNAP-9A) containing 17,000 curies (Ci) of ^{238}Pu (Harley, 1964). It would appear that the ^{238}Pu activity was almost completely released to the atmosphere upon reentry burnup of the satellite over

the southern Indian Ocean on 21 April, 1964. Subsequently nearly 80% of the ^{238}Pu was deposited within the southern hemisphere.

The total inventory of ^{239}Pu and ^{240}Pu (Table 4b) is an estimated 325 ± 36 kCi from all atmospheric nuclear tests up to late 1970, a total equivalent to ~ 5 metric tons of ^{239}Pu . About 80% of the total is deposited in the northern hemisphere, with the highest levels in surface soils between 30 and 70°N lat. (Table 4a).

The concentrations of ^{239}Pu (and ^{240}Pu) and ^{238}Pu have been measured in surface air only since 1964 and at only a few locations. Harley (1971) has reviewed the fragmentary evidence available. Between 1964 and 1970 the ^{239}Pu concentration in surface air ranged, typically, between 0.02 and 0.5 pCi/(10^3m^3) in both hemispheres (Harley, 1971; Schleien et al., 1970). Prior to 1964 the air concentrations of ^{239}Pu can be estimated from the ^{90}Sr (strontium) air concentration data, using a $^{239}\text{Pu}/^{90}\text{Sr}$ ratio of 0.017 (Harley, 1971). Surface air concentrations of Pu have been substantially higher, locally, in and near nuclear testing sites and plutonium processing facilities. Thus, for example, in areas east of the plutonium-contaminated spill area at the Rocky Flats plant the Pu air concentration ranged from 5 to 350 pCi/(10^3m^3) between 1964 and 1970 (Hammond, 1971). After the Pu spill area was paved with asphalt and an extensive area downwind was covered with clean gravel, the Pu air concentration in the eastern environs of the plant ranged between 0.2 and 6.0 pCi/(10^3m^3), still 10 to 100 times the concurrent Pu concentrations in surface air due to atmospheric nuclear tests (Volchok, 1971). These higher local air concentrations can be attributed to air reentrainment of surface soil contamination by the action of strong winds and other factors (see below).

Some perspective on relative magnitude of soil contamination levels is provided by the data in Table 5. Areas near and downwind of nuclear explosions show only one to two orders of magnitude higher levels than that due to cumulative fallout. By contrast much higher concentrations were deposited locally from the careless handling of Pu wastes at the Rocky Flats Pu plant west of Denver and by the high explosive dispersal of Pu from two nuclear devices near Palomares, Spain. There have been

Table 4a

Cumulative Plutonium in Fallout Distribution up to 1970
(Hardy et al., 1973)

Latitude	$^{239}\text{Pu} + ^{240}\text{Pu}$ (mCi/km ²)	^{238}Pu (Weapons) (mCi/km ²)	^{238}Pu (SNAP-9A) (mCi/km ²)
80 - 70° N	0.36 ± 0.05	0.009 ± 0.001	< 0.001
70 - 60° N	1.6 ± 1.0	0.038 ± 0.025	0.026 ± 0.015
60 - 50° N	1.3 ± 0.2	0.031 ± 0.004	0.013 ± 0.004
50 - 40° N	2.2 ± 0.5	0.053 ± 0.011	0.026 ± 0.011
40 - 30° N	1.8 ± 0.6	0.042 ± 0.014	0.025 ± 0.015
30 - 20° N	0.96 ± 0.07	0.023 ± 0.002	0.011 ± 0.004
20 - 10° N	0.24 ± 0.10	0.006 ± 0.002	0.011 ± 0.004
10 - 0° N	0.13 ± 0.06	0.003 ± 0.001	< 0.001
0 - 10° S	0.30 ± 0.20	0.007 ± 0.005	0.010 ± 0.007
10 - 20° S	0.18 ± 0.05	0.004 ± 0.001	0.036 ± 0.021
20 - 30° S	0.39 ± 0.16	0.009 ± 0.004	0.070 ± 0.042
30 - 40° S	0.40 ± 0.12	0.009 ± 0.003	0.061 ± 0.020
40 - 50° S	0.35 ± 0.21	0.008 ± 0.005	0.069 ± 0.038

Table 4b

Inventory of Plutonium Fallout (Hardy et al., 1973)

	$^{239}\text{Pu} + ^{240}\text{Pu}$ (kCi)	^{238}Pu (Weapons) (kCi)	^{238}Pu (SNAP-9A) (kCi)
Northern Hemisphere	256 ± 33	6.1 ± 0.8	3.1 ± 0.8
Southern Hemisphere	69 ± 14	1.6 ± 0.3	10.8 ± 2.1
Global Total	325 ± 36	7.7 ± 0.9	13.9 ± 2.2

a large number of accidents involving nuclear weapons in the United States and elsewhere (Leitenberg, 1970). How many of such accidents have resulted in a local dispersal of Pu and what measures, if any, have been taken to locate and remove the Pu contaminant in each case have not been disclosed.

Tables 4a and 4b indicate the cumulative Pu fallout up to the end of 1969. Atmospheric nuclear testing by France and China is continuing to add to these totals. The number of ^{238}Pu generators on satellites which are now orbiting the earth and which are destined to release ^{238}Pu to the atmosphere in the course of reentry burnup also has not been disclosed. Note that 80% of the ^{238}Pu from one SNAP-9A generator added about 15% to the total Pu activity in the southern hemisphere; this source may be an important one depending on how many orbiting Pu generators will reenter the earth's atmosphere.

D. The Size of PuO₂ Particles

Plutonium dioxide is the major product of Pu metal corrosion and of low temperature and high temperature combustion of plutonium and its compounds in air. It also is the product usually formed when oxygen compounds of Pu are heated in vacuo or inert atmospheres and when a wide range of Pu compounds are heated in air. Although PuO₂ is usually green, its color varies widely with purity, particle size, and method of preparation. It is a very refractory and highly insoluble material, the more so when prepared at high temperatures. The decreased reactivity of PuO₂ prepared at high temperatures is attributed to the increased perfection of its fluorite (CaF₂) type structure (face-centered cubic) with a calculated density of 11.46. The only other stable oxides of Pu, Pu₂O₃ and Pu₄O₇, are produced only under reducing or other special conditions. Plutonium dioxide is miscible with uranium dioxide, thorium dioxide and other actinide dioxides.

Because it is the size distribution of the insoluble PuO₂ or mixed oxide particles that to a large extent governs their interactions and more particularly, their role in inhalation carcinogenesis, it is pertinent to summarize the evidence on their size distribution for

Table 5

Ranges of Plutonium Contamination in Soils from Various Sources

Location	Event	Deposited $^{238}\text{Pu} + ^{240}\text{Pu}$ (mCi/ m ²)	Reference
20°N - 70°N	Cumulative fallout up to 1970	0.9 - 2.7	Hardy et al., 1973
Nagasaki	Nominal airburst	1.0 - 38	Sakanoue and Tush, 1971
New Mexico downwind of Trinity Shot	19-kiloton tower shot	1.0 - 250	Olafson et al., 1957
Offsite areas around Nevada Test Site	Local fallout from numerous nuclear tests	1.0 - 130	Bliss and Dunn, 1971
Offsite, east of Rocky Flats	Wind resuspension of Pu from spill area	1.5 - 1500	Poet and Martell, 1972 Krey and Hardy, 1970
Palomares, Spain	High explosive dispersal of Pu from two thermo- nuclear devices	> 3 x 10 ⁴ (3 acres) 3 x 10 ² -3 x 10 ⁴ (300 acres)	Langham, 1969

various sources and processes. For inhalation by man, particles of only a few microns diameter and smaller are of primary interest (Hatch and Gross, 1964), although there is some evidence for inhalation of particles as large as 30 μm in diameter (Stuart and Gaven, 1966).

For airborne particles resulting from the accidental combustion of Pu metal in laboratory air at the Rocky Flats plant, Mann and Kirchner (1967) report a PuO_2 particle mass median diameter (MMD) of 0.32 μm , with 97% of the activity associated with submicron sized particles. Stewart (1963) reported on the size distribution of PuO_2 particles generated by the oxidation of Pu metal at room temperature and 120C and on the surface of the liquid metal, under both dry and humid conditions. Slow oxidation at room temperature yielded particles of $\sim 0.5 \mu\text{m}$ MMD under dry conditions and several micrometers MMD and larger under humid conditions. At 123C the pure metal and its δ alloy yielded particles of 1.8 μm and $\sim 10 \mu\text{m}$ MMD, respectively. Of the oxide produced by the combustion of Pu at and above ignition only a small fraction becomes airborne, and the fraction of respirable size decreases with increasing temperature of oxidation. Ettinger et al. (1967) carried out similar experiments for various oxygen concentrations and humidity conditions. They report that size distributions of aerosols over burning Pu metal and Pu alloy ranged from 0.03 to 0.14 μm MMD and, over burning sodium aerosols, ranged from 0.53 to 16.2 μm MMD. The composition of aerosols from the combustion of Pu and Na mixtures varied widely.

Studies of the Pu aerosol size distribution in plutonium laboratory work areas at Los Alamos and Hanford yielded 0.1 to 4.2 μm MMD's, with geometric standard deviations of three or less (Moss et al., 1961; Anderson, 1964). The size distribution of Pu aerosols in normal stack effluents is governed by the exhaust filter system. Currently, multiple stages of HEPA (High Efficiency Particulate Aerosols) commercial filters are in use at the Rocky Flats and at other plutonium facilities in the United States. Evaluation of the effectiveness of such filter installations is incomplete. Little quantitative data is yet available on the efficiency vs particle size for multiple HEPA filter stages or on the changes in effectiveness vs flow rate and period of operation and less

is known about the degree of penetration of particles $\leq 0.1 \mu\text{m}$ in diameter.

The size distribution and radionuclide composition of airborne particles from a LMFBR accident are unpredictable. In a comprehensive discussion of some of the possible consequences of such an accident (Fish et al., 1972) the authors state:

The data from oxidation of plutonium metal and its compounds at and below 1200°C , which are essential to evaluations of hazards in the processing of plutonium fuel, may have very limited applicability to the prediction of aerosol behavior after a release in a sodium-cooled fast reactor. Airborne particles obtained in oxidation experiments have generally been at least 10 to 100 times as large as the agglomerated particles from vaporized PuO_2 or UO_2 . The corresponding mean masses of the particles obtained from the oxidations at 1200°C and below would be 10^3 to 10^6 times those from vaporized fuel. Data on accidental releases of plutonium from other events, such as fires and explosions, may also be of limited applicability.

Many of the complex and variable processes that influence the size distribution and properties of airborne particles resulting from fires, explosions, and other accidents have been discussed by Stewart (1969).

A few measurements of the size distribution of airborne PuO_2 resuspended from contaminated soils east of Rocky Flats (Volchok, 1972) show that from 20 to 40% of the activity is associated with particles of respirable size. The PuO_2 concentrations ranged up to $18 \text{ pCi}/(10^3 \text{ m}^3)$ for wind velocities between 10 and 17 km/h and the few data obtained suggest a possible increase in the respirable fraction with increased air concentration, which in turn increases very substantially at higher wind velocities (see discussion of wind resuspension, below). This evidence is seriously at odds with statements by Langham (undated) that particles of respirable size:

. . . are rare and short-lived in the deposition field . . . are carried down into the soil and fixed or cemented to larger particles by the action of water, . . . those that remain on the surface are difficult to resuspend because of their high threshold drag velocity . . . and significant concentrations will result only when the wind velocity is greater than 10 mph.

Resuspended soil particles and Pu, collected on filters 1 m above the ground surface east of Rocky Flats, were examined by alpha radioautograph techniques (Nathans et al., 1971). Results showed a log normal distribution of active particles with a median $^{239}\text{PuO}_2$ equivalent diameter of $\sim 1.15 \mu\text{m}$. However the PuO_2 on the filters appeared to be associated with soil particles, also log normal in size distribution, with $\sim 10 \mu\text{m}$ median diameter.

Because Pu inhalation exposure of the general population up to the present time has been limited principally to ^{239}Pu from global fallout and, to a lesser extent, to ^{238}Pu from SNAP-9A debris (Table 4a), it is of interest to know the size distribution and alpha activity per particle for these fallout sources. Holland and Volchok (1967) applied alpha radioautograph techniques to SNAP-9A debris samples collected in the stratosphere during the year following the April 1964 satellite failure. Their results showed $^{238}\text{PuO}_2$ equivalent diameters ranging from 5 to 58 μm . During the same period, stratospheric debris from nuclear test sources exhibited $^{239}\text{PuO}_2$ equivalent diameters $\leq 0.03 \mu\text{m}$. In both cases the activity is associated with submicron-sized particles in the lower stratosphere. Although there is appreciable coagulation of stratospheric fallout particles as they mix down into the lower atmosphere, most of the fallout activity in surface air is associated with particles of respirable size. Nuclear debris in local fallout, early fallout, "hot particles," etc., as well as the larger particles produced by the coagulation of fallout particles in the lower atmosphere, involve ^{239}Pu activities per particle corresponding to $^{239}\text{PuO}_2$ equivalent diameters ranging up to about 0.1 μm . If fractionation effects are taken into account (Freiling et al., 1965) the Pu activity of particulate matter from various nuclear debris sources can be estimated approximately from past measurements of fission products constituents and from known $^{239}\text{Pu}/^{90}\text{Sr}$ ratios for unfractionated fallout sources (Harley, 1971).

E. Meteorological Transport

The complex processes that govern the atmospheric transport and deposition of particles released to the atmosphere from normal operations

and accidents at various stages in the plutonium fuel cycle are relatively well understood and are well documented in the published literature. A full review is beyond the scope of this report. For a comprehensive treatment of transport and diffusion of instantaneous cloud sources or continuous plume sources in the lower atmosphere, the reader is referred to *Meteorology and Atomic Energy* (U.S.AEC, 1968). The global transport and deposition of radioactive fallout has been thoroughly documented in the published literature, (WMO, 1961, 1965; U.S.AEC-HASL, 1957-1973; U.S.AEC, 1965; Klement, 1965; American Chemical Society, 1970; CACR-IAMAP, 1966, 1970; UNSCEAR, 1958-1972; JCAE Hearings, 1957, 1959, 1962). The interactions of aerosols in the lower atmosphere, their deposition on surfaces and scavenging by precipitation have been widely discussed (Junge, 1963; Fuchs, 1964; Richardson, 1960; Styra et al., 1968; Mercer et al., 1972; U.S.AEC, 1970). Many aspects of the special problems involved in the release and atmospheric dispersion of radioactive aerosols from nuclear accidents also have been considered elsewhere (Fish et al., 1972; Stewart, 1969; U.S.AEC, 1965).

F. Resuspension of Deposited PuO₂

Among the more complex and less well understood aspects of environmental transport are the processes which govern the resuspension of contaminant deposited on various surfaces. Many aspects of the problem of adhesion and redispersion of particulate contaminants from solid surfaces in laboratory environments have been studied (Fish, 1967). The discussion below is addressed principally to a review of evidence pertinent to the redistribution and air reentrainment of PuO₂ particles deposited on soil surfaces.

The resuspension of deposited PuO₂ by the action of winds, vehicular traffic, and human activity gives rise to an inhalation hazard to occupants of the contaminated and downwind areas. Because PuO₂ particles are chemically stable and highly insoluble, they will accumulate and persist in the surface layer of soils until they are disturbed by physical action. Also, because PuO₂ is quite friable in nature, the larger particles continue to break down into smaller particles during the course of weathering

and aging in soils. The ultimate size distribution of aged PuO particles in soils has not been assessed. The particles will, of course, become attached to soil particles of various size, particularly under moist soil conditions. However it is evident from the size distribution of airborne PuO₂ resuspended from soil surfaces east of Rocky Flats (Volchok et al., 1972) that PuO₂ particles of respirable size remain unattached or become detached from larger particles during the reentrainment process (Smalley, 1970). Gillette et al. (1972) have shown that for particles between 0.6 and 12 μm in diameter the size distribution of airborne soil particles is very similar to that of the soil from which it was derived.

There is a considerable volume of published information on the physics of wind erosion of soil surfaces (Bagnold, 1960; Chepil, 1945, 1946, 1956; Chepil and Woodruff, 1963), the meteorology of dust storms (El-Fandy, 1953) and the size distribution of airborne soil particles (Gillette et al., 1972). Experimental evidence on the resuspension of particulate contaminants from various surfaces under natural and disturbed conditions has been discussed by Stewart (1967). The remarkable variations in the air concentration of PuO₂ resuspended from a Pu-contaminated spill area subjected to gusty winds and vehicular traffic are indicated in a report by Hammond (1971).

The wind reentrainment of soil particles in natural environments usually is initiated by the action of strong winds or gusty winds by the drag forces exerted on surface particles larger than 100 μm in diameter. This action is followed by saltation of the larger particles and air resuspension of the disturbed fines. Soil erosion proceeds more rapidly on dry, irregular surfaces that are relatively free of vegetation-- conditions characteristic of arid and semiarid regions, plowed fields, windswept slopes, etc. Surface irregularities and disturbances produced by grazing animals, vehicular traffic, and human activity all serve to enhance the rate of the soil erosion process.

In health physics surveillance it is common practice to express the relationship between the air concentration of a pollutant and its average

surface concentration in terms of a resuspension factor, K, which is defined as the pollutant concentration per cubic meter in air at a given height above the surface divided by the average surface contamination per square meter. This is useful and practical concept inside laboratories and buildings where one is concerned with contaminants on surfaces and their disturbance and redistribution in confined space. However such a relationship is highly artificial when applied to outside natural environments where the air concentration of a contaminant is unrelated to its local surface distribution. In outside environments air concentrations vary widely with levels and gradients of contamination in upwind areas; with the effects of weathering and aging; with wind velocity and vegetation cover; with disturbance of the contaminant by plowing or vehicular traffic, etc. Thus, for particulate contaminants in open areas resuspension factors range widely, from $K = 3 \times 10^{-9}/m$ for small areas of contamination on grass covered sandy soil under natural conditions to values approaching $10^{-3}/m$ for large areas of contamination disturbed by strong winds or by pedestrians and vehicles (Stewart, 1967). For very large areas with fairly uniform surface soil contamination levels of PuO_2 one can expect that the surface air concentration of PuO_2 will increase with wind velocity in the same manner as the soil dust concentration. The extreme variability of this relationship in semiarid areas typical of eastern Colorado and western Kansas and Nebraska is shown in Fig. 1.

G. Weathering in Soils

Plutonium accumulated in surface soils will usually be present as highly insoluble PuO_2 particles. Soluble Pu compounds will be highly adsorbed due to their chelating and ion exchange properties. Thus Pu will migrate extremely slowly through soils under the influence of percolating ground waters. The remarkable complexity of ground water transport processes and the influence of pH, other chemical factors, patterns of ground water movement, etc., are indicated in a study by Cearlock et al. (1970). Thus Pu can be expected to reside in the surface layer of undisturbed soils for a relatively long time and will persist as a source of Pu for uptake by vegetation and for resuspension as an airborne

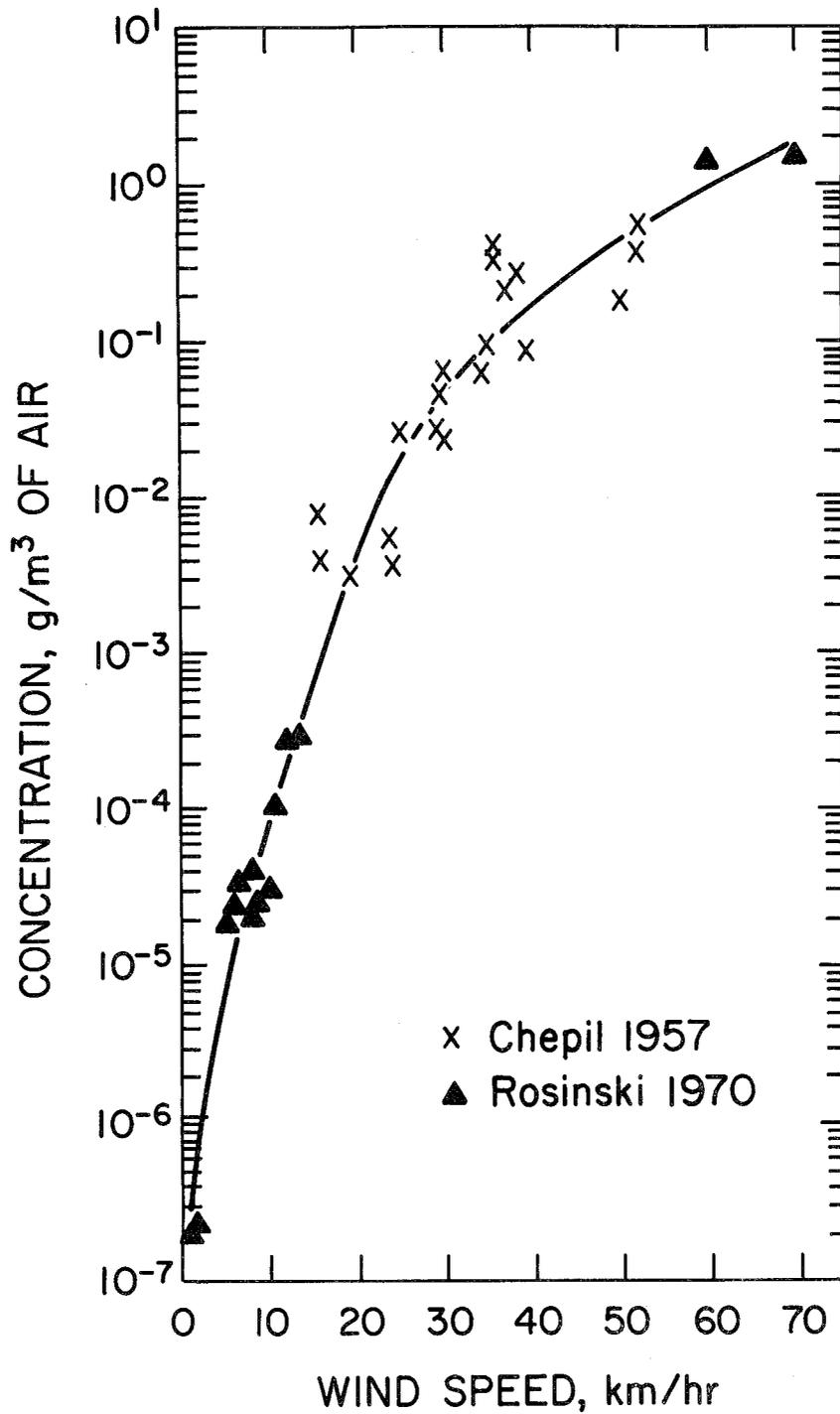


Fig. 1 Air concentration of soil dust 2-3 m above surface vs wind velocity in open grassland areas of eastern Colorado, western Kansas, and western Nebraska (Chepil and Woodruff, 1957; J. Rosinski, personal communication).

contaminant whenever it is disturbed. Aged fallout deposits show very limited downward mixing of Pu below the top few inches of soil. It is likely that such limited weathering of deposited Pu is due to physical transport of particulate PuO₂ or of adsorbed Pu by surface disturbances, by wind and water erosion, by earthworm stirring, by freezing and thawing, etc.

Far less can be said about the long term transport of Pu in underground systems on a time scale of hundreds and thousands of years. On such time scales even the very slow migration under the influence of ground water percolation may transport Pu over substantial distances. It is more likely that underground transport will involve the physical transport of particulate Pu along underground channels and streams or via fault zones, earth cracks, and zones of high porosity. Such possibilities raise serious questions regarding the practice of shallow burial of Pu wastes in steel drums that will disintegrate within decades. The accumulation of Pu in open bottom ditches at Hanford, in the sediments of holding ponds at Rocky Flats, and in underground nuclear explosion cavities may, in part, eventually find its way into underground water systems and aquifers and thus may contaminate surface and underground water supplies.

Less is known about the environmental chemistry and transport of the 460-year half-life ²⁴¹Am, the 18-year ²⁴⁴Cm, and the 163-day ²⁴²Cm. The limited observational data (Price, 1973) indicate a higher mobility for Cm and Am than for Pu in soils, due to more rapid ground water transport as well as to much more substantial uptake in plants, in the order Cm > Am > Pu. Although too little is known about the environmental chemistry and transport of Am and Cm, it is apparent that their uptake in the biosphere is rapid and substantial compared to Pu. Thus uptake of Am and Cm from soils and sediments via the food chain will be an important, if not the dominant, path of uptake of these elements by man.

H. Plutonium in Aquatic Systems

The chemistry of plutonium in natural waters has been reviewed by Andelman and Rozzell (1970), with particular attention to the effects

of pH, ionic strength, Pu concentration, the presence of complexing agents, and other variables. The most stable form of Pu in natural waters is the tetravalent hydroxide, which is highly insoluble and forms polymeric hydrolysis products that exhibit colloidal behavior. The colloidal particles vary in size, ranging between 10^2 and 10^{10} atoms of plutonium, and slowly undergo coagulation to form larger colloid particles. These Pu colloid particles are adsorbed on the surfaces of mineral and other substances and thus accumulate on suspended particles and sediments.

The long half-life of Pu and its concentration as insoluble colloid particles in lake and stream sediments give rise to very serious long-range consequences for the release of low-level liquid wastes containing Pu and other long-lived radionuclides. The special problems involved in the uptake and transport of radionuclides have been considered in detail by Sayre et al. (1963) who conclude:

Available evidence indicates that the sorption of waste materials from solution by stream sediments is the rule rather than the exception. Experiments have demonstrated that sorption of radioactive components from dilute solutions may exceed 90 percent, and that the concentration of radioactivity on the surface of sediment particles may become many thousands of times as great as the concentration in the surrounding liquid.

Because of the extreme insolubility and radiocolloid behavior of $\text{Pu}(\text{OH})_4$ colloid particles, the ultimate degree of concentration of Pu in stream sediments may be even more serious. The situation is similar in ponds and lakes that receive fallout PuO_2 over their surfaces as well as from stream sediments. The buildup of Pu contaminant in the sediments of such closed systems of limited size will continue indefinitely. The long-term rate of accumulation of natural sediments in lakes is a very slow process (Pennington et al., 1973; Krishnaswamy et al., 1971), typically only 0.3 to 0.6 mm per year. It also has been pointed out (Pennington et al., 1973):

. . . that biological activity (rotifers, worms, etc.) in the top layers of aerobic sediments may constitute an important mechanism for mixing (pseudo-diffusion) in the top few cm of sediments.

Thus the accumulation of Pu in sediments for a century or more will remain available for uptake by plants and microorganisms and, in turn, by the fish and wildlife that feed on them. It also is noted that microorganisms can be concentrated and transported up to the water surface by air bubbles and will be injected into the atmosphere when the bubbles burst (Blanchard and Syzdek, 1970; Wilkniss and Bressan, 1971). This process produces very high concentrations of microorganisms in water surface films and in airborne droplets. It also is possible that colloidal particles of Pu in sediments and in suspension, together with some of the particles to which they are attached may be concentrated and transported to surface air by the bubble-bursting process (Horne, 1969).

A recent review of published information on plutonium in marine environments has been carried out by Noshkin (1972) who summarizes the present state of knowledge as follows:

Essentially all the published work has been concerned with levels in the marine environment where plutonium is found widespread among planktonic, pelagic and benthic organisms. The concentrations are higher in organisms feeding on sediment or on surfaces than in those drawing largely on the water itself. Among the species where data are available are a variety of convenient "indicator organisms" for plutonium. There is some evidence that plutonium concentrations are increased in organisms of higher trophic levels. Bone and liver are major repositories for plutonium in marine vertebrates while muscle tissue of both marine vertebrates and invertebrates contain relatively lower concentrations. Plutonium is geochemically separated from both ^{90}Sr and ^{137}Cs in the water column and the sedimentation of ^{239}Pu may be more involved with biological processes than has been found for fallout rare earth isotopes. In marine sediments as in soils, plutonium is more mobile than was originally expected. What little is known of the behavior of plutonium in the marine environment should be used conservatively to assess the behavior and distribution of new plutonium additions derived from sources other than fallout, and even more conservatively in predicting the impact of other transuranics in the aquatic environment. Considerably more understanding of the aquatic radioecology of several of the elements is a major priority, especially since it now appears that when the relative biological effectiveness of alpha vs gamma or beta radiations is considered, fallout ^{239}Pu contributes more than fallout ^{90}Sr or ^{137}Cs to the artificial radiation exposure of many marine species.

SECTION II

Actinide Uptake by Man

Introduction

The problem of public exposure to plutonium and other actinides in the environment is primarily that of the chronic health effects of internal alpha emitters and, more particularly, of insoluble alpha-emitting particles. The principal paths to man are the inhalation of airborne particles of respirable size and the ingestion of particulate and soluble Pu and other actinides via the food chain. The long half-lives of the isotopes and the long-term accumulation of Pu isotopes and ^{241}Am in shallow layers of surface soils and of fresh water and marine sediments will give rise to a steadily increasing level of exposure. The resulting concentrated shallow layers of actinides pose special problems regarding the fate of critical microorganisms that reside in these boundary layers as well as the uptake of actinides in the food chain. The presence of insoluble contaminants in surface soils and their uptake by inhalation of resuspended Pu from soils and surfaces are problems of particular concern in arid and semiarid areas, in urban environments, and for certain occupational groups.

In this discussion attention will be given to some of the main unanswered questions, with suggested approaches to their resolution in some instances. A brief review of some of the pertinent literature, together with references to recent reviews of some aspects of the problems, is also provided.

A. Plutonium Inhalation

The inhalation of airborne PuO_2 appears to be the principal path of uptake of Pu contamination from the environment. Sources of airborne PuO_2 are numerous and include fallout from nuclear tests and from reentry burnup of ^{238}Pu in satellite power sources; airborne effluents from nuclear fuel processing facilities; accidental releases from Pu fires,

explosions and accidents; resuspended Pu from contaminated soils and surfaces; etc.

The physical, chemical, and anatomical factors that influence the inhalation of particulate aerosols and their distribution and clearance have been reviewed by Hatch and Gross (1964) and by Mercer et al., (1972). The ICRP Task Group on Lung Dynamics (1966) adopted a three-compartment model to characterize the pattern of deposition and clearance of particles: the nasopharyngeal (N-P) cavity; the tracheobronchial region (T-B) which includes the trachea and the bronchial tree down to and including the terminal bronchioles; and the pulmonary (P) region, including respiratory bronchioles, alveolar ducts, alveoli, etc. In a review of factors influencing alpha radiation dose estimates in the respiratory tract, Walsh (1970) compares this simplified Task Group model with earlier models and points out its limitations for purposes of detailed dose distribution calculations. Voilleque (1968) has applied the Task Group model to estimates of organ and tissue doses resulting from an acute exposure to inhalation of $^{239}\text{PuO}_2$ particles. The model is useful for showing the initial general pattern of distribution of inhaled particles vs size (Table 6).

The concept of "respirable size" of inhaled particles is somewhat vague but usually refers to the fraction of airborne particulate matter that penetrates to the lower lung, both the T-B and P compartments (Table 6). Based on conservative assumptions, which take into account the considerable variations in the actual lung distribution of particles $< 2 \mu\text{m}$ aerodynamic diameter, the respirable dust curve shown in Figure 2 was recommended at a Los Alamos conference, reported by Ettinger et al. (1972). These authors also point out that for occupational exposures the respirable fraction of PuO_2 particles generally range from 0.75 to 1.00, a considerably greater fraction than that assumed by the ICRP when defining MPCa (maximum permissible air concentration) levels (ICRP, 1960).

The fraction of PuO_2 deposited in the T-B region is rapidly cleared by ciliary action to the gastrointestinal tract. However, a large proportion of insoluble particle deposited in the pulmonary region persists in nonciliated areas, with PuO_2 -particle residence times of one or two

Table 6

Regional Lung Deposition Fractions vs Mass Median
Aerodynamic Diameter,* MMAD (ICRP, 1966; Voilleque, 1968).

MMAD (μm)	Nasopharyngeal (N-P cavity)	Tracheobronchial (T-B) region	Pulmonary (P) region
0.05	0.002	0.08	0.59
0.10	0.01	0.08	0.50
0.20	0.04	0.08	0.41
0.30	0.07	0.08	0.36
0.40	0.10	0.08	0.32
0.50	0.14	0.08	0.30
0.60	0.18	0.08	0.28
0.70	0.21	0.08	0.26
0.80	0.24	0.08	0.25
0.90	0.27	0.08	0.24
1.0	0.30	0.08	0.23
2.0	0.51	0.08	0.16
3.0	0.64	0.08	0.13
4.0	0.72	0.08	0.11
5.0	0.77	0.08	0.10
6.0	0.81	0.08	0.09
7.0	0.84	0.08	0.08

* Aerodynamic diameter: $d_a = (\rho d)^{1/2}$, where ρ is the density and d is the actual diameter of the inhaled particles.

years or more. In this region the insoluble particles undergo slow particle transport to other sites as well as slow dissolution in body fluids. The actual dissolution rates are functions of particle size, temperature conditions of particle formation, and other factors (Thomas, 1972). Raabe et al. (1972) indicate extremely low dissolution rates for submicron $^{239}\text{PuO}_2$ particles preheated to 1000°C , corresponding to dissolution half-lives of 1000 years or more. Thus, for such highly insoluble material, clearance of particles deposited on alveolar surfaces in the pulmonary region must take place almost completely by particle transport: partly by surface transport back to the upper respiratory tract, and partly by transport to the satellite lymph nodes which drain the lungs. Particles accumulated in the respiratory lymph nodes in part escape into the blood stream which, in turn, transports the particles to the liver, spleen, and bone marrow. The pattern of distribution of submicron PuO_2 particles in the liver, spleen and bone is likely to be very similar to that experienced for the colloidal ThO_2 which was injected into the blood stream of large numbers of Thorotrast patients (IAEA, 1968; N.Y. Acad.Sci. 1967).

Highly localized concentrations of PuO_2 (which accumulates in the pulmonary region, in the lymph nodes, and in other organs) are likely to have special significance in relation to the local radiation dose distribution and thus merit special attention. Hatch and Gross (1964) discuss pulmonary clearance processes in detail and point out that alveolar dust plaques or nodules serve as pulmonary dust depots and may account for most of the dust transported to lymph nodes. Regarding transport of dust to other sites, Hatch and Gross state:

Although excessive quantities of inhaled dust build up more-or-less permanent deposits in the lungs and in the satellite lymph nodes, these may not be the only repositories of such dust. Dust particles are found, for example, in the scalene lymph nodes situated at the base of the neck. This is so common that biopsy of these nodes has been utilized as an aid in the diagnosis of a definitive pneumoconiosis. The presence of dust in structures beyond the lymph nodes draining the lungs is an indication that the filtration of lymph by the nodes has been defective. It is therefore not surprising to find dust particles in the reticuloendothelial

cells of spleen, liver, and bone marrow in animals as well as man. Here one must assume that dust particles which somehow escaped the filtering action of various lymph nodes were carried by the lymph fluid into the thoracic duct which, in turn, emptied into the subclavian vein from where hematogenous distribution took place. In man there exists an additional mechanism by which inhaled dust may gain entrance into the blood stream and result in its widespread dissemination. Lymph nodes heavily infiltrated by dust particles may ulcerate into a nearby blood vessel and thereby expose the dust content of the node to the erosive action of the blood current.

Hatch and Gross (1964) also note that the magnitude of the pulmonary dust load influences the rate of clearance as well as the pattern of clearance. Thus the presence of heavy dust loads inhibits the early rapid clearance of particles. And the presence of fibrogenic dusts markedly increases the fraction of the pulmonary deposit which finds its way to the lymph nodes, and thus to the liver, spleen and bone as well. This experience with insoluble dusts would indicate that the retention of inhaled PuO_2 particles and the extent of their local accumulation and the burden of other organs may be significantly enhanced for some occupational groups (asbestos workers, miners, etc.) and for some population groups who work or reside in dusty environments.

The residence time for the P region used by the ICRP Task Group (1966) and Voilleque (1968) appears to be much too short and these authors do not take into account the actual distribution of particles and thus the dose distribution in the P region, in lymph nodes, liver, bone marrow, etc. Alveolar retention half-lives up to 1,500 days are reported for PuO_2 in beagles (Long, 1971). Thomas (1972) has developed an interspecies model to reconcile data from mice, rats and dogs and suggests a half residence time of $\sim 10,000$ days both for the phagocytic elements in the lung and for lymph nodes.

Attempts to estimate the relative PuO_2 concentrations in the lung and lymph nodes for both single and chronic exposures (Thomas, 1968; Long, 1971) are useful as guides to the general distribution of PuO_2 in man. However such estimates may be highly misleading as a basis of risk estimates which must depend on the radiation dose distribution and

thus on the size and specific activity of PuO₂ particles, and their local tissue distribution and persistence, all of which are influenced by the origin of the PuO₂, the age and activity of the individual, environmental and occupational considerations, the combination of exposure conditions involved, etc.

In the final analysis there is no substitute for direct measurements of PuO₂ distribution in man. Adequate techniques are available for radioautograph and radiochemical determination of human organ burdens vs age, occupation, environmental conditions, etc. in the general public, and for evaluation of organ distribution in occupational exposure cases. A comprehensive program of this kind is very much in order.

B. Uptake of PuO₂ from Surface Soils and Urban Dusts

The resuspension of PuO₂ particles accumulated in the surface layers of soils offers an inhalation exposure hazard that varies widely with environmental conditions and is of relatively greater significance in semiarid areas and for vegetation-free surfaces. Dry surface conditions, surface irregularities, strong winds, gusty winds, surface disturbances by human and vehicular activity, all serve to enhance the air concentration of the resuspended particles. However, deep plowing or the natural weathering of deposited PuO₂ particles will serve to reduce the surface soil concentration and diminish the airborne hazard. On roads and other paved surfaces any deposited PuO₂ will be repeatedly resuspended and redeposited, by vehicular traffic, sweeping, and other human activity. In urban areas, with a large proportion of paved surface area there is less opportunity for the weathering and dilution of deposited PuO₂. Thus the airborne concentration of pollutants in resuspended urban dusts will remain high unless the streets and highways are washed off frequently.

The air concentrations of resuspended particulate matter will vary by many orders of magnitude. In addition, the inhalation exposure conditions also will vary widely and will be highest for certain occupational groups and for urban dwellers. Because of the large variations

in inhalation exposure conditions there are serious limitations to the assessment of potential lung burdens for various population groups. Not only do resuspension factors range from 10^{-9} to 10^{-3} /m (see above), but differences in size distributions for PuO_2 from various sources determine the fraction deposited in the pulmonary region of the lungs. The persistence of pulmonary deposits of respirable particles and the pattern of their translocation to other organs are further influenced by the presence or absence of other insoluble dusts (Hatch and Gross, 1964). Thus, the presence of high lung concentrations of submicron fibrous particles will increase the residence time of PuO_2 particles in the P region of the lung and will also increase the fraction of PuO_2 transported to the lymph nodes, liver, spleen, and bone.

An alternative approach to the estimation of human organ burdens that will result from chronic inhalation exposure to PuO_2 contaminated soils and urban dusts is provided by experience with the physical and chemical nature and lung burdens of dust particles for various occupational groups and urban dwellers. Hatch and Gross (1964) have compared the magnitude of cumulative lung burdens vs daily intake of dusts for urban dwellers and for workers in dusty environments, and point out (p.69):

... the experience of a city dweller exposed to a moderately dirty community atmosphere which may contain as much as 1 mg of fine solid particles per cubic meter. Assuming a daily respiratory intake of only 10^3 m³ of air (very light activity) this individual will inhale 10 mg per day or more than 3 g of particulate matter in a year. Over a lifetime, the quantity inhaled may amount to several hundred grams. Despite this, only a few grams of mineral ash will be found in the lungs. Even the lungs of workers exposed to silica or other mineral dusts at the substantial concentrations encountered in the dusty trades are found to contain but a small fraction of the estimated quantity inhaled over a normal working life. Thus, among the South African gold miners exposed to the high levels of dustiness during the early years of operation ($\gg 10^3$ mg/m³), McCrae found the total mineral ash content of silicotic lungs to be no more than 20 g, which may be compared with an estimated intake of well over 1000 g. In contrast to this low percentage of retained mineral, one must assume from the experimental findings presented in Chapter 4 that the initial pulmonary deposition amounted to at least 20% of the inhaled particles.

The long-term accumulation obviously involves a very substantial

fractionation of the inhaled dust particles and thus the material accumulated in the lymph nodes, liver, and spleen is highly enriched in both the smaller and the more fibrogenic of the insoluble submicron particles present in the original airborne dust mixture (Hatch and Gross, 1964).

For these reasons it is the relative abundance of PuO_2 and other highly insoluble particles in airborne dusts of respirable size after resuspension, not the concentration of PuO_2 in surface soils, that provides a reasonably good index of the composition of persistent lung debris and its PuO_2 content that result from long-term inhalation exposure.

The size distribution of resuspended soil particles is appreciably different from that of the soils from which they were derived, as Gillette et al. (1972) have pointed out. For typical soils, less than 25% of the mass is in particles sufficiently small to be resuspended (i.e., particles $< 20 \mu\text{m}$ in diameter). And the size distribution of airborne soil particles is flattened for particles less than $2 \mu\text{m}$ diameter with less than 0.1% of the mass in airborne soil particles between 0.6 and $2.0 \mu\text{m}$ in diameter. The low abundance of the smaller particles is attributed to the high degree of agglomeration of the small clay particles in soils and the large forces necessary to separate them during the saltation and resuspension processes (Smalley, 1970). The cohesive forces between submicron particles in the dry state are poorly understood. However it appears that weak van der Waals-type chemical bonding at the points of particle contact is the principal mechanism, effective only for smaller particles and large contact areas. There are fundamental differences in the areas and strength of contact between quartz particles, clay particles, and other submicron constituents in soils. Particles of PuO_2 , which usually are very dense and spherical (Lippman, 1972), should be expected to exhibit appreciable differences in degree of agglomeration in different soil types and under various conditions. In general, particles near $0.1 \mu\text{m}$ in diameter or less should be almost completely attached to larger particles whereas larger particles of respirable size may remain unattached or become detached more readily.

It is clear from the above that the PuO_2 activity per gram of respirable size airborne dust can differ widely from its concentration in surface soils. Because most sources of PuO_2 produce particles of respirable size, PuO_2 agglomeration in surface soils and its subsequent resuspension will result mainly in a shift in size distribution towards the upper respirable size range and above. Due to size fractionation during the course of coagulation and reentrainment, a level of 1 pCi of PuO_2 per gram of soil may give rise to as much as 10 pCi of PuO_2 per gram of airborne dust particles of respirable size. Further fractionation with respect to particle size, shape, and solubility occurs during the course of deposition and clearance in the deep respiratory tract. Thus, for chronic exposure to surface soil levels of 1 pCi of PuO_2 per gram, the cumulative lung burden of insoluble particulate material may be expected to contain between 10 and 100 pCi of PuO_2 , with comparable burdens in the lymph nodes, liver, and bone.

On the basis of the above considerations it is suggested that, for chronic inhalation exposure, the concentration of PuO_2 per gram of insoluble airborne particles of respirable size may provide a much better criterion for standards than does the PuO_2 concentration of surface air per cubic meter or of deposited PuO_2 per gram of surface soil. Such a standard would relate more directly to ultimate organ burdens and thus, to whatever may be deemed to be acceptable organ burdens of PuO_2 . Standard techniques applicable to the collection of particles of respirable size have been adopted by the British Medical Research Council and by others (Lippman, 1972), (Fig.2).

Compared to PuO_2 fallout from atmospheric nuclear tests, the resuspension of deposited PuO_2 has been responsible for a larger fraction of past inhalation exposure than is generally appreciated. This stems from the fact that 80-90% of fallout Pu has been deposited by rains, with only the remaining 10-20% transported through surface air as dry fallout. By contrast, the resuspension of deposited PuO_2 takes place only with dry surface conditions and is entirely recycled through surface air. For this reason the inhalation exposure and radiation dose commitment for occupants of areas subjected to PuO_2 from stack effluents or resuspended

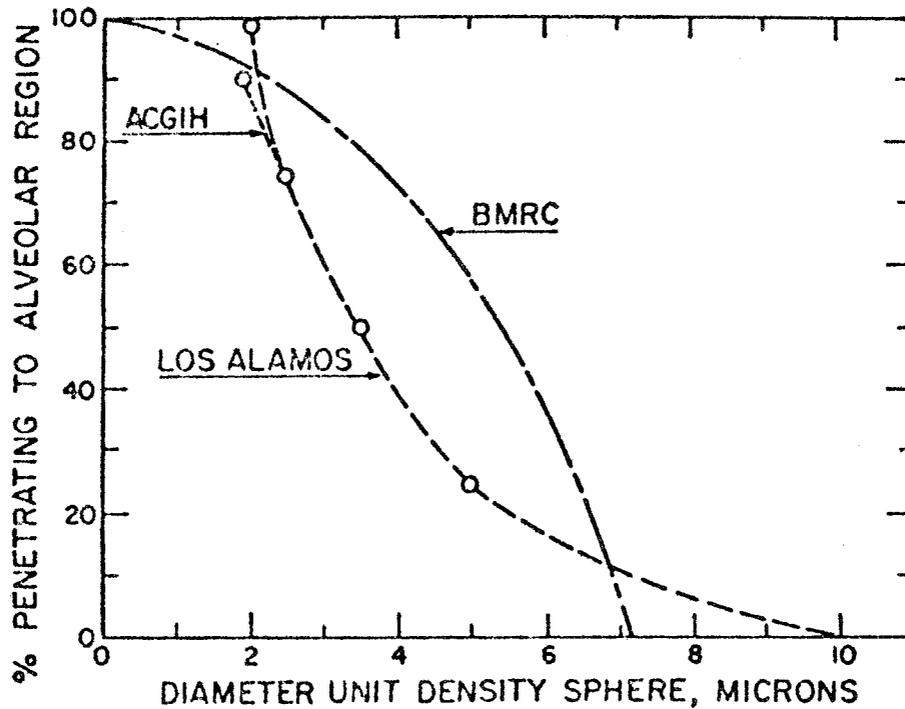


Fig. 2 Respirable fraction of dust particles vs aerodynamic diameter. The BMRC curve is the standard for "respirable dusts," based on the best match of that which passes an ideal horizontal elutriator to experimental data and adopted by the British Medical Research Council. The Los Alamos and ACGIH curves are standards for respirable particles adopted by the U.S. Atomic Energy Commission and the American Conference of Governmental Industrial Hygienists (Lippman, 1972).

PuO₂ is correspondingly much higher than that from fallout sources, per unit of accumulated PuO₂. Such comparisons are, of course, applicable only to the respirable size fraction of the PuO₂ from each source. Thus, for example, for the area east of Rocky Flats it is reasonable to assume that ~ 20% of fallout arrives as dry fallout with about two-thirds of fallout Pu in particles of respirable size, compared to about one-third for resuspended Pu from the Rocky Flats spill area (Volchok et al., 1972). On this basis, in an area east of Rocky Flats which has received ten times as much deposited Pu from the plant as from fallout, the residents have received about 25 times the inhalation dose commitment that the general population has received.

In urban areas the continued erosion of deposited fallout by vehicular traffic greatly enhances public exposure to respirable particles from fallout and other pollutant sources. This may be one of the most important factors contributing to the higher incidence of respiratory diseases, including lung cancer, in urban population groups.

C. Plutonium Uptake by Plants

The limited published information on the plant uptake of plutonium and other actinides from soils has recently been reviewed by Francis (1973) and Price (1973). However these authors have omitted consideration of the deposition and retention of airborne actinides on vegetation surfaces, an important or even dominant source of actinide contamination of plants in some situations and environments (see below).

In an early study of the uptake of Pu from soils by barley plants Jacobson and Overstreet (1968) showed a high degree of Pu uptake on root surfaces or in roots, but very limited translocation of Pu to the plant leaves, with uptake for various oxidation states in the order PuO₂⁺⁺ > Pu⁺⁴ > Pu⁺³. Rediske et al. (1955) found that the translocation to aerial portions of plants increased by an order of magnitude when the pH of soil moisture changed from 7 to 4. Wilson and Cline (1966) showed variations in plant uptake for various soil types, with uptake for acid soils three times greater than for calcareous soil. Romney et al. (1970) showed a seven-fold increase in foliar uptake of

Pu from soils with time over a five-year period and enhanced uptake with DTPA (diethylenetriamine pentaacetic acid) chelate in soils, with increases up to 20 fold.

Cline (1968) reported uptake of ^{241}Am in barley leaves substantially higher than Pu uptake, with concentration factors (i.e., ratio of microcuries per gram of plant and per gram of soil) of 0.003 for Am uptake and of 0.0002 and 0.0001 for Pu uptake from acid and alkaline soils respectively. Hale and Wallace (1970) and Wallace (1972) showed exceedingly large increases in Am uptake in the presence of DTPA and other chelating agents, ranging up to a 200-fold increase in soy beans, and an even higher uptake in bush beans. Price (1973) pointed out that available data on the uptake of curium and neptunium and other considerations indicate the order of plant uptake to be $\text{Cm} > \text{Am} > \text{Pu}$, with neptunium (Np) perhaps similar to Pu. The greater toxicity of americium than plutonium on plants also is noted (Cline, 1968).

The occurrence of natural chelating agents in soils as well as the widespread commercial use of chelating agents in agriculture to enhance plant uptake of essential micronutrients renders the prospect of possible widespread soil contamination by ^{241}Am , ^{244}Cm , and ^{242}Cm very disturbing. The evidence for a systematic increase in plant uptake of the actinides with time (Price, 1973; Romney et al., 1970) coupled with the long half-lives of ^{241}Am , ^{244}Cm , and the Pu isotopes will further magnify the problem. To account for the increased uptake with time, Price (1973) suggests the following mechanisms:

Evidence in the literature implicates three possible mechanisms leading to increased plant uptake with time: first, an increase in availability brought about by organic complexing or chelation by inherent soil properties, by-products of biological (microbial) activity, or products of partial decomposition of organic matter; second, increased root uptake and subsequent translocation to above-ground parts in response to a concentration buildup on root surfaces from an increase in radiocolloid size due to aging; third, a long-term increase in perennial plant concentration from a slow but continual uptake of non-metabolized elements. Each of these ideas is worthy of further study.

The direct deposition of airborne actinide aerosols on vegetation surfaces also is an important source of actinide contamination and, in view of the limited uptake of Pu from soils, relatively more so for Pu than for Am and Cm. The size distribution as well as the chemical properties of the particles influence their deposition and retention on plant surfaces. Particles about 0.3 μm and smaller will accumulate on plant surfaces by diffusive deposition due to their Brownian motion, and some larger particles will be deposited by inertial interception and gravitational sedimentation (Fuchs, 1964). The more soluble constituents of deposited aerosols in part may be adsorbed into the plant interior and in part may also be removed by precipitation. The smaller insoluble particles are likely to persist on plant surfaces unless they are re-suspended in air with the wax particle or plant fiber to which they are attached (Moorby and Squire, 1963). The residence times of deposited aerosols on plant surfaces varies widely with conditions from weeks to months for growing plants and for months on dormant plants and trees (Chamberlain, 1970). Many aspects of the complex and poorly understood processes of the deposition, retention, and regeneration of airborne contaminants on vegetation surfaces are discussed in the literature (Milbourn and Taylor, 1965; Martin, 1964; Buchauer, 1973; Fish, 1972; NAS-NRC, 1963; Bukovac and Wittwer, 1957; Rosinski and Nagamoto, 1965).

The relative magnitude of airborne deposits of Pu on vegetation will vary widely with the nature of the Pu source and local environmental conditions. The possible significance of such surface deposits could be evaluated by comparing the total concentration of Pu in and on the aerial portions of plants in areas subject to resuspension, with those levels expected on the basis of concentration factors (see above) applicable to root uptake alone. A sample of mixed grassy vegetation and a 1-cm-depth soil sample on which it grew were collected in October 1971 (Poet and Martell, 1972) and showed 0.89 ± 0.07 pCi $^{239}\text{Pu}/\text{g}$ of dry vegetation and 6.4 ± 0.9 pCi $^{239}\text{Pu}/\text{g}$ of dry soil. This result corresponds to an apparent concentration factor of 0.14, compared to an expected 10^{-4} for uptake from soils. Vegetation samples collected in off-site areas around Rocky Flats between 1 July and 31 December 1970 ranged

up to 2.5 pCi/g of dry vegetation within 8 km of the plant and up to 0.39 pCi/g beyond 8 km (Dow Chemical Co., 1972). At these greater distances soil concentrations lie below ten times the fallout level (or between 0.3 and 0.03 pCi ^{239}Pu /g of dry soil) in the top 1-cm depth, and lower concentrations averaged over the full root depth (Poet and Martell, 1972). The 1959 levels of ^{239}Pu due to atmospheric fallout on alfalfa and wheat grown in the United States were 0.43 ± 0.04 to 0.80 ± 0.08 pCi/g of alfalfa ash and 0.13 ± 0.02 to 0.67 ± 0.05 pCi/g of wheat ash (JCAE, 1959). Soil levels of Pu averaged over root depths at that time were an estimated 0.005 pCi/g of dry soil, or less.

On the basis of these few observations it is clear that the ^{239}Pu on vegetation due to fallout and to PuO_2 resuspended from contaminated soils contributed 10^3 to 10^5 times higher concentrations of ^{239}Pu in vegetation than that attributable to uptake from soils. The relatively much higher plant uptake of Am and Cm from soils (see above) indicates that both soil levels and airborne particles are important sources for these elements in vegetation.

Although the significance of picocurie quantities of plutonium per gram of dry vegetation is far from clear (see below) the acceptability of such levels is extremely doubtful. It is noted that no standards have been set by the ICRP or NCRP with respect to permissible levels of Pu and other actinides in vegetation as well as soils. Little is known and much research is needed on actinide effects on plant microorganisms essential to the growth and health of various crops, actinide uptake and translocation in animals, actinide distribution in meats and dairy products, actinide distribution in human diets, long-term accumulation in human organs, etc. The problems of the effects of the actinides on microorganisms, vegetation, animals, and man are complicated by the fact that the actinides will usually be present in the form of high specific-activity alpha-emitting particles of micron and submicron diameters, on vegetation surfaces as well as in the surface layers of soils and sediments.

D. Actinides in Food Chains

The presence of particulate actinides on plant surfaces would seem to be the principal source of actinide contamination of cereal grains and leafy vegetables. This certainly will be the case in areas involving airborne effluents from nuclear fuel reprocessing plants, accidents, etc., as well as in semiarid areas with actinide-contaminated soils and under conditions that give rise to the resuspension of particulates. In the case of Pu and Np isotopes, their deposition on plant surfaces in particulate form will usually be the only important source except for vegetation grown on soils with aged deposits of Pu and Np contamination at high concentrations, plants grown in greenhouses, and other very special situations.

For Am and Cm isotopes that are more readily taken up by plants from contaminated soils, both airborne particle deposits and uptake from soils are important sources. The latter source is greatly enhanced when DTPA or other effective chelating agents are present. However, leaching by precipitation and ground water will proceed more rapidly for Am and Cm isotopes than for Pu (Price, 1973). The result will be more rapid weathering of Am and Cm, offsetting the expected increase in plant uptake vs time to some extent.

Due to the interplay of the numerous factors which influence the uptake of Pu, Am, and Cm isotopes in vegetation, their individual and relative concentrations can be expected to vary widely with time and location. In addition, the relative proportions of the particulate component on surfaces and the component taken up from soils in the form of chelated organic complex molecules and as radiocolloids distributed within the interior of plants also can be expected to vary widely with conditions and time for each actinide. These latter variations may influence their uptake and distribution in mammalian systems, primarily because the insoluble particles will be eliminated more rapidly and to a larger extent via the gastrointestinal tract. Thus the biological significance of actinide contamination of vegetation may be highly dependent on the physical and chemical form of the contaminant.

Studies of the uptake and translocation of the actinides in animals show marked differences in the behavior of Pu, Am, and Cm. Taylor (1962) showed that, following oral administration of ^{241}Am and ^{239}Pu , the ^{241}Am was more rapidly cleared from the blood, had 5 to 10 times higher concentration in the liver, was somewhat higher in the kidney and spleen, and was deposited in skeletal tissue in approximately equal amounts. Following intravenous injection of Pu (IV) and Am (III) in several different chemical forms, the distribution and degree of uptake of Pu in various organs varied widely; however little variation was observed for Am (III) (Taylor, 1962). The differences in behavior may be attributed to the fact that at the pH of body fluids Pu (IV) exists as complex ions or in the colloidal state whereas trivalent ^{241}Am ions are very weakly hydrolyzed. For the widely variable proportions of surface particulate matter and root uptake components of Cm, Am, and Pu isotopes in vegetation, the relative rate of uptake of the actinides from the gastrointestinal tract of grazing animals and man will show much larger variations, with uptake in the order $\text{Cm} > \text{Am} > \text{Pu}$. The residence times for the individual actinides in the liver and the extent to which they are translocated into skeletal tissue also will vary widely with chemical form and colloidal state of the ingested contaminant (Lindenbaum and Rosenthal, 1972).

McClellan et al. (1962) have shown that after intravenous injection of various transuranic elements in mature sheep, ^{241}Am and ^{244}Cm followed ^{45}Ca from plasma to milk, with milk:plasma ratios of 10-40 for ^{45}Ca , 2-5 for ^{244}Cm and ^{241}Am , and only 0.02-0.15 for ^{239}Pu , ^{237}Np and ^{233}U . Miller et al. (1972) showed that Pu in milk is quantitatively transferred to cheese products, resulting in a Pu content per gram of cheese about five times that in milk. It would be of some interest to repeat these experiments using milk tagged with Am and Cm because these elements are very substantially enriched relative to Pu in milk.

On the basis of the foregoing it is evident that, although inhalation may be the principal path of Pu to man, there may be many situations in which the ingestion of Pu present as PuO_2 particles on vegetation surfaces may be an important contributing source of Pu uptake by man. This

will apply particularly when cereal grains, cereal grain products, leafy vegetables, etc., are significant components of the diet. By contrast, Cm isotopes and ^{241}Am will be enriched in vegetation due to substantial root uptake from soils as well as from dry deposition on plant surfaces. Cm and Am are further enriched in dairy products, animal liver, and to a lesser extent in other meat products. Thus assimilation via the food chain in a wide variety of foods appears to be the principal path of human uptake for Am and Cm isotopes. In the case of inhaled actinides which are deposited in the upper respiratory tract and transported to the gastrointestinal tract by ciliary activity and mucous flow, the pattern of uptake and accumulation in the liver and bone is similar. In addition, ^{241}Am deposited in the deep respiratory tract is much more rapidly cleared from the lungs than Pu, with greater translocation to the liver and less to the lymph nodes (Sanders, 1972). Undoubtedly the pattern for Cm isotopes is similar to that for ^{241}Am . Thus it would appear that the relative importance of organ burdens shifts from lung and lymph nodes for Pu to liver and bone for Am and Cm.

REFERENCES

The following abbreviations for corporate authors are used throughout the text and in the reference list:

CACR-IAMAP: Commission on Atmospheric Chemistry and Radioactivity, International Association for Meteorology and Atmospheric Physics

HASL: Health and Safety Laboratory, U.S. Atomic Energy Commission

IAEA: International Atomic Energy Agency

ICRP: International Commission on Radiological Protection

JCAE: Joint Committee on Atomic Energy, Congress of the United States

NAS-NRC: National Academy of Sciences, National Research Council

UNSCEAR: United Nations Scientific Committee on the Effects of Atomic Radiation

U.S. AEC: United States Atomic Energy Commission

WMO: World Meteorological Organization

American Chemical Society, 1970: *Radionuclides in the Environment*. Advances in Chemistry Series 93, Washington, D.C., 529 pp.

Andelman, J. B., and T. C. Rozzell, 1970: Plutonium in the water environment. In *Radionuclides in the Environment*, Advances in Chemistry Series 93, American Chemical Society, Washington, D.C., 118-137.

Anderson, B. V., 1964: Plutonium aerosol particle size distribution in room air. *Health Physics* 10, 899-907.

Bagnold, R. A., 1960: *The Physics of Blown Sand and Desert Dunes*. Methuen and Co., Ltd., London, 265 pp.

Bair, W. J., C. R. Richmond, and B. W. Wachholz, 1974: A radiobiological assessment of the spatial distribution of radiation dose from inhaled plutonium. U.S. AEC Report WASH-1320, U.S. Government Printing Office, Washington, D.C., 47 pp.

- Blanchard, D. C., and L. Syzdek, 1970: Mechanisms for the water-to-air transfer and concentration of bacteria. *Science* 170, 626-628.
- Bliss, W., and L. Dunn, 1971: Measurement of Plutonium in Soil around the Nevada Test Site. In *Proceedings of Environmental Plutonium Symposium*, Report LA-4756, Los Alamos Scientific Laboratory, Los Alamos, N. Mex., 89-92.
- Buchauer, M. J., 1973: Contamination of soil and vegetation near a zinc smelter by zinc, cadmium, copper and lead. *Environ. Sci. Technol.* 7, 131-135.
- Bukovac, M. J., and S. H. Wittwer, 1957: Absorption and mobility of foliar applied nutrients. *Plant Physiology* 32, 428-435.
- Cearlock, D. B., R. C. Routson, and C. A. Bryan, 1970: Mathematical simulation of groundwater transport of radionuclides at Hanford. BNWL-SA-3494, Battelle Northwest Laboratory Richland, Wash., 25 pp.
- Chamberlain, A. C., 1970: Interception and Retention of Radioactive Aerosols by Vegetation. *Atmos. Environ.* 4, 57-78.
- Chepil, W. S., 1945: Dynamics of wind erosion, Parts I to V. *Soil Science* 60, 305-320; *ibid.* 60, 397-411; *ibid.* 60, 475-480; *ibid.* 61, 167-177, 1946; and *ibid.* 61, 257-263.
- _____, 1956: Influence of moisture on erodibility of soil by wind. *Soil Sci. Soc. Proc.* 20, 288-292.
- _____, and N. P. Woodruff, 1957: Sedimentary characteristics of dust storms: II. Visibility and dust concentration. *Amer. J. Sci.* 255, 104-114.
- _____, and _____, 1963: The physics of wind erosion and its control. *Advances in Agronomy* 15, Academic Press, N.Y., 211-302.
- Cline, J. F., 1968: Uptake of ^{241}Am and ^{239}Pu by Plants. U.S. AEC Report BNWL-714, Pacific Northwest Laboratory, Richland, Wash. 8.24-8.25.
- CACR-IAMAP, 1966: Proceedings, International Symposium on Atmospheric Chemistry, Circulation and Aerosols. *Tellus* 18, Nos. 2 & 3.
- _____, 1970: Proceedings International Symposium on Atmospheric Trace Constituents and Atmospheric Circulation. *J. Geophys. Res.* 75: No. 9, 1700-1782; No. 12, 2217-2385; No. 15, 2877-3146; and No. 18, 3563-3691.
- Dow Chemical Company, Rocky Flats Division, 1972: *Annual Report: Environmental Safeguard '71*. RFP-ENV-71B, Golden, Colo., 38 pp.

- Edelstein, N., W. Easley, and R. McLaughlin, 1967: Optical and electron paramagnetic resonance spectroscopy of actinide ions in single crystals. In *Lanthanide/Actinide Chemistry*, Advances in Chemistry Series No. 71, American Chemical Society, Washington, D.C., 203-210.
- El-Fandy, M. G., 1953: On the physics of dusty atmospheres. *Quart. J. Roy. Met. Soc.* 79, 284-287.
- Ettinger, H. J., W. D. Moss, and H. Busey, 1967: Characteristics of the aerosol produced from burning sodium and plutonium. *Nuclear Sci. Eng.* 30, 1-13.
- _____, _____, and L. J. Johnson, 1972: Size selective sampling for plutonium-238. *Health Physics* 23, 41-46.
- Fish, B. R., 1967: Surface Contamination. Proceedings of a Symposium held at Gatlinburg, Tennessee, 1964, Pergamon Press, Inc., Elmsford, N.Y., 415 pp.
- _____, 1972: Electrical generation of natural aerosols from vegetation. *Science* 175, 1239-1240.
- _____, G. W. Keilholtz, W. S. Snyder and S. D. Swisher, 1972: Calculation of doses due to accidentally released plutonium from an LMFBR. Oak Ridge National Laboratory, Oak Ridge, Tenn., ORNL-NSIC-74, 123 pp.
- Francis, C. W., 1973: Plutonium Mobility in Soil and Uptake in Plants: A Review. *J. Environ. Qual.* 2, 67-70.
- Freiling, E. C., G. R. Crocker, and C. E. Adams, 1965: Nuclear debris formation. In *Radioactive Fallout from Nuclear Weapons Tests*, AEC Symposium Series 5, Clearinghouse, U.S. Dept. Commerce, Springfield, Va., 1-43.
- Fuchs, N. A., 1964: *The Mechanics of Aerosols*. The MacMillan Company, New York, N.Y., 408 pp.
- Geesaman, D. P., 1968: An Analysis of the Carcinogenic Risk from an Insoluble Alpha-Emitting Aerosol Deposited in Deep Respiratory Tissue. U.S. AEC Documents UCRL-50387, 19 pp. and UCRL-50387 Addendum, 11 pp.
- Gillette, D. A., I. H. Blifford, Jr., and C. R. Fenster, 1972: Measurements of aerosol size distributions and vertical fluxes of aerosols on land subject to wind erosion. *J. Appl. Meteor.* 11, 977-987.
- Hale, V. Q., and A. Wallace, 1970: Effects of chelates on uptake of some heavy metal radionuclides from soil by bush beans. *Soil Sci.* 109, 262-263.
- Hammond, S. E., 1971: Industrial-type operations as a source of environmental plutonium. In Proceedings of Environmental Plutonium

- Symposium, Report LA-4756, Los Alamos Scientific Laboratory, Los Alamos, N. Mex., 25-35.
- Hardy, E. P., P. W. Krey, and H. L. Volchok, 1973: Global inventory and distribution of fallout plutonium. *Nature* 241, 444-445.
- Harley, J. H., 1964: Possible ^{238}Pu distribution from a satellite failure. In U.S. AEC Report, HASL-149, 138-145.
- _____, 1971: Worldwide plutonium fallout from weapons tests. In Proceedings of Environmental Plutonium Symposium, Los Alamos Report LA-4756, 13-19.
- Hatch, T. F. and P. Gross, 1964: *Pulmonary Deposition and Retention of Inhaled Aerosols*, Academic Press, New York, N.Y., 184 pp.
- Holland, W. D., and H. L. Volchok, 1967: The determination of sizes of SNAP-9A abort particles collected at high altitudes. 12th Annual Mtg. Health Physics Society, Washington, D.C.; W. D. Holland, Final Report on AEC Contract No. AT(30-1)3803.
- Horne, R. A., 1969: Bubbles and chemical mass transport from the sea into the atmosphere. Chapter 11(8), *Marine Chemistry* (R. A. Horne, Ed.), John Wiley and Sons, Inc., New York, N.Y.
- IAEA, 1968: Proceedings of an IAEA-WHO Meeting on *The Dosimetry and Toxicity of Thorotrast*, Vienna, 4-7 October 1965, 173 pp.
- ICRP, 1960: Report of Committee II on permissible dose for internal radiation (1959). *Health Physics* 3, 1-380.
- ICRP Task Group on Lung Dynamics, 1966: Deposition and retention models for internal dosimetry of the human respiratory tract. *Health Physics* 12, 173-207.
- Jacobson, L., and R. Overstreet, 1968: The uptake by plants of plutonium and some products of nuclear fission adsorbed on soil colloids. *Soil Sci.* 65, 129-134.
- JCAE Hearings, 1957: *The Nature of Radioactive Fallout and its Effects on Man*, 2,065 pp; 1959: *Fallout from Nuclear Weapons Tests*, 2,618 pp; 1962: *Radiation Standard Including Fallout*, 1,040 pp; U.S. Government Printing Office, Washington, D.C.
- Junge, C. E., 1963: *Air Chemistry and Radioactivity*, Academic Press, Inc., New York, N.Y., 382 pp.
- Katz, J. J., and G. T. Seaborg, 1957: *The Chemistry of the Actinide Elements*, John Wiley and Sons, Inc., New York, N.Y., 508 pp.

- Klement, A. W., Jr., 1965: *Radioactive Fallout from Nuclear Weapons Tests*, U.S. AEC Symposium Series 5, CONF-765, Clearinghouse, U.S. Dept. of Commerce, Springfield, Va., 953 pp.
- Krey, P. W., and E. P. Hardy, 1970: Plutonium in soil around the Rocky Flats plant. U.S. AEC Report, HASL-235, 44 pp.
- Krishnaswamy, S., D. Lal, J. M. Martin, and M. Meybeck, 1971: Geochronology of lake sediments. *Earth Planet. Sci. Lett.* 11, 407-414.
- Langham, W. H., 1969: The problem of large-area plutonium contamination. U.S. Dept. of Health, Education and Welfare, Bureau of Radiological Health Seminar, Paper No. 002, 8 pp.
- _____, undated: *Biological Consideration of Nonnuclear Incidents Involving Nuclear Warheads*. Lawrence Radiation Laboratory Report, UCRL-50639, 37 pp.
- Leitenberg, M., 1970: So far so good. *Environment* 12, 2635.
- Lindenbaum, A., and M. W. Rosenthal, 1972: Deposition patterns and toxicity of plutonium and americium in liver. *Health Physics* 22, 597-605.
- Lippman, M., 1972: Aerosol sampling for inhalation hazard evaluations. In *Assessment of Airborne Particles* (T. T. Mercer, P. E. Morrow and W. Stober, Eds.), Charles C. Thomas, Publ., Springfield, Ill., 449-464.
- Long, A. B., 1971: Plutonium inhalation: The burden of negligible consequence. *Nuclear News*, 69-73.
- Mann, J. R., and R. A. Kirchner, 1967: Evaluation of lung burden following acute inhalation exposure to highly insoluble PuO₂. *Health Physics* 12, 877-882.
- Martell, E. A., 1975: Basic considerations in the assessment of the cancer risks and standards for internal alpha emitters. In *Proceedings*, U.S. Environmental Protection Agency, Hearings on Plutonium Standards (in press).
- Martin, W. E., 1964: Losses of Sr⁹⁰, Sr⁸⁹ and I¹³¹ from fallout contaminated plants. *Radiation Botany* 4, 275-284.
- McClellan, R. O., H. W. Casey, and L. K. Bustad, 1962: Transfer of some transuranic elements to milk. *Health Physics* 8, 689-694.
- Mercer, T. T., P. E. Morrow, and W. Stober, 1972: *Assessment of Airborne Particles*, Charles C. Thomas, Publ., Springfield, Ill., 540 pp.

- Milbourn, G. M., and R. Taylor, 1965: The contamination of grassland with radioactive strontium - I: Initial retention and loss. *Radiation Botany* 5, 337-347.
- Miller, C. L., J. G. Payne, Jr., E. W. Bretthauer, and A. A. Moghissi, 1972: Transfer of plutonium from milk into cheese. *Health Physics* 22, 563-565.
- Moorby, J., and H. M. Squire, 1963: The loss of radioactive isotopes from the leaves of plants in dry conditions. *Radiation Botany* 3, 163-167.
- Moss, W. D., E. C. Hyatt, and H. F. Schulte, 1961: Particle size studies on plutonium aerosols. *Health Physics* 5, 212-218.
- Nathans, M. W., H. C. Shaw, and W. D. Holland, 1971: The size distribution and plutonium concentration of particles from the Rocky Flats Area. Final Report on AEC Contract AT(30-1)4290, TLW 6101R, 42 pp.
- NAS-NRC, 1963: *The Behavior of Radioactive Fallout in Soils and Plants*. Publication 1092, National Science Foundation, Washington, D.C., 32 pp.
- New York Academy of Science, 1967: Proceedings of conference on distribution, retention and late effects of thorium dioxide. In *Annals N.Y. Acad. Sci.* 145, Art. 3, 523-858.
- Noshkin, V. E., 1972: Ecological aspects of plutonium dissemination in aquatic environments. *Health Physics* 22, 537-549.
- Olafson, J. H., H. Nishita, K. H. Larson, 1957: *The Distribution of Plutonium in Soils of Central and Northeastern New Mexico as a Result of the Atomic Bomb Test of July 16, 1945*. University of California Report on Contract AT(904-1)-GEN-12, UCLA-406 (declassified), 25 pp.
- Owen, J. B., 1968: *Control of Personnel Exposures to External Radiations in a Plutonium Chemical Plant*, Dow Chemical Company, RFP-1254, 9 pp.
- Pennington, W., R. S. Cambray, and E. M. Fisher, 1973: Observations on lake sediments using fallout ^{137}Cs as a tracer. *Nature* 242, 324-326.
- Pigford, T. H., 1974: *Radioactivity in Plutonium, Americium and Curium in Nuclear Reactor Fuel* (A Ford Foundation, Energy Policy Project Study), Dept. of Nuclear Engineering, University of California, Berkeley, Calif. (94720), 58 pp.
- Poet, S. E., and E. A. Martell, 1972: Plutonium-239 and americium-241 contamination in the Denver area. *Health Physics* 23, 537-548.

- Polzer, W. L., 1971: Solubility of plutonium in soil/water environments. In *Proceedings of the Rocky Flats Symposium on Safety in Plutonium Handling Facilities*, AEC Report CONF-710401, Health and Safety, TID-4500, 411-429.
- Price, K. R., 1973: A review of transuranic elements in soils, plants and animals. *J. Environ. Qual.* 2, 62-66.
- Raabe, O. G., H. A. Boyd, G. M. Kanapilly and R. K. Latven, 1972: A study of the in vitro solubility of $^{239}\text{PuO}_2$. *Annual Report on the Fission Product Inhalation Program*, Lovelace Foundation, Albuquerque, N. Mex., 29-35.
- Rediske, J. H., J. F. Cline and A. A. Selders, 1955: The adsorption of fission products by plants. U.S. AEC Report HW-36734, 1-17.
- Richardson, E. G., 1960: *Aerodynamic Capture of Particles*, Pergamon Press, Inc., Elmsford, N.Y., 200 pp.
- Romney, E. M., H. M. Mork, and K. H. Larson, 1970: Persistence of plutonium in soils, plants and small mammals. *Health Physics* 19, 487-491.
- Rosinski, J., and C. T. Nagamoto, 1965: Particle deposition on and reentrainment from coniferous trees. *Kolloid-Z. und Z. Polymere* 204, 111-119.
- Sanders, C. L., 1972: Deposition patterns and the toxicity of transuranium elements in the lung. *Health Physics* 22, 607-615.
- Sakanoue, J., and T. Tshu, 1971: Plutonium content of soil at Nagasaki. *Nature* 234, 92-93.
- Sayre, W. W., H. P. Guy, and A. R. Chamberlain, 1963: Transport of radionuclides by streams. Geological Survey Professional Paper 433-A, U.S. Government Printing Office, Washington, D.C., 35 pp.
- Schleien, B., J. A. Cochran, and P. J. Magno, 1970: Sr-90, Sr-89, Pu-239 and Pu-238 concentrations in ground-level air, 1964-1969. *Environ. Sci. Technol.* 4, 598-602.
- Seaborg, G. T., and M. L. Perlman, 1948: Search for elements 94 and 93 in nature; presence of $^{94}\text{Pu}^{239}$ in pitchblende. *J. Am. Chem. Soc.* 70, 1571-1573.
- Smalley, I. J., 1970: Cohesion of soil particles and the intrinsic resistance of simple soil systems to wind erosion. *J. Soil Sci.* 21, 154-161.
- Stewart, K., 1963: The particulate material formed by the oxidation

- of plutonium. In *Progress in Nuclear Energy*, Series LV, Vol. 5, 525-579.
- _____, 1967: The resuspension of particulate material from surfaces. In *Surface Contamination* (B. R. Fish, Ed.), Pergamon Press, Inc., Oxford, 63-74.
- _____, 1969: Principal characteristics of radioactive contaminants which may appear in the atmosphere. In *Progress in Nuclear Energy Series, XII, 2, Part 1*, 321-360.
- Stuart, B. O., and J. C. Gaven, 1966: Large particle inhalation studies. In *Pacific Northwest Laboratory 1965 Report in the Biological Sciences*, BNNL-280, Battelle Northwest Laboratory, Richland Wash., 49-52.
- Styra, B., C. Garbaliuskas, and V. Lujanas, 1968: *Atmospheric Scavenging of Radioisotopes*, Vilnyas (trans., D. Lederman), Keter Press, Jerusalem, 1970 (Avail., U.S. Dept. of Commerce), 250 pp.
- Tamplin, A. R., and T. B. Cochran, 1974: *Radiation Standards for Hot Particles*. National Resources Defense Council Report, Washington, D.C., 52 pp.
- Taylor, D. M., 1962: Some aspects of the comparative metabolism of plutonium and americium in rats. *Health Physics* 8, 673-677.
- Thomas, R. G., 1968: Transport of relatively insoluble materials from lung to lymph nodes. *Health Physics* 14, 111-117.
- _____, 1972: In vivo solubility of intramuscularly injected ^{51}Cr - ^{239}Pu oxide particles. *Annual Report on the Fission Product Inhalation Program*, LF-45, Lovelace Foundation, Albuquerque, N. Mex., 55-60.
- _____, 1972: An interspecies model for retention of inhaled particles. In *Assessment of Airborne Particles*, (T. T. Mercer, P. E. Morrow and W. Stober, eds.), Charles C. Thomas, Publ., Springfield, Ill., 405-425.
- UNSCEAR Reports, 1958-1972: United Nations, New York and Geneva.
- U.S. AEC, Health and Safety Laboratory, 1957-1973: Reports HASL-1 through HASL-274, Clearinghouse, U.S. Dept. of Commerce, Springfield, Va.
- _____, 1965: *International Symposium on Fission Product Release and Transport under Accident Conditions*, Oak Ridge, Tenn., 5-7 April 1965; U.S. AEC Report CONF-650407, Vols. 1 & 2, TID-4500, 43rd edn., U.S. Dept. of Commerce, Springfield, Va., 1,228 pp.
- _____, 1965: *Radioactive Fallout from Nuclear Weapons Tests*. AEC Symposium Series 5, CONF-765, U.S. Dept. of Commerce, Springfield, Va., 22151, \$6.50, 953 pp.

- _____, 1968: *Meteorology and Atomic Energy*, (D. H. Slade, Ed.). TID-24190, U.S. Dept. of Commerce, Springfield, Va., 22151 (\$3.00), 455 pp.
- _____, 1970: *Symposium Series 22 Precipitation Scavenging*. U.S. Dept. of Commerce, Springfield, Va. 22151, 499 pp.
- _____, 1972: *Environmental Statement, Contaminated Soil Removal Facility*, Richland, Wash., WASH-1520, 36 pp.
- _____, 1973: *Enewetak Radiological Survey*, Report NVO-140, Vol. I, October; U.S. AEC, Nevada Operations Office, Las Vegas, Nev., 736 pp.
- Voilleque, P. G., 1968: Calculation of organ and tissue burdens and doses resulting from an acute exposure to a radioactive aerosol using the ICRP Task Group *Report on the Human Respiratory Tract*. U.S. AEC Idaho Operations Office, IDO-12069, 44 pp.
- Volchok, H. L., 1971: Resuspension of plutonium-239 in the vicinity of Rocky Flats. In *Proceedings of Environmental Plutonium Symposium*, Report LA-4756, Los Alamos Scientific Laboratory, Los Alamos, N. Mex., 99-103.
- _____, R. Knuth, and M. T. Kleinman, 1972: *Plutonium in the Neighborhood of Rocky Flats, Colorado: Airborne Respirable Particles*. U.S. AEC, HASL-246, I-2 to I-17.
- Wallace, A., 1972: Increased uptake of ^{241}Am by plants caused by the chelating agent DTPA. *Health Physics* 22, 559-562.
- Walsh, P. J., 1970: Radiation dose to the respiratory tract of uranium miners--A review of the literature. *Environ. Res.* 3, 14-36.
- Wick, O. J., 1967: *Plutonium Handbook, A Guide to the Technology*, Vol. I and II, Gordon and Breach, Sci. Publ. Inc., 966 pp.
- Wilkniss, P. E., and D. J. Bressan, 1971: Use of radionuclides to evaluate the transport of pollutants from the hydrosphere into the atmosphere. In *Nuclear Techniques in Environmental Pollution*, International Atomic Energy Agency, Vienna, 297-307.
- Wilson, D. O., and J. F. Cline, 1966: Removal of plutonium-239, tungsten-185 and lead-210 from soils. *Nature* 209, 941-942.
- WMO, 1961: Technical Note No. 43, *Meteorological Factors Influencing the Transport and Removal of Radioactive Debris*. WMO-No.111.TP.49, Geneva, Switzerland, 171 pp.
- _____, 1965: Technical Note No. 68, *Meteorological Aspects of Atmospheric Radioactivity*. WMO-No. 169.TP.83, Geneva, Switzerland, 194 pp.

