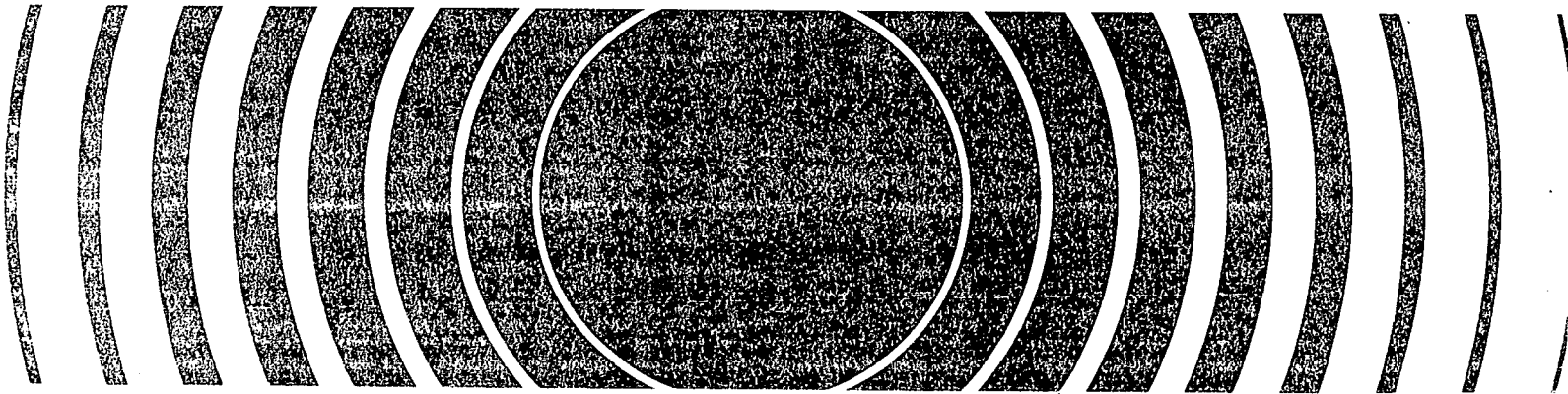




Transuranium Elements

Volume 1 Elements Of Radiation Protection



TRANSURANIUM ELEMENTS

VOLUME I

ELEMENTS OF RADIATION PROTECTION

BY

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PREFACE

This document was developed over a number of years as the technical basis for guidance or criteria for addressing the problem of environmental contamination by plutonium and other transuranium elements. It is now published independent of such guidance or criteria, and is intended to serve as a useful compendium of information relevant to the making of informed decisions on how to deal with such contamination. It reflects the author's personal views and is not intended to provide a comprehensive summary of the subject.

Volume I presents an overview of selected information on the chemical and physical properties of the transuranium elements, the biological effects and risks of exposure, the major environmental transport mechanisms, and the contamination in the environs of a number of sites in the United States. Several of these chapters were prepared during the 1970's and have not been extensively revised, because relatively little new and different information has been published since that time.

Volume II presents the general background relevant to developing criteria for site-specific remedial action options. The information is intended to provide an overall perspective on the considerations necessary for the decision-making process and should not be viewed as giving specific instructions on how to address a specific situation of environmental contamination. The information should be used in an advisory context only and not for purposes of implementation of regulations.

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1. INTRODUCTION

The chemical elements with atomic number greater than uranium (atomic number 92) are known as transuranium elements. They are entirely man-made or present in nature in only infinitesimally small amounts. The most important of these elements is plutonium. Other transuranium elements of importance include neptunium, americium, curium, and californium.

The transuranium elements, starting with neptunium (atomic number 93) are members of the actinide series, which also includes the naturally occurring radioactive elements actinium, thorium, protactinium and uranium, and consists of elements with properties that stem from partial vacancies in the 5f electron shell (Fig 1-1). The chemical behavior of the series is somewhat similar to the lanthanide series which is characterized by a similar electron structure. However, the shielding of the 5f electrons by outer electrons is less effective than that of 4f electrons; thus the chemical properties of the actinides are more complicated than those of the lanthanides. Although the latter exist primarily in the III oxidation state and exhibit ionic bonding, the actinides (through plutonium) can exist in multiple oxidation states. Because of their extreme reactivity, the II and VII oxidation states are not likely to be encountered in the environment. The oxidation-reduction behavior of the triad U-Np-Pu is complicated, and multiple oxidation states can coexist in solution. From the point of view of geochemical behavior, the actinides can be considered the higher analogue of the lanthanides.

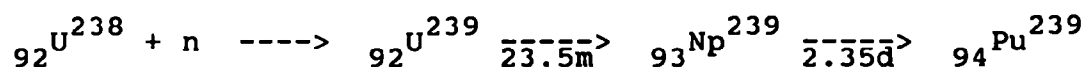
Plutonium is formed by a nuclear reaction, in which neutrons captured by uranium-238 atoms cause the formation of uranium-239, which then decays radioactively to form plutonium-239.

FIGURE 1-1

PERIODIC CHART OF THE ELEMENTS																	
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57-71 La* Series	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89-103 Act Series	(104)	(105)	(106)	(107)	(108)										

*Lanthanide Series	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
†Actinide Series	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 Lw	(103)

These nuclear reactions are represented by the following equations:



The sequence is initiated by fissioning of a uranium-235 nucleus. Elements with atomic numbers greater than plutonium are formed by successive neutron capture reactions. The amounts of the higher isotopes produced depend on the type of reactor and how it is operated, but generally do not exceed a small fraction of the plutonium.

The complex interactions between the various oxidation states of neptunium and plutonium are partly governed by their total concentration in solution. When concentrations are sufficient, disproportionation reactions between oxidation states are common. However, such concentrations are unlikely to be found in the environment, and the stable oxidation states will be a function of the chemical environment, e.g., the presence of oxidizing or reducing agents and complexing ligands. Standard oxidation-reduction potentials can be used to predict the stability field for various plutonium species.

The element plutonium is a silvery-white metal, atomic number 94 in the actinide series. There are at least sixteen different isotopes of plutonium, of which the four most important are: plutonium-238 (half-life, 86.4 years), plutonium-239 (half-life, 24,390 years), plutonium-240 (half-life, 6580 years), and plutonium-241 (half-life, 13.2 years). Plutonium has a melting point of 640°C and a boiling point of 3327°C. It oxidizes rapidly on exposure to air to form plutonium dioxide (PuO_2). Plutonium metal exhibits six different crystalline forms, each in a well-defined temperature range, with densities between 16.00 and 19.86 grams per cubic centimeter.

At room temperature, the most stable oxide is plutonium dioxide. Plutonium dioxide is a highly refractory material which melts at 2200-2400°C and is difficult to dissolve by normal methods. The behavior of PuO₂ particles in environmental and biological systems is greatly influenced by their size and temperature of formation.

The chemistry of plutonium is very complex and many different chemical species often co-exist. Plutonium has four oxidation or valence states, ranging from III to VI, and forms stable compounds with all the nonmetallic elements except the rare gases. In aqueous solutions, Pu(III) is oxidized into Pu(IV), which is the most stable state. The compounds PuF₄, Pu(IO₃)₄, Pu(OH)₄ and plutonium oxalate are insoluble in water. The chlorides, nitrates, perchlorates, and sulfates are soluble in water. The most important compounds in processing of plutonium are PuF₄, Pu(NO₃)₄, and PuO₂.

Plutonium (IV) ions complex readily with organic and inorganic compounds. The stability of complexes decreases in the order Pu(IV) > Pu(III) > Pu(VI) > Pu(V). It is, therefore, probable that most if not all plutonium in environmental and biological systems is in the Pu(IV) state. Plutonium ions in solution rapidly hydrolyse and form polymers. The tendency to hydrolyse decreases in the order Pu(IV) > Pu(VI) > Pu(III) > Pu(V). Hydrolysis of Pu(IV) can result in the formation of relatively insoluble polymers, a process which is only slowly reversible.

In biological systems, ionic plutonium can be expected to undergo three kinds of reaction:

- o Hydrolysis to yield colloidal or polymeric species
- o Complexing by proteins and other macromolecules
- o Complexing (chelating) with cell components of small molecular weight (such as citrate or amino acids)

The fissionable property of plutonium makes it an important source of nuclear energy. When a neutron is absorbed by the plutonium nucleus, it splits (fissions) to form two atoms of about half the atomic number of plutonium and releases an enormous amount of energy in the form of heat. This energy is equivalent to the difference in mass of the reactants and the products of the reaction, and is given by Einstein's mass-energy equation

$$E = mc^2$$

The nuclear energy released from one pound of plutonium is equivalent to the thermal energy from burning of 3,000,000 pounds of coal.

The formation-decay scheme for the most important transuranium isotopes is shown in Fig 1-2. With the exception of Pu-241, the nuclides are all alpha-emitters and most of them undergo spontaneous fission. Particle emissions are usually accompanied by X-ray and gamma-ray emissions of varying energies, which can be utilized for the detection of transuranium nuclides in a wide range of circumstances.

The transuranium elements are formed in nuclear power reactors. The amount of radioactivity and the mass of these radionuclides present in spent fuel from a light-water reactor are shown in Table 1-1. The total quantities of the more significant transuranium elements generated by the end of 1978 were estimated as: neptunium - 4.5 tons; plutonium - 77 tons; americium - 1 ton; and curium - 260 kg. A projection of total plutonium inventories is shown in Figure 1-3. Reliable projections are difficult because of the present uncertain status of the nuclear power industry, but it has been estimated that more than 2400 tons of plutonium would be produced by the commercial industry by the year 2000. Additional amounts are produced by specially operated reactors for nuclear weapons production.

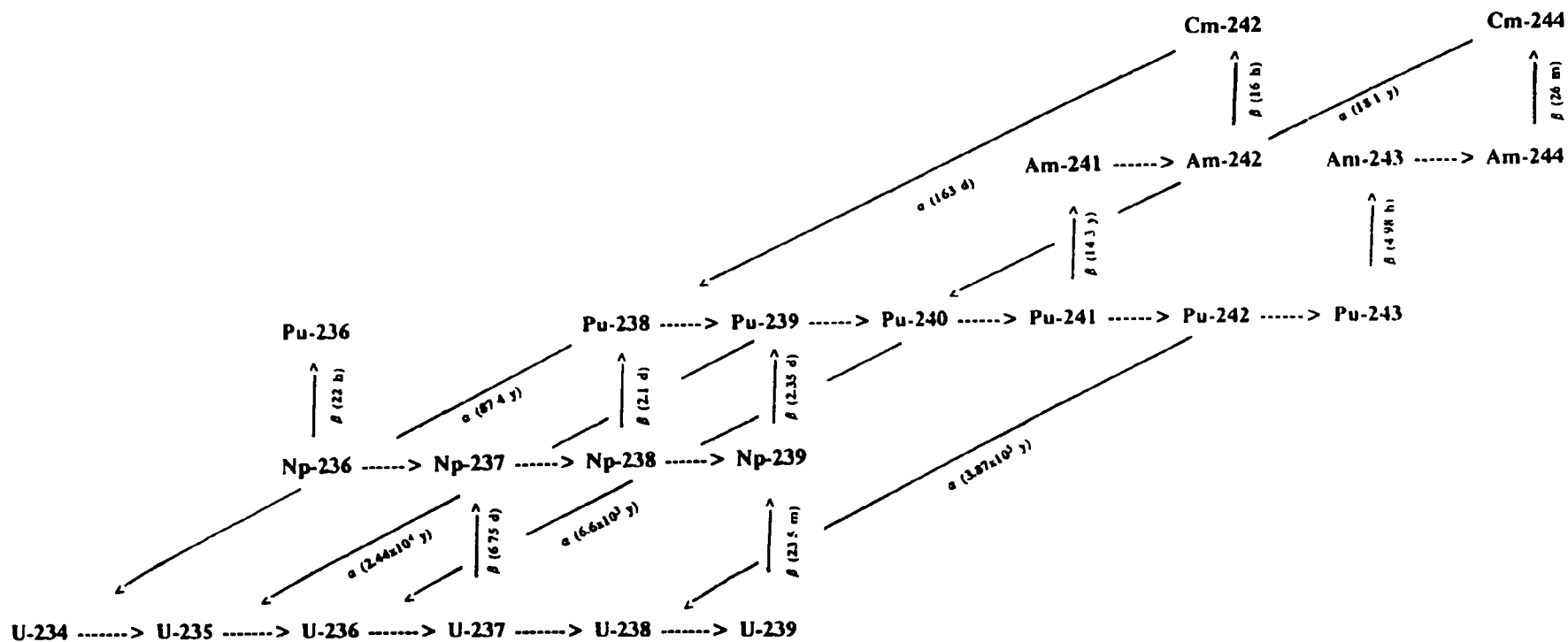


FIGURE 1-2

FORMATION-DECAY SCHEME FOR MAJOR TRANSURANIUM ELEMENTS

TABLE 1-1

RADIOACTIVITY AND MASS OF IMPORTANT TRANSURANIUM ELEMENTS IN LWR FUELS AT DISCHARGE FROM A REACTOR

	Curies	Grams/Tonne
Np-237	3.04 E 01	4.32 E 02
Np-239	2.21 E 07	9.51 E 01
Pu-238	2.19 E 03	1.28 E 02
Pu-239	3.07 E 02	4.94 E 03
Pu-240	5.26 E 02	2.31 E 03
Pu-241	1.26 E 05	1.22 E 03
Pu-242	1.78 E 00	4.60 E 02
Am-241	1.02 E 02	2.98 E 01
Am-243	1.71 E 01	8.55 E 01
Cm-242	3.87 E 04	1.17 E 01
Cm-244	1.53 E 03	1.89 E 01

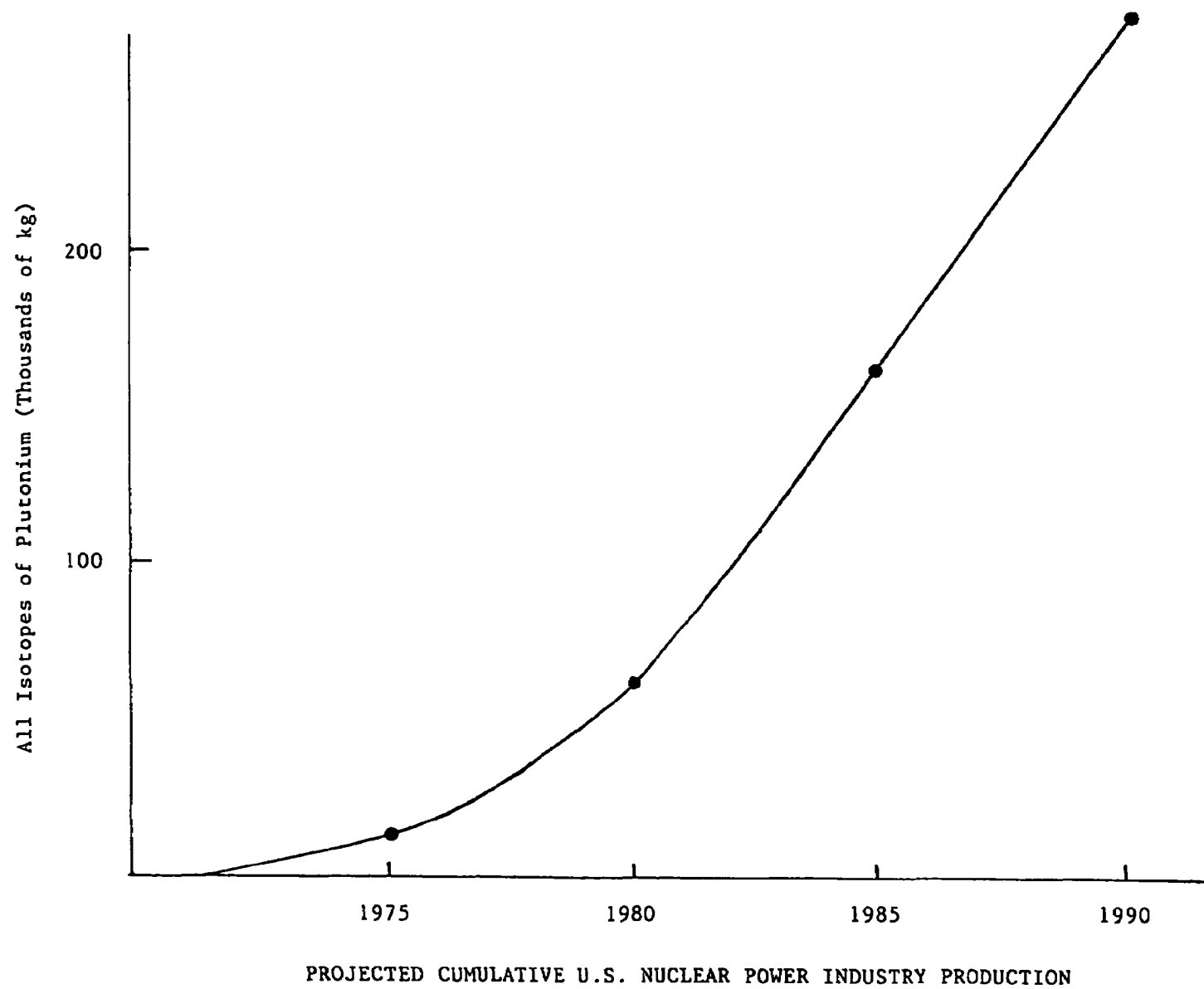


FIGURE 1-3

Plutonium-239 is the most common plutonium isotope. A light water reactor contains about 600 kilograms; the liquid metal fast breeder reactor proposed for commercial operation would contain about 3000 kilograms. Several tens of tons of plutonium-239 have been produced for weapons purposes. Reactor-grade plutonium is roughly 7% plutonium-239 and 20% plutonium-240, whereas weapons-grade plutonium is roughly 93% and 7%, respectively.

Plutonium-238 is very useful as an energy source for remote or isolated power supplies, such as in satellites or for cardiac pacemakers. About 40 kilograms of plutonium-238 have already been specially produced for such purposes. It is not a significant component of normal nuclear fuel.

Present levels of the transuranium elements in the environment have resulted from several sources - regional and worldwide fallout from the testing of nuclear weapons in the atmosphere, accidents involving military and related operations, and local releases from nuclear facilities. The major portion of the transuranium elements in the environment is the result of surface and atmospheric nuclear weapons tests during the period 1945-1963. Atmospheric tests injected radioactivity into the stratosphere which has since then been slowly deposited more or less uniformly over the lands and oceans of the earth. An estimated 4.2 tons of Pu-239-240 (about 320 kilocuries, 60% of which is Pu-239) have been globally dispersed as the result of atmospheric testing of nuclear weapons mainly before 1963, and another 110,000 curies of Pu-239-240 have been deposited locally around the sites of nuclear weapons testing. About 120,000 curies of Am-241, which were produced in nuclear weapons fallout debris, account for an additional alpha activity equivalent to about 1/4 of that from the alpha-emitting plutonium isotopes. A beta-emitting plutonium isotope, Pu-241, will roughly double the amount of Am-241 originally produced during testing. As a result of these earlier weapons tests, the existing level of

transuranium element contamination in soils of the United States is about 0.002 uCi/m^2 (Fig 1-4). More recent weapon tests have not added significant amounts to this level.

The accumulated deposition of globally dispersed plutonium produced from early testing is almost complete. The annual plutonium deposition rate measured in the New York area shows that of the 0.3 MCi of Pu-239 globally dispersed by nuclear weapons testing through 1962, about 90% of it had returned to the earth by 1965. Of the 320 kCi of Pu-239-240 globally deposited up to the end of 1973, 250 kCi were in the northern hemisphere and 70 kCi in the southern hemisphere, in agreement with the distribution of nuclear explosions.

It is useful to compare the environmental levels of plutonium from fallout with the quantities of natural alpha emitters present in the environment. For example, it has been estimated that the continental U.S.A. contains about 4.4 MCi of natural alpha emitters in the upper 2 cm layer of soil. Of this amount, 1.6 MCi are alpha-emitting radionuclides of uranium and thorium. Thus, the amount of alpha activity from plutonium radionuclides in the upper 2cm of soil (0.016 MCi) is about 1% of that from naturally occurring actinide radioactivity and 0.36% of the total natural alpha radioactivity.

In contrast to global fallout, local contamination is generally associated with areas close to sites of nuclear weapons explosions and with areas exposed to low-level releases from the nuclear fuel cycle or activities related to the production of nuclear weapons. The local fallout is deposited onto the soil surface soon after release and enters into the soil weathering processes immediately.

Areas where there is substantial localized contamination above the general background level are well documented and extensive environmental analyses have been carried out at all

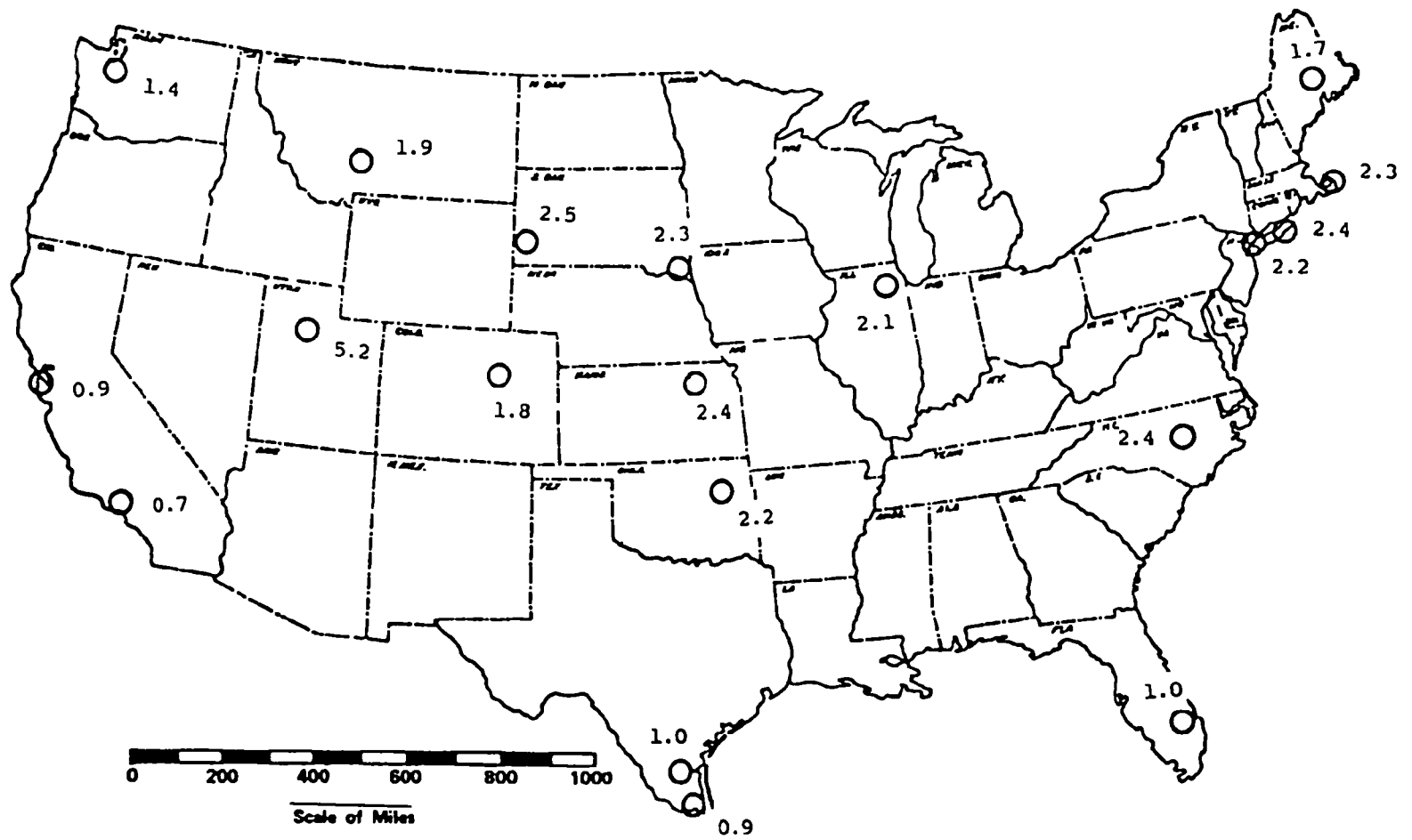
TABLE 1-2

INVENTORY OF PLUTONIUM FOR SELECTED SITES IN THE UNITED STATES

LOCATION	APPROX INVENTORY	REMARKS
U.S. (Fallout)	20,000 Ci	Worldwide Pu-238= 17,000 Ci Pu-239=440,000 Ci U.S. average=1.5 mCi/km ²
Nevada Test Site (near Las Vegas, NV)	> 155 Ci	Nuclear Test Site Surface and Subsurface Tests
Rocky Flats Plant (near Denver, CO)	8-10 Ci	Weapons Fabrication Plant (limited cleanup in progress)
Mound Laboratory (Miamisburg, OH)	5-6 Ci	Pu-238 in sediments in canals
Savannah River Plant (SW part of SC)	3-5 Ci	Pu and higher isotopes production
Los Alamos Lab (NW of Santa Fe, NM)	1-2 Ci	Weapons Development (high levels in remote canyons)
Hanford Site (central WA)	—	Pu Production—Research Facility (high levels in trenches on site)
Oak Ridge Laboratory (east TN near Knoxville)	—	Research & Development Facility
Trinity Site (near Alamogordo, NM)	45 Ci	Site of first atomic bomb test

FIGURE 1-4

CUMULATIVE DEPOSIT OF Pu²³⁹ IN mCi PER km²

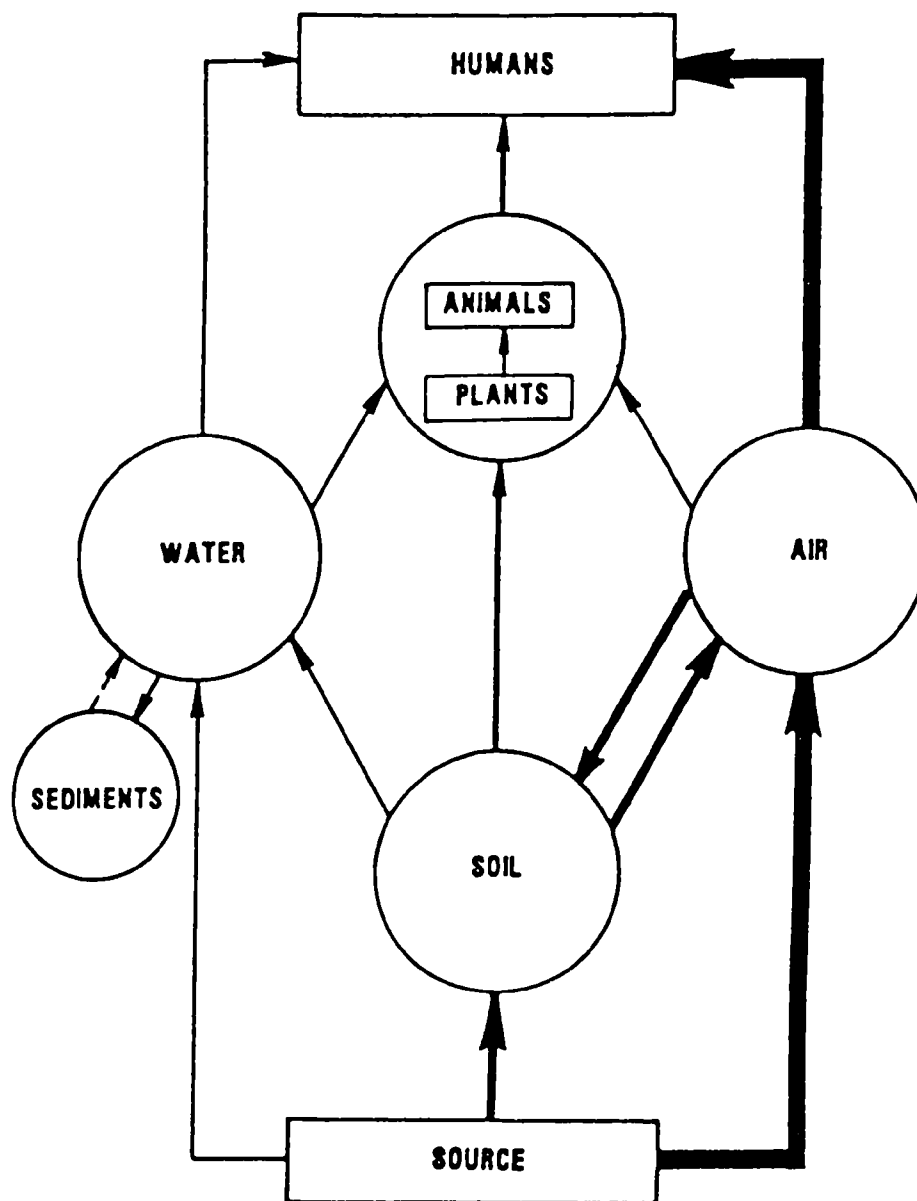


these sites. Areas of highest contamination are, for the most part, on Federally owned property and therefore are under the direct control of the Federal government and access to these may be restricted. Table 1-2 shows estimates of the amount of plutonium in the environment at several known United States locations.

Plutonium and other transuranium elements can move through the environment by a variety of transport mechanisms and pathways. These are determined by the chemical and physical form of the deposited material, the characteristics of the surface, local land use patterns, and other factors such as wind or rainfall. Principal environmental pathways to humans are shown in Fig. 1-5.

Transuranium elements released to the environment may exist as discrete particles or they may become attached to other materials. The principal modes of transport of these elements from a source to man are by direct airborne movement from the source or by resuspension of previously deposited small particles by the action of wind or other disturbance. Resuspension is a complex phenomenon affected by a number of factors, including the characteristics of the surface, type of vegetative cover, meteorological conditions, and age of the deposit. In general, resuspension will be relatively high immediately after initial deposition, gradually decrease with time, and approach a long-term constant within about one year after deposition.

Plutonium suspended by wind can be redeposited on soil or intercepted by biological surfaces. Redeposition of plutonium on soils can lead to major changes in the distribution of the element within an ecosystem. Wind redistributes plutonium in soil, as inferred from sampling of contaminated sites. Field studies, primarily in arid regions, imply that wind transport of soil is highly seasonal and is relatively more important in dry, sparsely vegetated areas than in heavily vegetated areas. Soil



**PRINCIPAL PATHWAYS OF THE TRANSURANIUM ELEMENTS
THROUGH THE ENVIRONMENT TO MAN**

FIGURE 1-5

particle sizes and plutonium concentrations in soil affect the importance of wind as a plutonium transport vector. Plutonium concentrations of various soil size fractions can differ by several orders of magnitude and, depending on source characteristics, are generally highest in the smaller size fractions.

Plutonium in air is deposited on vegetation or on soil, contaminating plants by direct deposition or by root uptake. There is no significant evidence of bioaccumulation through succeeding trophic levels since metabolic discrimination usually acts against the transfer of plutonium. Soil chemical reactions influence the behavior of the various forms of plutonium. Plutonium in soil may exist in ionic form (positive or negative) or as a neutral compound depending on the pH and redox potential (Eh) of the respective soil type. The major factor governing plutonium availability will be the solubility of the compound. A number of organic and inorganic substances form complexes with plutonium, thereby increasing its solubility.

Downward transport of plutonium through the soil will occur via leaching of soluble plutonium, by transport of soluble oxide particles and by transport of soil particles onto which plutonium is adsorbed. The latter two processes are active mainly in the top soil and leaching will usually terminate at lower depths.

Transuranium elements in terrestrial environments can enter plants by foliar absorption and root uptake. The route of entry into plants will depend on the nature of the source; climatic conditions affecting deposition, retention, and chemistry of particles on leaf surfaces; the foliar surface area exposed; and soil conditions affecting resuspension and solubility.

Plants remove very little plutonium from soil. Concentrations in plants generally range from 0.001 to 0.00001 of the amount in soil. Part of the large reported variation in

plant uptake is related to the wide variety of experimental and field conditions encountered. Plutonium concentrations in food are generally low, and a very low transfer across the gastrointestinal tract may make the ingestion pathway in man relatively unimportant.

There is little evidence to suggest that differing sources of transuranic elements affect their chemical properties when the elements are moderately well dispersed in aquatic systems. Transuranic elements are soluble, to a limited extent, in both freshwater and marine systems and are therefore available for transfer across biological membranes.

Transport of plutonium and other transuranium elements through the food chain and subsequent ingestion is generally of lesser importance than the air pathway. Transuranium elements may be deposited on plant surfaces or assimilated through the plant root system. The uptake by plants is relatively small and most animals, including humans, have a high discrimination factor against transfer of these elements into body tissues. The solubility of plutonium in water is very low and nearly all plutonium released into lakes and streams is ultimately deposited and sorbed onto sediments. Other possible routes of entry into humans include direct ingestion of contaminated soils and subcutaneous contamination of wounds.

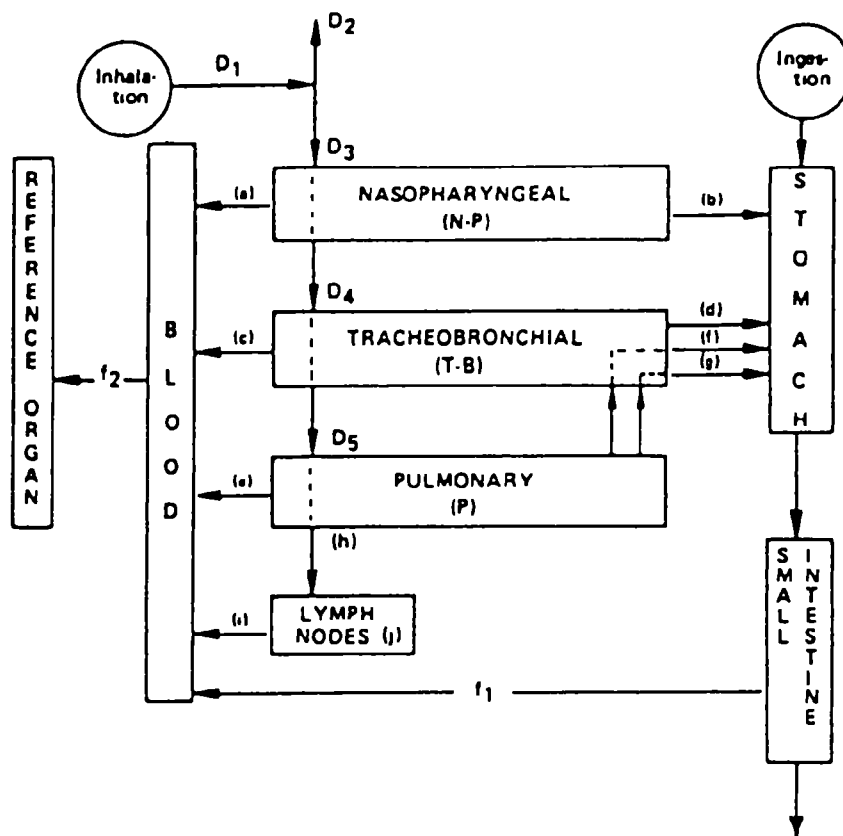
Inhalation is generally the principal pathway to man and arises from radioactive materials directly injected into the air and from resuspension of previously deposited materials. Ingestion is a less significant hazard, because of the cumulative dilution in transfer from soils or water to food; ingested plutonium is very poorly absorbed from the gastrointestinal tract. Contamination of wounds is not a likely occurrence.

Inhaled particles are initially deposited in various regions of the respiratory tract, where they remain until either cleared

or translocated to other body organs. Much of the material deposited in the lung is cleared within a few days, but some of the smaller particles which diffuse into the pulmonary regions of the lung are removed much more slowly and have a biological half-life of a year or more (Fig 1-6). This may lead to an increase in the risk of lung cancer in exposed individuals, but this has not been conclusively demonstrated. Inhaled transuranium elements may also transfer and be retained in other body organs, and cause cancers of the bone and liver. For the less soluble transuranium compounds, such as plutonium oxide, this will contribute only marginally to the total risk for the inhalation pathway.

Potential health effects caused by the transuranium elements are a function of several biological and physical parameters including the biological retention time in tissue, the type of radioactive emission, and the half-life of the nuclide. For the more important transuranium nuclides, such as Pu-238 or Pu-239, biological retention times are very long and radioactive decay occurs at such a slow rate that uptake of these materials in the human body will result in prolonged exposure of body organs. Many of the transuranium nuclides decay by emission of an alpha particle (ionized helium atom), in a manner similar to radium and other naturally occurring alpha emitting nuclides. Alpha particles are highly ionizing and damaging, but their penetration in tissue is very small (about 40 μm). Thus, biological damage is limited to tissue in the immediate vicinity of the radioactive material, and a potential health hazard from transuranium elements in the environment can only result when these materials are inhaled or ingested into the body.

Ingestion of transuranium elements generally represents a smaller environmental risk to humans than inhalation. A relatively small fraction of any ingested transuranium element may be transferred to the bloodstream from the digestive tract and deposited in bone, liver, gonadal tissue, and other organs.



D_1 IS THE TOTAL AEROSOL INHALED; D_2 IS THE AEROSOL IN THE EXHALED AIR, D_3 , D_4 , AND D_5 ARE THE AMOUNTS DEPOSITED IN THE NASOPHARYNGEAL, TRACHEOBRONCHIAL, AND PULMONARY LUNG RESPECTIVELY. THE LETTERS (a) THROUGH (j) INDICATE THE PROCESSES WHICH TRANSLOCATE MATERIAL FROM ONE COMPARTMENT TO ANOTHER

FIGURE 1-6

In most cases, less than one part in ten thousand of the ingested material is absorbed by the body, with the remainder excreted. The cancer risk to individuals as a result of ingestion of transuranium elements is mainly due to potential bone and liver cancers.

A potential risk of genetic damage to the progeny of exposed individuals exists because of possible accumulation of transuranium elements in gonadal tissues. At the dose rates for other organs provided by the interim guidance, this risk is very small compared to the natural incidence of genetic damage.

2. ENVIRONMENTAL CHEMISTRY OF PLUTONIUM

2.1 GENERAL PROPERTIES

Plutonium is a silvery white metal which readily oxidizes in air. The oxidation rate of plutonium depends upon the temperature and the relative humidity of the air and increases as the relative humidity increases. In going from 0% to 50% relative humidity, the oxidation rate will increase by several orders of magnitude.

Plutonium is capable of forming many compounds with oxygen, including the simple binary oxides, the peroxide, the hydroxide (or hydrated oxides), and a series of ternary and quaternary oxides. Plutonium dioxide (PuO_2) is the most stable of the oxides and is formed under most conditions, especially when plutonium metal is ignited in air. Calcined PuO_2 is unreactive under normal environmental conditions, is extremely insoluble in water, and if ingested would be highly insoluble in body fluids. Because of its desirable properties such as high melting point (2240°C), irradiation stability, chemical stability ($F_{298} = -253 \text{ kcal}$), compatibility with metals, and low vapor pressure, plutonium dioxide has found widespread use as a fuel either alone or in combination with other compounds. For example, plutonium-238 is used as a heat source for thermoelectric generators employed in devices such as heart pacemakers and communication satellites. When used as a heat source, the $^{238}\text{PuO}_2$ is fashioned into pellet form and then encapsulated in a refractory metal. Other solid forms of plutonium are possible but are of little concern from the standpoint of being likely environmental contaminants.

2.2 CHEMICAL BEHAVIOR IN AQUEOUS ENVIRONMENTS

Plutonium dioxide is a highly refractory material when prepared at high temperatures and reacts slowly with aqueous solutions. The rate at which dissolution proceeds depends on such factors as the pH and temperature of the solution and the presence of oxidizing, reducing, and complexing agents. As the dioxide dissolves, plutonium ions are formed which can undergo complexation and/or hydrolysis with other species present in the aqueous phase. Hydrolysis can be viewed as a form of complexation in which the hydroxyl ion is the ligand. The most stable hydrolysis product of plutonium in the pH range generally encountered in natural waters is $\text{Pu}(\text{OH})_4$ (Andelman, 1970). This hydroxide has been reported (Taube, 1964) to have a solubility product constant of 7×10^{-56} which is indicative of a high degree of insolubility.

Potentially four oxidation states of plutonium (III, IV, V, VI) can exist in solution in equilibrium with $\text{Pu}(\text{OH})_4$. The predominant soluble form of plutonium in a soil/water environment will be determined by the oxidizing or reducing capability of the solution. The availability of plutonium for uptake by plants and other biological systems will vary depending upon the valence state (Adams, 1975 and Price, 1973). Generally, the availability to plants follows the valence order $\text{V} > \text{VI} = \text{III} > \text{IV}$.

Most efforts to ascertain the valence of soluble plutonium have been theoretical because of the difficulty encountered in determining valence states at typically low environmental concentrations. One means of predicting the predominant plutonium species under specified conditions is the use of stability diagrams (Polzer, 1971). These diagrams examine the phase relationships between possible plutonium species as a function of the redox potential of the solution. Their use is limited, however, to systems which are in equilibrium and for

which the concentration of additional ions is insignificant. This approach only considers the thermodynamics of the system and not the kinetics, thereby indicating which reactions are possible but not how fast they will occur. Within these limitations, stability diagrams can be useful in evaluating the observed environmental behavior of plutonium.

From stability diagrams, Polzer (1971) and Andelman (1970) both concluded that, when the concentration of complexing anions is insignificant, the predominant form of soluble plutonium will be Pu^{+3} . However, natural environments contain complex materials from the decomposition of plant and animal matter which may affect the chemical equilibrium of the soil/water environment. Bondietti (1975) stated that the presence of a complexing agent such as EDTA can increase the susceptibility of Pu^{+3} to oxidation to Pu^{+4} due to the activity-lowering effect that complexation produces. His studies have attempted to ascertain the probable valence state of plutonium in the presence of various organic substance commonly encountered in the environment. He concluded that the Pu(IV) valence state would be the most important and that higher oxidation states would be reduced to the IV state through the action of phenolic materials like humic substances and reducing sugars. Further valence reduction to Pu(III), although possible, was not felt to be likely under the redox conditions normally encountered. Bondietti observed that only a small fraction (<20% of the plutonium in a soil contaminated 30 years previously) was desorbable and, therefore, available for uptake by biological systems. The majority of the plutonium was strongly associated with the solid phase and, as a result, leaching losses were concluded to be insignificant compared to the physical movement of contaminated soil particles.

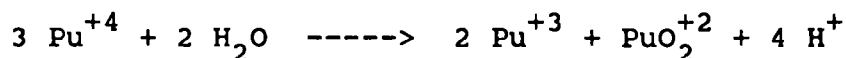
2.3 FRESHWATER ENVIRONMENT

The above observation is consistent with studies of plutonium released into aquatic systems. For example, Wahlgren

(1973) has shown that 95% of the plutonium added to Lake Michigan as a result of atmospheric fallout from weapons testing is rapidly removed from the water column to the sediments. Further studies of Lake Michigan by Alberts (1974) have demonstrated that plutonium is strongly associated with the sediments and is not easily solubilized under aerobic conditions. Stagnant, polluted systems however could have a greater solubilizing effect due to high concentrations of complexing organic material.

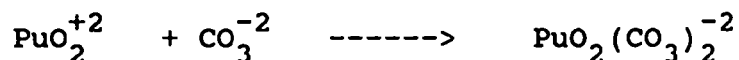
Studies of the fate of plutonium in flowing water systems have been conducted in the Great Miami River (Ohio) where discharges of plutonium from Mound Laboratory have occurred. These studies (Bartelt, 1975 and Muller, 1977) have attempted to elucidate the mobility of plutonium in aquatic ecosystems. Consistent with the Lake Michigan studies, plutonium was found associated with the sediments and at least 90% of the radioactivity which moves down the river does so as a result of the resuspension/entrainment of bottom sediments.

The predominant chemical form of soluble plutonium is, however, still open to question. For example, the Lake Michigan study indicates that most of the soluble fraction is in an anionic form with a particle diameter of less than 30 Å. This may indicate sorption of plutonium to colloidal silica and other negatively charged colloidal minerals. There has also been speculation that the VI valence state can be an important environmental species. The VI valence state can be generated by the disproportionation of PuO_2 during dissolution (Cleveland, 1970):



Although simple stability diagrams do not predict the VI valence to be important, the presence in natural waters of complexing agents (e.g., carbonates) can stabilize this valence state (Andelman, 1970). One such reaction which might associate

plutonium in anionic form is:



Even though the chemical behavior of plutonium in fresh water is not completely understood, the many studies to date have consistently demonstrated that plutonium will quickly associate with the solid phase and, under the redox conditions normally encountered in the environment, solubilization will be minimal. In general, the plutonium activity committed to fresh water will be bound there in a form which is relatively insoluble in biological systems.

2.4 MARINE ENVIRONMENT

Because of its long radiological half-life, it must be assumed that some of the plutonium in any freshwater body will eventually be carried to an ocean or sea. Knowledge of the behavior of this radionuclide in a marine environment is therefore important when evaluating the long term impact of any release to the environment.

In a study conducted off the coast of Massachusetts by Bowen et al., the bioturbation and sedimentation rate effects on plutonium sediment profiles were examined (Bowen, 1975). Analyses of cores taken prior to 1964 and from 1968 to early 1975 showed a progressive migration of Pu from deeper sediment layers to areas nearer the surface. It appears that primary amines emanate from the deeper anoxic or nearly anoxic sediment layers. These amines seem to be involved in a mechanism for releasing Pu from the sediments, but when they reach the oxygenated upper sediment layers what appears to be microbiological consumption immobilizes the Pu again. In this study the remobilization of Pu is attributed to a reducing mechanism, as it was in other studies (Edgington, 1975 and Alberts, 1975). This would indicate a redistribution of Pu

in the sediments without a release to the water column. The distribution of Pu in sediment samples with respect to particle size was noted as being uniform. No preference for the finer fractions was observed and concentrations of these nuclides per gram of sediment varied only by a factor of two from the finest to the coarsest fractions measured.

The activities of bottom feeders can also have an effect on sediment Pu profiles. These fish tend to enhance the mixing of the sediments and the water column. This will either increase the deposition rate of the Pu from the water column to the sediments or, in times of relatively low fallout rates, it can enhance possible resolubilization of the sediment-bound Pu. However, extremely low and variable Pu levels in the water itself seem to indicate that the Pu inventory is mostly removed to the sediment bed.

In another study, the fate of plutonium released to the Irish Sea over approximately 20 years has been examined. This study (Hetherington, 1975) has noted that >96% of the plutonium released was removed to the sediments in a relatively short time period. The mechanism responsible for carrying the Pu to the sediment bed does not appear to be biological nor does it seem to be a simple diffusion mechanism. Sorption to particles in the water column and subsequent deposition in the sediment bed appears to be the primary mechanism for removing Pu from the water. Once these sediments are consolidated, there appears to be no detectable remobilization of the plutonium.

Similarly, a study of Bombay Harbor Bay (Pillai, 1975) observed that 99% of the released Pu was localized in the sediments around the discharge point, but areas of higher activity were found in locations with particularly high siltation rates. The Pu-containing sediments were examined to determine the conditions necessary to release the Pu from the sediment. Violent agitation for 8 hours did not produce any detectable

release of Pu to seawater. Also, extraction tests using hydrochloric acid at 80°C resulted in no release of Pu to the solution.

As with freshwater systems, the marine studies have shown that over 90% of the Pu entering either as fallout or in the form of liquid effluents was found trapped in the sediments. This behavior is mostly attributed to the bonding of Pu to the organic material associated with sediment particles. A very small fraction of the Pu entering a body of water remains unfilterable. Most of this is assumed to be Pu in some colloidal form.

Even though Pu is strongly bound to the sediment bed in most water bodies it can still move with water currents and other physical actions. The effects of biota on Pu behavior are seen mostly in the sediments. Both microorganisms and more complex species can alter the depth profiles. Primary amines released deep in the sediment by microbes can solubilize the plutonium in anaerobic regions and carry it towards the surface. Microbes in the interface regions, along with chemical reactions, bind the Pu to the sediment again. Larger biota can physically alter profiles by digging into the bed and allowing the overlying water to penetrate. The most significant role of biota is the concentration of Pu in their systems. Seaweeds, with concentration factors of up to 1000, are by far the most notable. Shellfish, even though they have a lower concentration factor, are important because they may be a direct food source.

2.5 CHEMICAL BEHAVIOR IN SOILS

Similar to its aqueous behavior, the interaction of plutonium with soils is not completely understood and has been explained to date only qualitatively. This is in part due to the difficulty in characterizing plutonium at very low environmental concentrations. Oftentimes the predominant valence state of

plutonium cannot be established - much less its chemical form. However, several studies conducted at sites where plutonium has been in the environment for as long as 30 years have yielded some knowledge of its behavior. One such series has been conducted by Tamura at four sites having different ecosystems. The selected sites were: the Nevada Test Site (Nevada), Oak Ridge National Laboratory (Tennessee), Mound Laboratory (Ohio), and Rocky Flats (Colorado). The source terms were different at the four sites: at NTS the source was a "safety shot" series which dispersed plutonium as the oxide, at ORNL an earthen dike gave way releasing material from a holdup pond, at Mound Laboratory an acid solution of plutonium leaked from a waste transfer line, and at Rocky Flats cutting oil contaminated with plutonium leaked from storage drums. The association of plutonium with the soils of these various sites will be affected by factors associated with the source term and the prevailing local environmental conditions.

The ease with which plutonium is extracted from the solid phase can indicate its availability for biological uptake. Studies by Tamura (1976) have shown low extraction (10-15%) from soils of the Nevada Test Site and Rocky Flats, while ORNL and Mound Laboratory soils showed much higher extraction (60-85%). These results are evidence of the existence of different chemical species at the various sites. Experiments by other investigators have provided further evidence of such differences. Specifically, autoradiographic analyses of soil samples from Rocky Flats (Hayden 1974; Nathans 1974; Sehmel 1975) have shown the presence of discrete particles of plutonium (probably the oxide) attached to larger soil particles, while the same technique indicated a general dispersion of plutonium in the soil of Mound Laboratory (Rodgers 1975).

In order for plutonium to be incorporated into the soil matrix, it must be available in ionic form and capable of displacing some other cation from the matrix. This could explain

the difference in the extraction and autoradiography studies at the various sites. At NTS and Rocky Flats the plutonium was originally released as metal or oxide particles which require aqueous dissolution to generate plutonium ions. In contrast, the Mound release was as an acid solution already containing plutonium in ionic form which probably reacted quickly with the solid phase and was incorporated into the soil matrix. The Rocky Flats and NTS soils, on the other hand, could contain hydrated or polymerized plutonium oxide attached to soil particles via adhesion. Once polymerized, plutonium is slowly depolymerized by strong acids (Cleveland 1970) and this could account for the low extractability of the Rocky Flats and NTS samples.

Rodgers (1975) in his studies around Mound Laboratory proposed a mechanism for the bonding of plutonium to soils and the following has been excerpted from his review:

Proposed bonding mechanisms: Basically, soils are made up of silicate materials and other minerals. Quartz sand has a continuous silicate structure where each silicon atom is bound to two silicons. This continuous structure is interrupted at the surface and in natural systems (where water is abundant), the surface is composed of unsaturated oxygen bonds. That is, each surface oxygen is bound to only one silicon atom. The remaining bond is usually occupied by other cationic species. Clays are more complicated, but are also based on the continuous silicate structure except that some silicon atoms have been replaced by Mg^{2+} , Al^{3+} , Fe^{3+} , and Fe^{2+} (mostly Al^{3+}). These substituted atoms in the silicate structure result in variations in long range crystal structure. Rather than forming three-dimensional silicate networks as in quartz, many clays form two-dimensional sheets which cleave easily to form plates. The surface of these layered sheets that make up the clay particles also exhibit unsaturated oxygen bonds. This, in part, accounts for the higher sorption capacity in clays (relative to the same size silicate particle) since sorption can take place between the silicate layers within the clay particles.

The unsaturated oxygen bonds in natural soils and clays are occupied by cations such as H^+ , K^+ , Ca^{2+} , Mg^{2+} , or other available cations. The bonding strength order of these cations is:



In order to bond to the silicate, an ion must displace or exchange with the cation already bonded:



The extent of the exchange depends on the relative strength of the bonds and the relative solution concentrations of the two cations.

Some cations form silicate bonds that are fairly weak (such as Na^+ and K^+) and may be only electrostatic while other metal cations may even develop covalent character.

Tetravalent plutonium ions are well noted for the formation of strong bonding (complexing) with oxygenated ligands. The strength of plutonium-oxygen bonds is also indicated by the acidic character of plutonium hydroxide forming hydrous plutonium oxide.

Plutonium ions can compete with hydrogen ions for the bonding sites on the silicates even when the $\text{Pu}^{4+}/\text{H}^{+}$ concentration ratio is 10^{-11} or less. The very large bonding potential of plutonium suggests that sorbed plutonium cannot be significantly displaced from soil by the concentrations of cations existing in nature.

Chemicals that complex the plutonium compete with the silicate particles for the plutonium and tend to reduce the extent of sorption of the plutonium on soil. For example, the formation of plutonium hydrolytic species $\text{Pu}(\text{OH})_3^{3+}$, $\text{Pu}(\text{OH})_2^{2+}$, $\text{Pu}(\text{OH})_3^{+}$, and $\text{Pu}(\text{OH})_4$ (as well as "polymeric" forms) tends to reduce ion exchange sorption. Some of the hydrolytic plutonium "polymeric" forms may adsorb to the surface of the soil particles and the precipitation of $\text{PuO}_2 \cdot (\text{H}_2\text{O})$ may be nucleated by the colloidal soil particles when plutonium concentrations are relatively high (greater than 10^{-6}M). Some moderately strong organic complexing agents, such as citric acid, can reduce the sorption of plutonium on soil.

2.6 INTERACTION WITH PLANTS

Consideration of the oxidation-reduction potentials of the valence state couples in the presence of organic material generally found in soils indicates that $\text{Pu}(\text{IV})$ is the predominant valence state in the environment (Bondietti 1975). Since the availability to plants generally follows the order $\text{VI} > \text{VI} = \text{III} > \text{IV}$, a low uptake of plutonium by plants would be expected. This expectation has been confirmed by the studies conducted in the laboratory and in the environmental where many varieties of plants have been grown in contaminated soils. Several reviews (Bernhardt 1976; Bulman 1976; Mullen 1976; and Thomas 1976) describe such details as the types of crops grown, the amount and chemical form of the plutonium used, and the method by which plutonium was added to the growing medium.

The results of these studies are usually expressed in terms of a "concentration factor" defined as the ratio of the plutonium concentration per gram of plant material to the plutonium concentration per gram of soil. This index indicates the degree to which plutonium is concentrated or discriminated against in the soil to plant pathway. Even though a variety of

methods and conditions have been used in these studies, the concentration factors are generally of the order of 10^{-4} to 10^{-5} . On occasion some higher values, ranging as high as 10^{-1} , have been reported, but these are usually attributed to external contamination of the plant surface rather than incorporation into the plant tissue.

Such small concentration factors tend to support the conclusion that the terrestrial food chain does not constitute a significant exposure pathway. However, the observation has been made that these experiments were all of a short-term nature and, as such, did not consider the possibility of increases in concentration factors over time due to a variety of possible influencing factors. As a result of preliminary investigations, several mechanisms of increased uptake have been reported (Cleveland 1976; Bondietti 1975; Romney 1970; Lipton 1976; Price 1973; and Beckert 1975). Included among these are: 1) chelation by organic constituents in the soil or through the addition of fertilizers, 2) increased root absorption zone contact as plutonium migrates down through the soil, 3) increased radiocolloid size due to aging, and 4) the long-term action of microorganisms present in the soil.

Much of the research into these mechanisms is in the early stages; however, the preliminary data have not demonstrated that substantial changes will occur under conditions generally encountered in the environment. It is most likely that, if such changes take place, that they will occur slowly over the order of years to decades.

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3. DOSE AND RISK TO HEALTH DUE TO INHALATION AND INGESTION OF TRANSURANIUM NUCLIDES

3.1 INTRODUCTION

Most of the transuranium nuclides decay by emission of an alpha particle (ionized helium atom). Alpha particles transfer much of their energy for each collision (high-LET) and are very damaging to tissue, but their range is very small (less than 100 microns). Thus, biological damage is limited to tissue in the immediate vicinity of the radioactive material, and a potential health hazard from transuranium elements in the environment can only result from inhalation or ingestion into the body. For the more important transuranium nuclides, such as Pu-238 or Pu-239, biological retention times are very long and radioactive decay occurs at such a slow rate that uptake into the body will result in prolonged exposure of body organs.

Risk estimates for internal organs are in accord with the recommendations published by the International Commission on Radiological Protection and by the NAS-BEIR Committee (popularly known as the BEIR-3 and BEIR-4 Reports). Lung inhalation dosimetry is in accord with the recommendations of ICRP Reports 19 and 30.

3.2 CURRENT STATUS

3.2.1 SOMATIC RISK

At the low levels of radiation exposure attributed to radionuclides in the environment, the principal health detriment is the induction of cancers (solid tumors and leukemia), and the expression, in succeeding generations, of genetic effects.

Most of the observations of radiation-induced carcinogenesis in humans are on groups exposed to low-LET radiations. These groups include the Japanese A-bomb survivors and medical patients treated with x-rays for ankylosing spondylitis in England from 1935 to 1954. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and the National Academy of Sciences Committee on the Biological Effects of Ionizing Radiations (NAS-BEIR) have provided knowledgeable reviews of these and other data on the carcinogenic effects of human exposures (Table 3-1).

Observed excess cancers have occurred, for the most part, following relatively high doses of ionizing radiation compared to those likely to occur due to environmental contamination from controllable sources of radiation. Therefore, a number of assumptions must be made about how observations at high doses should be applied at low doses and low dose rates for radiation of a given type. These assumptions include the shape of the dose response function, the risk projection model, and possible dose rate effects.

The 1980 BEIR-3 report examined three dose response functions in detail: (1) linear in which effects are directly proportional to dose at all doses; (2) linear quadratic in which effects are very nearly proportional to dose at very low doses and proportional to the square of the dose at high doses; and (3) a quadratic dose response function where the risk varies as the square of the dose at all dose levels. These define a range of predicted somatic effects and represent an envelope of risk estimates. The first two of these functions appear to be compatible with most of the data on human cancer.

TABLE 3-1

A COMPARISON OF ESTIMATES OF THE RISK OF FATAL CANCER
FROM A LIFETIME EXPOSURE AT 1 RAD/YEAR
(Low-LET Radiation)

	Cases per 10 ⁶ person rad	Projection Model
BEIR-3 (a) (b)	158	Absolute Risk
BEIR-3 (a) (c)	403	Relative Risk
BEIR-3 (a) (d)	270	Absolute/Relative Risk
UNSCEAR (e)	240-440*	"Multiplicative"
UNSCEAR (e)	160-200*	"Additive"
ICRP (f)	200	None
BEIR-5 (g)	770/810	Absolute/Relative Risk
ICRP (h)	500	None

- (a) Based on Tables V-3 and 4 in BEIR-3 Report (1980)
 (b) Absolute Risk (All Organs) - Linear Dose Response Model
 (c) Relative Risk (All Organs) - Linear Dose Response Model
 (d) L-L Absolute Risk Model for Bone Cancer and Leukemia;
 L-L Relative Risk Model for All Other Cancer
 (e) Based on Table 10 in UNSCEAR Report (1988)
 (f) ICRP Como Statement (1987)
 (g) Appendix 4 in BEIR-5 Report - Male/Female (1989)
 (h) ICRP (1990)

* Dose Rate Effectiveness Factor (DREF): UNSCEAR (1988) = 2.5
 ICRP (1990) = 2.0

The BEIR-3 committee limited its risk estimates to a minimum dose rate of 1 rem per year and stated that it "does not know if dose rates of gamma rays and x-rays of about 100 mrad/y are detrimental to man." At dose rates comparable to the annual dose that everyone receives for naturally occurring radioactive materials, a considerable body of scientific opinion holds that the effects of radiation are reduced. The National Council on Radiation Protection and Measurement (NCRP) Committee 40 has suggested that carcinogenic effects of low-LET radiations may be proportionally less for small doses and dose rates by a factor of from 2 to 10 times than have been observed at high doses. The International Commission on Radiological Protection and the United Nations Scientific Committee on Atomic Radiation have used a dose rate effective factor of about 2.5 to estimate the risks from occupational and environmental exposures. Their choice is fully consistent with and equivalent to the reduction of risk at low doses obtained by substituting the BEIR-3 linear-quadratic response model for the linear model.

To estimate the risk of radiation exposure that is beyond the years of observation, either a relative risk or an absolute risk projection model (or suitable variations) must be used. These models are described in detail in Chapter 4 of the 1980 BEIR-3 report. The relative risk projection model projects the currently observed percentage increase in cancer risk per unit dose into future years. An absolute risk model projects the average observed number of excess cancers per unit dose into future years at risk. Because the underlying risk of cancer increases rapidly with age, the relative risk model predicts a larger probability of excess cancer towards the end of a persons lifetime. In contrast, the absolute risk model predicts a constant incidence of excess cancer across time. Therefore, given the incomplete data we have now, with less than lifetime followup, a relative risk model projects somewhat greater risk than that estimated using an absolute risk model.

The National Academy of Sciences BEIR Committee and other scientific groups, e.g. UNSCEAR, have not concluded which projection model is the appropriate choice for most radiogenic cancers. However, evidence is accumulating which favors the relative risk projection model for most solid cancers. As pointed out by the 1980 NAS BEIR Committee:

"If the relative-risk model applies, then the age of the exposed groups, both at the time of exposure and as they move through life, becomes very important. There is now considerable evidence in nearly all the adult human populations studied that persons irradiated at higher ages have, in general, a greater excess risk of cancer than those irradiated at lower ages, or at least they develop cancer sooner. Furthermore, if they are irradiated at a particular age, the excess risk tends to rise pari passu (at equal pace) with the risk of the population at large. In other words, the relative-risk model with respect to cancer susceptibility at least as a function of age evidently applies to some kinds of cancer that have been observed to result from radiation exposure."

Leukemia and bone cancer are exceptions to the general validity of a lifetime expression period for radiogenic cancers. Most, if not all, of the leukemia risk has apparently already been expressed in both the A-bomb survivors and the spondylitics. Similarly, bone sarcoma from acute exposure appears to have a limited expression period. For these diseases, the BEIR-3 Committee believed an absolute risk projection model with a limited expression period to be appropriate for estimating lifetime risk.

A life table analysis can be used to estimate the number of fatal radiogenic cancers in an exposed population of 100,000 persons. This analysis considers not only death due to radiogenic cancer but also the probabilities of other competing causes of death which are, of course, much larger and vary considerably with age. The use of life tables in studies of risk due to low-level radiation exposure is important because of the time delay inherent in radiation risk. After a radiation dose is received, there is a minimum induction period of several years

(latency period) before a cancer is clinically observed. Following the latency period, the probability of occurrence of a cancer during a given year is assumed to be constant for a specified period, called a plateau period. The length of both the latency and plateau periods depends upon the type of cancer.

Estimates of the cancer risk from low-LET, whole-body, lifetime exposure can be based on different assumptions. If one assumes linearity and no dose rate effects, and a relative risk projection (the BEIR-3 L-L model) for solid cancers and an absolute risk projection for leukemia and bone cancer, this yields an estimated 400 fatalities per million person rad whole-body exposure. The International Commission on Radiological Protection provided estimates of 100 fatalities per million person rad exposure in 1977, and revised this to 200 fatalities per million person rad in 1987.

For environmental contamination by transuranium elements, inhalation and ingestion are the most common modes of exposure. In many cases, depending on the chemical and physical characteristics of the radioactive material, this may result in a nonuniform distribution of radioactivity within the body so that some organ systems receive much higher doses than others. For example, plutonium isotopes concentrate in the bone and liver, and the doses to these organs can be orders of magnitude larger than the average dose to the body.

Information on the proportion of fatal cancers due to cancer in a particular organ is not precise. One reason is that the data in BEIR-3 are based on whole-body exposures and it is possible that the incidence of radiogenic cancer varies, depending on the number of organs that are exposed. Except for breast and thyroid cancer, very little information is available on radiogenic cancer due to exposure of only one region in the body. Another reason is that most epidemiology studies use mortality data from death certificates, which often provide

questionable information on the site of the primary cancer. Moreover, when the existing data are subdivided into specific cancer sites, the number of cases becomes small, and sampling variability is increased. The net result of these factors is that numerical estimates of the total cancer risk are more reliable than those for most single sites.

The probability that fatal cancer occurs at a particular site has been derived from life table analyses for each cancer type using the information on cancer incidence and mortality in the BEIR-3 report, and normalized to the fraction of fatal cancer due to radiogenic cancer at a particular site. Results are listed in Table 3-2 and compared with similar estimates published by UNSCEAR and ICRP.

3.2.2 EXPOSURE TO HIGH-LET RADIATION

In some cases, ingestion and inhalation of alpha particle emitting radionuclides can result in a relatively uniform exposure of the body organs by high-LET radiations. Unlike exposures to x-rays and gamma rays, where the resultant charged particle flux results in linear energy transfers (LET) of the order of 0.2 to 2 KeV per micron in tissue, 5 MeV alpha particles result in energy deposition at a track average rate of more than 100 KeV per micron. High-LET radiations have a larger biological effect per unit dose (rad) than low-LET radiations. For cell killing and other readily observed endpoints, the relative biological effectiveness (RBE) of high-LET alpha radiations is often ten or more times greater than low-LET radiations.

Charged particles have been assigned quality factors, Q , to account for their efficiency in producing biological damage. Unlike an RBE value, which is for a specific and well defined endpoint, a quality factor is based on an average overall assessment by radiation protection experts of potential harm. In 1979, the ICRP assigned a quality factor of 20 to alpha

TABLE 3-2

COMPARISON OF PROPORTION OF THE TOTAL RISK OF
RADIOGENIC CANCER FATALITIES BY BODY ORGAN

Site	BEIR-3(a)	UNSCEAR	ICRP-26(b)	ICRP(1990)
Lung	.21	.24	.16	.13
Breast	.13	.16	.20	.05 (f)
Red Marrow	.16	.13	.16 (c)	.12
Thyroid	.10	.065	.04	.02
Bone	.010	.023	.04	.01
Liver	.085	.081	(.08) (d)	.03
Stomach	.084	.081	(.08)	.16
Intestine	.039	.12	(.08)	--
Pancreas	.058	.023	(.08)	--
Urinary	.025	.023	(.08)	.04
Colon	--	--	--	.18
Gonads	--	--	--	.13
Other	.11 (e)	.046	-	.13

(a) Lifetime exposure and cancer expression.

(b) Normalized for risk of fatal cancer.

(c) Leukemia

(d) Five additional organs which have the highest dose are assigned 0.08 for a total of 0.4.

(e) Total risk for all other organs including the esophagus, lymphatic system, pharynx, larynx, salivary gland, and brain.

(f) Population average (male-female)

particle irradiation from radionuclides. The reasonableness of this numerical factor for fatal radiogenic cancers at a particular site is not well known, but it is probably conservative for all sites and highly conservative for some.

The dose equivalent, in units of rem, is the dose, in units of rad, times the appropriate quality factor for a specified kind of radiation. For the case of internally deposited alpha particle emitters the dose equivalent from a one rad dose is equal to 20 rem. It should be noted that prior to ICRP Report 26

the quality factor for alpha particle irradiation was ten, that is, the biological effect from a given dose of alpha particles was estimated to be ten times that from an acute dose of low-LET x-rays or gamma rays of the same magnitude in rad. The ICRP decision to increase this quality factor to 20 followed from its decision to estimate the risk of low-LET radiations, in occupational situations, on the assumption that biological effects were reduced at low dose rates for low-LET radiation. There is general agreement that dose rate effects do not occur for high-LET (alpha) radiations. The new ICRP quality factor for alpha particles of 20 largely compensates for the fact that the low-LET risks are now based on an assumed dose rate effectiveness factor (DREF) of 2.5.

The NAS BEIR-3 Committee has stated that, "For high-LET radiation, such as from internally deposited alpha-emitting radionuclides, the linear hypothesis is less likely to lead to overestimates of the risk and may, in fact, lead to underestimates". However, at low doses, departures from linearity are small compared to the uncertainty in the human epidemiological data and assumption of a linear response probably provides an adequate model for evaluating risks in the general environment.

In 1980 the ICRP published a task group report "Biological Effects of Inhaled Radionuclides" which compared the results of animal experiments on radiocarcinogenesis following the inhalation of alpha particle and beta particle emitters. The task group concluded that "the experimental animal data tend to support the decision by the ICRP to change the recommended quality factor from 10 to 20 for alpha radiation."

The estimates of the risk of fatal cancer due to a uniform organ dose from internally deposited alpha particles have been calculated for both the data from the BEIR-3 report and Report 26 of the International Commission on Radiological Protection and compared with the estimates of the BEIR-4 report. A comparison of results is shown in Table 3-3.

The organ risk estimates based on the ICRP model assume an overall risk of 200/million person-rad as given in the Como Statement, a quality factor of 20, and the organ weighting factors of ICRP Report 26. The organ risk estimates based on BEIR-3 were prepared by multiplying the relative risk estimate of 400/million person-rad for a uniformly distributed whole body dose of low-LET radiation by a quality factor of 20, dividing by a dose rate effectiveness factor of 2.5 (except for endosteal bone which is based on data for high-LET (alpha) radiation), and then apportioning this risk by organ. The risk coefficient for leukemia was derived directly from the data in BEIR-3 as 43.5 fatalities per 10^6 person rad of low-LET radiation, or 360 fatalities per 10^6 person rad of high-LET radiation.

Comparison of risk estimates indicates that the several models all agree within better than a factor of two, and the choice of a calculational model may be dictated by an effort to establish uniformity. The BEIR-4 Committee noted that the risk of leukemia from intake of transuranium elements is not well established and all the estimates are subject to considerable uncertainty. It further stated that "dose calculations that

TABLE 3-3

ESTIMATED NUMBER OF FATALITIES FROM EXPOSURE
TO INTERNALLY DEPOSITED ALPHA (HIGH-LET) EMITTERS

Target Organ/Cancer	Fatalities per 10 ⁶ Person-Rad		
	BEIR-3*	ICRP-26**	BEIR-4
Lung	670	480	700
Liver	274	240	260-300
Red Bone Marrow/Leukemia	350	480	400
Skeletal Bone/Bone Cancer	240	120	300

* Relative Risk Model and Tables V-14/15 (BEIR-3 Report)
Endosteal Bone = 12 x Skeletal Bone (ICRP-26/30 models)

** Risk = 2×10^{-4} /rem (ICRP 1987 Como Statement)
Organ Weighting Factors (ICRP Report 26)

may be appropriate for radiation protection purposes may be entirely misleading for projecting risks of cancer mortality".

3.2.3 UNCERTAINTIES IN RISK ESTIMATES FOR RADIOGENIC CANCER

Numerical estimates of risks due to radiation are neither accurate nor precise. A numerical evaluation of radiogenic cancer risks depends both on epidemiological observations and a number of ad hoc assumptions which are largely external to the observed data set. These assumptions include such factors as the expected duration of risk expression and variations in radiosensitivity as a function of age and demographic characteristics. A major assumption is the shape and slope of the dose effects response curve, particularly at low doses where there is little or no epidemiological data. In 1971, the BEIR Committee based its estimates of cancer risk on the assumption that effects at low doses are directly proportional to those observed at high doses, the so called linear-nonthreshold hypothesis. The BEIR-3 Committee considered three dose response models and indicated a preference for the linear quadratic model. The risk coefficients derived for their linear quadratic model, and to a lesser extent their linear model, are subject to considerable uncertainty primarily because of two factors: 1) systematic errors in the estimated doses of the individual A-bomb survivors and 2) statistical uncertainty due to the small number of cancers observed at various dose levels. In addition to a dose response model, a "transportation model" is needed to apply the risks from an observed irradiated group to another population having different demographic characteristics.

3.2.4 GENETIC RISK

Genetic harm, or the genetic effects of radiation exposure, are those effects induced when radiation damages the nucleus of the cells of exposed individuals which become the eggs or sperm. The damage, in the form of a mutation or chromosome aberration,

is transmitted to and may be expressed in a child conceived after the radiation exposure and in subsequent generations.

One of the first estimates of genetic risk was made in 1956 by the National Academy of Science Committee on the Biological Effects of Atomic Radiation (BEAR Committee). Based on fruit fly (*Drosophila*) data and other considerations, the BEAR Genetics Committee estimated that 10 Roentgens per generation continued indefinitely would lead to about 5,000 new instances of "tangible inherited defects" per 10^6 births, and about one-tenth of them would occur in the first generation after the irradiation began. UNSCEAR estimated that one rad of low-LET radiation would cause a 1% to 10% increase in the spontaneous incidence of genetic effects.

In 1980, an ICRP Task Group summarized recommendations that formed the basis for the genetic risk estimates published in ICRP Report 26. These risk estimates are based on data similar to that used by the BEIR and UNSCEAR Committees, but with slightly different assumptions and effect categories. The 1980 NAS BEIR Committee revised genetic risk estimates. Estimates for the first generation are about a factor of two smaller than reported in the BEIR-1 report. A summary of genetic risk estimates is shown in Table 3-4.

Although genetic risk estimates are made for low-LET radiation, some radioactive elements, deposited in the ovary or testis can irradiate the germ cells with alpha particles. The ratio of the dose (rad) of low-LET radiation to the dose of high-LET radiation producing the same endpoint is called RBE and is a measure of the effectiveness of high-LET compared to low-LET radiation in causing the same specific endpoint.

Studies of the RBE for alpha-emitting elements in germinal tissue have only used plutonium-239. The estimated RBE for plutonium-239 alpha radiation versus chronic gamma radiation for

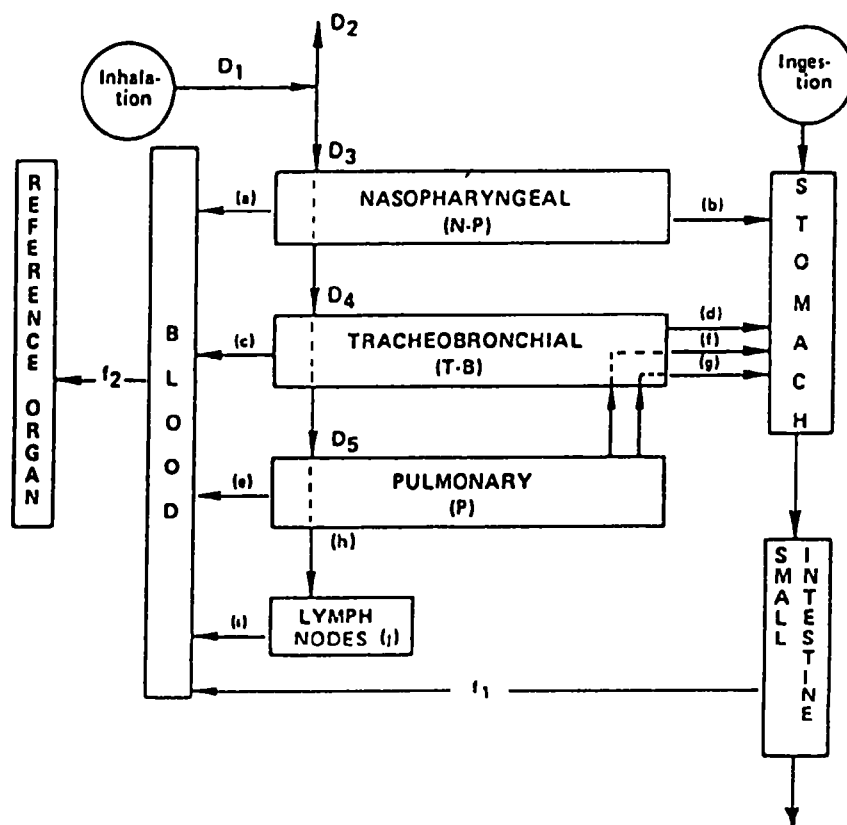
TABLE 3-4

SUMMARY OF GENETIC RISK ESTIMATES PER 10^6 LIVE-BORN
(Average Population Exposure of 1 Rad of Low Dose Rate
Low-LET Radiation in a 30-Year Generation)

Source	Serious Hereditary Effects	
	First Generation	Equilibrium (All Generations)
BEAR, 1956	---	500
BEIR-I, 1972	49* (12-200)	300* (60-1500)
UNSCEAR, 1972	9* (6-15)	300
UNSCEAR, 1977	63	185
ICRP-TG, 1980	89	320
BEIR-3, 1980	19* (5-75)	257* (60-1100)
UNSCEAR, 1982	22	149

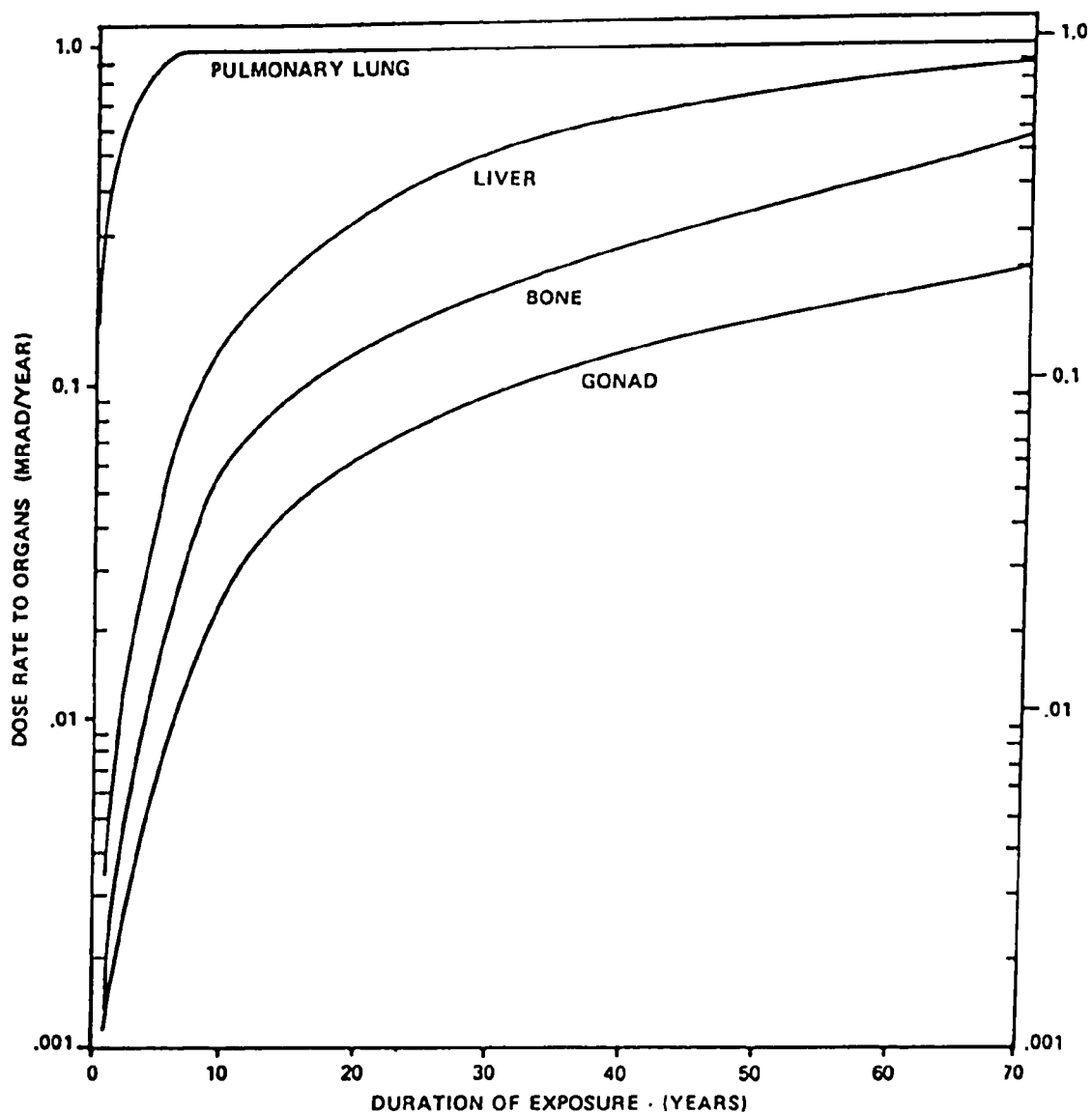
Numbers in parentheses () are the range of estimates

* Geometric Mean



D^1 IS THE TOTAL AEROSOL INHALED; D^2 IS THE AEROSOL IN THE EXHALED AIR; D^3 , D^4 , AND D^5 ARE THE AMOUNTS DEPOSITED IN THE NASOPHARYNGEAL, TRACHEOBRONCHIAL, AND PULMONARY LUNG RESPECTIVELY. THE LETTERS (a) THROUGH (j) INDICATE THE PROCESSES WHICH TRANSLOCATE MATERIAL FROM ONE COMPARTMENT TO ANOTHER

FIGURE 3-1



DOSE RATE TO ORGANS AS A FUNCTION OF TIME DUE TO CHRONIC INHALATION OF ONE MICRON (AMAD) PLUTONIUM PARTICLES AT A CONCENTRATION OF 2.6 fCi/m^3 . EQUILIBRIUM DOSE RATE TO PULMONARY LUNG 1 MRAD PER YEAR ADULT REFERENCE MAN — BREATHING RATE 2.3×10^4 LITERS PER DAY.

FIGURE 3-2

reciprocal translocations as determined by cytogenetic analyses is between 23 and 50. However, the observed RBE for single locus mutations in developing offspring of male mice given plutonium-239 compared to those given x-ray irradiation is 4. The average RBE for reciprocal translocations and for specific locus mutations is 20. Since reported neutron RBE's are similar to those listed above for plutonium-239 alpha radiation, we used an RBE of 20 to estimate genetic risks for all high-LET radiations. This is consistent with the RBE for high-LET particles recommended for estimating genetic risks associated with space flight.

Human genetic risk estimates are limited by the lack of confirming evidence of genetic effects in humans, and depend on a presumed resemblance of radiation effects in animals to those in humans. The study with the largest data base, the Japanese A-bomb survivors, appears, at best, to provide only an estimate of the minimum doubling dose for calculating the maximum genetic risk in man. However, doubling dose estimates are also uncertain since the number of human disorders having a recognized genetic component is constantly increasing, and the type of genetic damage implicated in a specific disorder may change. The combined uncertainties in doubling dose estimates and the magnitude of genetic contributions to various disorders probably introduces an overall uncertainty of about an order of magnitude in the risk estimates.

3.2.5 CALCULATIONAL MODELS

Estimates of the annual dose rate to organs and tissues of interest are calculated using models recommended by the International Commission on Radiological Protection, supplemented by comparable models proposed by the British National Radiation Protection Board. These dose rates are used in conjunction with a life table analysis of the increased risk of cancer due to

radiation. This analysis takes account of both competing risks and the age of the population at risk.

Internal exposures occur when radioactive material is inhaled or ingested. The RADRISK code developed by the U.S. Environmental Protection Agency implements current dosimetric models to estimate the dose rates at various times to specified reference organs in the body from inhaled or ingested radionuclides. The dosimetric methods in RADRISK are adapted from those of the INREM II code, based primarily on models recommended by the International Commission on Radiological Protection and the British National Radiation Protection Board. The principal qualitative difference is that RADRISK computes dose rates to specified organs separately for high and low linear energy transfer (LET) radiations, whereas INREM II calculates the committed dose equivalent to specified organs. The direct intake of each nuclide is treated as a separate case. For chains, the ingrowth and dynamics of daughters in the body after intake of a parent radionuclide are considered explicitly in the calculation of dose rate. Consideration is also taken of different metabolic properties for the various radionuclides in a decay chain.

The ICRP Task Group Lung Model (Figure 3-1) is used to calculate particulate deposition and retention in the respiratory tract. This model has four major regions: the naso-pharyngeal, tracheobronchial, pulmonary, and lymphatic tissues. A fraction of the inhaled activity is initially deposited in each of the naso-pharyngeal, tracheobronchial, and pulmonary regions. The material clears from the lung to the blood and the gastrointestinal tract. Deposition and clearance of inhaled particulates in the lung are controlled by the particle size and solubility classification. The size distribution of the particles is specified by the activity median aerodynamic diameter (AMAD); in this study, all particulates are assumed to have an AMAD equal to 1.0 micron. The model employs three solubility classes, based on the chemical properties of the

nuclide; classes D, W, and Y correspond to rapid (days), intermediate (weeks), and slow (years) clearance, respectively, of material deposited in the respiratory passages.

Activity absorbed by the blood from the GI or respiratory tract is assumed to be distributed immediately to systemic organs. The distribution of activity to these organs is specified by fractional uptake coefficients. The list of organs in which activity is explicitly distributed (herein termed source organs) is element-dependent, and may include such organs as bone or liver.

Parameters for the uptake factors across the intestinal (gut) wall as recommended in ICRP Report 48 have been used. These are larger than those given in ICRP Report 30, and are generally similar to f_1 values for transuranium elements in the general environment published by the British National Radiation Protection Board. Use of these larger f_1 values results in greater estimated doses and risks from ingestion of transuranic materials than those calculated by use of the earlier ICRP recommendations, but has little effect on doses following inhalation.

3.3 RADIATION RISK - A PERSPECTIVE

To provide a perspective on the risk of fatal radiogenic cancers and the hereditary damage due to radiation, we have calculated the risk from background radiation to the U.S. population using the risk coefficients presented in this chapter. The risk due to background radiation is very largely unavoidable; therefore, it is a good basis of comparison with the estimated risks of doses from radionuclides in the environment. Moreover, if there is bias in the estimated risk of radionuclides, the same bias is present in the risk estimates for background radiation.

Low-LET background radiation has three major components: cosmic radiation, which averages to about 28 mrad per year in the United States, terrestrial sources, such as radium in soil, which contribute an average of 26 mrad per year, and the low-LET dose due to internal emitters. The latter differs between organs, to some extent, but for soft tissues is about 24 mrad per year. Fallout from nuclear weapons tests, naturally occurring radioactive materials in buildings, etc. contribute about another 10 mrem for a total low-LET whole body dose of about 90 mrad per year. The lung and bone receive somewhat larger doses due to high-LET radiations. Although extremes do occur, the distribution of this background annual dose to the U.S. population is relatively narrow, and analysis indicates that 80% of the U.S. population would receive annual doses that are between 75 millirad per year and 115 millirad per year.

The BEIR-3 linear models yield an average lifetime risk of fatal radiogenic cancer of 400 per 10^6 person rad for low-LET radiation. The average is for a group having the age and sex specific mortality rates of the 1970 U.S. population. This is reduced by a dose rate effectiveness factor of 2.5 to give a risk of 160 per 10^6 person rad. More recent evaluations, such as the BEIR-5 Report and the 1989 UNSCEAR evaluation have provided similar estimates of risk. The average lifetime risk to an individual due to low-LET background radiation is calculated as follows:

The average duration of exposure in this group is 70.7 years and at 9×10^{-2} rad per year, the average lifetime dose is 6.36 rad. The risk of fatal cancer per person in this group is:

$$\frac{160 \text{ fatalities}}{10^6 \text{ person rad}} \times 6.36 \text{ rad} = 1.0 \times 10^{-3}$$

The probability of death from cancer in the U.S. is about 0.16, i.e. 16%. Thus the calculated percentage of deaths due to low-LET background radiation indicates that about 0.1% of all deaths, or about 0.6% of all cancer deaths, may be due to low-LET background radiation.

The spontaneous incidence of serious congenital and genetic abnormalities has been estimated to be about 105,000 per 10^6 live births, about 10.5% of live births. The low-LET background radiation dose of about 90 mrad/year in soft tissue results in a genetically significant dose of 2.7 rad during the 30 year reproductive generation. Since this dose would have occurred in a large number of generations, the genetic effects of the radiation exposure are thought to be an equilibrium level of expression. Since genetic risk estimates vary by a factor of 20 or more, the Agency uses a log mean value of this range to obtain an average value for estimating genetic risk. Based on this average value, the background radiation may cause 700 to 1000 genetic effects per 10^6 live births, depending on whether or not the oocyte is as sensitive to radiation as the spermatogonia. This result indicates that about 0.67% to 0.95% of the current spontaneous incidence of serious congenital and genetic abnormalities may be due to the low-LET background radiation.

3.4 TRANSURANIUM ELEMENTS - EXPOSURE PATHWAYS AND BIOEFFECTS

3.4.1 INHALATION

In general, the most important pathway for human exposure from plutonium oxide and other transuranium radionuclides in the environment is expected to be inhalation. This route provides a direct pathway for alpha particles to enter a sensitive organ, the pulmonary lung. Subsequently, a fraction of the inhaled material is redistributed via the blood to such important organs as the bone, liver and gonadal tissues. This is in contrast to the ingestion pathway, where the gut walls act as a barrier to plutonium absorption by blood. The dose to the gut wall itself is not a major cause of concern because plutonium alpha particles have a short finite range in tissue, 41 microns (μm), i.e., less than two-thousandths of an inch. Radiosensitive dividing cells in the gut wall are over 100 μm distant from the gut contents and are effectively isolated from the alpha radiation.

The dose to various tissues from inhaled plutonium is highly time dependent. Insoluble material deposited in the pulmonary lung is removed fairly rapidly; half is assumed to be cleared within 500 days. Clearance from other organs is much slower; the estimated biological half-life in the liver is assumed to be 40 years, and 100 years for bone. The dose delivered to an organ is directly related to the residence time of the radioactive material. Following a single acute exposure to airborne plutonium, the lung (pulmonary) dose rate decreases due to the clearance of particles from the lung so that almost all of the dose is received in a few years. In contrast, the dose rates to the liver and bone are relatively constant over this time span.

In the case of chronic environmental contamination leading to a constant annual intake, the temporal pattern of the dose rate to various organs due to inhalation is different. For pulmonary tissues, a constant (equilibrium) dose rate is realized

within relatively few years of the start of exposure. The dose rate to liver increases more slowly and does not equal that to lung tissue until after about 70 years of exposure. The dose rate to bone never approaches that to the lung and liver. Therefore, the total risk from chronic inhalation will vary with the duration of exposure.

3.4.2 DOSE TO LUNG TISSUES

In most cases, the lung is the organ of primary concern when assessing the risks from plutonium and other transuranium elements in soil. Animal studies, particularly those with dogs which have a relatively long life span, indicate that lung cancer can result from inhaled plutonium aerosols. Even so, the assessment of the risk to humans cannot be directly inferred from animal evidence. Almost all lung cancers in dogs exposed to plutonium occur in a different location than radiogenic cancers in humans following exposure to radon daughters. The animal cancers are in the peripheral parts of the lung and often to different cell types than human lung cancers. They are histologically classified as bronchilar carcinomas (a type of adenocarcinoma). In humans, the inhalation of alpha particle emitters (but not transuranium elements) has usually resulted in bronchial cancers (hilar bronchogenic carcinomas). These are primarily epidermoid and anaplastic carcinomas, but include some adenocarcinomas. Cancers histologically similar to those in animals (peripheral adenocarcinomas) are found much less frequently. This difference may be due to differing exposure conditions but must, in part, be due to differences in tissue sensitivity between species.

A National Academy of Sciences committee concluded that "the risk from alpha irradiation of the deep lung tissues would not be underestimated by applying risk factors from human experience with cancer induced by irradiation of the bronchial tree". Therefore, the risk estimates are based on the highest dose rate

received by any lung tissues (pulmonary lung) and risk estimates appropriate for the most sensitive tissues within the lung, the site of bronchial cancers. It should be noted that the dose to bronchial tissues following plutonium inhalation is small compared to that received in the pulmonary lung; we therefore believe this is a very conservative approach.

In assessing the dose and risk due to the inhalation of transuranium elements, only retention in the pulmonary region is of primary importance. Residence time of inhaled materials in the nasopharyngeal and tracheo-bronchial regions is short compared to that in pulmonary tissues.

Dosimetric models for projecting the average distribution of ionizing radiation within body organs due to the inhalation of radioactive aerosols are still somewhat crude. A general model was published in ICRP Report 19, "The Metabolism of Compounds of Plutonium and Other Actinides", as an amended version of a model developed earlier for the ICRP.

Inhaled aerosols are considered on the basis of particle size and other aerodynamic parameters. The fraction of inhaled materials initially retained or exhaled, and the deposition in various portions of the respiratory tract, is a given function of the activity median aerodynamic diameter (AMAD) of the aerosol particles. The rate at which deposited material is removed from the lungs is considered to be a function only of the chemical state of the inhaled material, and not of size or radioactive content. Environmental sources of plutonium and other transuranium elements are likely to be in the oxide or hydroxide form. Actinides in either form are currently classified as Class Y (insoluble) materials by the ICRP. The dose estimates made below apply only to Class Y compounds.

Radioactivity is assumed to leave the pulmonary tissues by three routes; elevation up the tracheobronchial tree via the

mucus elevator into the gut by way of the esophagus and stomach, transport of particulate materials into the lymphatic system and lymph nodes and, most importantly, dissolution into the blood stream. Most of the activity in blood is redeposited into the liver and bone.

The dose to different organs from inhaled aerosols of various sizes has been considered by a number of workers who have written computer codes to quantify the ICRP model. Since the ICRP model is not described in the form of unambiguous equations, there are minor differences between the results obtained with various codes. The EPA code PAID has been used to calculate the annual dose rates due to isotopes of plutonium, americium, and curium for a number of particle sizes. Typical results for five micron, one micron, and five-tenths micron AMAD plutonium-239 aerosols (1 fCi/m^3) are shown in Table 3-5 for the various compartments in the lung for the case of lifetime exposure (70 years). It is seen that most of the radiation dose is received by pulmonary tissue. Given equal concentrations in air, the dose to the pulmonary lung is not very sensitive to particle size; for example, in the size range from 0.05 to 5.0 micron (AMAD) the pulmonary dose decreases by a factor of about five. The aerosol concentration, as a function of AMAD, required to produce a 1 millirad equilibrium dose to Reference Man is shown in Table 3-6.

The ICRP Lung Model classifies inhaled aerosols into three groups depending on the length of time they are retained in the lung: Class Y compounds are retained for years, Class W for weeks, and Class D for days. The retention times are largely controlled by the solubility of the inhaled material in body fluids. There is some evidence from animal experiments that high specific activity plutonium oxides such as $^{238}\text{PuO}_2$ are more soluble than the long half-life transuranium nuclides and should be considered Class W compounds. Americium-241 and curium-244 may in some cases be in a chemical form that leads to Class W

behavior. Because of their shorter residence time in lung tissues, inhaled Class W transuranics results in a relatively small lung dose compared to that received by other organs, due to the translocation and deposition of dissolved material. Therefore, the dose rate to other organs, rather than the dose rate to pulmonary lung may become limiting.

For most transuranium elements (except plutonium-241) the limiting concentration in air of a Class W material is greater than that for a Class Y material containing the same radionuclide. For 1 micron (AMAD) particles the limiting concentration of Class W compounds is about 3 times greater than for Class Y compounds. The aerosol concentration of several transuranium nuclides which would produce a 3 mrad skeletal bone dose in the 70th year of continuous exposure to transuranium radionuclides in the Class W form are shown in Table 3-7, and the annual dose rates for particles with concentration of 1 fCi/m³ and 1 micron (AMAD) following 70 years of inhalation exposure are shown in Table 3-8. More than 99 percent of the equilibrium dose rate to pulmonary lung tissue is achieved after five years of exposure. For other organs the dose rate increases with the duration of exposure. Lifetime exposure to a constant concentration of curium-244 aerosols is unlikely because of its relatively short half-life (17 years).

The inhalation of Class W compounds results in both somatic and genetic risks. Although the limiting dose rate for inhaled Class W compounds is in terms of the dose to bone, the dose to lung tissue is not negligible. The genetic risk for inhaled Class W transuranium materials is larger than for inhaled Class Y aerosols, and nearly the same as the genetic risk for ingestion.

TABLE 3-5

ANNUAL DOSE RATE TO VARIOUS LUNG COMPARTMENTS FROM CHRONIC EXPOSURE TO PLUTONIUM-239 AEROSOLS

Concentration: 1.0 fCi/m³

Particle AMAD: 0.05, 1.0 and 5.0 Microns

Duration of Exposure (Years)	Pulmonary mrads/yr. x 10 ⁻¹			Tracheobronchial mrads/yr. x 10 ⁻²			Nasopharyngeal mrads/yr. x 10 ⁻⁶		
	0.05u	1.0u	5.0u	0.05u	1.0u	5.0u	0.05u	1.0u	5.0u
1	3.9	1.5	.7	2.7	1.1	6.1	.04	11.	30.
5	9.1	3.5	1.7	3.7	1.5	7.9	.04	11.	30.
10	9.8	3.8	1.8	3.8	1.6	8.1	.04	11.	30.
70	9.9	3.8	1.8	3.8	1.6	8.1	.04	11.	30.

TABLE 3-6

AEROSOL CONCENTRATIONS IN fCi/m³ PRODUCING A 1 MRAD/YEAR
EQUILIBRIUM DOSE RATE TO THE PULMONARY REGION OF REFERENCE MAN
(Class Y Clearance)

Aerosol AMAD (u)	Pu-238	Pu-239	Pu-240	Pu-241*	Am-241	Cm-244
0.05	1.0	1.0	1.0	330	1.0	1.0
0.10	1.1	1.2	1.2	390	1.1	1.1
0.30	1.6	1.7	1.7	540	1.6	1.6
0.50	1.8	1.9	1.9	630	1.8	1.8
1.0	2.5	2.6	2.6	850	2.4	2.5
2.0	3.4	3.5	3.5	1,100	3.3	3.4
3.0	4.1	4.3	4.3	1,400	4.0	4.1
5.0	5.2	5.4	5.4	1,800	5.1	5.2

* only alpha dose rate due to Am-241 daughter is considered

TABLE 3-7

AEROSOL CONCENTRATIONS IN fCi/m³ PRODUCING
A 1 MRAD/YR SKELETAL BONE DOSE IN THE 70th YEAR
(Lifetime Inhalation by Reference Man - Class W Solubility)

Size (AMAD)	²³⁸ ₉₄ Pu	²³⁹ ₉₄ Pu	²⁴¹ ₉₄ Pu*	²⁴¹ ₉₅ Am	²⁴⁴ ₉₆ Cm
0.05	1.9	1.6	65	1.6	3.7
0.10	2.1	1.8	75	1.8	4.0
0.30	2.9	2.1	85	2.1	4.7
0.50	2.9	2.2	90	2.1	5.0
1.0	2.6	2.2	90	2.1	5.0
2.0	2.4	2.0	85	2.0	4.7
3.0	2.2	1.9	80	1.8	4.3
5.0	2.1	1.8	75	1.8	4.0

Beta Emitter - includes only alpha dose rate due to the Am-241 daughter

TABLE 3-8

ANNUAL DOSE RATES TO VARIOUS ORGANS OF REFERENCE MAN
 FOR LIFETIME EXPOSURE TO TRANSURANIUM RADIONUCLIDES
 Class W Concentration = 1 fCi/m³ Size = 1 micron (AMAD)

Nuclide	Equilibrium Dose Rate (microrad/y)		Dose rate in the 70th year (microrad/y)	
	Pulmonary Lung	Tracheobronchial Lung	Liver	Bone Marrow
Pu-238	41	16	930	400
Pu-239	39	15	930	400
Pu-241	0.04	0.01	20	9
Am-241	41	41	960	410
Cm-244	44	17	460	180

3.4.3 DOSE TO BONE, LIVER, AND THE TOTAL BODY

A portion of the aerosols initially deposited in lung tissue is soluble, eventually enters the bloodstream and is redeposited into other body organs. The current ICRP model assumes that 45% of plutonium is redeposited in bone, 45% in liver, and a small fraction in gonads. Release from these organs is slow; 40 years is the assumed half-life in liver and 100 years the assumed half-life in bone and perhaps longer (no observed release) in gonadal tissue. For this assessment it is assumed that a 100 year half-life is appropriate for both bone and gonadal tissue. Percent deposition of other transuranium elements is similar to that for plutonium and ICRP Report 19 has recommended that the percentages shown above be used for all transuranium elements. However, ICRP Report 48, based on more recent data, suggests that the distribution may differ substantially for the other transuranium elements. There is also considerable evidence that the retention times in body organs are shorter than earlier estimates.

The dose rates to various body organs from continuously inhaled plutonium-239 (oxide), americium-241, curium-244 and plutonium-241 as a function of years of exposure are shown in Table 3-9, based on the assumption that all are Class Y particulates. Dose rates have been calculated by averaging the energy absorbed over the total organ: 1800 grams for liver; 5000 grams for bone; 15 grams for the respiratory lymph nodes. Almost all of the dose to liver and bone is from material in the lungs and lymph nodes that has been dissolved and transferred via the blood. Transfer to blood of swallowed materials is much less important.

3.4.4 INGESTION

The magnitude of calculated doses due to the ingestion of transuranium elements is directly proportional to the fraction of

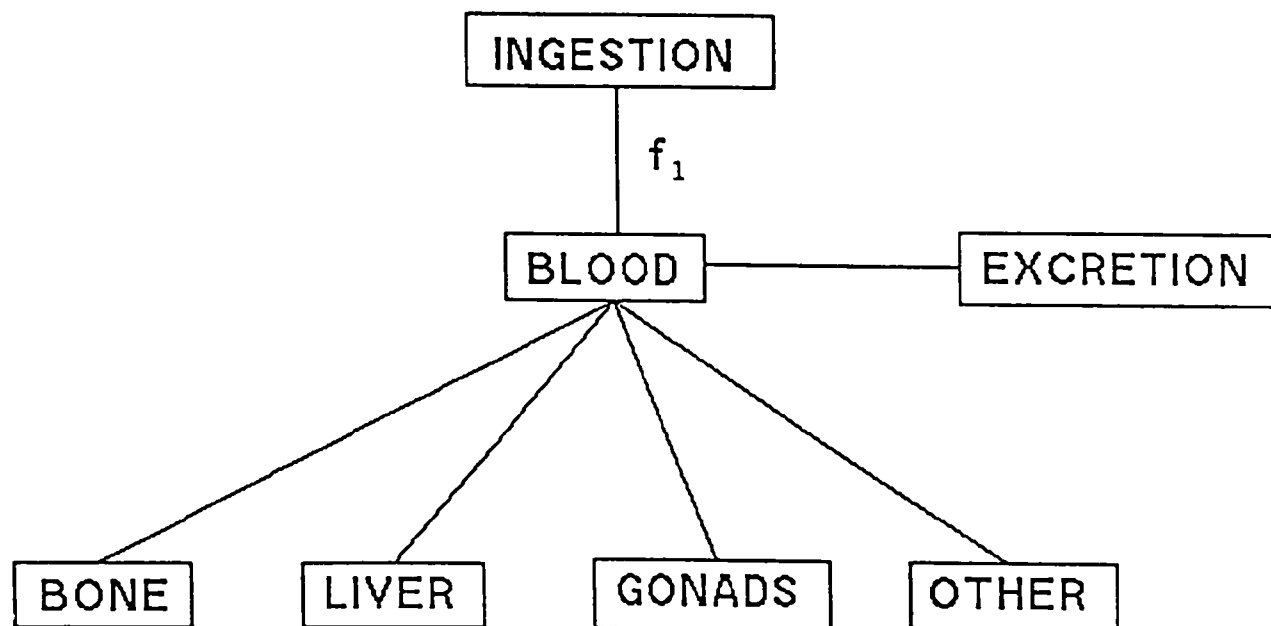


FIGURE 3-3

TABLE 3-9

**ANNUAL DOSE RATES TO VARIOUS ORGANS
FROM CHRONIC INHALATION OF TRANSURANIUM RADIONUCLIDES
(In Millirad/Year)**

Aerosol AMAD: 1 μ mConcentration: 1 fCi/m³ $f_1=10^{-3}$

Nuclide: Pu-239					Nuclide: Pu-241/Am-241*				
<u>Duration of Exposure</u>	<u>Liver</u>	<u>Skeletal</u>	<u>Bone Red Marrow</u>	<u>Endosteal</u>	<u>Liver</u>	<u>Skeletal</u>	<u>Bone Red Marrow</u>	<u>Endosteal</u>	
1 year	1.0 E-3	5.0 E-4	4.8 E-4	6.6 E-3	1.4 E-6	5.0 E-7	4.6 E-7	5.8 E-6	
5 years	1.8 E-2	6.5 E-3	6.2 E-3	8.6 E-2	8.7 E-5	3.2 E-5	3.0 E-5	3.7 E-4	
10 years	5.2 E-2	1.9 E-2	1.8 E-2	2.5 E-1	4.6 E-4	1.7 E-4	1.6 E-4	2.0 E-3	
15 years	8.9 E-2	3.4 E-2	3.2 E-2	4.5 E-1	1.1 E-3	4.1 E-4	1.6 E-4	2.0 E-3	
20 years	1.3 E-1	4.9 E-2	4.7 E-2	6.5 E-1	1.8 E-3	7.1 E-4	6.6 E-4	4.8 E-3	
30 years	1.9 E-1	7.8 E-2	7.4 E-2	1.0 E-0	3.4 E-3	1.4 E-3	1.3 E-3	1.6 E-2	
40 years	2.4 E-1	1.1 E-1	1.1 E-1	1.5 E-0	5.0 E-3	2.2 E-3	2.0 E-3	2.6 E-2	
50 years	2.9 E-1	1.3 E-1	1.2 E-1	1.7 E-0	6.4 E-3	3.0 E-3	2.8 E-3	3.5 E-2	
70 years	3.6 E-1	1.7 E-1	1.6 E-1	2.3 E-0	8.0 E-3	4.4 E-3	4.1 E-3	5.1 E-2	
Nuclide: Am-241					Nuclide: Cm-244/Pu-240				
<u>Duration of Exposure</u>	<u>Liver</u>	<u>Skeletal</u>	<u>Bone Red Marrow</u>	<u>Endosteal</u>	<u>Liver</u>	<u>Skeletal</u>	<u>Bone Red Marrow</u>	<u>Endosteal</u>	
1 year	1.5 E-3	5.0 E-4	4.6 E-4	5.8 E-3	1.6 E-3	6.0 E-4	5.6 E-4	6.4 E-3	
5 years	1.9 E-2	7.0 E-3	6.3 E-3	8.1 E-2	1.8 E-2	1.6 E-3	1.5 E-3	1.7 E-2	
10 years	5.5 E-2	2.1 E-2	1.9 E-2	2.4 E-1	4.7 E-2	1.7 E-2	1.6 E-2	1.8 E-1	
15 years	9.5 E-2	3.6 E-2	3.3 E-2	4.2 E-1	7.3 E-2	2.8 E-2	2.6 E-2	3.0 E-1	
20 years	1.3 E-1	5.2 E-2	4.8 E-2	6.0 E-1	9.4 E-2	3.7 E-2	3.4 E-2	3.9 E-1	
30 years	2.0 E-1	8.2 E-2	7.5 E-2	9.5 E-1	1.2 E-1	4.9 E-2	4.5 E-2	5.2 E-1	
40 years	2.6 E-1	1.1 E-1	1.0 E-1	1.3 E-0	1.4 E-1	5.7 E-2	5.3 E-2	6.1 E-1	
50 years	3.0 E-1	1.4 E-1	1.3 E-1	1.6 E-0	1.5 E-1	6.3 E-2	5.8 E-2	6.7 E-1	
70 years	3.7 E-1	1.8 E-1	1.7 E-1	2.1 E-0	1.6 E-1	6.8 E-2	6.3 E-2	7.3 E-1	

*Alpha dose only - 70th year beta dose rates: liver = 0.11 urad;
bone = 0.049 urad.

the ingested material that is assumed to cross the gut wall and enter the blood stream. This fraction is not well known and it is reasonable to assume that it varies considerably depending on the solubility of the ingested material. Animal experiments to measure gut transfer, which used highly insoluble laboratory prepared materials in the oxide form, yield transfers in the parts per million range. However, plutonium oxide found in the environment has been shown to be much more soluble than the refractory oxides utilized in animal experiments. This is in agreement with recent experiments showing plutonium oxides formed at low temperatures are more soluble than those formed at high temperatures. Therefore, it is reasonable to assume that plutonium oxidized in the environment will not be as insoluble as the materials which have been used to determine plutonium gut transfer in animals. A tabulation of recently published values for the transfer coefficient of transuranium elements across the gut wall is shown in Table 3-10.

In contrast to inhalation, the ingestion dose to liver and the resultant risk is a direct function of the amount of radioactivity transferred to blood via the gastrointestinal tract. For transuranium elements having a physical half-life comparable to the residence time in liver, the risk will be somewhat greater since the dose approaches the equilibrium value earlier in life. Averaging the estimates of early death yields an estimated risk of two deaths per 100,000 exposed persons, somewhat less than for cancer of the bone. For Am-241, Pu-241, and Cm-244 this average lifetime risk is 2.1, 1.2 and 3.2 per 100,000 exposed, respectively.

The transfer factors from gut to blood have been utilized to calculate annual dose rates as functions of the duration of ingestion. Dose rates to bone, liver, and the total body of reference man due to the chronic ingestion of 1000 pCi/year of plutonium-239 oxide, americium-241, and curium-244 are shown in Table 3-11. Pu-241, half life 14.8 years, and Cm-244, half life

TABLE 3-10

FACTORS FOR ABSORPTION FROM THE GASTRO-INTESTINAL TRACT FOR TRANSURANIUM ELEMENTS

Element/Chem Form		ICRP-30*	ICRP-48**
Pu-238	Oxide	10^{-5}	10^{-5}
	Nitrate	10^{-4}	10^{-4}
	Other	10^{-4}	10^{-3}
Pu-239	Oxide	10^{-5}	10^{-5}
	Nitrate	10^{-4}	10^{-4}
	Other	10^{-4}	10^{-3}
Am		5×10^{-4}	10^{-3}
Cm		5×10^{-4}	10^{-3}
Np		10^{-2}	10^{-3}

* ICRP-30 = occupational exposures

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** ICRP-48    general population exposure (via food pathway)
               for plutonium                      = 10-3
               for all other transuranium elements = 10-3
               children under one year = 10 x value for adults.

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17.9 years, are the most rapidly decaying transuranium elements capable of causing chronic exposures in a contaminated environment. Because of their short half lives, the occurrence of lifetime ingestion of these radionuclides is remote. This is even more true of curium-242, half life 0.045 years, where only acute intake is a plausible mode of exposure.

The organ dose rates were calculated on the basis of organ masses appropriate for reference man, not children. However, there is enough proportionality between food intake and organ mass as a function of age so that the results are applicable to a lifetime exposure situation. Because there is some evidence from animal studies that the newborn have a particularly high transfer of transuranium elements across the gut wall to blood, the average transfer fractions may not be applicable to infants, (less than one year old). The duration of this increased transfer is unknown for humans, perhaps a few weeks or considerably longer. A comparison of average skeletal dose rates with and without increased infant uptake is shown in Table 3-12.

3.4.5 RISK OF BONE CANCER

Unlike radium-226, which is distributed throughout the bone volume following long-term ingestion, plutonium is preferentially deposited and retained on endostial bone surfaces, principally in the organic matrix. In some cases as much as 30-50% of the endostial plutonium has been shown to be retained on osteogenic cells. Americium and curium are also retained on bone surfaces. Alpha particle emitters which are retained on bone surfaces have been shown to be more tumorigenic than radium-226 and other bone volume seekers. Surface seekers deliver a higher dose to osteogenic cells adjacent to bone surfaces, and such doses are thought to be the cause of radiogenic bone sarcomas.

TABLE 3-11

ANNUAL DOSE RATE DUE TO CHRONIC INGESTION OF
PLUTONIUM-239 OXIDE, AMERICIUM-241, PLUTONIUM-241, & CURIUM-244
(In Microrad/Year)

Annual Intake = 1000 pCi/Year

$$f_1 = 10^{-3}$$

Duration of Ingestion (Years)	Plutonium-239 Oxide		
	<u>Red Marrow</u>	<u>Bone</u> <u>Endosteal</u>	<u>Liver</u>
1	7.6	1.1 E+2	24
5	36.5	5.0 E+2	116
10	71.2	9.8 E+2	220
15	102	1.4 E+3	320
20	136	1.9 E+3	410
30	204	2.8 E+3	560
40	254	3.5 E+3	690
50	314	4.3 E+3	810
70	407	5.6 E+3	980

Duration of Ingestion (Years)	Americium-241		
	<u>Red Marrow</u>	<u>Bone</u> <u>Endosteal</u>	<u>Liver</u>
1	7.7	9.8 E+1	25
5	38	4.8 E+2	120
10	74	9.4 E+2	230
15	110	1.4 E+3	340
20	140	1.8 E+3	430
30	200	2.6 E+3	590
40	260	3.3 E+3	720
50	320	5.2 E+3	830
70	410	5.2 E+3	990

Duration of Ingestion (Years)	Plutonium-241/Americium-241		
	<u>Red Marrow</u>	<u>Bone</u> <u>Endosteal</u>	<u>Liver</u>
1	0.006	0.08	0.02
5	0.14	1.8	0.45
10	0.51	6.5	1.6
15	1.0	13	3.1
20	1.7	22	4.9
30	3.2	41	8.7
40	4.9	62	12
50	6.5	82	16
70	9.4	119	21

Duration of Ingestion (Years)	Curium-244/Plutonium-240		
	<u>Red Marrow</u>	<u>Bone</u> <u>Endosteal</u>	<u>Liver</u>
1	7.9	9.0 E+1	2.6 E+1
5	37	4.2 E+2	1.2 E+2
10	65	7.5 E+2	2.1 E+2
15	92	1.1 E+3	2.8 E+2
20	108	1.2 E+3	3.3 E+2
30	133	1.5 E+3	3.9 E+2
40	150	1.7 E+3	4.3 E+2
50	160	1.8 E+3	4.6 E+2
70	175	2.0 E+3	4.8 E+2

TABLE 3-12

AVERAGE SKELETAL DOSE RATES DUE TO CHRONIC INGESTION OF
OF PLUTONIUM-239 OXIDE

WITH AND WITHOUT INCREASED INFANT UPTAKE

Annual Intake = 1000 pCi/Year

Average Skeletal Dose Rate in Microrad/Year

Age	Without Enhanced Infant Absorption*	With Enhanced Infant Absorption**
1	9	86
5	43	76
10	84	108
15	124	133
20	162	170
30	235	242
40	303	310
50	366	372
70	480	485

*GI tract to blood transfer 10^{-3} all ages.

**GI tract to blood transfer 10^{-2} first year of life.

There is no clinical evidence of bone cancer being caused by plutonium. The most relevant human data is for medical patients treated with radium-224, which, because of its short half-life (3.64 days), is retained mainly on bone surfaces. A large number of patients (approximately 900) who were treated with radium-224 for tuberculosis and ankylosing spondylitis have been followed for bone cancer incidence. The dose to these patients has been calculated in terms of the average skeletal dose, defined as total alpha energy emitted divided by bone mass, even though the dose distribution is very nonuniform. On this basis, it has been estimated that for chronic irradiation due to Pu-239, 200 bone cancers will be produced per 10^6 rad to a 7 kg skeletal mass. In terms of the dose to mineral bone, these results yield 140 bone cancers per rad to osseous tissue.

Because of uncertainties in the redistribution of Ra-224 following its initial deposition on bone surfaces, the above estimate of the average skeletal dose delivered in the Ra-224 cases may be too high, leading to an underestimate of the risk per rad for radium-224. While the above analysis assumes that half of the skeletal radium-224 decays on bone surfaces and half in the bone volume, others have stated that only 1.5% of the skeletal radium-224 decays within the bone volume away from bone surfaces. This would increase the risk per rad by 174%. Furthermore, this model predicts that for radium-224 on bone surfaces, the dose rate to osteogenic cells near the bone surface is 8.9 times the average skeletal dose rate; for plutonium-239, 12.8 times. In terms of average to osseous tissues as calculated for these risk estimates, plutonium-239 would therefore deliver 1.44 times as much alpha particle dose to osteogenic cells as radium-224. This estimate is likely to be too high, since it assumes all the plutonium is retained on bone surfaces and none is buried in the remodeling or bone growth process.

The residence time of plutonium on bone surfaces depends on age. In rapidly growing animals, it is relatively short, while

there is a prolonged Pu-239 bone surface residence time in adult bone and they accumulate more Pu-239 with time. Since almost all of the body burden is assumed during adult life, the exposure regime due to chronic plutonium inhalation and ingestion may favor a surface dose distribution. After giving due consideration to the smaller number of bone precursor cells in adults, Jee, et al. have characterized the plutonium-239 injury to bone as low in rapidly growing beagles, moderate for young adults, and high for adults. A similar characterization for degrees of insult would appear to hold for humans, particularly when subject to chronic exposure.

3.4.5 CANCER OF THE LIVER

The magnitude of the potential risk of inducing liver cancer by means of plutonium and other transuranium elements has been recognized only recently. Earlier regulations were based on the 1959 NCRP-ICRP assumption that the critical organ for plutonium deposited from blood is bone. More recently, it has been recognized that deposition in liver is as likely as in bone, and ICRP Report 19 assumes that 45% of the plutonium and other transuranium elements dissolved in blood is deposited in the liver and an equal amount in bone. Based on the results of animal studies, deposition in liver may be somewhat lower for plutonium and somewhat higher for americium and curium.

The risk to humans from alpha emitters deposited in the liver can be assessed on the basis of rather limited information obtained from epidemiological studies of medical patients. Earlier in this century, a low specific activity alpha particle emitting contrast medium called Thorotrast was utilized in some diagnostic x-ray procedures. In subsequent years patients who were treated with it, mainly European, have been followed clinically and shown to have a higher than expected incidence of liver cancer. These data are pertinent although they do have limitations. Because the amount of material injected into the

blood in these studies was quite large, its deposition in the liver was uneven. Liver cancer incidence in this group would not necessarily be higher than might be expected for a more uniform deposition. On the contrary, there is a general consensus that highly localized concentrations of alpha particle emitters are likely to be less carcinogenic than a more uniform distribution. Another possible limitation of these data is that the relatively large quantity of Thorotrast deposited in the liver could lead to a foreign body response, which might in turn result in cancer. While the quantitative applicability of the human experience with Thorotrast to the prediction of plutonium risks to liver is tentative, there is an abundance of experimental animal data showing that liver cancers can be induced by plutonium. Liver cancers are seen less frequently than bone cancers in most experiments with animals, but since liver cancers have a longer latent period than bone cancers, they may be more important in a longer-lived species such as man. The primary source of data on Thorotrast patients is Faber's review, also cited by the British Medical Research Council (MRC) as a basis for its assessment of plutonium toxicity. Estimates of risk for liver cancer are: absolute risk, 4.2×10^{-6} liver cancer for each organ rad per year at risk; relative risk, an 11% increase per rad.

3.4.6 LEUKEMIA DUE TO BONE MARROW IRRADIATION

Several authors have pointed out that there is a potential risk of leukemia from plutonium incorporated into bone tissues. Alpha particles originating in trabeculae may irradiate a significant fraction of the bone marrow, and the plutonium in marrow itself will act as a source. Based on auto-radiographic studies of bone and bone marrow, it has been estimated that the dose to trabecular marrow is 88 percent of the average skeletal dose due to plutonium-239.

The International Commission on Radiological Protection states in ICRP Report 26 that "The red bone marrow is taken to be the tissue mainly involved in the causation of radiation-induced leukemia; other blood-forming tissues are thought to play a minor role in leukemogenesis." Observations on humans irradiated for therapeutic purposes or on Japanese survivors of nuclear explosions indicate that the incidence of radiation-induced leukemia reaches its peak within a few years of irradiation, and returns to pre-irradiation levels after about 25 years. For radiation protection purposes the risk factor (for gamma radiation) is taken to be 2×10^{-3} per Sievert. [This is equivalent to 4×10^{-4} /rad, or 400 per 10^6 person year rad, for alpha radiation with an RBE of 20] Using this factor, the incremental risk due to leukemia for the inhalation mode of exposure is comparable to the risk to pulmonary tissue. However, the BEIR-3 Report states that "It should be recognized that risk (of leukemia) estimated at a selected point in the high- dose region may overestimate the magnitude of hazards of low-dose exposures by a factor of 2-10, depending on the type of radiation, its rate of delivery, and the high-dose point at which the observations were made."

The estimated risk of leukemia due to the ingestion of transuranium elements is somewhat greater than for the inhalation mode. For a 3 mrad limiting dose in the 70th year to skeletal bone, the leukemia risk ranges from 0.4 to 1.6 cases per 100,000 exposed for the absolute and relative risk models respectively and is comparable to the estimated risk of liver cancer.

3.5 COMPARISON OF INHALATION AND INGESTION RISKS

The total somatic risks to persons from exposure at dose rates in accord with the objectives of the Recommendations on Dose Rates from Transuranium Elements have been calculated in accord with the above discussion. A summary of estimated lifetime risks from inhalation and ingestion of plutonium-239 is shown in Table 3-13.

TABLE 3-13

ESTIMATED RISKS IN THE 70th YEAR
FROM INHALATION AND INGESTION OF PLUTONIUM-239

	<u>Inhalation</u> Class Y *	<u>Ingestion</u> ** Class W
Lung Cancer	0.67×10^{-6}	-
Liver Cancer	0.25×10^{-6}	0.62×10^{-6}
Leukemia	0.15×10^{-6}	0.34×10^{-6}
Bone Cancer	0.12×10^{-6}	0.26×10^{-6}
Total	1.19×10^{-6}	1.22×10^{-6}

* Inhalation: Class Y is normalized to an equilibrium dose rate to pulmonary lung of 1 mrad/yr (AMAD = 1 μ m and air conc = 2.6 fCi/m³)

** Ingestion: Class W is normalized to a dose rate of 1 mrad/yr to skeletal bone in the 70th year of continuing exposure

3.6 GENETIC DAMAGE

The degree to which transuranium elements are translocated from human blood to gonadal tissues is not well known due to the analytical difficulty of making reasonably precise measurements at the low activity levels usually involved, and the large variability between individuals in the general population. In addition to limited information from studies of laboratory animals, there are three sources of post-mortem human data: the general population exposed to fallout plutonium, industrially exposed radiation workers, and a few clinical studies with hospital patients. Richmond and Thomas reported that for the five animal species considered in their 1974 review, an average of 0.03% of the plutonium in blood was transferred to gonadal tissue. The data on which this average is based varied by a factor of about ten. A review of clinical data, based on only four persons, also leads to an estimate of about 0.03% for transfer from blood to gonadal tissue.

The British Medical Research Council (MRC) also reviewed this problem in its 1975 analysis of plutonium toxicity and concluded that 0.05% of the plutonium in blood would be transferred to gonadal tissue. Since the mass of the ovary is 11 grams, the MRC estimate on transfer from blood is equivalent to 0.005% per gram of ovary. The mass of testes is greater than that of the ovary by a factor of about 3. The MRC assumed equal quantities of plutonium in each, so that the concentration (percent per gram) in the testes would be about one-third of that in ovarian tissue, or 0.002% per gram. Other authors have estimated that the amount of plutonium in the smaller female gonad was a factor of five to ten less than males, a somewhat less conservative assumption. Recent data reported for plutonium in beagle gonads indicates that the concentration per gram is about 0.0055% in ovaries and 0.0012% in testes.

Risks due to transuranium element are not only to persons directly exposed to the radiation but also to their progeny. Alpha particles can damage the male progenitor cells producing sperm and the egg cell (oocyte) in the female. The expression of this damage is either genetic impairment of the live-born offspring or fetal death.

Risk estimates for genetic damage are based on the gonadal dose received in a reproductive generation, i.e., the first 30 years of life. The 30-year gonadal doses due to chronic ingestion of transuranium elements that would also cause a 3 millirad per year dose rate to red bone marrow after 70 years, are shown in Table 3-14. Chronic ingestion of Cm-244 would cause genetic risks about two times larger. For the given recommendations, genetic effects due to inhalation are substantially less important than those due to ingestion.

It is estimated that a 30-year dose of one millirad due to alpha-emitting transuranium elements in gonadal tissue may cause between 0.1 and 2 genetic effects per 100,000 live births in the first generation. If this gonadal dose were to continue indefinitely so that a new equilibrium of genetic damage was established in the population, the risk might increase to 0.6 to 15 per 100,000 live births. Currently, the rate of observed genetic effects in the U.S. is about 6000 per 100,000 live births.

TABLE 3-14

OCCUPATIONAL EXPOSURE

ANNUAL LIMIT OF INTAKE (ALI) AND DERIVED AIR CONCENTRATION (DAC)
FOR THE MORE IMPORTANT TRANSURANIUM NUCLIDES

(From Federal Guidance Report No. 11 EPA 520/1-88-020)

Nuclide/ $t_{1/2}$	Class/ f_1	Inhalation		Ingestion	
		ALI μCi	DAC $\mu\text{Ci}/\text{m}^3$	f_1	ALI μCi
NEPTUNIUM					
Np-236 5 10^4 y	W 10^{-1}	0.02	$9 \cdot 10^{-1}$	0.001	3
Np-237 2.14 10^6 y	W 10^{-3}	0.004	$2 \cdot 10^{-12}$	0.001	0.5
Np-238 2.117 d	W 10^{-1}	60	$3 \cdot 10^{-6}$	0.001	1000
Np-239 2.355 d	W 10^{-1}	2000	$9 \cdot 10^{-7}$	0.001	2000
PLUTONIUM					
Pu-238 87.74 y	W 10^{-3}	0.007	$3 \cdot 10^{-12}$	0.001	0.9
	Y 10^{-3}	0.02	$8 \cdot 10^{-11}$	$1 \cdot 10^{-4}$	9
Pu-239 24065 y	W 10^{-1}	0.006	$3 \cdot 10^{-11}$	0.001	0.8
	Y 10^{-1}	0.02	$7 \cdot 10^{-12}$	$1 \cdot 10^{-4}$	8
Pu-240 6537 y	W 10^{-3}	0.006	$3 \cdot 10^{-11}$	0.001	0.8
	Y 10^{-1}	0.02	$7 \cdot 10^{-11}$	$1 \cdot 10^{-4}$	8
Pu-241 14.4 y	W 10^{-1}	0.3	$1 \cdot 10^{-10}$	0.001	30
	Y 10^{-1}	0.8	$1 \cdot 10^{-10}$	$1 \cdot 10^{-4}$	400
Pu-242 3.76 10^5 y	W 10^{-1}	0.007	$3 \cdot 10^{-12}$	0.001	0.8
	Y 10^{-1}	0.02	$7 \cdot 10^{-12}$	$1 \cdot 10^{-4}$	8
AMERICIUM					
Am-241 432.2 y	W 10^{-1}	0.006	$3 \cdot 10^{-11}$	$1 \cdot 10^{-4}$	0.8
Am-242 152 y	W 10^{-3}	0.006	$3 \cdot 10^{-12}$	$1 \cdot 10^{-4}$	0.8
Am-242m 16.02 h	W 10^{-1}	80	$4 \cdot 10^{-6}$	$1 \cdot 10^{-4}$	4000
Am-243 7380 y	W 10^{-3}	0.006	$3 \cdot 10^{-12}$	$1 \cdot 10^{-4}$	0.8
Am-244 10.1 h	W 10^{-1}	200	$8 \cdot 10^{-6}$	$1 \cdot 10^{-4}$	3000
Am-244m 26 m	W 10^{-1}	4000	$2 \cdot 10^{-6}$	$1 \cdot 10^{-4}$	$6 \cdot 10^4$
CURIUM					
Cm-242 162.8 d	W 10^{-1}	0.3	$1 \cdot 10^{-10}$	$1 \cdot 10^{-3}$	30
Cm-244 18.11 y	W 10^{-1}	0.01	$5 \cdot 10^{-12}$	$1 \cdot 10^{-4}$	1

TABLE 3-15

THIRTY-YEAR GONADAL DOSE DUE TO AN INGESTION PATTERN
RESULTING IN A 3 MILLIRAD/YEAR ALPHA PARTICLE DOSE RATE
TO BONE IN THE 70th YEAR
(Chronic Lifetime Ingestion At A Constant Rate)

Transuranium Element	Gonadal Dose (millirad)
Pu-238	4.3
Pu-239	3.7
Pu-240	3.7
Pu-241	2.0
Am-241	4.0
Cm-244	7.0

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4. TRANSURANIUM ELEMENTS IN THE ENVIRONMENT

[Reprinted with Minor Changes from "Proposed Guidance on Dose Limits for Persons Exposed to Transuranium Elements", EPA/520/4-77/016]

4.1 INTRODUCTION

Plutonium and other transuranium elements have been released into the general environment primarily from four sources. In order of decreasing importance, these include:

- a. Aboveground testing of nuclear weapons
- b. Accidents involving nuclear weapons and satellite power sources
- c. Accidents at nuclear facilities
- d. Planned discharges of effluents from nuclear facilities

The aboveground testing of nuclear weapons during 1945-1963 caused a worldwide dispersal of plutonium and americium. For the most part, this radioactivity was injected into the stratosphere and has been redeposited more or less uniformly over the earth's lands and waters. This redeposited plutonium and americium is available to people through inhalation and ingestion pathways and exists as an ubiquitous source in the general environment upon which is superimposed the releases of transuranium elements from other sources. Fallout plutonium is primarily a mixture of Pu-239 and Pu-240 with lesser amounts of Pu-238, Pu-241 and Pu-242. About 58% is Pu-239 and 39% Pu-240; because these two radionuclides are essentially identical with respect to chemical behavior and alpha energies, the sum of their activities in the environment will be referred to as "Pu-239" (1). The daughter of Pu-241 ($t_{1/2} = 12y$) is Am-241, so that as the Pu-241 continues to decay, the concentration of Am-241 in the environment increases relative to the amount of Pu-239. The Am-241/Pu-239 activity

ratio in soil is now about 0.25 and will eventually increase to 0.40 (2).

Aboveground nuclear weapons testing produced approximately 430 kilocuries of Pu-239 over the period 1945 to 1974. About 105 kilocuries deposited quickly near the various detonation sites. Of the 325 kilocuries injected into the stratosphere, 250 kilocuries have deposited in the mid-latitudes of the northern hemisphere, 70 kilocuries have deposited in other latitudes and about 5 kilocuries still remained in the stratosphere as of 1974. This has led to cumulative depositions of Pu-239 on ground surfaces in the United States that range from 0.001 to 0.003 $\mu\text{Ci}/\text{m}^2$. Since 1967, sporadic aboveground nuclear tests have held the air concentration level of plutonium to relatively constant values, currently ranging from 0.01 to 0.1 fCi/ m^3 in ground level air. These levels are not believed to be the result of resuspension from soil surfaces.

The cumulative deposition of fallout Pu-239 at selected locations in the United States (3) is shown in Table 4-1. The distribution in soil with respect to depth at both an undisturbed site and a cultivated site is shown in Table 4-2 (4.5). Table 4-3 is a summary of fallout Pu-239 levels in New York City for both air and ground deposition as a function of time from 1954 to 1975 (6). The total amount of Pu-238 injected into the stratosphere from aboveground nuclear tests is about 9 kilocuries (3). In addition, 17 kilocuries of Pu-238 were released in the high stratosphere of the southern hemisphere when a satellite containing a nuclear power source (SNAP-9A) failed to orbit and disintegrated (9). As a result, there are measurable amounts of Pu-238 in most environmental media. An estimated 90 curies of curium (Cm-245 and Cm-246) have been produced as the result of weapons tests (1).

A potential source for future release of the transuranium elements to the general environment are operations associated

TABLE 4-1

Cumulative Deposit of Fallout Pu-239 at Selected
Locations in the United States

Approximate Location	Pu-239 Concentration (a) ($\mu\text{Ci}/\text{m}^2$)
Richland, Washington	0.0014
San Francisco, California	0.0009
Los Angeles, California	0.0007
National Test Site, Montana	0.0019
Rapid City, South Dakota	0.0025
Topeka, Kansas	0.0024
Tulsa, Oklahoma	0.0022
Corpus Christi, Texas	0.0010
Chicago, Illinois	0.0021
Augusta, Maine	0.0017
Cape Cod, Massachusetts	0.0023
Long Island, New York	0.0024
Raleigh, North Carolina	0.0024
Miami, Florida	0.0010
Average ($\pm 2\sigma$)	0.0018 \pm 0.0006

(a) Top 30 cms of soil

TABLE 4-2

Concentration of Fallout Pu-239 in Soil as a
Function of Depth at North Eastham, Massachusetts

Depth (cm)	Undisturbed Site	
	Concentration of Pu-239 ($\mu\text{Ci}/\text{m}^2$)	% of Total
0-2 (Includes Vegetation) (a)	0.91×10^{-3}	52 (a)
2-4	0.37	21
4-6	0.15	8
6-8	0.12	7
8-10	0.052	3
10-12	0.035	2
12-14	0.028	2
14-16	0.027	1
16-21	0.047	3
21-26	<0.004	<0.1
Depth (Cm)	Cultivated Site(5)	
	Concentration of Pu-239 ($\mu\text{Ci}/\text{m}^2$)	% of Total
0-5	0.42×10^{-3}	19
5-10	0.45	20
10-15	0.34	16
15-20	0.37	17
20-25	0.32	15
25-30	0.20	10
30-35	0.02	0.01
35-40	0.02	0.01
40-45	0.02	0.01
45-50	0.02	0.01
50-55	0.02	0.01

(a) 12% of the total plutonium was associated with vegetation

TABLE 4-3

Fallout Pu-239 in New York City

Year	Deposition ($\mu\text{Ci}/\text{m}^2$)	Cumulative deposit ($\mu\text{Ci}/\text{m}^2$)	Surface air concentration (fCi/m^3)
1954	0.07×10^{-3}	0.00007	0.14
1955	0.09	0.00016	0.18
1956	0.12	0.00028	0.23
1957	0.12	0.00040	0.23
1958	0.16	0.00056	0.32
1959	0.23	0.00078	0.45
1960	0.04	0.00082	0.081
1961	0.06	0.00089	0.13
1962	0.32	0.0012	0.63
1963	0.62	0.0018	1.68
1964	0.41	0.0022	0.91
1965	0.14	0.0024	0.33
1966	0.05	0.00245	0.12
1967	0.04	0.0025	0.051
1968	0.04	0.0025	0.08
1969	0.06	0.0026	0.06
1970	0.03	0.0026	0.068
1971	0.03	0.0026	0.06
1972	0.02	0.0027	0.027
1973	0.01	0.0027	0.013
1974	0.02	0.0027	0.039
1975	0.01	0.0027	0.02

with the Light-Water Reactor Fuel Cycle. About 250 kg of plutonium, which is inside the spent fuel rods, is removed per year from a 1000 Mw(e) light-water reactor. The isotopic composition of this plutonium is typically 59% Pu-239, 29% Pu-240, 11% Pu-241, 4% Pu-242 and 2% Pu-238 (9). Table 4-4 is a listing of the nuclear properties of the more significant transuranium nuclides (7).

Sections that follow briefly discuss specific controlled sites and other areas in the general environment which have become contaminated with transuranium elements significantly above levels attributed to atmospheric fallout. These include: local fallout from aboveground nuclear tests, accidents at nuclear facilities, and effluent releases from nuclear facilities. Data were selected to be representative of conditions at these sites during the mid-1970's. Additional information is available in the documents that have been referenced. Table 4-5 provides a summary of these sites, their locations, inventories, and approximate onsite and offsite maximum soil concentration levels.

This chapter was prepared in the mid-70's and has not been revised. The environmental data represent conditions at that time and do not necessarily represent current levels of environmental contamination. In general, concentrations in near-surface soils are depleted with time and the hazards to exposed persons are diminished.

4.2 MAJOR SITES OF ENVIRONMENTAL CONTAMINATION

4.2.1 The Nevada Test Site (NTS)

The Nevada Test Site (10) is an area of about 3500 km² located in Nye County, Nevada, 90 kilometers northwest of Las Vegas. It is surrounded by an exclusion area 25 to 100 kms wide between the test site itself and public lands. The climate is

TABLE 4-4

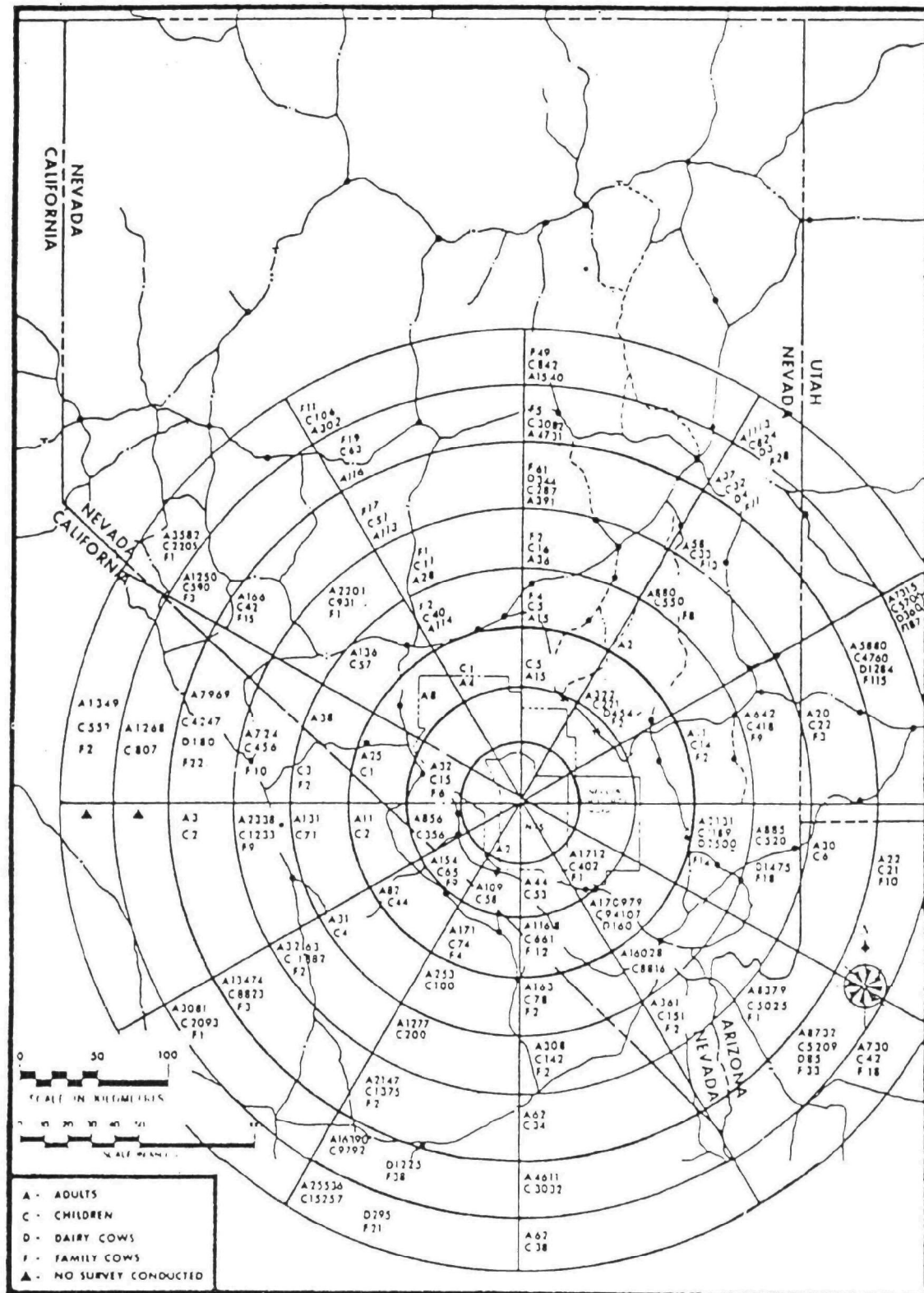
Nuclear Properties of Environmentally Significant Transuranium Radionuclides

<u>Radionuclide</u>	<u>Radiological half-life (y)</u>	<u>Mode of Decay</u>	<u>Energy of Major Radiations (MeV)</u>	<u>Daughter Radionuclide</u>
Pu-238	87.4	alpha	5.50; 5.46	U-234
Pu-239	2.4×10^4	alpha	5.16; 5.11	U-235
Pu-240	6.6×10^3	alpha	5.17; 5.12	U-236
Pu-241	14.3	beta	0.021 (max)	Am-241
Pu-242	3.9×10^5	alpha	4.90; 4.86	U-238
Am-241	443.	alpha	5.49; 5.44	Np-237
Am-243	7.4×10^3	alpha	5.28; 5.23	Np-239
Cm-242	0.45	alpha	6.12; 6.07	Pu-238
Cm-244	18.1	alpha	5.81; 5.77	

TABLE 4-5

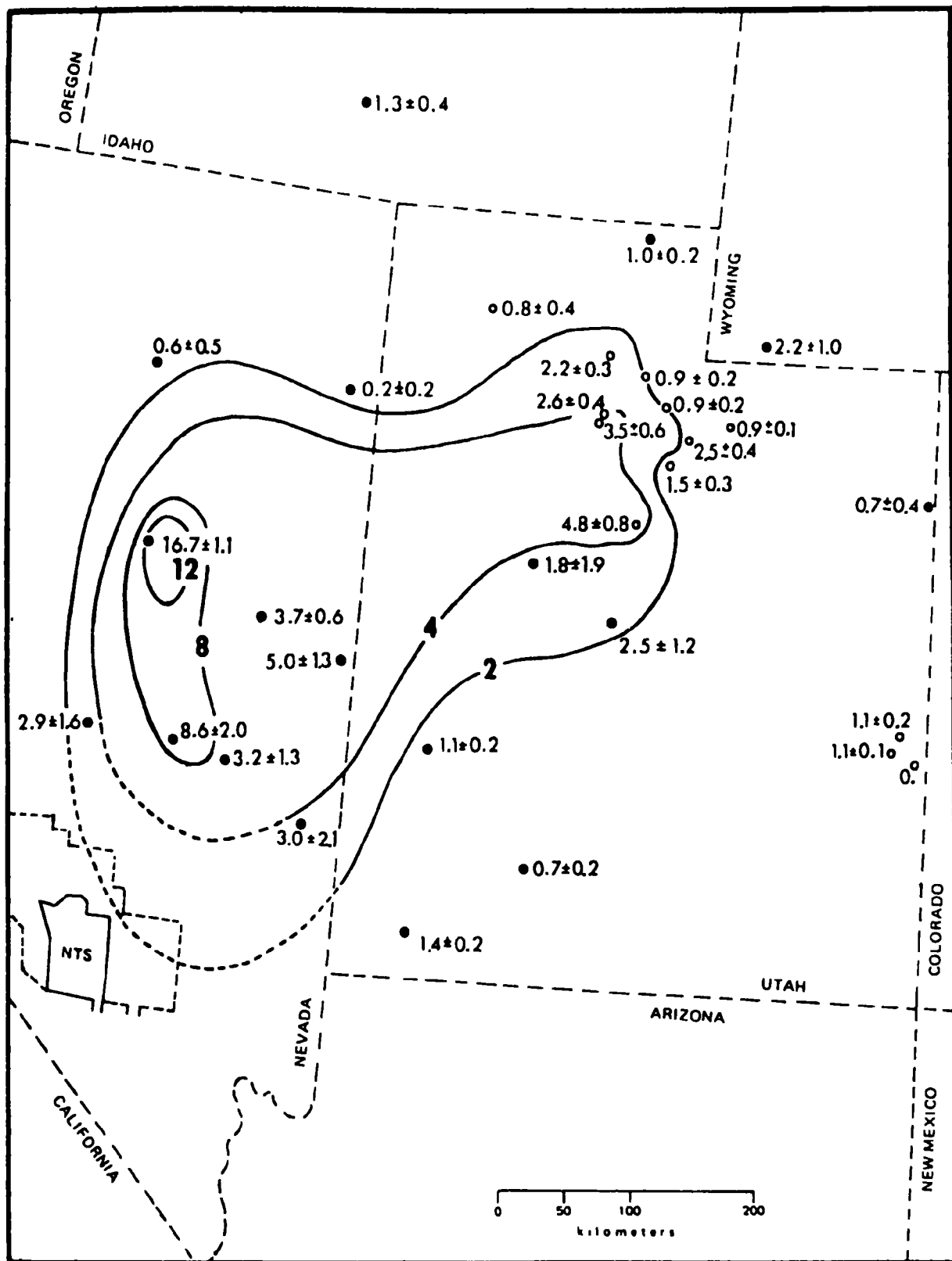
INVENTORY OF PLUTONIUM FOR SELECTED SITES IN THE UNITED STATES

LOCATION	APPROX INVENTORY	REMARKS
U.S. (Fallout)	20,000 Ci	Worldwide Pu-238 = 17,000 Ci Pu-239 = 440,000 Ci U.S. Average = 1.5 mCi/km ²
Nevada Test Site (near Las Vegas, NV)	>155 Ci	Nuclear Test Site Surface and Subsurface Tests
Rocky Flats Plant (near Denver, CO)	8-10 Ci	Weapons Fabrication Facility
Mound Laboratory (Miamisburg, OH)	5-6 Ci	Pu-238 Processing Facility
Savannah River Plant (SW area of SC)	3-5 Ci	Pu Production Facility (Pu and higher isotopes)
Hanford Site (central WA)	*	Pu Production-Research Facility (high levels of Pu on site)
Los Alamos Laboratory (NW of Santa Fe, NM)	*	Weapons Development (Pu-239 in remote canyons)
Oak Ridge Laboratory (east TN near Knoxville)	*	Research and Development Facility
Idaho National Engineering Lab (central ID)	*	Separation, Test, and Research Facility (Pu-239 in soil/groundwater)
Trinity Site (near Alamogordo, NM)	>45 Ci	Site of first atomic bomb test



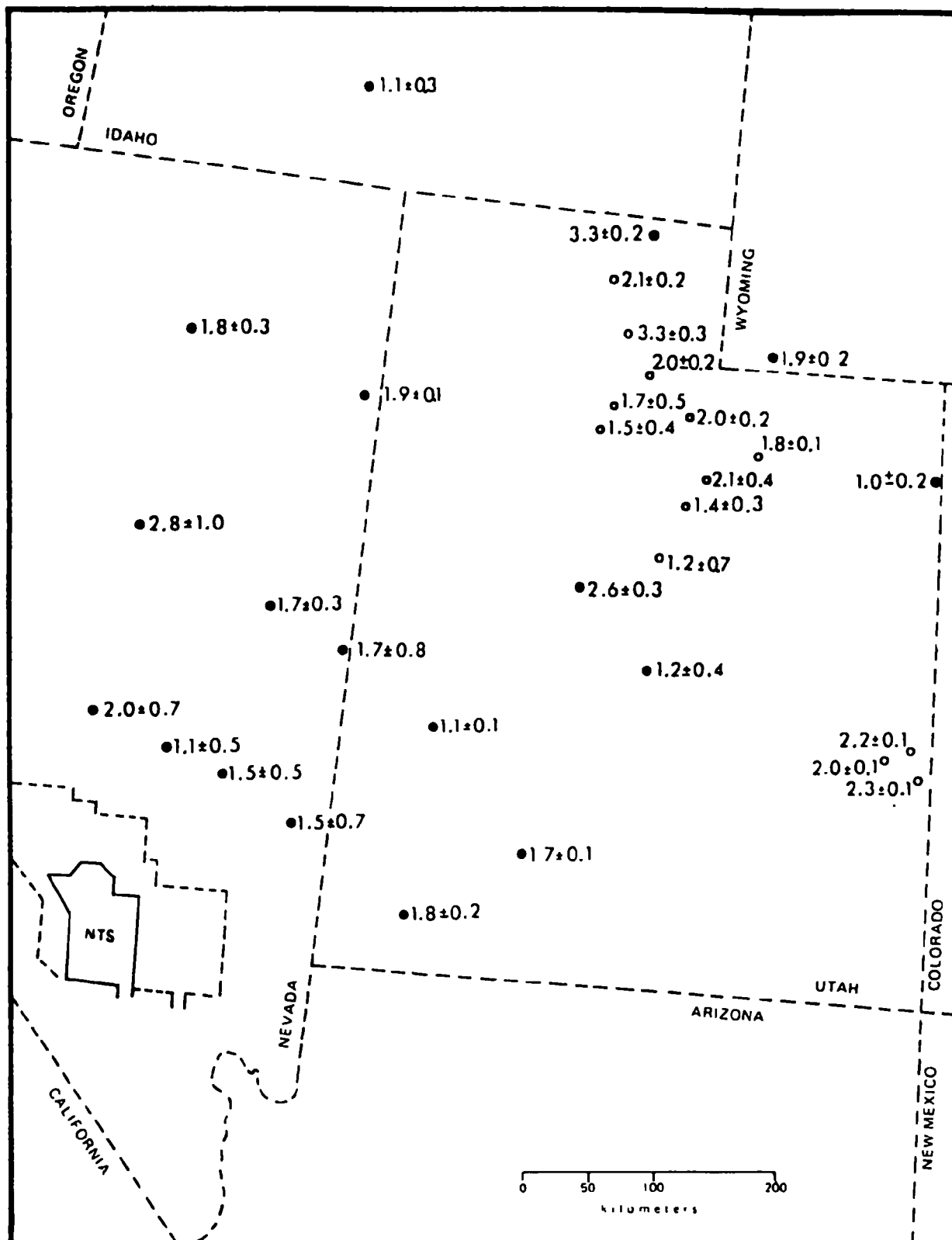
**POPULATION DISTRIBUTION BY AZIMUTH AND DISTANCE
AROUND NTS (10)**

FIGURE 4-1



CUMULATIVE NTS DEPOSIT OF Pu-239,240 (12)
(mCi per km²)

FIGURE 4-2



CUMULATIVE GLOBAL FALLOUT DEPOSIT OF Pu-239,240 (12)
(mCi per km²)

FIGURE 4-3

TABLE 4-6

Estimated Inventory of Plutonium in Surface Soil at Specific
Areas within the National Test Site and Tonopah Test Range

Area	Size of Area (m ²)	Estimated Inventory (curies Pu-239) ^a	Range of Soil Concentrations ^b (μ Ci/m ²)	Pu/Am Activity Ratios
13	3.6x10 ⁶	44 \pm 9	2 to 840	9
5 (GMX)	2.4x10 ⁵	3 \pm 0.3	3 to 530	10
Double Track	3.0x10 ⁵	5 \pm 1	7 to 2,800	} 22 to 26
Clean Slate 1	2.2x10 ⁵	5 \pm 2	15 to 120	
Clean Slate 2	7.9x10 ⁵	29 \pm 6	4 to 260	
Clean Slate 3	1.3x10 ⁵	30 \pm 5	12 to 370	
Plutonium Valley	4.8x10 ⁶	39 \pm 4	1 to 6,200	5 to 8
Total		<u>155</u>		

(a) Inventory as measured to 5 cm soil depth.

(b) Soil concentration range values refer to concentrations within sub regions of each site as selected by stratified random sampling.

TABLE 4-7

Pu-239 in Air Samples Near the NTS

Location	Date	Downwind Pu-239 Concentration (fCi/m ³)		Upwind Pu-239 Concentration (fCi/m ³)		Concentration Downwind vs Upwind
			Date			
Furnace Creek, CA	2/20/71	0.05	4/20/71	0.20		No Difference
	10/30/72	0.04	12/7/72	0.051		
Death Valley Jct. California	2/20/71	0.20	5/2/71	0.20		No Difference
	3/31/71	0.12	5/3/71	0.20		
	10/24/72	0.10	10/2/72	0.065		
	10/28/72	0.07	12/6/72	0.048		
	12/13/72	0.03	12/7/72	0.055		
Beatty, NV	2/03/71	0.09	6/28/70	0.40		Significant Difference
	2/25/71	0.08	4/20/71	0.17		
	3/01/71	0.06	5/2/71	0.20		
	3/17/71	0.19	5/20/71	0.12		
	10/28/71	0.08	10/16/71	0.20		
	4/23/72	0.088	4/16/72	0.075		
Diablo, NV	4/20/71	0.20	2/20/71	0.06		No Difference
	5/04/71	0.20	3/1/71	0.07		
	5/26/71	0.40	3/31/71	0.30		
Hiko, NV	3/26/71	0.20	9/25/70	0.07		Significant Difference
	4/25/71	0.20	2/20/71	0.07		
	6/26/71	0.20	3/18/71	0.09		
	9/25/71	0.70	10/28/71	0.06		
Indian Spring, NV	3/13/71	0.10	4/20/71	0.15		Probable Difference
	4/23/72	0.087	4/16/72	0.19		
Lathrop Wells, NV	3/18/70	0.06	4/20/71	0.20		No Difference
	9/25/70	0.17	6/26/71	0.30		
	3/18/71	0.20	9/25/71	0.20		
	10/24/72	0.04	10/2/72	0.042		
Pahrump, NV	5/17/71	0.30	4/20/71	0.10		No Difference
	10/30/72	0.12	10/2/72	0.044		
Scotty's Jct., NV	2/19/70	0.15	4/20/71	0.17		No Difference
	3/31/71	0.20	5/3/71	0.20		
			5/27/71	0.20		
	10/24/72	0.068	12/4/72	0.035		
	10/29/72	0.15	12/6/72	0.056		
	12/13/72	0.059	12/7/72	0.042		
Warm Springs, NV	3/12/71	0.14	2/19/70	0.09		No Difference
	3/26/71	0.08	3/18/70	0.23		
	4/25/71	0.13	3/31/70	0.10		
			2/3/71	0.60		
			2/20/71	0.60		
			3/17/71	0.20		
			3/31/71	0.12		

for the most part arid, with insufficient rainfall (from 10 to 25 centimeters/year precipitation) to support trees or crops without irrigation. Winds blow primarily from the north, except for May through August when they are predominantly from the south-southwest. Fig. 4-1 shows the population distribution around this site by azimuth and by distance.

Major programs conducted at NTS have included nuclear weapons tests, tests for peaceful uses of nuclear explosives, nuclear reactor engine development, basic high energy nuclear physics research, and seismic studies. As the result of these activities, the test site exclusion area and, to a much smaller extent, areas outside the exclusion areas have become contaminated with plutonium.

Although the total inventory of plutonium in soils within the NTS is not known, detailed surveys have been made of certain specific locations in the site and the Tonopah Test Range (TTR) which are believed to be the areas most highly contaminated with plutonium and americium (11). As shown in Table 4-6, the inventory of plutonium in these areas is about 155 curies. Estimates of offsite plutonium concentration in soil as the result of activities at NTS have been made (12) in units of mCi/km^2 ($1 \text{ mCi}/\text{km}^2 = 0.001 \mu\text{Ci}/\text{m}^2$). Fig. 4-2 shows isopleths for this material; Fig. 4-3 shows, for comparison, the additional amounts of plutonium at the same locations due to fallout. Offsite levels of plutonium in soil are less than $0.1 \mu\text{Ci}/\text{m}^2$, most areas being far lower. A limited special study was made of plutonium concentrations in air at locations close to the NTS (13). Results are shown in Table 4-7 and indicate that, while resuspended plutonium from NTS has probably been detected at three locations, the air concentration level has not exceeded $0.5 \text{ fCi}/\text{m}^3$. Long-term air surveillance from 1966 to 1972 at eight major centers of population in western states was performed for ambient plutonium levels in ground level air (13). Within annual cycles, these levels, which are ascribed entirely to

fallout plutonium from the stratosphere, indicated concentrations varying from 0.01 to 0.5 fCi/m³ with mean annual values of approximately 0.1 fCi/m³. The milk surveillance network around the NTS does not analyze samples for plutonium. Water samples from wells both onsite and offsite in surrounding communities were analyzed for Pu-239 and Pu-238. Plutonium was not detected, indicating levels less than about 0.1 pCi/l Pu-239 or Pu-238.

4.2.2 Rocky Flats Plant (RFP)

The Rocky Flats Plant (10) is located in Jefferson County, Colorado, 26 kilometers northwest of Denver. Currently, the site consists of 6,500 acres of Federally owned land of which 385 acres is enclosed within a security fence. The area is arid (40 cm/y precipitation) with predominant winds from the northwest. Figure 4-4 is a wind rose for the site; Figures 4-5 and 4-6 provide population densities around the site by azimuth and distance. Less than 10 μ Ci of plutonium was released from plant stacks and vents to the atmosphere in 1975 (10). Data on the total amount of plutonium released to surface waters from 1973 to 1975 have not been published. The plant produces components for nuclear weapons, which involves the processing of plutonium. As the result of leakage from barrels of plutonium-contaminated cutting oil, parts of the site and, to a lesser degree, the general environment around the site have been contaminated with plutonium and americium. The total amount of plutonium released to the environment is estimated to be about 11 curies of which 3.4 \pm 0.9 curies is estimated to be at offsite locations (14). Of the approximately 8 curies onsite, more than half is stabilized by cover with an asphalt pad and remedial measures have been taken to control the remainder to the extent practicable. Figures 4-7 and 4-8 show values for Pu-239 concentrations in soils (5 cm depth) around the site (15, 16). Concentrations of Am-241 within the site boundaries are about 10% of Pu-239 values (17). Table 4-8 provides selected

TABLE 4-8

Plutonium Concentration in Ambient Air at Selected Locations-
Rocky Flats Site, 1975

Location	Station	Average Plutonium Concentration (fCi/m ³)	Station Location with Respect to the Plant
Onsite	S-14	<0.02	West
	S-16	<0.06	Northwest
	S-4	0.1	North
	S-6	1.	East
	S-11	0.01	South
Three to six Kilometers Distance from Plant	S-31	<0.03	West
	S-34a	<0.04	North
	S-37a	0.06	East
	S-41	<0.03	South
Boulder		<0.03	Northwest
Marshall		<0.03	North
Superior		<0.04	North
Walnut Creek		<0.03	East
Wagner		<0.04	East
Leyden		0.04	South

(a) Site Boundary

yearly average plutonium concentrations in ambient air within the plant boundary, at distances of 3 to 6 kilometers from the plant and in nearby communities (10). Liquid effluents released from the Plant may eventually reach the Great Western Reservoir, while storm water runoff from the site tends to collect both there and in Standley Lake. Both reservoirs are sources of drinking water. Table 4-9 gives estimates of plutonium and americium concentrations in these water supplies as well as in finished drinking water for nearby communities (10). Modifications of the Rocky Flats Plant operations have been made that will eventually halt all of its liquid effluent discharges. In summary, in offsite areas around the Rocky Flats plant, the plutonium concentration in ambient air is 0.06 fCi/m^3 , plutonium in finished drinking water is 0.03 pCi/l , and plutonium in soil is $0.1 \text{ } \mu\text{Ci/m}^2$ for 5 cm deep samples (17).

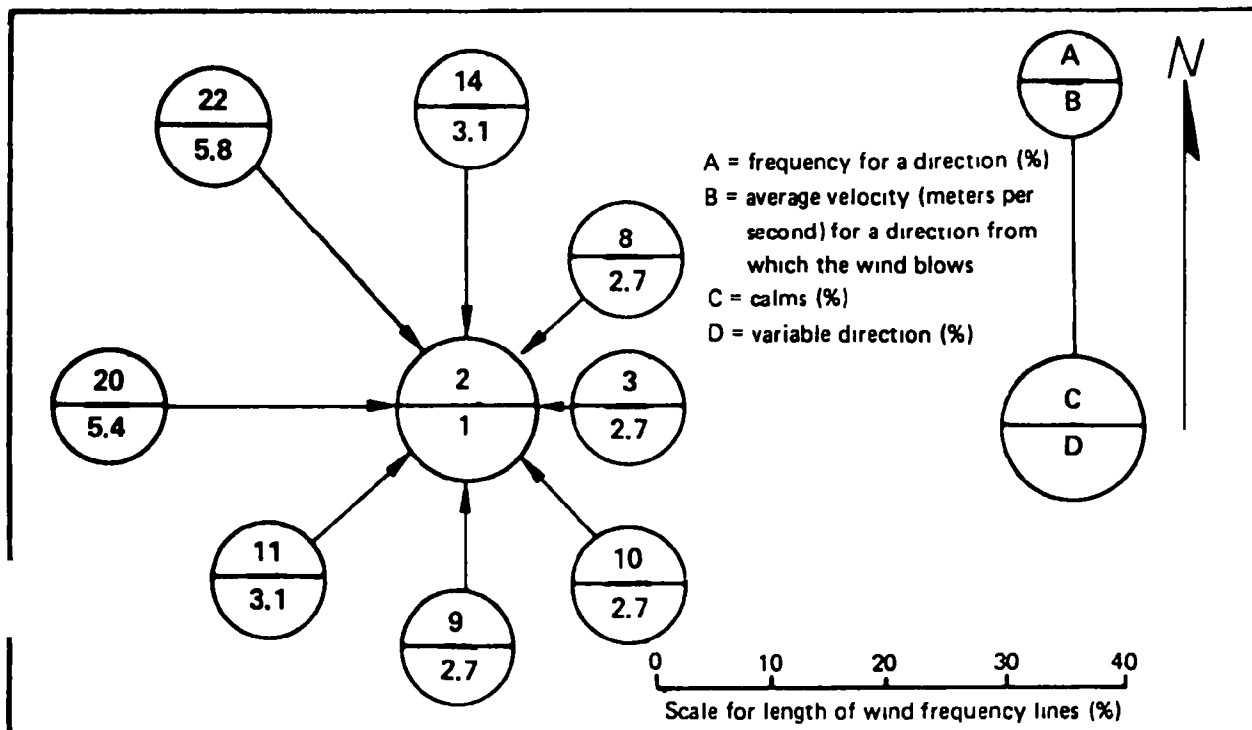
4.2.3 Mound Laboratory (ML)

Mound Laboratory (10) is located in Miamisburg, Ohio, 16 kilometers southwest of Dayton. The 180 acre site is within an industrialized river valley in a region that is predominantly agricultural. Corn and soy beans are major crops and livestock is pastured. Winds are predominantly from the south or west; average precipitation is 91 cm/yr. The population distribution around the site is given in Figures 4-9 and 4-10. The mission of the laboratory includes research, development and production of components for the nuclear weapons program, and fabrication of radioisotopic heat sources for medical applications and space operations. This latter operation involves processing large quantities of Pu-238 which has become the plutonium radionuclide of primary concern associated with this site. Pu-238 in airborne effluent discharges from the plant has, over time, contaminated the site and, to a lesser degree, offsite areas. Figure 4-11 shows estimates of the levels of Pu-238 in soil around the site. Approximately 0.5 curies of Pu-238 have been released to the offsite environment. The concentration of Pu-238 in various

TABLE 4-9

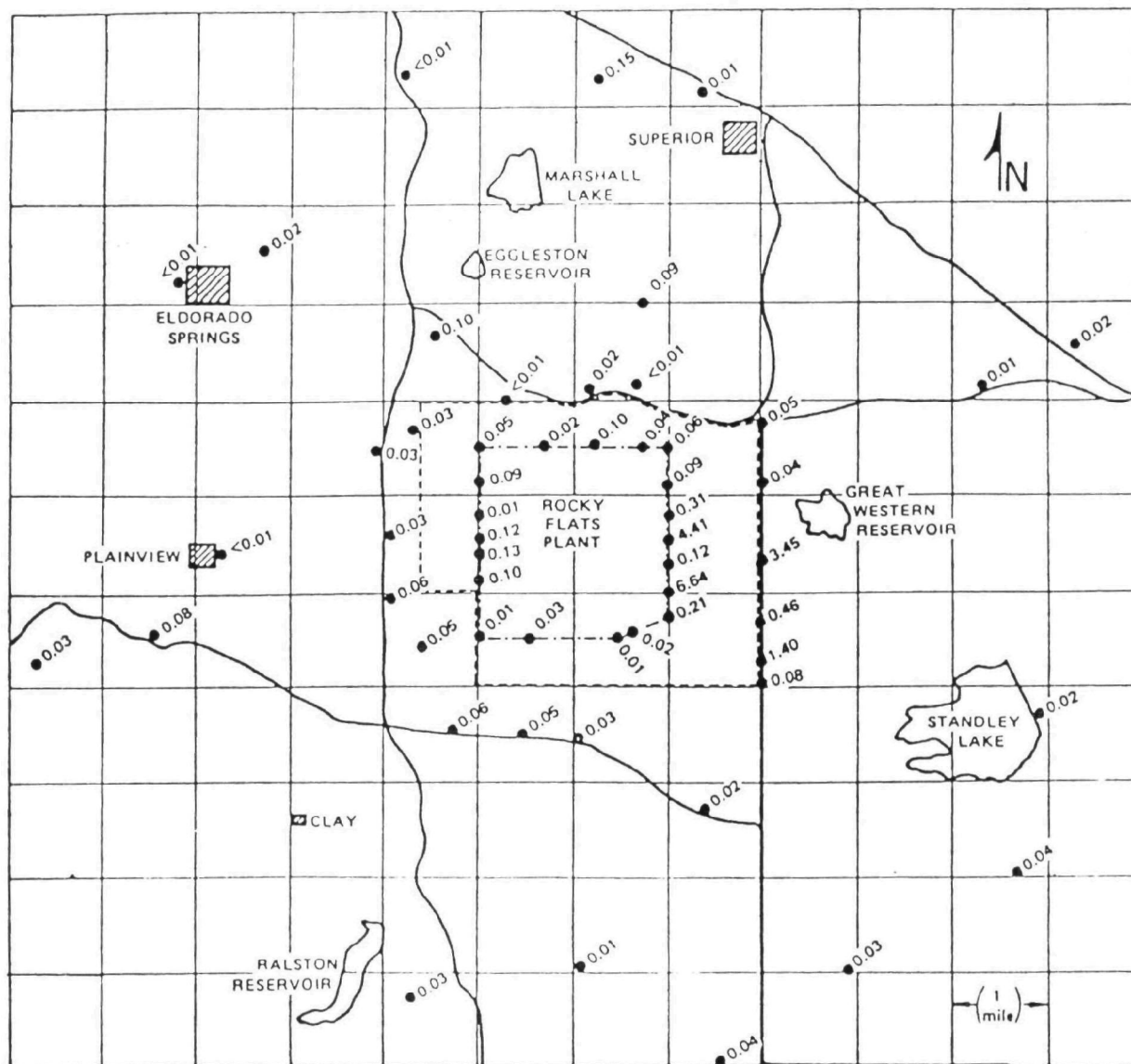
Concentrations of Plutonium and Americium in Water Supplies
and in Finished Drinking Water - Rocky Flats Site, 1975

Location	Water Supply	Concentration	
		Plutonium (pCi/l)	Americium (pCi/l)
Great Western	Reservoir	< 0.1	< 0.03
Standley Lake	Reservoir	< 0.04	< 0.03
Boulder	Drinking Water	< 0.007	< 0.006
Broomfield	"	< 0.04	< 0.03
Denver	"	< 0.008	< 0.04
Golden	"	< 0.009	< 0.009
Lafayette	"	< 0.007	< 0.007
Westminister	"	< 0.04	< 0.03
(Walnut Creek at Indiana Street)	(Discharge to Great Western Reservoir)	(0.6)	(0.2)



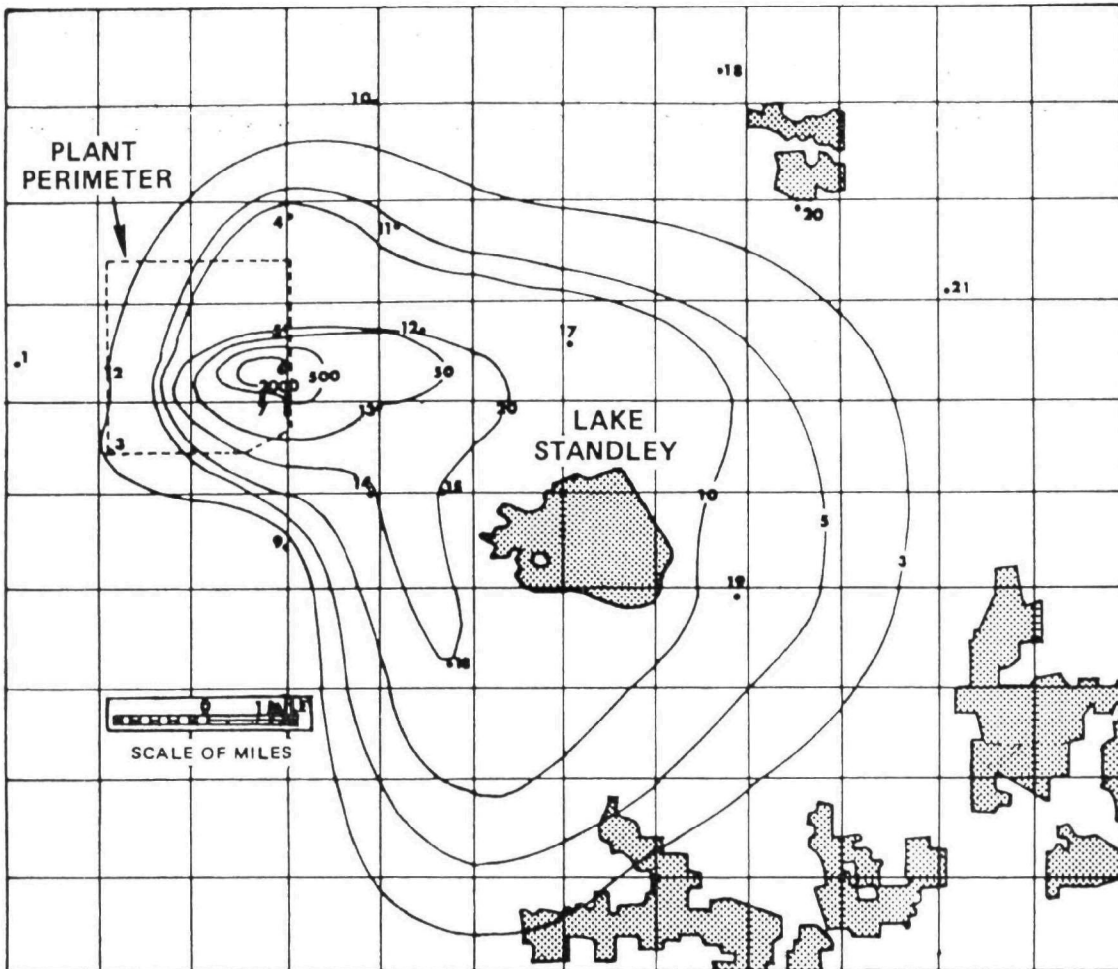
WIND ROSE FOR THE ROCKY FLATS SITE (10)

FIGURE 4-4



**ROCKY FLATS 1974
PLUTONIUM CONCENTRATIONS IN SOIL.
(VALUES IN PICOCURIES PER GRAM. (15))**

FIGURE 4-7



ROCKY FLATS
PLUTONIUM—239 CONTOURS mCi/km² (16).

FIGURE 4-8

environmental media around the Mound Laboratory is given in Table 4-10 for 1975 (10). The annual average concentration of Pu-238 in offsite ambient air did not exceed 0.03 fCi/m³; in surface waters it was as high as 1.4 pCi/l (in an off-site pond) but in water supplies it did not exceed 0.05 pCi/l. In 1969, an underground pipe carrying acid radioactive waste solutions ruptured. During repair work on this pipe, heavy rains eroded the radioactive soil, and carried about 5 curies of Pu-238 off-site into waterways adjacent to the Laboratory. This plutonium now is in sediments that are mostly buried under approximately 1-3 feet of additional non-contaminated sediments added by normal processes later in time (18). A special study of this incident was also conducted by the U.S. Environmental Protection Agency; results are given in Fig. 4-12 and in Table 4-11.

4.2.4 Savannah River Plant (SRP)

The Savannah River Plant (10) is located on a 790 km² Federally owned site along the Savannah River in Aiken and Barnwell Counties. South Carolina, about 100 kilometers southwest of Columbia. The surrounding area is predominantly forested with some diversified farming, the main crops being cotton, soy beans, corn, and small grains, and the production of beef cattle. The climate is mild, with an annual rainfall of 115 cm/y. Population density around the site ranges from 10 to 400 people per square mile. SRP produces plutonium, tritium and other special nuclear materials. Facilities include nuclear reactors, nuclear fuel and target fabrication plants, nuclear fuel reprocessing plants, a heavy water production plant and various supporting laboratories. Two airborne releases, in 1955 and 1969, from fuel reprocessing plant operations are believed to have caused the detectable plutonium contamination of soil that is found within a 2 km radius around those facilities within the site perimeter. Approximately 1 curie of plutonium is estimated to be within the isopleths shown in Fig. 4-13 (20). During the 1960's, radioactive liquid effluents were released from SRP such that

TABLE 4-10

Concentration of Pu-238 in Environmental Media
Mound Laboratory

Ambient Air

<u>Sample Location</u> <u>(Location Number)</u>	<u>Average Concentration of Pu-238</u> <u>(fCi/m³)</u>
onsite (211)	0.2
(212)	0.05
(213)	1.0
(214)	0.06
North of Plant (101)	0.02
East of Plant (103)	0.01
South of Plant (104)	0.01
West of Plant (105)	0.009
Miamisburg (122)	0.02
Dayton (108)	0.008
(National Average from Fallout)	(0.003)

Waters

<u>Sample Location</u> <u>(Location number)</u>	<u>Average Concentration of Pu-238</u> <u>(pCi/l)</u>
Great Miami River	
Above the Plant (1)	0.019 ± 0.0022
Below the Plant (4)	0.052 ± 0.004
Canal/pond area	
North Pond	0.22
South Pond	1.4
Miamisburg Drinking Water	0.043 ± 0.003
Private Well J	0.020 ± 0.002
Private Well B	0.006 ± 0.00003

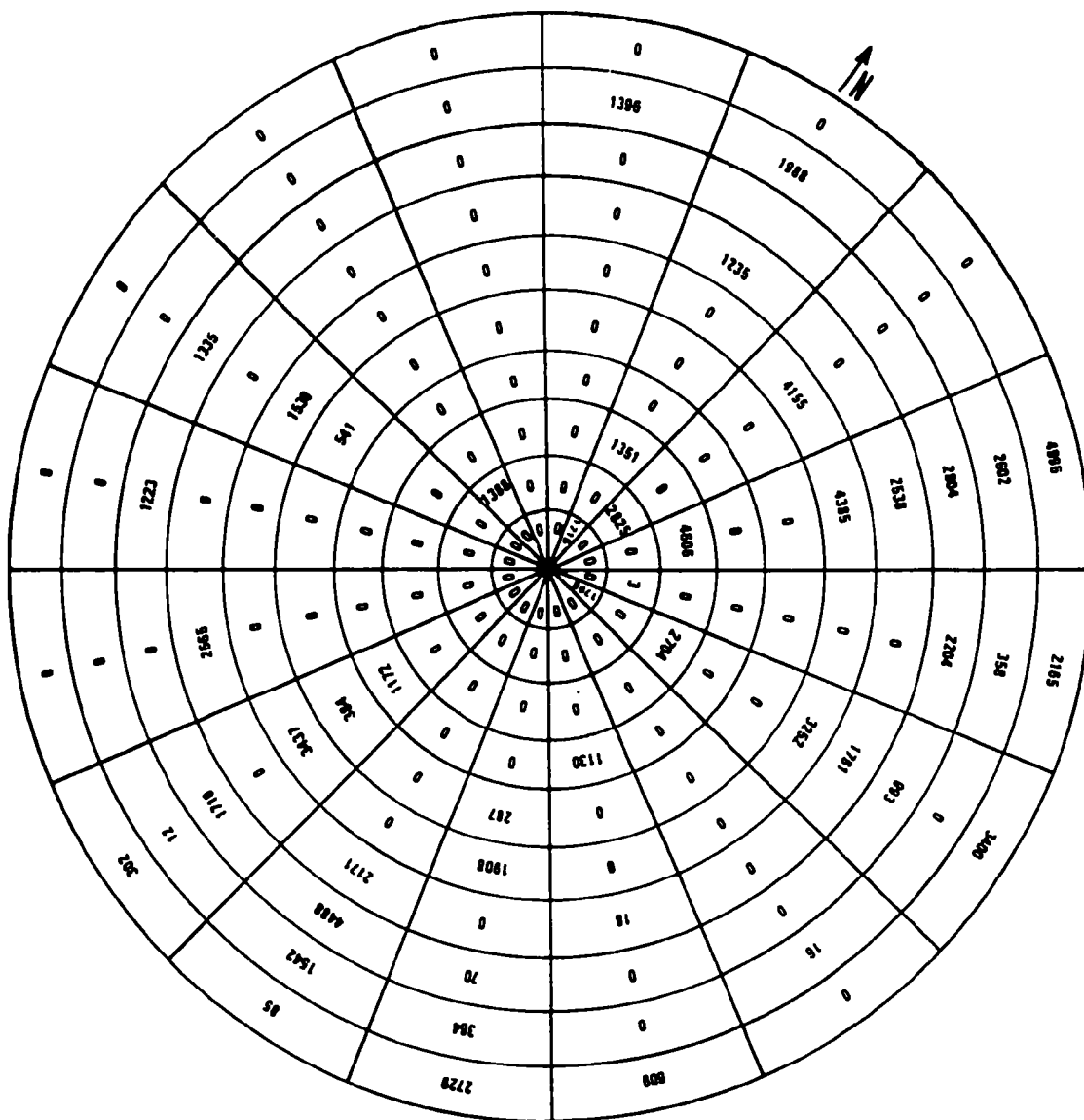
Foodstuff Collected Close to the Plant

<u>Sample</u>	<u>Average Concentration of Pu-238</u> <u>(pCi/g)</u>
Milk	2×10^{-4}
Fruits & Vegetables	$< 6 \times 10^{-4}$
Grass	1×10^{-2}
Field Crops	1×10^{-3}
Aquatic life	$< 3 \times 10^{-4}$

TABLE 4-11

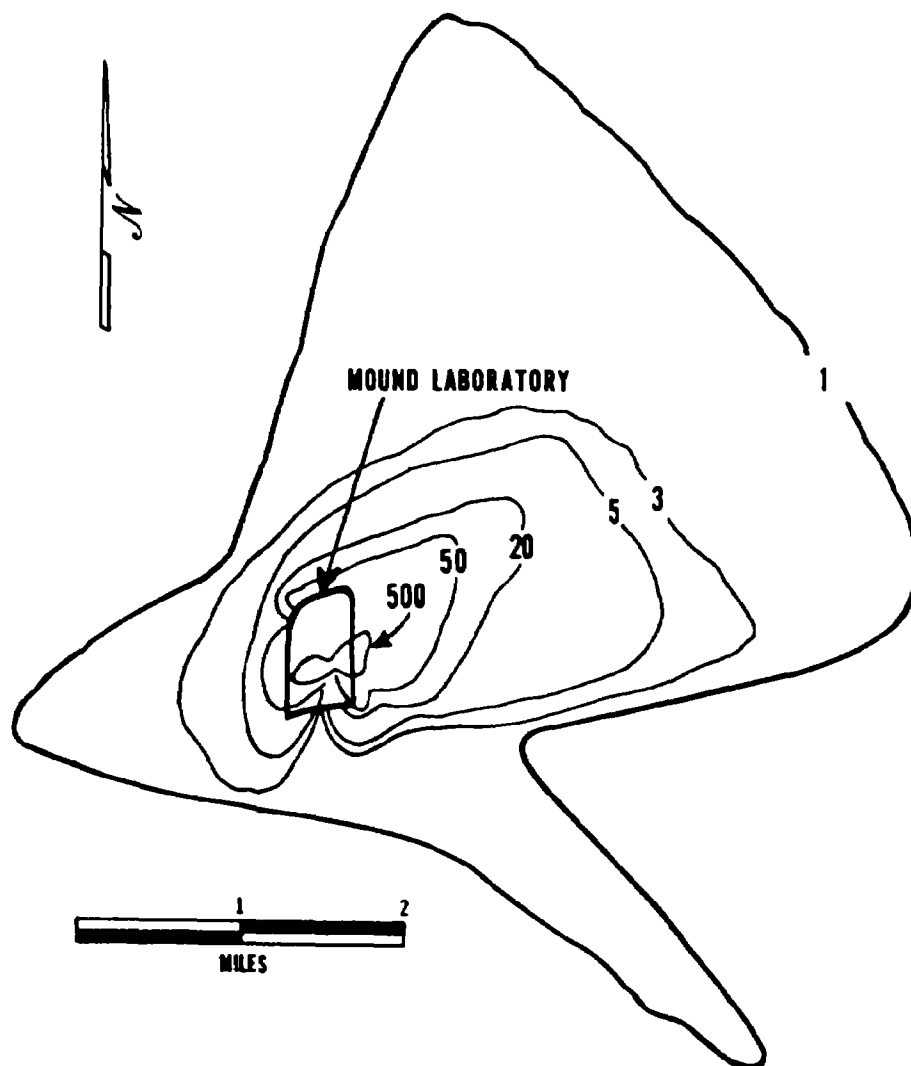
MOUND LABORATORY
U.S. ENVIRONMENTAL PROTECTION AGENCY
1974 SURVEY

Plutonium in Samples from the Vicinity of Mound Laboratory			
I.D. #	Location	Concentration	
		²³⁸ Pu	²³⁹ Pu
<u>Core sediment samples collected by Mound Laboratory, pCi/g dried weight</u>			
EA-1	North Canal at south end of South Pond	0.13 0.09	< 0.02 < 0.02
EB-1	"	0.13 0.11	< 0.02 < 0.02
EC-1	"	4.8	0.07
ED-1	"	1.1	< 0.02
EE-1	"	440	2.3
EF-1	"	1170 1090	15.4 7.9
EG-1	"	10.8	0.18
EH-1	"	26	0.16
EI-1	"	0.98	0.06
EJ-1	"	0.89 1.30	0.05 0.06
FA-1	North end of North Canal	8.9 7.5 9.1	0.51 0.44
FE-1	"	16.9 19.2	0.18 0.05
GA-1	North end of North Pond	0.48	< 0.02
HA-1	Middle of North Pond	5.1	0.07
IA-1	South end of North Pond	2.5	0.06
JA-1	North end of South Pond	0.70	0.02
KA-1	Middle of South Pond	27	0.27
LA-1	South end of South Pond	10.9	0.30
CE-1	South Canal at west drainage ditch	24	0.46
QE-1	South Canal where it crosses US 25	920	10.9
<u>Sediment samples, top 1 inch, pCi/g dried weight</u>			
EPA-1	South Canal at west drainage ditch	230	3.54
EPA-20	East drainage ditch, ~200 ft south of Mound Rd culvert	1.9	0.06
EPA-17	South drainage ditch, 15 ft from junction with South Canal	47	0.84
EPA-18	South Canal, 10 ft from junction with south drainage ditch	60	0.77
<u>Surface soil and mud samples, top 1 inch, pCi/g dried weight</u>			
EPA-2	Railroad cut south of control box	0.12	0.02
EPA-3	Railroad cut north of control box	0.39	0.13
EPA-6	Run-off hollow	3.8	0.11
EPA-7	At shelter house SE of South Pond	0.44	0.07
EPA-13	NE of Lab, at fence between tennis court and Harmon Field	0.10	0.02
EPA-14	SE of Lab, at SW corner of Mound Park	0.44	0.04
EPA-15	SW of Lab, at junction of US 25 with South Canal	0.96	0.04
EPA-12	NW of Lab, at alley south of Mound Rd	0.19 0.17	0.05 0.05



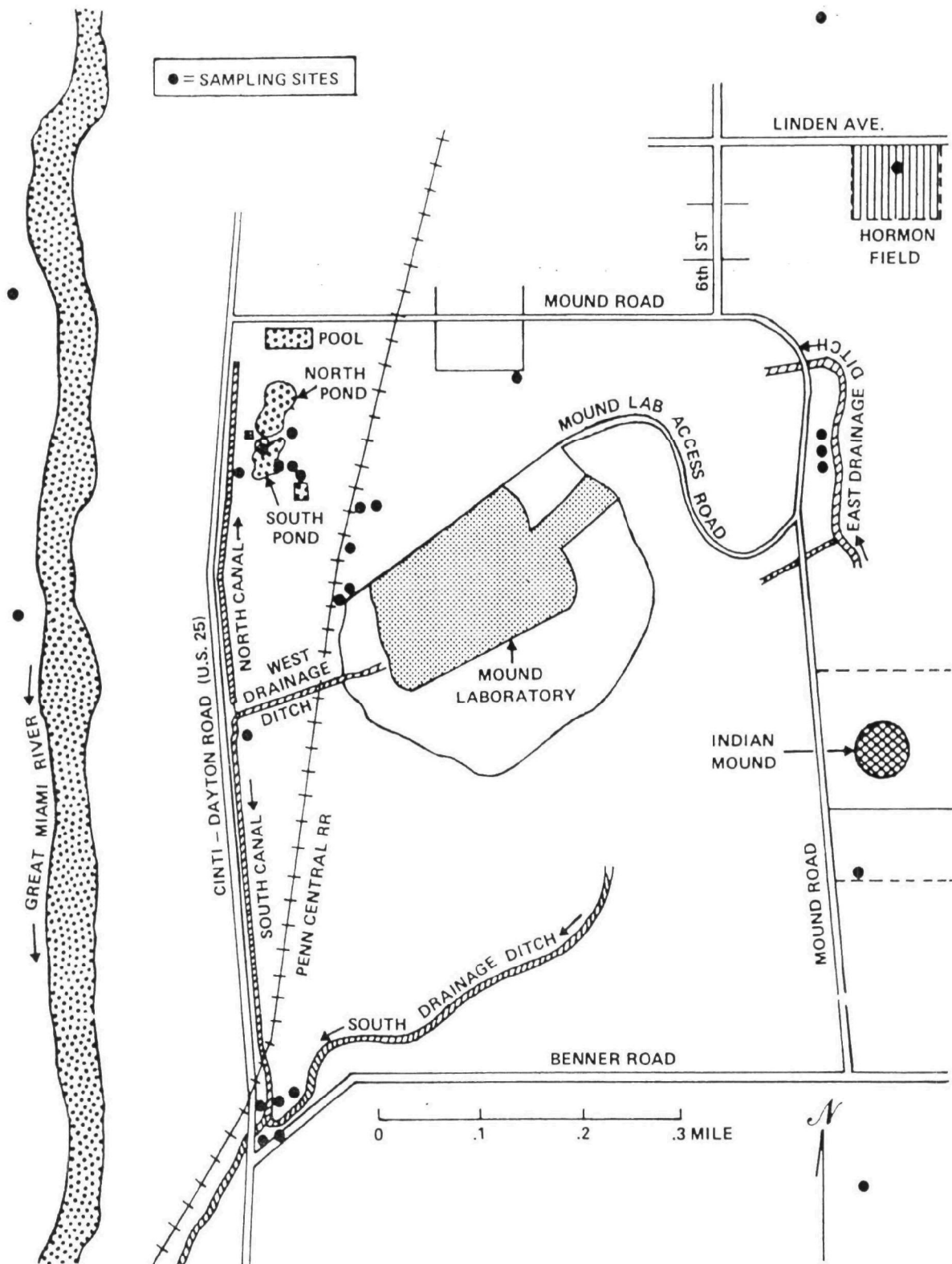
POPULATION DISTRIBUTION AROUND MOUND LABORATORY
 (0 to 10 km) (LAT 39 6305 LONG 84 2897)

FIGURE 4-9



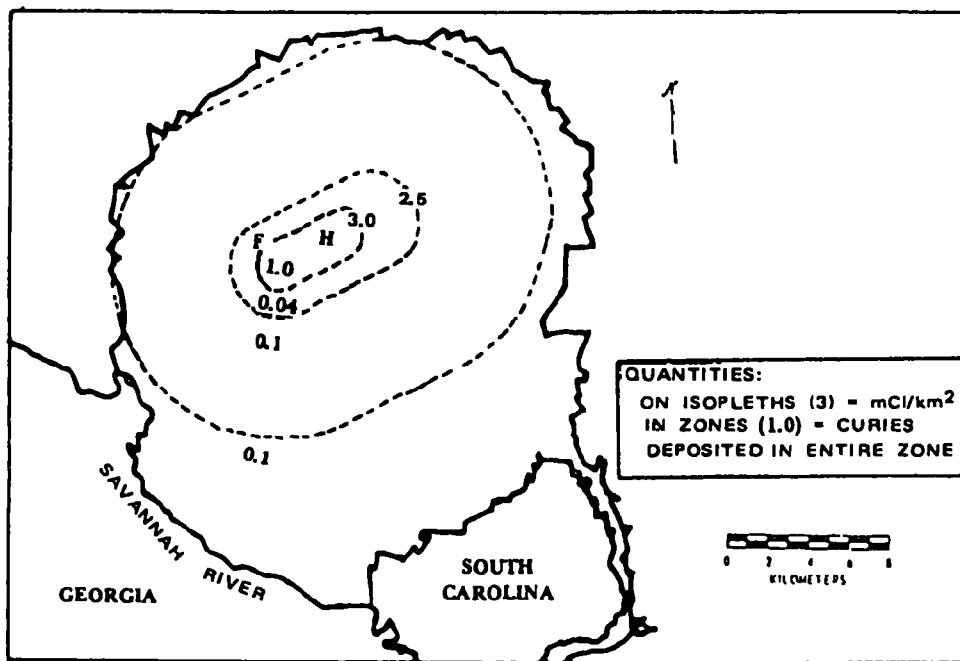
MOUND LABORATORY
PRELIMINARY ESTIMATE OF PLUTONIUM - 238 AIRBORNE DEPOSITION
(m Ci/km²) (10)

FIGURE 4-11



**U.S. ENVIRONMENTAL PROTECTION AGENCY SAMPLING SITES
MOUND LABORATORY (19)**

FIGURE 4-12



SAVANNAH RIVER PLANT PLUTONIUM DEPOSITION (10)

FIGURE 4-13

TABLE 4-12

**PLUTONIUM CONCENTRATION IN ENVIRONMENTAL MEDIA
AROUND THE SAVANNAH RIVER PLANT - 1975**

AMBIENT AIR

<u>Sample Location</u>	<u>Average Plutonium Concentration</u>	
	<u>Pu-239₃</u> <u>(fCi/m)</u>	<u>Pu-238₃</u> <u>(fCi/m)</u>
Plant Perimeter (Locations not specified)	0.02	0.001
25 mile radius (Locations not specified)	0.02	0.001

RAIN WATER

<u>Sample Location</u>	<u>Average Plutonium Concentration</u>	
	<u>Pu-239₂</u> <u>(pCi/m²)</u>	<u>Pu-238₂</u> <u>(pCi/m²)</u>
Plant Perimeter	2.2	0.11
25 mile radius	1.9	0.15

SOIL (0-5 cm depth)

<u>Location</u>	<u>Average Plutonium Concentration</u>	
	<u>Pu-239₂</u> <u>(uCi/m²)</u>	<u>Pu-238₂</u> <u>(uCi/m²)</u>
Plant Perimeter		
NW quadrant	0.0009	5×10^{-5}
NE	0.0014	8
SE	0.0010	6
SW	0.0012	8
Springfield, SC	0.0003	4×10^{-5}
Aiken Airport, SC	0.0010	7
Clinton, SC	0.0008	3
Savannah, GA	0.0005	2

radioactive materials, including Cs-137, Co-60, and plutonium deposited in offsite swamp areas. The plutonium concentrations in these remote areas range from about 3 to $11 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ Pu-239 and 0.3 to $6 \times 10^{-3} \text{pCi}/\text{m}^2$ Pu-238. The amount attributed to fallout sources is approximately $1 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ Pu-239 and $0.1 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ Pu-238 (10). Levels of plutonium in various environmental media in the general environment around SRP are given in Table 4-12 for 1975 (10). Resuspended plutonium from the contaminated areas within the site was not detected in offsite ambient air. Values for plutonium concentration levels in ambient air at onsite locations have not been published. In 1975, the plant released 2 mCi Pu-238 and 0.5 mCi Pu-239 to the atmosphere; the plant released 8 mCi Np-239 and 19 mCi Pu-239 in liquid effluents.

4.2.5 Los Alamos Scientific Laboratory (LASL)

The Los Alamos Scientific Laboratory (10) is located on a 110 km^2 site in Los Alamos County in North Central New Mexico about 40 kilometers northwest of Santa Fe. The site is on a series of mesas separated by canyons that run eastward from the Jemez Mountains to the Rio Grande Valley. The climate is semi-arid with rainfall of 46 cm/y. While the land around the site is undeveloped, about 16,000 people reside in the immediate area. The primary mission of LASL is associated with nuclear weapons research and development. Industrial effluents from these operations have for some time been discharged onsite into canyons, where the transuranium nuclides in these effluents soon become attached to soil particles. It is estimated that less than 1 Ci of transuranic waste has been disposed of to Pueblo, DP-Los Alamos, and Montandad canyons. Liquid effluents are usually absorbed in the soil so they do not flow beyond the site boundaries, but, during periods of heavy runoff, storm waters have carried detectable amounts of transuranium elements down the canyons and offsite. Plutonium concentrations in sediments in the canyons receiving liquid waste are given in Table 4-13 (21). Concentration levels of plutonium and americium in various

TABLE 4-13

**PLUTONIUM IN SEDIMENTS IN THE LIQUID WASTE RECEIVING
CANYONS ON THE LASL SITE - 1975**

Average Plutonium Concentration in dry soil ^(a)			
Distance from outfall (kms)	Acid Pueblo Canyon (pCi/g)	DP-Los Alamos Canyon (pCi/g)	Mortandad Canyon (pCi/g)
0	3	40	220
0.6	10	1	20
1.3	2		9
2.6	0.4	0.2	11
5.1	1	0.4	0.1
10.2	0.2		0.03
Estimated Canyon Inventory		0.1 to 0.3 curies	

(a) top 5 cm of soil

TABLE 4-14

**PLUTONIUM AND AMERICIUM CONCENTRATIONS IN ENVIRONMENTAL
MEDIA AT THE LASL SITE - 1975**

Ambient Air

		Average Radionuclide Concentration		
Station Location (Station Number)		Pu-239 ₃ (fCi/m ³)	Pu-238 ₃ (fCi/m ³)	Am-241 ₃ (fCi/m ³)
On Site	22	0.02	-	-
	23	0.02	1x10 ⁻³	3x10 ⁻³
	24	0.02	5x10 ⁻⁴	-
	25	0.02	5x10 ⁻⁴	7x10 ⁻³
	26	0.02	5x10 ⁻⁴	-
Perimeter	12	0.02	6x10 ⁻⁴	0.02
	14	0.02	9x10 ⁻⁴	-
	18	0.02	1x10 ⁻⁴	1x10 ⁻³
	20	0.02	5x10 ⁻⁴	-
Off Site (Santa Fe)	1	0.02	6x10 ⁻⁴	-
	4	0.02	7x10 ⁻⁴	4x10 ⁻³
	8	0.02	9x10 ⁻⁴	4x10 ⁻³
	11	0.02	4x10 ⁻⁴	4x10 ⁻³

Surface Water and Water Supplies

		Average Radionuclide Concentration	
Sample Location		Pu-239 (pCi/l)	Pu-238 (pCi/l)
Regional Surface Waters		9x10 ⁻⁴	6x10 ⁻⁴
Perimeter Surface and Ground Waters		8x10 ⁻³	2x10 ⁻³
Los Alamos Water Supply		3x10 ⁻⁴	3x10 ⁻⁴

Soils

		Average Radionuclide Concentration ^(a)	
Sample Location		Pu-239 (pCi/g)	Pu-238 (pCi/g)
On site		40x10 ⁻³	1x10 ⁻³
Site Perimeter and Regional areas		12x10 ⁻³	0.5x10 ⁻³

(a) Top 5 cms of soil.

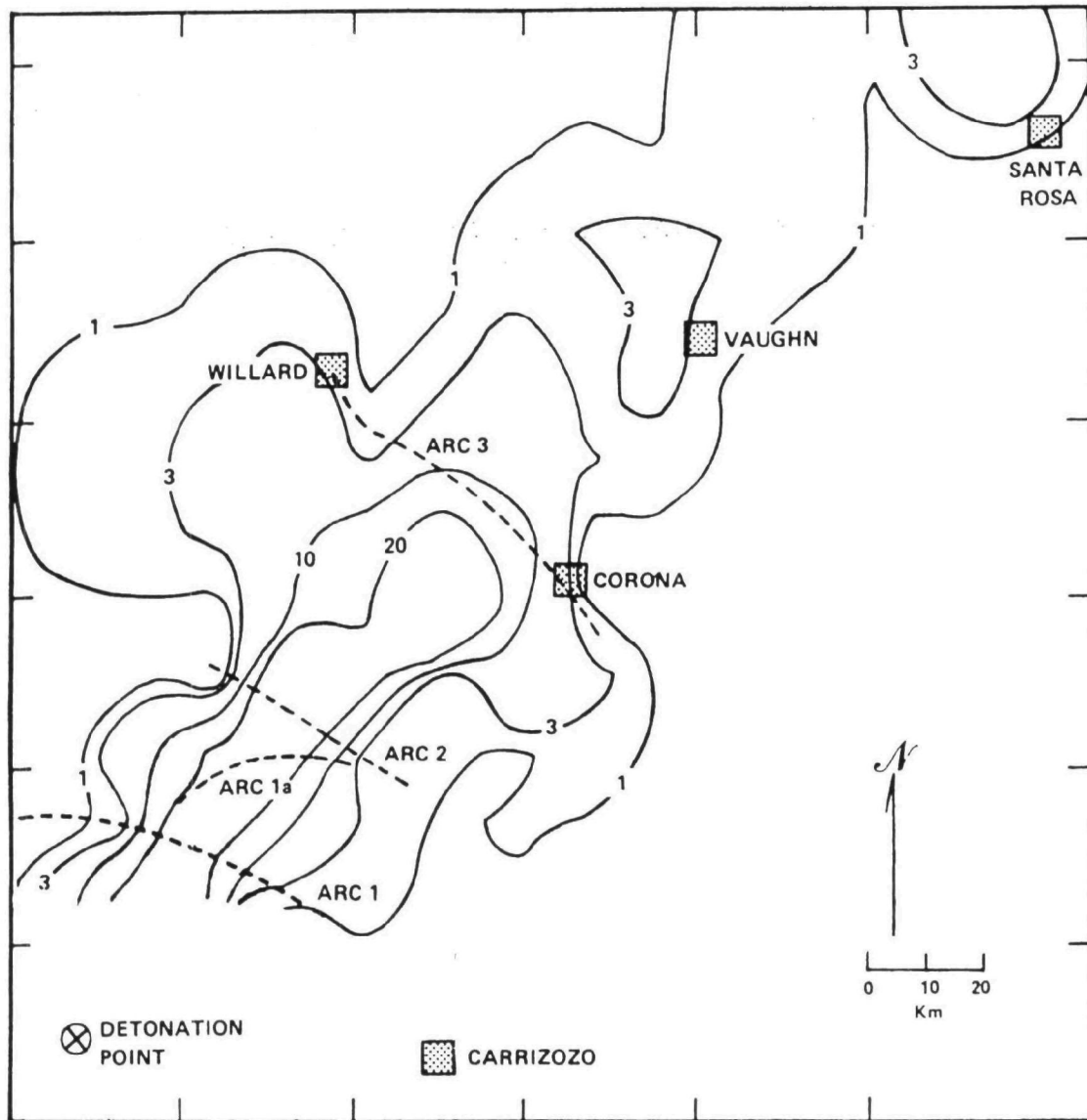
environmental media around the LASL site are given in Table 4-14 for 1975 (10). During the same year, the Laboratory discharged less than 0.3 mCi of plutonium to the air; the amount of transuranium elements discharged in liquid effluents was not published.

4.2.6 The Trinity Site

The first nuclear device was tested at the Trinity Site, 100 kilometers northwest of Alamogordo, New Mexico, on July 16, 1945. During 1973 and 1974, the site was surveyed by the U.S. Environmental Protection Agency to determine the extent of resulting plutonium contamination (22). Figure 4-12 shows plutonium contours based on this study. Highest soil activity levels were 0.05, 0.09 and 0.02 $\mu\text{Ci}/\text{m}^2$ found along arcs IA, 2 and 3, respectively. The total amount of plutonium estimated to be within the 3 nCi/ m^2 contour is approximately 45 curies (1 nCi = 10^{-9} curies).

4.2.6 Other Sites of Underground Nuclear Detonations

A listing of major underground tests conducted at locations other than the Nevada Test Site is shown in Table 4-15. All transuranium elements are believed to be contained at the site of detonation.



TRINITY SITE
1973-1974 PLUTONIUM
SOIL SAMPLING RESULTS
(nCi/m²)·(14)

FIGURE 4-14

TABLE 4-15

Underground Testing Conducted Off the Nevada Test Site

Name of Test, Operation or Project	Date	Location	Yield ^d (kt)	Depth m (ft)	Purpose of the Event ^e
Project Gnome/ Coach ^a	12/10/61	48 km (30 mi) SE of Carlsbad, N.M.	3.1 ^f	360 (1184)	Multi-purpose experiment.
Project Shoal ^b	10/26/63	45 km (28 mi) SE of Fallon, Nev.	12	766 (2510)	Nuclear test detonation re- search experi- ment
Project Dribble ^b (Salmon Event)	10/22/64	34 km (21 mi) SW of Hattiesburg, Miss.	3.3	823 (2700)	Nuclear test detonation re- search experi- ment.
Operation Long Shot	10/29/65	Amchitka Island, Alaska	~80	716 (2350)	DOD nuclear test detection experiment.
Project Dribble ^b (Sterling Event)	12/03/66	34 km (21 mi) SW of Hattiesburg, Miss.	0.38	823 (2700)	Nuclear test detonation re- search experi- ment.
Project Casbuggy ^a	12/10/67	88 km (55 mi) E of Farmington, N.M.	19	1292 (4240)	Joint Government- industry gas stimulation ex- periment.
Faultless Event ^c	01/19/68	Central Nevada Test Area 96 km (60 mi) E of Tonopah, Nev.	200- 1000	914 (3000)	Calibration test.
Project Miracle Play (Diode Tube) ^b	02/02/69	34 km (21 mi) SW of Hattiesburg, Miss.	Non- nuclear explosion	823 (2700)	Detonated in Salmon/Sterling cavity. Seismic studies.
Project Milson ^a	04/10/69	19 km (12 mi) SW of Hillie, Colorado	40	2568 (8425)	Gas stimulation experiment.
Operation Milrow ^c	10/02/69	Amchitka Island, Alaska	~1000	1219 (4000)	Calibration test.
Project Miracle Play (Humid Water) ^b	04/19/70	34 km (21 mi) SW of Hattiesburg, Miss.	Non- nuclear explosion	823 (2700)	Detonated in Salmon/Sterling cavity. Seismic studies.
Operation Cannikin ^c	11/06/71	Amchitka Island, Alaska	~5000	1829 (6000)	Test of war- head for Spartan missile.
Project Rio Blanco	05/17/73	48 km (30 mi) SW of Meeker, Colorado	3x30	1780 2040 (5860 to 6690)	Gas stimula- tion experi- ment.

^aPlowshare Events^bVela Uniform Events^cWeapons Tests^dInformation from "Revised Nuclear Test Statistics," distributed on September 20, 1976, by David G. Jackson, Director, Office of Information Services, U.S. Atomic Energy Commission, Las Vegas, Nevada.^eNews release AL-62-50, AEC Albuquerque Operations Office, Albuquerque, New Mexico, December 1, 1961^f"The Effects of Nuclear Weapons" Rev. Ed. 1964.

4.3 PRODUCTION/UTILIZATION FACILITIES

A number of other sites in the United States have utilized plutonium in various aspects of weapons production. Tables 4-16 to 4-22 show levels of the transuranium elements in the general environment several of these facilities known to use the transuranium elements in their operations. At this time, the total amount of the transuranium radionuclides in the environs of these sites is believed to be less than 1 curie.

TABLE 4-16

Environmental Monitoring for the Transuranium Elements
at the Pantex Plant Site

Site: Pantex Plant
 Location: 25 kilometers northeast of Amarillo, Texas
 Mission: Atomic Weapons Assembly involving significant quantities
 of uranium, plutonium, tritium

Transuranium Elements Released to the Environment

No releases during the period 1973-1975

<u>Media</u>	<u>Location of Sample Collection</u>	<u>Sample Station</u>	<u>Average Plutonium Concentrations - CY 1975</u>
Air ^(a)	10 kilometers from plant in various directions	1	0.03 fCi/m ³
		2	0.07
		3	0.03
		4	0.02
		5	0.00 ± 0.01
		6	0.01
		8	0.4
		9	0.00 ± 0.01
		10	0.09
25 kilometers from plant		11	0.5 (1 sample only)
		12	0.00 (1 sample only)
Soil ^(b)	Offsite in various directions from the plant	31	0.00 ± 0.02
		Different Stations	to 0.05 ± 0.02 pCi/g
Jackrabbits	Onsite	11 samples	0.00 ± 0.02 pCi/g (wet) in kidney, liver, lung, flesh, and bone

(a) Average air concentrations of plutonium in 1973 ranged from 0.4 to 2 fCi/m³ (10), which is higher than any other site. These high levels are believed to have been caused by analytical errors.

(b) Soil samples collected to a depth of 5 cm.

TABLE 4-17

Environmental Monitoring for the Transuranium Elements at Argonne National Laboratory

Site: Argonne National Laboratory
 Location: DuPage County, Illinois, 43 kilometers southwest of Chicago
 Mission: Research and Development including chemical and metallurgical plutonium laboratories

Transuranium Elements Released to the Environment (CY 1975)

To air - not published
 To surface waters (Sawmill Creek) - 0.1 mCi Pu-239;
 0.5 mCi Np-237;
 0.05 mCi Am-241;
 <0.05 mCi Curium and Californium

<u>Media</u>	<u>Location of Sample Collection</u>	<u>Number of Stations</u>	<u>Average Plutonium Concentrations - CY 1975</u>
Air	Site Perimeter Offsite	Av. of 2 Stations 1 station	0.02 fCi/m ³ 0.02
Surface Water (Sawmill Creek)	Downstream from Outfall		$< 5 \times 10^{-4}$ pCi/l Pu-239 $< 3 \times 10^{-3}$ Pu-238 4×10^{-3} Am-241 4×10^{-2} Np-237 $< 1 \times 10^{-3}$ Cu-242 5×10^{-3} Cu-244
Sawmill Creek Phytoplankton	Upstream from Outfall Downstream from Outfall		2.4×10^{-5} pCi/g Pu-239 1.5×10^{-5} Pu-239
Des Plaines River	Upstream from Sawmill Creek Downstream from Sawmill Creek		5×10^{-4} pCi/l Pu-239 8×10^{-4} Pu-239
Illinois River	McKinley Woods State Park Below Dresden Power Station		2×10^{-4} pCi/l Pu-239 3×10^{-4} Pu-239
Soil ^(a)	Site Perimeter Offsite	Av. of 10 Locations Av. of 10 Locations	2×10^{-3} μ Ci/m ² Pu-239 1×10^{-4} Pu-238 2×10^{-3} Pu-239 2×10^{-4} Pu-238

^(a) Soil samples collected to depth of 30 cm.

TABLE 4-18

Environmental Monitoring for the Transuranium Elements at Battelle-Columbus Laboratories (West Jefferson Site)

Site: Battelle Laboratories (West Jefferson Site)
Location: Columbus, Ohio
Mission: Reactor Fuel Research (Plutonium Laboratory)

Transuranium Elements Released to the Environment (CY 1975)

To air - 1.5 μCi Pu-239
 To surface waters - not published

<u>Media</u>	<u>Sample Collection Location</u>	<u>Average Plutonium Concentrations - CY 1975</u>
Air	(Site Boundary concentration as calculated using atmospheric dispersion equations)	$(4 \times 10^{-3} \text{ fCi/m}^3)$
Silt	Above and Below Outfall	$< 2 \times 10^{-2} \text{ pCi/g (dry)}$
Grass	Onsite and Various Locations Onsite, 3-8 kilometers	$< 2 \times 10^{-2} \text{ pCi/g (dry)}$
Food Crops	Corn, Soybeans, Rye, Vegetables 0.4 to 8 kilometers in Various Directions around Site	$< 2 \times 10^{-2} \text{ pCi/g (dry)}$

TABLE 4-19

Environmental Monitoring for the Transuranium Elements at the Idaho National Engineering Laboratory

Site Idaho National Engineering Laboratory
Location Southeastern Idaho; 35 kilometers west of Idaho Falls
Mission Includes - Fuel reprocessing, calcining liquid radio-
active waste, and storage and surveillance of solid
transuranic waste

Transuranium Elements Released to the Environment (CY 1975)

To air - 2 mCi Pu-238, Pu-239, and Np-237
To disposal well - "very small amounts"

<u>Media</u>	<u>Sample Collection Location</u>	<u>Average Plutonium Concentrations-CY 1975</u>
Air	Boundary Stations	0.02 fCi/m ³ Pu-239 0.01 fCi/m ³ Am-241
Surface Soils	Boundary Stations 18 Samples	$2 \pm 2 \times 10^{-2}$ pCi/g (a)
	Distant Location 12 Samples	$3 \pm 4 \times 10^{-2}$ pCi/g (a)

(a) Soil Samples Collected to Depth of 5 cm.

TABLE 4-20

Environmental Monitoring for the Transuranium Elements at the Oak Ridge Facilities

Site Oak Ridge Facilities
Location Oak Ridge, Tennessee
Mission Multipurpose Research Laboratory, Gaseous Diffusion Plant,
and Nuclear Weapons Operations (Y-12 Plant)

Transuranium Elements Released to the Environment (CY 1975)

To air - 4 μCi sum of all transuranium elements
To Clinch River - 20 mCi sum of all transuranium elements
(CY 1973 - 80 mCi; CY 1974 - 20 mCi)

<u>Media</u>	<u>Sample Collection Location</u>	<u>Average Plutonium Concentrations-CY 1975</u>
Air	Perimeter Stations	0.014 fCi/m ³ Pu-239 < 0.001 fCi/m ³ Pu-238
	Remote Stations	0.013 fCi/m ³ Pu-239 < 0.001 fCi/m ³ Pu-238
Soil	Perimeter Stations ^(a)	4x10 ⁻² pCi/g ^(a) Pu-239
Water	White Oak Creek	Not published
	Clinch River	Not published

(a) Soil Samples Collected to Depth of 1 cm.

TABLE 4-21

Environmental Monitoring for the Transuranium Elements at Hanford

<u>Site</u>	Hanford
<u>Location</u>	Southeastern Washington, 320 kilometers east of Portland, Oregon
<u>Mission</u>	Includes Fuel fabrication, liquid waste solidification and radioactive waste burial. Originally, plutonium for nuclear weapons was produced here.

Transuranium Elements Released to the Environment (CY 1975)

To air	1 mCi sum of all plutonium elements
To surface waters -	0.9 mCi sum of all plutonium elements

<u>Media</u>	<u>Sample Collection Location</u>	<u>Average Plutonium Concentrations-CY 1975</u>
Air	Perimeter Stations	<0.03 fCi/m ³ Total Pu
	Distant Stations	<0.04 Total Pu
Soil	Perimeter Stations	<7x10 ⁻³ pCi/g (dry) Pu-239
		<4x10 ⁻⁴ Pu-238
Water	Columbia River-Upstream	<0.03 pCi/l Pu-239
	-Downstream	<0.02 Pu-239
Vegetation	Perimeter Stations	<2x10 ⁻³ pCi/g (dry) Pu-239

TABLE 4-22

ENVIRONMENTAL MONITORING - LAWRENCE LIVERMORE LABORATORY

Media	Sample Collection Location	Average Plutonium	
		Concentrations - 1975	
Air	Site 300	Pu-238	9.0×10^{-4} fCi/m ³
		Pu-239	0.28 fCi/m ³
	Perimeter	Pu-238	$2 \cdot 10 \times 10^{-4}$ fCi/m ³
		Pu-239	0.02-0.035 fCi/m ³
Soil (top 1 cm)	Site 300	Pu-239	0.001-0.03 pCi/g (dry)
	Livermore Valley	Pu-239	0.001-0.1 pCi/g (dry)
Water	Reclamation Plant (effluent)	Pu-239	0.6 pCi/l

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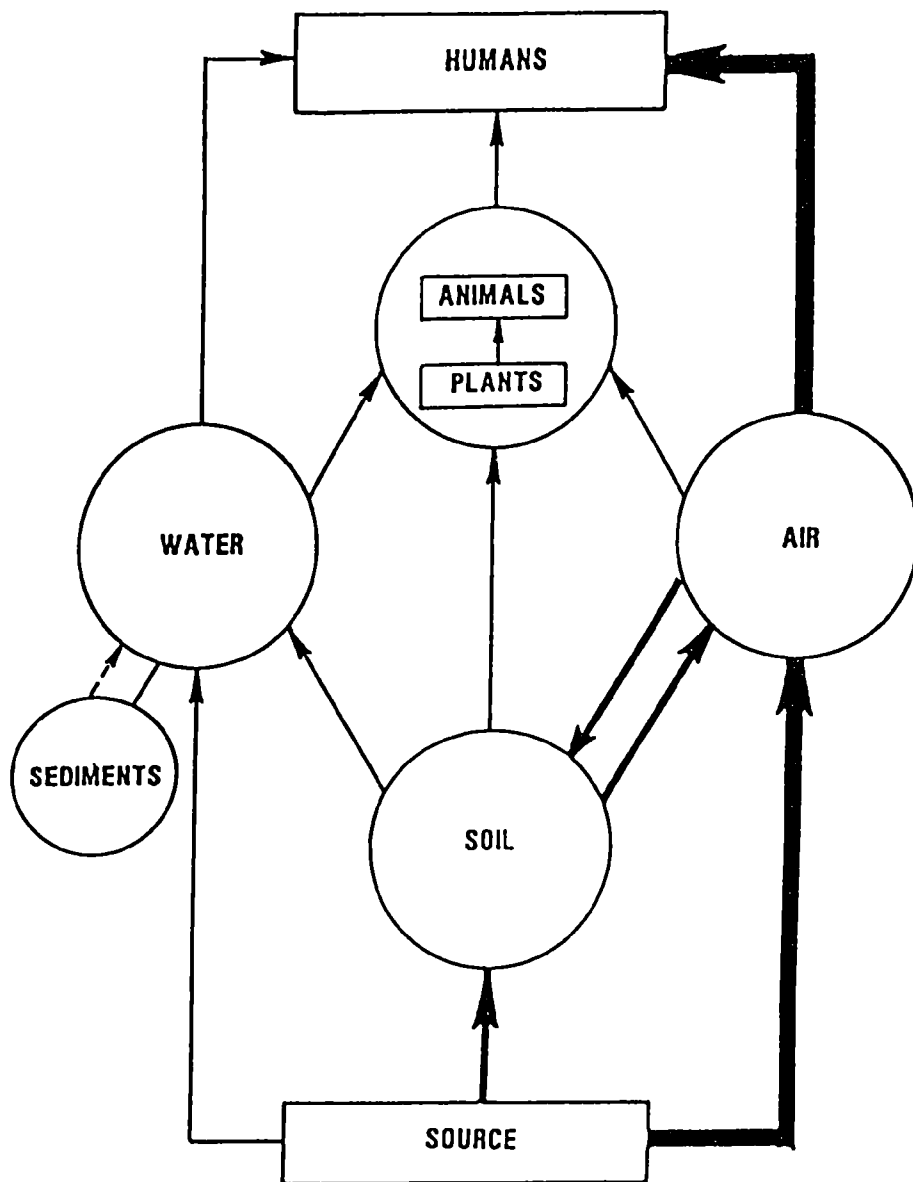
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5. ENVIRONMENTAL TRANSPORT & EXPOSURE PATHWAYS

5.1 Introduction

This chapter provides a brief overview of the transport of the transuranium elements through the environment and the potential exposure pathways through the biosphere. Availability, uptake, and translocation of the transuranium elements within the ecosystem depend upon many factors, including: the mode of release (e.g., accidental fire or spillage), the physical form upon release (e.g., particulate or liquid), the chemical form (e.g., elemental, oxide, or nitrate), and the nature of the environment where the contamination occurs (e.g., desert soil or aqueous media). Also important is use of the environment, which can significantly effect the mobility of the transuranium elements and, subsequently, their effect upon exposed populations. Because there are so many variables, potential pathways of exposure should be evaluated on a site-by-site basis. However, some general conclusions can be made based upon present knowledge of the environmental behavior of these elements. For a terrestrial ecosystem, the major environmental transport pathways are illustrated in Figure 5-1. These pathways include:

1) exchange between air and soil, water, and vegetation as a result of deposition and resuspension, 2) exchange between soil and water by erosion, leaching, absorption, and precipitation, and 3) uptake from air, soil, and water by plants, animals, and man. A comprehensive summary of the environmental transport models has recently been published by the National Council on Radiation Protection and Measurement as NCRP Report No. 76, Radiological Assessment: Predicting the Transport, Bio-accumulation, and Uptake by Man of Radionuclides Released to the Environment. The pathways expected to produce the principal exposures to people are discussed in greater detail in the following sections.



**PRINCIPAL PATHWAYS OF THE TRANSURANIUM ELEMENTS
THROUGH THE ENVIRONMENT TO MAN**

FIGURE 5-1

5.2. Environmental Transport

5.2.1 Aerosol Transport

Airborne releases of the transuranium elements result from both normal and accidental occurrences. Normal operational releases are small and are expected to decrease in the future due to improvements in containment. Accidental releases - such as those resulting from transportation accidents and fires, can lead to localized contamination, and would probably necessitate some form of protective and remedial action. Generally accidental releases of the transuranium elements will be as the element, the oxide, or in soluble form. When the release is in the form of the element, it will convert rapidly to the oxide form, which is relatively insoluble and stable thermodynamically. Airborne releases of the transuranium elements will generally be in the oxide form and contain a substantial percentage of particles within the respirable size range. Because of the small gravitational settling velocities associated with such particles, they can be transported long distances by air currents before depositing on the ground. These particles then will become incorporated into soil and aquatic systems. When deposited on soil, the aerosol particles can attach themselves to the larger, less mobile soil particles. For example, Mork (1) found that Plutonium at most sites was usually bound to soil particles with a diameter greater than $44\text{ }\mu\text{m}$. Likewise, Tamura (2) has analyzed the plutonium bound to soil particles at the Nevada Test Site and showed that the plutonium bound to coarse particles ($5\text{-}20\text{ }\mu\text{m}$) was present as PuO_2 , while the plutonium bound to fine particles ($2\text{-}5\text{ }\mu\text{m}$) was present as hydrated PuO_2 . Subsequent transport will be as a result of wind and mechanical forces which transfer their energy to the surface particles causing them to roll, slide or even become airborne. The smaller the particle diameter the greater will be the tendency for the particle to stay airborne

and the greater will be the distance that it will travel before returning to the surface. Some of the many factors which can influence the redistribution of surface particles by wind are listed in Table 5-1. The multiplicity of factors and their complex interrelationship makes the prediction of soil resuspension and transport a very complex problem. Accordingly, the resuspension of soil particles has been the focus of much research over the past several years.

One of the more commonly used indexes of resuspension has been the concept of a resuspension factor which is defined as the ratio of the air concentration to soil concentration. A wide range of values has been reported for resuspension factors for a variety of surfaces and modes of disturbance. Table 5-2 is a summary of some values reported (3) for newly deposited PuO_2 released during weapons testing. Such a wide range of values makes the prediction of the resuspension factor for a particular set of conditions a difficult task. In general, values for newly deposited material seem to fall in the range $10^{-5}(\text{m}^{-1})$ to $10^{-7}(\text{m}^{-1})$ under conditions of low mechanical disturbance. In areas where the surface is rocky or paved, the resuspension factor may range up to $10^{-3}(\text{m}^{-1})$ due to these smoother, harder surfaces and because little mixing with noncontaminated surfaces occurs (4). Mechanical disturbances, such as vehicular traffic, will also increase the resuspension factor by as much as a factor of 10 to 100 (5). For planning purposes, Stewart (3) and others (6) have recommended a resuspension factor of $10^{-6}(\text{m}^{-1})$ for freshly deposited material under quiescent conditions but recommended increasing this value to $10^{-5}(\text{m}^{-1})$ if there is moderate vehicular or other disturbing activity. As the freshly deposited material becomes aged, fixed to the soil, or mixed with the soil, the characteristics of the contaminant approach the resuspension characteristics of the soil itself. On the basis of empirical information, a model has been proposed (7) in which the resuspension factor decreases from $10^{-5}(\text{m}^{-1})$ to $10^{-9}(\text{m}^{-1})$ within two years. Bennett has reportedly (8) estimated that in a humid

TABLE 5-1

FACTORS AFFECTING RESUSPENSION

METEOROLOGICAL FACTORS

Wind Frequency Distribution:
 Mean Wind Speed
 Wind Direction
Intensity of Gusts
Vertical Turbulent Exchange
Air Density (Temperature-Pressure)
Frequency and Amount of Rain/Snowfall

GROUND-SURFACE PROPERTIES

Soil Characteristics
 Soil Type
 Moisture Content
 Density
 Texture
Particle Characteristics
 Shape
 Density
 Size Distribution
 Cohesiveness
Land Use
Surface Roughness
Type and Amount of Vegetative Cover
Topography/Terrain Irregularities
Soil Disturbance Activities

TABLE 5-2

SUMMARY OF SELECTED EXPERIMENTAL RESULTS
FOR RESUSPENSION (after Stewart)

Measurement Conditions	Resuspension Factor, R_f (m^{-1})	
	Range	Mean
Plutonium sampled at 1 ft above ground (1)		
Vehicle traffic	3×10^{-4} to 7×10^{-4}	
Pedestrian traffic	1.5×10^{-6} to 3×10^{-4}	
Particle size: Mainly 20-60 μm , with 1% in hazardous range ($\sim 3 \mu m$ for PuO_2)		
Uranium sampled downwind from a crater (1)		
At 1 ft. above ground (dust stirred up)		1×10^{-3}
At 1 ft. above ground		3×10^{-4}
At 2 ft. above ground		1×10^{-5}
Brick/plaster dust sample contaminated with I-131 (2)		
Enclosed space	2×10^{-4} to 4×10^{-5}	
Open space		2×10^{-6}
Sample in cab of Landrover, after a test (1)		
Round 1 (H + 18 hr)		2.5×10^{-5}
Round 2 (H + 5 hr)		6.4×10^{-5}
Airborne material without artificial disturbance of ground, consisting of limestone rock and sand with coarse grass and small bushes (3)	1×10^{-6} to 8×10^{-5} (12 results)	1×10^{-5}
Random samples following a tower shot, without artificial disturbance, near crater (3)	1×10^{-8} to 1×10^{-6} (9 results)	2×10^{-7}
On two roads formed by soil grading -no artificial disturbance (3)	1.5×10^{-6} to 1×10^{-8} (14 results)	2.5×10^{-7}
At back of a moving Landrover (3):		
D-Day + 4 (21 results)	8×10^{-7} to 3×10^{-5}	1.4×10^{-5}
D-Day + 7 (21 results)	6×10^{-7} to 4×10^{-6}	1.5×10^{-6}
D-Day + 7 over tailboard	1.6 and 3.1×10^{-5}	2.5×10^{-5}
(1) From nuclear weapon and other tests at Maralinga		
(2) From Civil Defense trial at Falfield, Gloucester		
(3) From Hurricane Trial		

eastern climate the resuspension factor reaches $10^6(\text{m}^{-1})$ to $10^7(\text{m}^{-1})$ after the first good rain or wetdown and then rapidly decreases to $10^9(\text{m}^{-1})$. If the transuranium material is released as a solution rather than as the oxide, its resuspension will probably be in the low range (see Section 2.2). Experiments (3) with yttrium chloride solution sorbed onto soil have indicated a resuspension factor of $10^9(\text{m}^{-1})$, while measurements at Mound Laboratory (4) of Pu-238 released from a waste transfer line produced resuspension factors in the range of $10^8(\text{m}^{-1})$ to $10^9(\text{m}^{-1})$. Because of the propensity for greater mobility on the part of freshly deposited material, stabilization of newly contaminated land should be undertaken as soon as possible after the initial accident in order to reduce the resuspension and inhalation exposure.

5.2.2 Soil Transport

Soil contamination by plutonium has been the most prevalent situation encountered and, therefore, is the most widely studied. Plutonium dispersed onto soil has demonstrated a tendency to bond chemically and/or physically with the soil rather than exist as a separate entity (4,9,10,11,12). Plutonium oxide is relatively inert and initially attaches itself to the soil matrix as a result of adhesive forces established between the plutonium particle and the soil substrate. Over a period of time, weathering processes such as freezing, thawing, and precipitation, will begin to "solubilize" the oxide.

Although generally considered to be insoluble, plutonium oxide can undergo dissolution in a neutral aqueous media. The plutonium oxide particle dissolves, producing plutonium ions until the formation of a hydrated coating inhibits further dissolution. The rate and degree of dissolution depends on many factors including pH, temperature, the presence of oxidizing, reducing, and complexing agents, as well as the specific activity of the radionuclide. The dissolution rate of $^{238}\text{PuO}_2$, for

example, has under certain circumstances been found (13) to be 100 times greater than that of $^{239}\text{PuO}_2$. Plutonium ions formed during the dissolution can undergo ion exchange reactions with the oxygenated ligands commonly found in soil (e.g., silicates) and become sorbed onto the soil, or react with other agents present in the aqueous phase and form soluble complexes. Chemicals that complex the plutonium compete with the silicate particles for the plutonium and tend to reduce the extent of plutonium sorption on soil.

When plutonium is released to soil in soluble form (e.g., as a nitrate) it will already be in ionic form and, in such situations, has been shown (4) to react rapidly with soil. Plutonium ions are capable of displacing most cations (e.g., calcium, magnesium, sodium, etc.) generally found in soils and of forming strong chemical bonds. Several studies (2,14,15) have shown that, after sorption of plutonium has occurred, it will not be readily displaced from the soil by natural processes. Once in the soil, the transuranium elements can be depleted through the migration of particles down through the surface or through the resuspension of a fraction of the material back into the air stream. Of these two mechanisms, the resuspension of soil particles, with which these nuclides have associated in one form or another, will be the principal mode of further environmental transport. The resuspension of soil particles occurs as a result of wind action and more intermittently as a result of mechanical forces, such as plowing and vehicular disturbances. The size of the particle will determine its distance and mode of transport. Particles with diameters greater than $1000\ \mu\text{m}$ generally slide or roll along the surface (creep), while particles with diameters in the range of $50\ \mu\text{m}$ to $1000\ \mu\text{m}$ move in short hops along the surface, usually at a small distance from the surface (saltation). The suspension of particles is

generally restricted to those below 50 μm , which will be carried along with the air stream.

5.2.3 Aqueous Transport

Studies have been conducted on various water bodies, streams, rivers, lakes, estuaries and oceans to determine the final disposition of plutonium in these environs. The following behavior has been noted:

(1) More than 90% of the plutonium becomes bound to suspended sediments and carried to the sediment bed.

(2) Situations where reducing and complexing agents are both present can lead to resolubilization of the plutonium in the sediment bed

(3) Seaweeds generally have the ability to concentrate plutonium with concentration factors of >1000 .

(4) Benthic biota can alter the plutonium concentration profiles in the sediment beds.

Specifically, plutonium oxide exposed to an aqueous medium undergoes slow dissolution, producing various complex ions of plutonium as well as polymers in colloidal form and the hydrous oxide as a precipitate (13). In 1972 Langham (16) studied the fate of PuO_2 following the Thule incident and found that the majority of the plutonium agglomerated into inactive debris with only about 1% suspended as fine particulates in the water. Further studies (17) after the Thule incident showed 95% of the plutonium to be associated with the bottom sediments to a depth of at least 10 cm. In addition, a study (18) of nuclear waste discharged into the Irish Sea from Windscale has found most of the Pu-239 and Am-241 to be associated with the sediments close to the discharge area. Similar findings were observed (4) around Mound Laboratory where plutonium was accidentally discharged into a freshwater canal. Again the plutonium was found to be largely associated with the bottom sediments. Therefore, although the movement of the transuranium elements through aqueous systems is

not yet well defined, the information now available would indicate a limited mobility for environmental transport via such systems.

5.3. Exposure Pathways

The principal hazard that arises as a consequence of soil being contaminated with the transuranium elements is exposure to radiation through the inhalation and ingestion pathways. For a detailed discussion of the entry of plutonium and other actinides into animals and man and the resultant biological behavior, the reader is referred to ICRP Publication 19 (19) and the chapter entitled "Dose and Risk to Health due to the Inhalation and Ingestion of Transuranium Radionuclides". The following section will be limited to a description of the environmental factors affecting the inhalation and ingestion pathways.

5.3.1 Inhalation

Inhalation exposures arise from direct injection of transuranium radionuclides into the atmosphere (e.g., normal emissions and accidental fires) and also from the resuspension of previously deposited material. For the latter pathway, only a very small fraction of the material on the surface actually becomes airborne and available to man. In general, the respirable size is considered to be that range of particles with aerodynamic diameters less than 10 μm . An assessment can be made, using dosimetry models, of the potential health hazard resulting from the inhalation of airborne particles. Such a model requires knowledge of the total airborne activity and of the activity median aerodynamic diameter associated with it. (Aerodynamic diameter is the diameter of a sphere of unit density having the same settling velocity as the particle in question of whatever shape and density). The assumption made by most

dosimetry models is that the aerosol distribution is log normal and can, therefore, be described through the use of two parameters - the activity median aerodynamic diameter (AMAD) and the geometric standard deviation of the distribution, σ_g .

Sampling should be conducted so that 1) the total airborne distribution is being measured, and 2) the AMAD determined actually describes the corresponding distribution of airborne activity. Healy (20) and others (21) have emphasized the necessity of considering the resuspension of soil by mechanisms other than normal wind activity. The possibility exists that other mechanisms could, under certain circumstances, produce exposures exceeding those normally received via the resuspension pathway. Although this possibility has been recognized, relatively little experimental data is currently available to determine quantitatively the importance of the many possible secondary resuspension mechanisms. Two commonly encountered disturbances (agricultural operations and vehicular disturbance) have recently been investigated, however, and some conclusions can be drawn from these studies.

For the agricultural situation, the vicinity of a field contaminated over a period of twenty years was monitored. Increase in airborne activity was measured during such activities as plowing, disking, and planting (22). During these operations, the air activity was found to increase by a factor of approximately 30 at the location of the tractor operator and by a factor of 6 at a distance 30 meters away from the edge of the field. Assuming that these activities take place 30 days of the year for 8 hours each day (i.e., 1/36 of the year), it can be calculated that the average yearly air activity will increase by 80 percent for the tractor operator and 10 percent for an individual in the vicinity of the field. This level of increased air concentration will occur for only the first year. Subsequent agricultural activities should generate lower air concentrations, because the activity originally on the surface will be diluted

through mixing with soil previously below it. The conclusion that can be drawn from such an analysis is that these agricultural operations would pose an increased inhalation hazard to the vehicle operator during the first cultivation cycle, and some protective action might be in order during that time. Subsequent cultivation, however, should not lead to significant increases in the inhalation hazard. For surrounding areas, no significant inhalation hazard would be predicted during any of these operations.

Regarding vehicular disturbances, Sehmel (5) has examined the importance of auto and truck traffic in increasing resuspension. It was concluded that such disturbances, in the case of an asphalt surface with newly deposited material, will lead to increased resuspension, with a fraction resuspended of the order of 10^{-5} to 10^{-2} per vehicle passage. The higher rates occurred at speeds typical of freeway driving; after the passage of about 100 cars only a small fraction of the original contamination remained on the road surface. The material resuspended from the road surface deposited on the ground at various distances from the road and was again available for resuspension, but at a much lower rate.

The potential for increased exposure from such situations will depend upon many factors in addition to the quantity of contaminating material, including the time of exposure, the frequency of vehicular passage and the speeds, and the distance from the road to the receptor. Based upon Sehmel's experiment, it can be expected that the integrated inhalation exposure due to the vehicular disturbance will be smaller than the chronic exposure received from daily living within the generally contaminated area. The material deposited on the road surface will be depleted quickly and, once it is removed from there, its resuspension will be orders of magnitude lower. Sehmel's results indicate that the material transferred to the road parking strip is resuspended at a rate only one tenth of that on the road

itself. In addition, the total quantity of material resuspending at the higher rate would be small relative to the surrounding area; once redistributed over the larger area, it should show little increase in the average air concentration.

5.3.2 Ingestion

Under normal circumstances, exposures via ingestion will arise from the consumption of crops and animals grown on land contaminated by the transuranium elements. Studies to date have made assessments of the ingestion pathway by two methods: some studies have looked at the uptake factors for various plant species grown in contaminated soils, while others have measured the residual amounts of fallout plutonium in processed foods.

Two publications (23,24) have tabulated the results of uptake studies performed on plutonium and other transuranium elements. These studies have shown plant and animal uptakes to be very small, with the concentration in plants (fresh weight basis) being generally less than 10^{-4} of that in dried soil and the concentration in animal tissue (fresh weight) being about 10^{-5} of that in the plants they eat. Preliminary studies (23) for transuranium elements other than plutonium produced uptake factors somewhat higher than comparable studies with plutonium. Initial studies (25,26) indicate an increase in uptake with time, possibly as a result of bacterial action or increased solubilization. The use of chelating agents as a part of agricultural practices may also increase the uptake of the transuranium elements with time (27).

Measurements of plutonium in "market basket" food samples, in which proportions of processed foods are chosen to represent the annual total diet, can be used as indicators of the quantity of plutonium ingested (28). Such findings apply to a given soil contamination level when all consumed food is grown on

contaminated lands. The methodology gives an overestimate of activity ingested, because foods from other areas not as highly contaminated will make up part of the diet. These studies have observed uptake factors for plutonium in the range of 10^4 plus or minus an order of magnitude. Based upon such uptake factors and food consumption estimates, the annual estimated intake during 1972 of fallout plutonium was 1.6 pCi while the intake for 1965 was estimated to be 2.6 pCi. There is no reason to believe that the uptake factors for crops grown on land with concentrations higher than fallout levels should vary significantly from those obtained through this "market basket" sampling technique. Some evidence indicates that americium is concentrated in certain species of plants relative to plutonium. Preliminary analyses (29) of the "market basket" samples indicate that the Am 241/Pu 239 ratios in diets are not greatly different from current Am 241/Pu 239 ratios in soil. The ingestion of plutonium through drinking water is another possible pathway to humans. The concentration of fallout plutonium in finished drinking water has been found (28) to be low (<3 fCi/l). However, in areas of elevated levels, plutonium could migrate over time into cisterns and wells, thus increasing the activity in drinking water. It has been suggested (20) that a significant ingestion pathway could be the accidental ingestion of contaminated soil by adults or the deliberate ingestion of soil by children (pica). However, assumptions of very large soil consumption rates would be required for this pathway to become as significant as the inhalation pathway, because of the small transfer rate of most transuranium nuclides from the gastrointestinal tract to blood.

5.4. Methods of Relating Soil Concentration to Airborne Activity

The relationship between soil and air concentrations is affected by many complex factors (see Table 5-1). Attempts to derive values for them have resulted in many different approaches; each uses different concepts and methods of

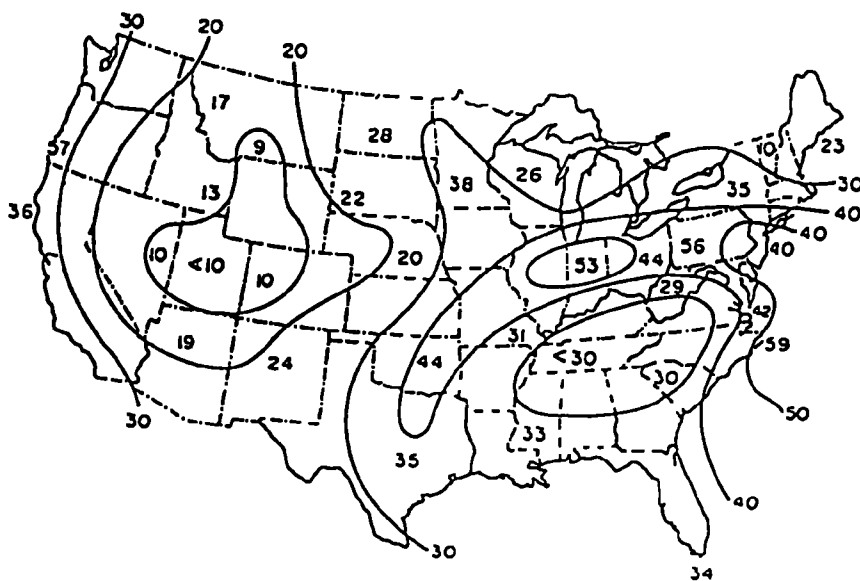
measurement, selecting some physical factors as important and tending to neglect others or include them as constants. The purpose of this section is to describe briefly some of the more commonly used approaches to relate soil contamination levels to airborne radioactivity.

5.4.1 Resuspension Factor

One of the earliest and still most widely used methods of predicting the relationship between soil and air contamination is the resuspension factor. It is defined as the ratio of the plutonium concentration in air, measured at some distance above the ground, to that of plutonium in the soil:

$$K = \frac{\text{concentration in air (activity/m}^3\text{)}}{\text{concentration in soil (activity/m}^2\text{)}}$$

The resuspension factor has limitations in its application. In the first place, it assumes that the air concentration above a contaminated surface is directly proportional to the surface contamination level, rather than on the extent of ground contamination upwind of the sampling site which is indeed the case. In the second place, the resuspension factor is an empirically determined value which can be applied only to prevailing conditions at a given site and at a given time. Most resuspension experiments have been conducted for a relatively short duration of time and do not necessarily represent the long-term average situation for a particular area. Finally, applying a resuspension factor derived at one particular site to predict airborne contamination levels at another site would be a questionable extrapolation. However, for areas where the resuspension factor has been measured over a period of time, sufficiently long to average out the variability of the local meteorology, then this approach can be useful in assessing the potential hazard from existing soil contamination.



ANNUAL MEAN MASS CONCENTRATIONS ($\mu\text{g}/\text{m}^3$) OF AIRBORNE PARTICLES FROM NON-URBAN STATIONS OF THE U.S. NATIONAL AIR SAMPLING NETWORK. 1964 - 1965

FIGURE 5-2

TABLE 5-3

OBSERVED AIR CONCENTRATIONS
 COMPARED WITH PREDICTIONS BY MASS LOADING MODEL
 [Adapted from Anspaugh et al (1974)]

Location, etc.	Radionuclide	Air Concentration	
		Predicted ^a	Measured ^b
<u>GMX site, USAEC Nevada</u>			
<u>Test Site</u>			
NE, 1971-1972	²³⁹ Pu	7200 aCi/m ³	6600 aCi/m ³
GZ, 1972, 2 weeks	²³⁹ Pu	120 fCi/m ³	23 fCi/m ³
<u>Lawrence Livermore</u>			
<u>Laboratory</u>			
1971	²³⁸ U	150 pg/m ³	52 pg/m ³
1972	²³⁸ U	150 pg/m ³	100 pg/m ³
1973	²³⁸ U	150 pg/m ³	86 pg/m ³
1973	⁴⁰ K	1000 aCi/m ³	980 aCi/m ³
<u>Argonne National</u>			
<u>Laboratory</u>			
1972	²³² Th	320 pg/m ³	240 pg/m ³
1972	nat _U	215 pg/m ³	170 pg/m ³
<u>Sutton, England</u>			
1967-1968	nat _U	110 pg/m ³	62 pg/m ³

^a Predicted value is equal to the soil concentration (activity/g) × 10⁻⁴ g/m³.

^b Most values are annual averages.

5.4.2 Mass Loading

One attempt to increase the capability of predicting soil resuspension has been the mass loading approach. This technique assumes the mass loading of the air with particulates to be an index of resuspension and derives the airborne concentration of a specific radionuclide by a comparison with its concentration on the adjacent surface. Specifically:

$$\text{Air Concentration} = \text{Soil Concentration} \times \text{Mass Loading} \times \text{C.F.}$$

where C.F. is the units conversion factor based upon the depth of sampling and the soil density.

Airborne particulate mass loading is one of the criteria for clean air standards and measurements are widely available for urban and nonurban locations through the National Air Surveillance Network (NASN). The data recorded at nonurban stations are a better indicator of the levels of resuspended material than are urban measurements. In general, annual mean mass concentrations of airborne particulate material at the nonurban stations range from 5-50 micrograms per cubic meter (see Fig. 5-2); the mean arithmetic average for 1966 of all 30 nonurban NASN stations was 38 ug/m^3 (30). Anspaugh (30,31) employed this model to predict air concentrations at a number of sites. Predicted values did not exceed measured values by more than a factor of roughly five (Table 5-3). The fallacy of the model is in assuming that the resuspendible fraction of the soil would carry with it an equal fraction of the activity, which implies essentially that: 1) activity is distributed homogeneously in the top soil and, 2) activity exists independent of particle size. For instance, if the specific ground activity is associated mostly with particles of size greater than $50 \text{ }\mu\text{m}$, a very small air concentration would result, although the model would predict the same air concentration for this case as it would for all the activity being distributed among particles of

resuspendible size. In either case the model would fail. Data obtained (2,4,15) at sites of present contamination have shown a nonuniform distribution of activity with particle size, probably caused by such factors as: 1) the chemical form of the Plutonium when released, 2) the ion exchange capacity of the soil, and 3) the surface area of the soil particles. It would seem reasonable, however, that the error associated with using the mass loading approach would be least for soils in which the contaminant has been present for some time and in making predictions of average annual air concentrations.

5.4.3 Resuspension Rate and Other Approaches

Other approaches of a more sophisticated nature have been developed to describe the resuspension of particles from a soil surface. These approaches have attempted to include in their formulations as parameters some of the physical forces which control the resuspension phenomenon. One such technique proposed by Healy and Fuquay (32) is the resuspension rate approach. This model combines atmospheric transport and diffusion along with particle resuspension to calculate airborne concentration. This is achieved by assuming that the rate of pickup of particles from a surface is directly proportional to the ratio of wind forces to gravity forces on individual particles. Taking the wind force on a particle as proportional to the square of the wind velocity and to the particle area exposed to wind, the model develops a formulation for the resuspension rate, i.e., the rate at which particles will be resuspended by wind from a soil surface. Once the resuspension rate has been determined it can be used as the source term in a standard atmospheric diffusion equation to predict the resultant air concentration at some distance from the contaminated site. Healy (20) has refined the model formulations to be capable of handling various geometric configurations of the contaminated area and the variability of surface concentration within the contaminated area. The advantages of a model of this type are that it recognizes some of the physical conditions and

processes which affect resuspension as well as providing a method to calculate air concentration at various distances away from the contaminated area.

One assumption by Healy in the original formulation of the resuspension rate model was that the pickup rate for particles is a function of the square of the wind velocity. Studies have been conducted to establish the relationship between resuspension rate and wind velocity. One such study conducted by Sehmel (33) at Rocky Flats, for only short time periods and for one sampling station, found the air concentration to be a function of the square of the wind velocity, implying that the pickup rate was a function of the cubic power of the wind speed. However, other experiments by Sehmel (34) have shown resuspension rates to increase with wind speed to the 6.5 power. Studies are continuing to better elucidate this functional relationship.

Other approaches (35,36) have also been proposed which attempt to relate particle resuspension to such factors as the soil erodibility index, surface roughness factor, and quantity of vegetative cover. These models generally require the determination of several empirical constants in their application. Although these constants may be applicable for the conditions under which they have been measured, the general applicability of these formulations in predicting air concentration has not been demonstrated at this time.

5.4.4 Enrichment Factor

In order to take into consideration the non-uniform distribution of activity with soil particle size as well as the non-uniform resuspension of particle sizes, an "enrichment factor" can be derived which is included in the mass loading calculation. Potential exposure due to contaminated soil depends largely on the amount of activity associated with particles in the respirable size range (generally 10 μm). It has been suggested by several investigators that sampling of only those particles in a soil sample which are within the inhalable size range would give the best measure of risk to the public health. However, the weight fraction of particles in the less than 10 μm range is small in most soils, and sampling, separation, and analysis techniques are correspondingly more difficult and inaccurate. There is also considerable evidence that some of the larger particles really consist of aggregates and are relatively easily broken down into smaller ones, so that an instantaneous measurement of a single size range may not give a good picture of long-term trends. Another important objection to limited sampling is that larger particle sizes may make a substantial contribution to other possible pathways (e.g., ingestion), and hence should be measured. To evaluate the potential hazard of the inhalable fraction of soils, while retaining the advantages and conveniences of analyzing the entire soil sample, the mass loading approach can be modified by use of an "enrichment factor". The proposed method weights the fraction of the activity contained within the respirable range in terms of its deviation from the activity to mass ratio for the entire sample and, at the same time, addresses the problem of the nonuniform resuspension of particle sizes mentioned in the previous section. The inhalable fraction of the soil is weighted by considering the relative distribution of activity and soil mass as a function of particle size for representative samples of soil. To accomplish this, the sample of contaminated soil is segregated into size increments, and the activity and mass contained within each size

increment is determined. The factor g_i is then defined as the ratio of the fraction of the total activity contained within a size increment i to the fraction of the total mass contained within that increment. A value greater than 1 for g_i implies an enrichment of activity in relation to mass within that incremental fraction, while a value less than 1 indicates a dilution of the activity with respect to mass relative to the average for the sample.

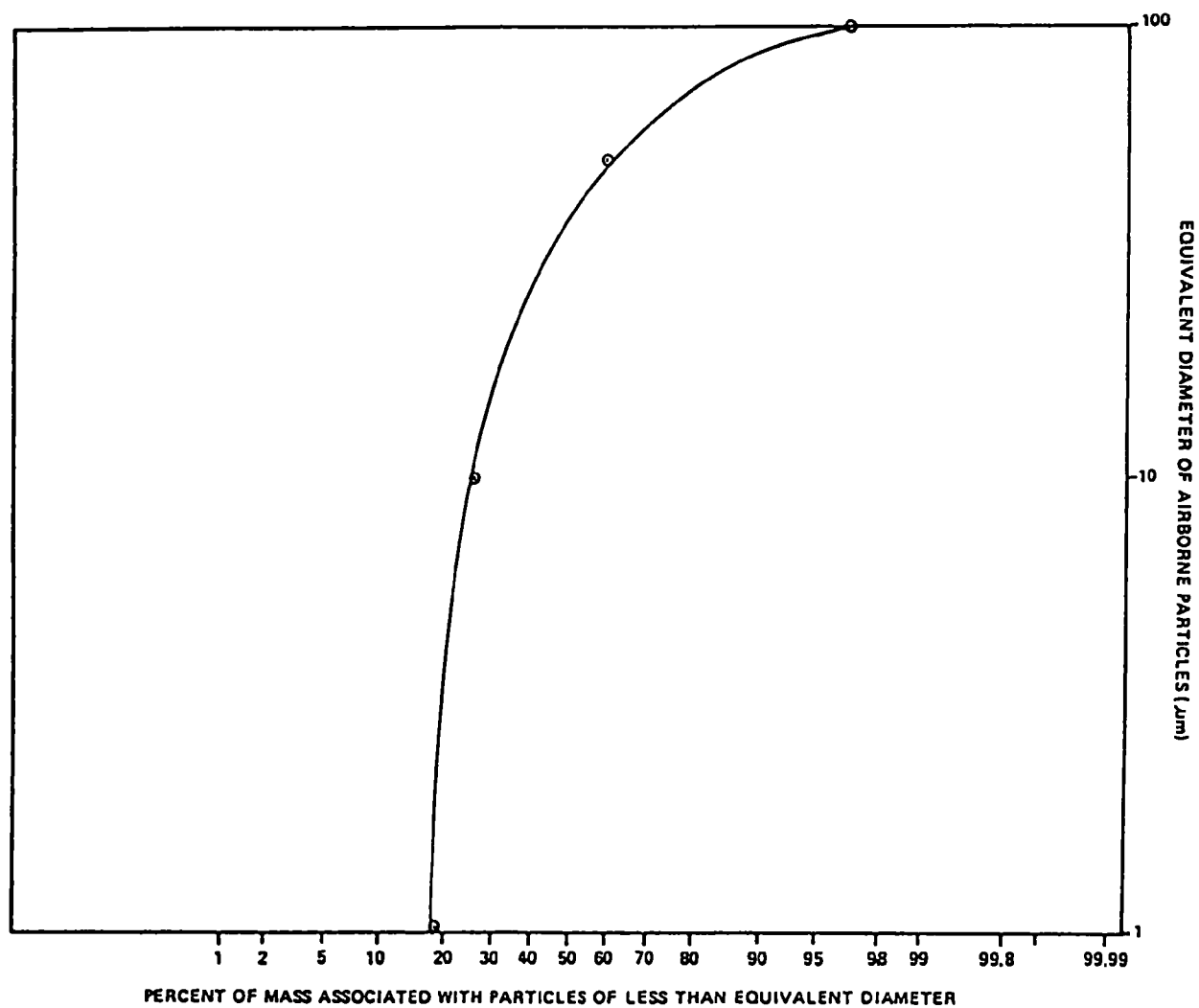
In order to evaluate the potential for inhalation of resuspended plutonium, the nonuniform resuspension of particle sizes in each size increment of the surface soil must also be considered. Accordingly, the mass loading can be derived as a function of the measured particle size spectrum. The fraction of the airborne mass contained within each size increment, i , is calculated and designated as f_i . The factors of f_i and g_i can then be incorporated into the mass loading formulation as follows:

$$\text{Air Activity}_i = \text{Air Mass Loading} \times f_i \times \text{Soil Activity} \times g_i$$

Summation over all the size increments results in the total air concentration:

$$\text{Air Activity} = \text{Air Mass Loading} \times \text{Soil Activity} \times \sum f_i g_i$$

The term $\sum f_i g_i$ weights the contribution of the plutonium from each soil size fraction to the total resuspended material, thereby taking into account both the nonuniform resuspension of particle sizes as well as the nonhomogeneous distribution of activity. The summation of $f_i g_i$ will be referred to as the "enrichment factor", where f_i accounts for the distribution of airborne mass as a function of particle size and g_i accounts for the variability of both soil activity and soil mass as a function of particle size.



PARTICLE SIZE DISTRIBUTION OF RESUSPENDED SOIL

FIGURE 5-3

TABLE 5-4

EXPERIMENTAL DATA FOR WEIGHT AND ACTIVITY FRACTIONS
 FOR SOILS IN THE ENVIRONS OF THE ROCKY FLATS PLANT
 [Sampling and Analysis by US Environmental Protection Agency]

<u>Sample</u>	<u>Size Increment (um)</u>	<u>Wgt Fract</u>	<u>Act Fract</u>	<u>g₁</u>	<u>f₁</u>	<u>$\sum f_1 g_1$</u>
RF 1A	2000-105	.62	.07	.12	-	
	105-10	.18	.40	2.21	.7	
	<10	.20	.53	2.65	.3	2.34
RF 1B	2000-105	.63	.39	.63	-	
	105-10	.17	.06	.34	.7	
	<10	.20	.55	2.74	.3	1.06
RF 1C	2000-105	.64	.43	.68	-	
	105-10	.16	.07	.46	.7	
	<10	.20	.49	2.47	.3	1.06
RF 2A	2000-105	.46	.13	.28	-	
	105-10	.34	.37	1.10	.7	
	<10	.20	.50	2.48	.3	1.51
av.						1.49

5.4.5 Correction for Area Size

Use of the mass loading approach implies that the air concentration is at equilibrium with the ground surface, i.e., a steady state situation exists in which the amount of material coming up from the surface is balanced by the amount of material depositing back onto the surface. In the strictest sense this limit can only be achieved for source areas approaching infinite dimensions. For source areas of finite dimensions a fraction of the airborne mass loading can be arising from an uncontaminated area upwind which, although contributing dust to the atmosphere, contributes no radioactivity. The smaller the size of the contaminated area the less it will contribute to the mass loading level and the greater the uncertainties involved in applying the mass loading model.

Healy (37) has attempted to quantify the relationship between the size of the contaminated area and the air concentration that would result from it. His calculations show that, for a contaminated area which is 50 meters in horizontal depth, the air concentration would be approximately a factor of one hundred smaller than from an area 5000 meters in depth (based upon certain assumptions regarding meteorological conditions). Therefore, a precise calculation requires a correction for area size when applying the mass loading approach to small areas of contamination. However, since one cannot predict a priori the extent of a contamination incident nor the prevalent meteorology, a conservative estimate can assume that the area contaminated is sufficiently large that a correction for area size is not necessary.

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