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TRANSURANIC ELEMENTS IN THE ENVIRONMENT

A Summary of Environmental Research on Transuranium
Radionuclides Funded by the U. S. Department of Energy
Through Calendar Year 1979

Wayne C. Hanson, Editor
Pacific Northwest Laboratory

Prepared for the U. S. Department of Energy
Assistant Secretary for Environment
Office of Health and Environmental Research

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Foreword

Before 1973 environmental research into the behavior of the transuranium elements was conducted on an ad hoc basis. It was usually prompted by some contamination event, such as the loss of nuclear material in the military aircraft accidents at Palomares, Spain, and Thule, Greenland, or the discovery of plutonium concentrations that exceeded fallout levels at such locations as the Rocky Flats Plant near Golden, Colo., and the Nevada Test Site. These research activities were usually aimed at describing the distribution of plutonium and appraising the health hazard at the individual site. Because this information was gathered at specific sites, it was not sufficient for generalized statements about environmental movement. In about 1970 the Nevada Applied Ecology Group began an integrated program at the Nevada Test Site in an attempt to provide a broader information base on transuranium elements. This program, however, was applicable primarily to desert environments. Some experimental studies at other locations were concerned with the uptake of transuranium elements by vegetation, but most of these dealt with western soils of high pH. No concerted effort was made to study transuranic radionuclide behavior in the marine environment except for studies at Thule, Greenland, and the Pacific Testing Grounds in the Marshall Islands.

In 1973 the U. S. Atomic Energy Commission, Division of Biomedical and Environmental Research (BER) (now U. S. Department of Energy, Office of Health and Environmental Research), performed an intensive study of its research efforts in support of the development of nuclear power with special emphasis on the Liquid Metal Fast Breeder Reactor (LMFBR). The environmental team reviewed information gathered up to that time on transuranic cycling in various environments and concluded that a comprehensive description of the environmental hazards of plutonium and the other transuranium elements relative to the LMFBR could not be made with the available data nor would it be forthcoming with the established research by BER contractors. It was obvious that too much of the past research had been centered on studies in the western regions, which were arid or semiarid, and essentially no studies had been made of soil movement and plant uptake in the humid eastern regions where fuel reprocessing plants were scheduled to operate. In addition, very little information was available on the cycling of plutonium through aquatic food webs inclusive of the marine studies in Greenland and the Marshall Islands. Essentially no research on the environmental behavior of transplutonic elements was under way, and the question of the long-term behavior and fate of the transuranium elements had not been addressed in any effective way. Further, the question of biological modification of the transuranium elements, which might lead to increased mobilization in the environment and possible underestimations of the dose to man, was not addressed.

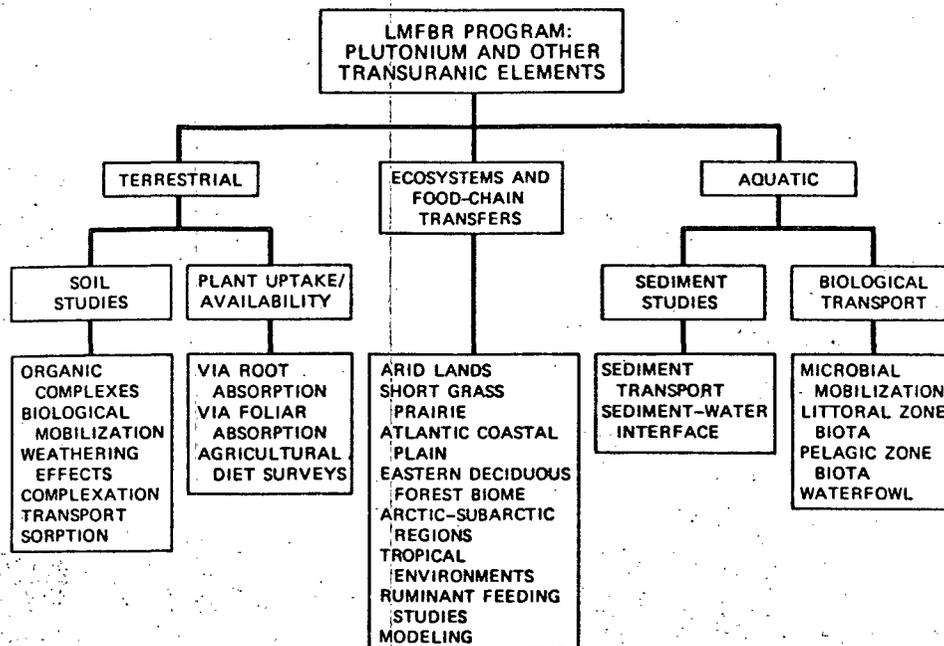


Fig. 1 United States Department of Energy, Office of Health and Environmental Research, transuranium elements research program.

laboratory reports, government documents, and refereed journals. This volume is an attempt to assemble the accumulated information as a synthesis document to provide an up-to-date interpretation of the environmental behavior of the transuranium elements.

R. L. Watters
Office of Health and Environmental Research
U. S. Department of Energy

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The conclusions of the environmental team prompted AEC to develop a research program which would develop the information that was not available and which would be as comprehensive as possible for future assessments on the impacts of transuranic radionuclides from all stages in the nuclear fuel cycle. The program was designed to take advantage of the high-quality research that was already under way at the Health and Safety Laboratory, the Nevada Test Site, Lawrence Livermore Laboratory, Pacific Northwest Laboratory, the University of California at Los Angeles, Woods Hole Oceanographic Institute, and the University of Washington and to complement this research with research projects in other geographies and climates. Research activities were selected to cover all aspects of environmental transport from soil processes to ecosystem cycling. The objectives of the research program were to understand the cycling behavior of the transuranium elements in our environment and to determine to what degree these elements would be transported to us through food chains and aerial pathways. A further objective was to develop a satisfactory description of the degree to which the transuranium elements persist in the environment as a first step in assessing the potential hazard of these species on a historical and geological time scale.

To achieve the broad objectives of this program, we must answer many questions. If the transport of the transuranium elements is to be described, we must know to what degree these elements can be mobilized in the soils and aquatic sediments where they reside. A compendium of concentration ratios for plant uptake into food crops on various agricultural soils must be assembled. The transport through aquatic and terrestrial food chains must be quantified and appraised for the potential for human ingestion. Changes in the availability of transuranic elements due to resuspension from soils also must be better understood. Although the thrust of this work is on environmental transport, all the research scientists are alert for unusual concentration processes that might lead to radiological effects within environmental systems.

The areas of research just mentioned are of immediate concern, but beyond these near-term considerations are those related to the possible long-term persistence of the transuranic elements in available form on the scale of hundreds and thousands of years. Such considerations are very difficult to address adequately with contemporary research. However, two approaches are under way which may provide reasonable first approximations to the prediction of long-term behavior. One is the theoretical approach to studying the chemical and physical processes in soil of these radionuclides with the objective of developing good thermodynamic data. We need information on the equilibrium concentrations of the various oxidation states in different environments, on complexation processes, and on diffusion coefficients for various species. We can then apply this type of information to predictive modeling. An empirical approach would be to study the distribution and environmental behavior of naturally occurring elements that have properties analogous to those of the transuranium elements. For instance, the availability for plant uptake of the rare earth neodymium, which has been subjected to weathering for thousands of years, may provide a basis for predicting the uptake of americium after long periods of time because americium and neodymium have quite similar chemical and physical properties. Other rare earths, uranium, and thorium are also candidates as analogs for some of the transuranic elements in environmental studies.

The general areas of this program are outlined in Fig. 1. Research has now been conducted for periods of time ranging from 2 yr for new work to 6 yr for work that preceded this comprehensive program. Some of the results have been published in

Ecological Relationships of Plutonium in Southwest Ecosystems

T. E. HAKONSON and J. W. NYHAN

A comprehensive summary of results was prepared on plutonium distribution and transport in Los Alamos and Trinity Site study areas. Despite differences in ecosystems and plutonium source, there are several similarities in plutonium distribution between Los Alamos and Trinity Site study areas. First, the soils/sediment component contains virtually all the plutonium inventory, with vegetation and rodents containing less than 0.1% of the total in all cases.

Plutonium has penetrated to considerable soil depths at both locations, although it has occurred much more rapidly and to a greater degree in the alluvial soil at Los Alamos than in the arid terrestrial system at Trinity Site. However, in all cases less than 50% of soil-column plutonium inventories was found in the surface 2.5 cm. The plutonium penetration depth appears to correspond to the moisture penetration depth at Trinity Site. This is probably the governing factor at Los Alamos, although storm runoff and accompanying turbulent mixing processes complicate the process. In Acid-Pueblo Canyon, the bulk of the soil column inventory lies in the lower profiles, an indication of the loss of the plutonium from surface layers due to sediment transport.

Soil plutonium, in most cases, was associated with relatively coarse-size fractions. The silt-clay (<53 μm) fraction contained relatively little (<15%) of the plutonium; this reflects the small amounts of this size fraction in study area soils. An exception was in Area 21 at Trinity, where the <53- μm soil-size fraction contained about 73% of soil plutonium inventories. The importance of these distributional differences was demonstrated for Trinity Site, where Bagnold dust samples from Area 21 contained 54% silt-clay material and samples from Area Ground Zero (GZ) contained less than 10% of this material.

Concentrations in herbaceous vegetation were generally related to those in soils from all sites. Our belief, although unsubstantiated, is that external contamination of the plant surfaces is the major contaminating mechanism in these arid systems. The plutonium concentrations in certain rodent tissues from all study areas were related to corresponding soil concentrations. Over 95% of the plutonium body burden in rodents was associated with pelt and gastrointestinal tract samples, indicating the dominance of physical processes as the contaminating mechanism.

Horizontal transport of soil plutonium is dominated by physical processes. At Los Alamos water governs the downstream transport of soil plutonium, and indications are that wind is a relatively more important transport vector at Trinity Site.

In no case was there evidence for trophic-level increase due to physiological processes as plutonium passes from the soil to vegetation to the rodents, although food habits of rodents are not known sufficiently to preclude a trophic-level increase. We believe, however, that rodents most likely come into contact with environmental plutonium directly from the soil and not through a food-web intermediary.

Several reviews on environmental plutonium distribution and transport indicated a general lack of published field data from representative areas of the United States (Francis, 1973; Price, 1973; Romney, 1977; Hanson, 1975; Hakonson, 1975). Several field studies of plutonium have been initiated in the last few years to address informational needs at a number of locations which encompass a wide spectrum of climatic conditions ranging from deserts to humid forests and contain plutonium from industrial, weapons, or accidental-release sources.

The comparison of plutonium data from two southwest ecosystems in this chapter is one step in the total synthesis of information from various regions of the United States where types of ecosystems and sources of plutonium differ. The southwest United States is an important study locale because of the energy activities that may develop and the lack of understanding of the processes in arid systems which govern distribution and transport of contaminants. In this regard studies on environmental plutonium are useful to develop an understanding of patterns that are applicable to the transport and fate of other materials.

The objective of this chapter is to use existing plutonium contamination in the canyon waste areas at Los Alamos and in the grasslands in the fallout zone at Trinity Site

- To evaluate the role of environmental transport processes in distributing and redistributing surface inputs of plutonium.
- To evaluate the transport of environmental plutonium to the biosphere and the relationships that lead to the potential for human exposure.
- To compare plutonium behavior in these two major southwest ecotypes.

The tasks in this study were to (1) document plutonium inputs where possible, (2) develop an understanding of distributions by inventory of major environmental components, and (3) evaluate transfers as functions of ecological variables. Plutonium, as used in this chapter, denotes ^{238}Pu and/or $^{239,240}\text{Pu}$.

Site Descriptions

Los Alamos

The canyons at Los Alamos, in north central New Mexico (Fig. 1), are typical of those in the southwest plateau region of New Mexico, Arizona, Colorado, and Utah. They vary from 10 m to over 200 m in depth and were formed by water erosion of the volcanic substrate of the Pajarito Plateau. The area has a semiarid continental mountain climate (Table 1) with annual precipitation ranging from about 20 to 50 cm as elevation increases from 1650 to 2200 m; rainfall accounts for about 75% of the annual precipitation. Drainage from the 113-km² Laboratory site is via the many canyons that bisect the plateau. The biotic resources of the canyons are diverse (Miera et al., 1977); total vegetative ground cover is variable but generally high and approaches 100% in some areas owing to the dense overstory, which is partly due to the industrial liquid effluents.

Nearly all the liquid wastes generated by the Laboratory since 1943 have been collected by industrial waste lines, treated (since 1951), and released into one of three canyons (Fig. 1). The results of studies in two of these canyons are emphasized in this chapter since they represent the extremes in temporal use history. The oldest waste-receiving area is Acid-Pueblo Canyon, which was used from 1943 to 1963 and

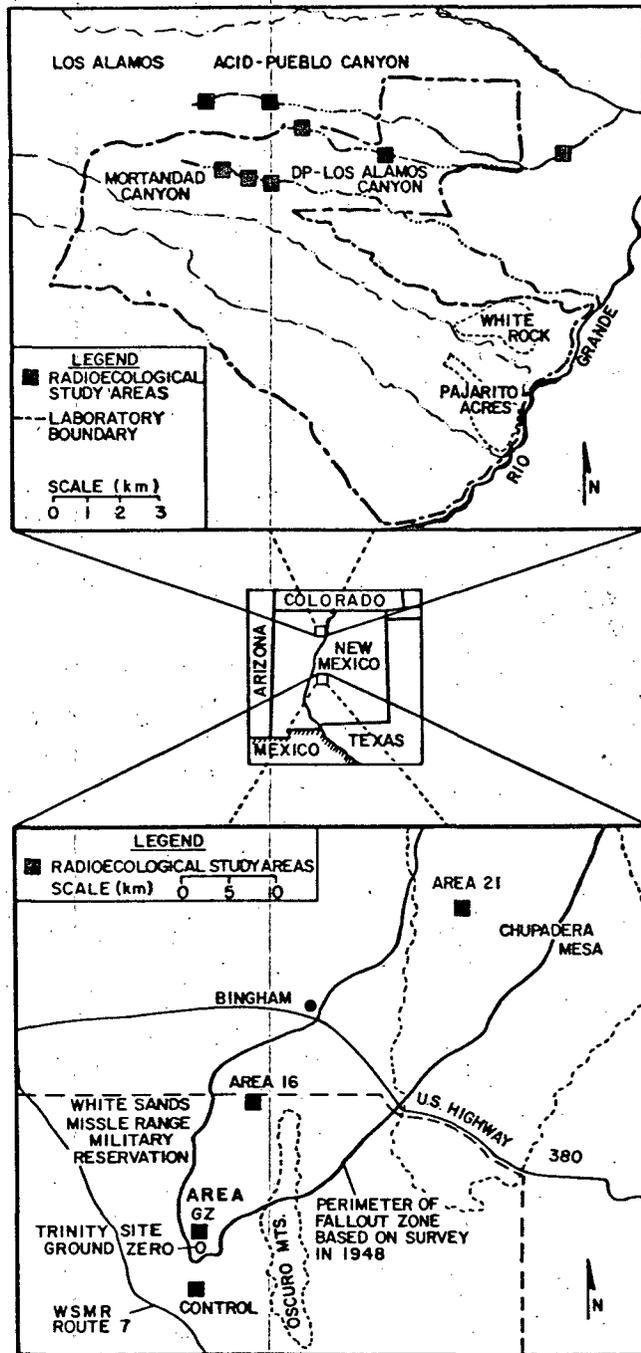


Fig. 1 Plutonium study areas at Los Alamos and Trinity Site, New Mexico.

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TABLE 1 Some Characteristics of Plutonium Study Areas at Los Alamos and Trinity, New Mexico

	Mortandad	Acid-Pueblo	Area GZ	Area 21
Annual precipitation, cm	43 to 52	39 to 54	12 to 25	30 to 40
Average annual temperature, °C	7.5	7.1	15	12
Range	-26 to 36	-23 to 31	-5 to 39	-4 to 38
Soil	Sandy alluvium	Sandy alluvium	Sandy loam	Loam
Soil pH	8.6 to 9.2	7.1 to 7.9	7.5 to 8.4	8.2 to 8.4
Soil cation exchange capacity, equivalents/kg soil	0.06 to 0.09	0.05 to 0.10	0.02 to 0.02	0.02 to 0.03
Soil organic carbon, %	0.10 to 0.40	0.04 to 0.54	0.40 to 0.70	0.75 to 1.5
Clay mineralogy	Amorphous	Amorphous	Mixed	Mixed
Plutonium source	Industrial liquid effluent	Industrial liquid effluent	Weapon fallout	Weapon fallout
Soil $^{239,240}\text{Pu}/^{238}\text{Pu}$ concentration ratio, (pCi/g)/(pCi/g)	0.35*	100*	12†	21†
Weathering time of plutonium in environment (yr) as of 1973, the year most of the data in this paper were collected	0 to 11	14 to 30	28	28

*See Miera et al. (1977); Nyhan, Miera, and Peters (1976a).

†See Neher and Bailey (1976).

received an estimated 173 mCi of plutonium; Mortandad Canyon has been used for the least amount of time (from 1963 to present) and currently receives most of the Laboratory's liquid waste plutonium. As of 1973 and 1974, the years from which data in this chapter were collected, Mortandad Canyon had received about 61 mCi of plutonium.

Surface water exists in the upper reaches of both canyons as a result of Laboratory effluents and/or domestic sewage-treatment effluent; the lower portions of the canyons are normally dry. Surface water, including the pulse releases of plutonium-contaminated liquid effluent, rapidly percolates into the alluvium and generally disappears about 1 km downstream. Relatively large flows occur in both canyons during storm runoff events. Storm runoff reaches the Rio Grande via Acid-Pueblo and Los Alamos Canyons (Fig. 1), but the runoff water in Mortandad Canyon rapidly soaks into the thick alluvial deposits and seldom reaches postoutfall distances beyond 3 km. Many rainstorms at Los Alamos are intense, of short duration, and result in dramatic flash floods in the canyons.

Trinity

Trinity Site and the associated fallout zone is located in the northern end of the Tularosa basin in south central New Mexico (Fig. 1). The region is characterized (Table 1) by low rainfall (12 to 40 cm), high summer temperatures (commonly greater than 37°C), and severe wind and water erosion on exposed and disturbed ground surfaces. Rainfall

accounts for about 90% of the annual precipitation. The area supports a relatively dense vegetation cover, considering the region; total vegetative ground cover ranges from about 15 to 25% (Neher and Bailey, 1976).

On July 16, 1945, a 20-kt atomic bomb was detonated 31 m above the ground surface at Trinity Site during a relatively unstable climatic regime when winds were to the northeast and were accompanied by intermittent thundershowers. Fallout from the cloud deposited in a northeast direction in the general pattern outlined in Fig. 1 (Larson et al., 1951). Relatively high fallout deposition occurred on Chupadera Mesa about 35 to 55 km from the crater. The reasons for the heavy deposition in this zone are unknown but may be related to weather or topographic factors. The elevation increases from about 1500 m at the crater to 2100 m on Chupadera Mesa. The fallout zone within 15 km of the crater is on the White Sands Missile Range, which is under U. S. Army jurisdiction. Beyond 15 km the fallout zone is on mixed private and public (Bureau of Land Management, State; and U. S. Forest Service) lands that are used heavily for domestic livestock grazing.

Plutonium Distribution

General

The chronic release of treated liquid effluents to the Los Alamos canyons has resulted in soil plutonium concentrations that are generally much higher than those at Trinity Site. Concentrations of a few hundred picocuries per gram (dry weight) are found in soils from the canyons, whereas those in Trinity soils average less than 1 pCi/g (Table 2). Worldwide fallout concentrations of ^{239,240}Pu in Los Alamos and Trinity Site soils average about 0.01 pCi/g (Apt and Lee, 1976; Nyhan, Miera, and Neher, 1976b).

**TABLE 2 Ranges in Plutonium Concentration and Variability
Estimates in Some Los Alamos and Trinity Ecosystem
Components in 1973 and 1974**

Component*	Los Alamos	Trinity
Soil (0 to 15 cm)		
pCi Pu/g	1-290†	0.02-0.32
CV‡	0.32-2	0.52-0.88
nCi Pu/m ²	190-80,000	2.8-63
Vegetation		
pCi Pu/g	0.08-76	0.002-0.37
CV‡	0.65-2.2	0.38-1.1
pCi Pu/m ²	0.7-600	0.07-5
Rodents		
fCi Pu/g	7-300	3-100
CV‡	0.16-1.3	0.52-1.3
fCi Pu/m ²	0.2-20	0.03-2

*Dry-weight concentrations.

†Includes ²³⁸Pu and ^{239,240}Pu.

‡Coefficient of variation (CV = standard deviation/mean).

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Vegetation at both study locations contains the highest plutonium concentrations of any biotic component yet examined (Hakonson and Bostick, 1976). Plutonium concentrations in native grasses and forbs ranged from 0.08 to 76 pCi/g (dry weight) at Los Alamos and from 0.002 to 0.37 pCi/g in the Trinity fallout zone; levels in vegetation generally do not exceed those in corresponding soil samples. Additionally, the highest plutonium concentrations were associated with plants growing closest to the ground surface; taller growth forms, such as shrubs and trees, contained the lowest concentrations (Hakonson and Bostick, 1976; Hakonson and Johnson, 1974).

Plutonium concentrations in rodents, as representatives of the primary consumer trophic level, reflect the low physiological availability of the element. Pooled samples of internal organs from rodents generally do not contain measurable levels of plutonium, even though habitat soils may contain up to a few hundred picocuries per gram. Plutonium concentrations in whole rodents ranged from analytical detection limits of about 5 fCi/g to a few hundred femtocuries per gram; most of this radioactivity was associated with samples of pelt and gastrointestinal (GI) tract and contents.

Plutonium concentration variability, as characterized by the coefficient of variation in soils, plants, and animals, was uniformly high at all study sites. It commonly varied up to 2.0, with extreme values approaching 3.0 (Hakonson and Bostick, 1976; Nyhan, Miera, and Neher, 1976b). Variability of this magnitude has been observed at several environmental plutonium study sites in the United States (Little, 1976; Gilbert and Eberhardt, 1976) and results in the need for very large sample sizes in field experiments (Gilbert and Eberhardt, 1976; White and Hakonson, 1978).

Soils

Horizontal Distribution. Horizontal plutonium concentration gradients are evident in both study areas, reflecting the dispersion from point sources of plutonium. Concentrations in the Los Alamos stream channels decrease one to two orders of magnitude in a predictable fashion (Nyhan, Miera, and Peters, 1976a; Hakonson and Bostick, 1976) within 10 km of the effluent sources, whereas similar differences occur over much greater distances at Trinity and do not necessarily decrease with distance. For example, plutonium concentrations in Trinity soils gradually increase from a minimum just outside the crater to a maximum at about 50 km from the crater (Nyhan, Miera, and Neher, 1976b; Larson et al., 1951).

Liquid effluent radionuclides at Los Alamos have been transported laterally into the stream banks as well as to downstream areas. Stream-bank soils accumulate radionuclides to levels equivalent to adjacent channel soil (Anonymous, 1977), and they serve as a long-term source of these materials to stream-bank biota. The stream banks, which are heavily vegetated, retard the downstream movement of radionuclides since they are not subject to the severe erosion encountered in the channel.

Although plutonium concentrations average much higher in the canyons than at Trinity, the extent of the contamination in the canyons is confined to less than 0.1 km², whereas the low-level contamination at Trinity Site covers several thousand square kilometers. Consequently the ecosystems at risk at Los Alamos are exposed to higher concentrations of plutonium than those at Trinity; however, the areas involved are smaller, and corrective action could be taken more easily should it ever be necessary.

Vertical Distribution. Some data from Area 21 (see Fig. 1) at Trinity Site indicate that the plutonium originally deposited on those environs in 1945 has been depleted from the soil surface over a 23-yr period (Table 3). Area 21 soils contained about 700 nCi/m² in 1950 (Olafson, Nishita, and Larson, 1957) and 18 nCi/m² in 1973 (Nyhan, Miera, and Neher, 1976b).

The depletion of plutonium from the soil surface is primarily due to the vertical transport of the element into the soil profile rather than to horizontal transport away from the study site by wind or water. Evidence that plutonium has migrated into the soil profile at the two Trinity Site locations is illustrated in Table 4 and is presented in detail by Nyhan, Miera, and Neher (1976b). In 1973 plutonium was detected at the 28- and 35-cm depths at Areas GZ and 21, respectively, whereas in 1950 plutonium was confined exclusively to the surface 2.5 cm (Olafson, Nishita, and Larson, 1957). The patterns of distribution with depth were typical of those observed in terrestrial soils in that plutonium concentrations decreased with depth.

TABLE 3 Comparison of Plutonium Concentrations in Surface (0 to 2.5 cm) Soils from Chupadera Mesa as a Function of Time After the Atomic Bomb Test at Trinity Site in 1945

Plutonium concentration, nCi/m ²		
1950*	1951*	1973†
746(0.31)‡ n = 6	341(0.82)‡ n = 3	18(0.48)‡ n = 8

*Data for 1950 and 1951 from Larson et al. (1951), and Olafson, Nishita, and Larson (1957).

†Data for 1973 from Nyhan, Miera, and Neher (1976b).

‡Parenthetic value is coefficient of variation (CV = standard deviation/mean).

TABLE 4 Mean Percent Plutonium Inventory in Soil Profiles from Los Alamos and Trinity Site Study Locations in New Mexico

Trinity Site*			Los Alamos*		
Depth, cm	Area GZ	Area 21	Depth, cm	Mortandad	Acid-Pueblo
0-2.5	29(0.78)‡	41(0.46)‡	0-2.5	20(0.44)‡	4.0(0.76)‡
2.5-5.0	18(0.72)	19(0.63)	2.5-7.5	36(0.23)	10(0.48)
5-10	21(0.81)	6.0(0.88)	7.5-12.5	21(0.55)	20(1.3)
10-15	15(0.67)	8.0(0.92)	12.5-30	24(0.79)	67(0.18)
20-25	17(1.3)	16(1.0)			
25-33	ND‡	10(1.2)			

*n = 8 for Trinity Site data; n = 10 for Los Alamos data.

‡Parenthetic value is coefficient of variation (CV = standard deviation/mean).

‡Not detectable.

The depth of plutonium transport into channel and bank soil profiles in the Los Alamos canyons is much greater than that at Trinity. In areas where permanent surface water exists (i.e., Mortandad Canyon), elevated plutonium concentrations are found at depths of 100 cm in the channel and at depths of 50 cm in the stream bank. Plutonium concentrations in channel soils do not show any consistent patterns with sampling depth, whereas decreasing concentrations with depth are evident in bank soils. In downstream areas, which are dry except during periods of storm runoff, plutonium occurs at depths of at least 30 cm (Nyhan, Miera, and Peters, 1976a).

The transport of plutonium into the channel alluvium and stream-bank soil has been rapid, as shown by the presence of elevated ^{238}Pu at the lower sampling depths. Elevated ^{238}Pu was observed at soil depths of 30 cm in Mortandad Canyon in 1972, about 4 yr after the first significant release of this element to the canyon (Hakonson and Bostick, 1976). In contrast, fallout $^{239,240}\text{Pu}$ in Trinity soils 5 yr after the bomb test was confined to the upper 2.5 cm of soil (Olafson, Nishita, and Larson, 1957).

A common feature of plutonium distribution in soils from both locations was that in 1974 less than one-half the total plutonium in the soil column was present in the surface 2.5 cm (Table 4) despite differences in soils and source of plutonium. In Acid-Pueblo Canyon 10 yr after the decommissioning of those facilities for waste disposal, an average of 67% of the soil column inventory was below the 12.5-cm depth, which reflects depletion of plutonium from the surface layers by vertical and horizontal transport processes. Previous studies in the canyons have shown that horizontal transport of soil during storm runoff events is an important mechanism in the downstream transport of plutonium (Purtymun, 1974; Hakonson, Nyhan, and Purtymun, 1976).

The depletion of plutonium from the soil surface decreases the probability of horizontal transport by wind and water but may increase the probability of uptake by vegetation during the time that the element is distributed within the plant rooting zone. However, over long periods of time, continued movement of plutonium into the soil profile may remove the element from the biologically active zone of the soil.

Particle Size Relationships. The highest concentrations of plutonium in soil at the Los Alamos and Area 21 locations were associated with the silt-clay fraction, whereas this fraction at Area GZ, 1 km from the crater, contained the lowest concentrations of plutonium (Table 5) (Nyhan, Miera, and Neher, 1976b; Nyhan, Miera, and Peters, 1976c). At the GZ location, the highest concentrations were measured in the 1- to 2-mm soil particles, which perhaps reflects the physical characteristics of the fallout debris near the detonation site and/or depletion of the plutonium from smaller size fractions by wind or water transport vectors. Decreasing plutonium particle sizes with increasing distance from the crater were also noted at weapons test sites in Nevada (Romney, 1977).

The inventory of plutonium among the various soil size fractions in surface soils at the Los Alamos and Area GZ Trinity study sites was similar in that the silt-clay size fraction ($<53\ \mu\text{m}$) comprised less than 10% of the soil mass and contained less than 15% of the plutonium (Table 5), whereas over 80% of the plutonium was associated with soil particles greater than $53\ \mu\text{m}$ (Nyhan, Miera, and Neher, 1976b; Nyhan, Miera, and Peters, 1976c). The reverse was true at Area 21, Trinity Site, where the $<53\text{-}\mu\text{m}$ fraction comprised 36% of the soil mass and contained over 70% of the soil plutonium inventory.

TABLE 5. Comparative Distribution of Plutonium in Surface Soil (0 to 2.5 cm) Size Fractions at the Los Alamos and Trinity Study Areas

	Soil size fraction*					
	<53 μm*	53-105 μm	105-500 μm	500-1000 μm	1-2 mm	2-23 mm
Mortandad Canyon						
pCi Pu/g†	1500.0	1300.0	610.0	310.0	87.0	69.0
Soil weight, %	2.2	1.8	14.0	21.0	26.0	35.0
Pu in fraction, %	14.0	6.0	27.0	21.0	16.0	16.0
Acid-Pueblo Canyon						
pCi Pu/g†	85.0	60.0	25.0	8.8	7.9	25.0
Soil weight, %	3.0	3.0	16.0	26.0	28.0	24.0
Pu in fraction, %	7.0	7.0	31.0	19.0	17.0	19.0
Trinity Site, Area GZ						
pCi Pu/g†	0.07	0.05	0.92	2.1	5.3	0.01
Soil weight, %	8.9	11.0	49.0	23.0	6.1	2.0
Pu in fraction, %	0.78	0.43	36.0	38.0	25.0	0.01
Trinity Site, Area 21						
pCi Pu/g†	3.8	1.7	0.42	0.64	1.6	0.23
Soil weight, %	36.0	18.0	25.0	4.2	2.9	14.0
Pu in fraction, %	73.0	16.0	5.5	1.4	2.4	1.8

*Size fraction data based on composite samples.

†Pu denotes primarily ²³⁹Pu in Mortandad Canyon and ^{239,240}Pu at all other study locations.

Vegetation

Plant-Soil Relationships. The concentrations of plutonium in the study area vegetation were related to the levels of plutonium in associated soils (Fig. 2). The relationship between plutonium concentrations in vegetation and in soils was predictable over a range of five orders of magnitude in concentrations; this relationship is similar to relationships that were observed in the Rocky Flats environs (Little, 1976).

Plant-soil plutonium concentration ratios (CR = picocuries per gram of vegetation/picocuries per gram of soil) are a convenient means of estimating the plutonium levels in vegetation growing on contaminated soils. Ratio estimates for native grasses in the Los Alamos and Trinity Site study areas (Table 6) ranged from 0.05 to 1.2, whereas the values for forbs ranged from 0.04 to 1.1. All these ratios are high relative to those derived from experimental studies where root uptake was the contamination mechanism (Romney and Davis, 1972; Wilson and Cline, 1966), which indicates that either plutonium is much more available to plants under field conditions or that mechanisms other than root uptake are responsible for the plutonium measured in plant samples from the field.

The relative amounts of plutonium associated with the internal and external portions of the vegetation are difficult to assess under field conditions, although we contend that

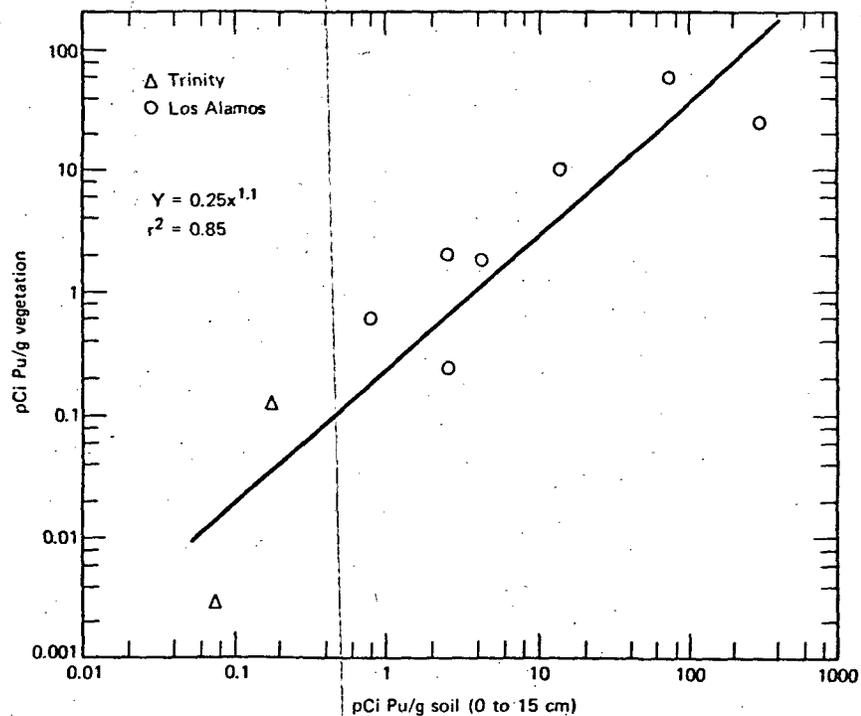


Fig. 2 Relationship of average plutonium concentration in herbaceous vegetation (grasses and forbs combined) and in corresponding soils in Los Alamos and Trinity Site study areas.

TABLE 6 Plutonium Concentration Ratios for Vegetation and Associated Soils from Los Alamos and Trinity Study Sites

Component	Plutonium concentration ratio*							
	Los Alamos				Trinity Site			
	n	Mortandad Canyon	n	Acid-Pueblo Canyon	n	Area GZ	n	Area 21
Grass	24	0.93(0.94)	19	0.13(1.2)	13	0.05(0.54)	16	1.2(0.74)
Forb	16	0.31(1.3)	11	0.23(0.28)	17	0.04(0.97)	21	1.1(0.92)

*Ratio calculated as $\{pCi Pu/g (dry weight) plant\} / \{pCi/g (dry weight) soil (0 to 15 cm depth)\}$. Parenthetic value is coefficient of variation (CV = standard deviation/mean).

most of the plutonium in our study areas is externally deposited on plant surfaces. Information supporting this conclusion includes:

- The high plant/soil plutonium concentration ratios compared to greenhouse studies.
- The obvious presence of soil in vegetation samples.

In addition, other investigators have shown that some of the plutonium associated with native vegetation samples can be removed by a wash treatment (Aldredge, Arthur, and Hiatt, 1977).

Rodents

Rodent-Soil Relationships. Plutonium in internal organs (i.e., liver, bone, and muscle) of rodents sampled within our study areas generally could not be measured. However, concentrations of plutonium in pelt and GI tract samples were readily measured and were

TABLE 7 Inventory of Plutonium in Small Mammal Tissues from Mortandad Canyon

Tissue	Percent of total body weight	Total plutonium,* pCi/g	Percent total plutonium
Pelt	23	0.85	50.0
GI tract	10	1.8	46.0
Lung	2	0.034	0.02
Liver	5	0.035	0.5
Carcass	60	0.018	2.8

*Based on six pooled samples.

directly correlated with levels in the study area soils ($r^2 = 0.90$). Over 95% of the plutonium body burdens in rodents was associated with these two tissues, as shown by the data for Mortandad Canyon in Table 7. Thus we conclude that, in our study areas, physical and biological processes (i.e., contamination of the pelt or ingestion of soil) dominate in the transport of plutonium to rodents.

Plutonium Inventories

The fractional distribution of plutonium in Los Alamos and Trinity ecosystem components (Table 8) is based on quantitative estimates of ecosystem component mass (grams per square meter) and corresponding plutonium concentrations (picocuries per square meter) in those compartments. The distribution of plutonium among five components was generally quite similar between sites in that over 99% of the plutonium was associated with soil and less than 1% with biota. Live vegetation contained 10^{-6} to 10^{-8} % of the plutonium inventory. We conclude that very little of the environmental plutonium present in our study areas has appeared in the biological components of the ecosystem even after 30 yr of exposure. These results are essentially the same as those observed at Rocky Flats and Oak Ridge (Little, 1976; Dahlman, Garten, and Hakonson, this volume).

000

ion site

soils

Area 21
1.2(0.74)
1.1(0.92)
depth).

TABLE 8 Plutonium Inventory Ratios for Some Components of Los Alamos and Trinity Study Areas in New Mexico

Component*	Plutonium inventory ratio*							
	Los Alamos				Trinity			
	n	Mortandad Canyon	n	Acid-Pueblo Canyon	n	Area GZ	n	Area 21
Grass	24	4.1×10^{-5} (0.90)	20	5.6×10^{-4} (1.6)	13	2.0×10^{-3} (0.99)	16	1.3×10^{-4} (0.76)
Forb	16	4.8×10^{-5} (1.2)	11	1.7×10^{-4} (1.4)	17	1.7×10^{-4} (1.0)	21	3.5×10^{-5} (0.77)
Litter					5	1.6×10^{-4} (2.0)	3	1.1×10^{-4} (0.81)
Rodents	33	1.5×10^{-9} (0.77)	48	4.5×10^{-10} (0.99)	40	3.7×10^{-9} (1.7)	20	2.3×10^{-9} (0.47)
Soil	29	0.99(0.00009)	23	0.99(0.001)	8	0.99(0.0003)	8	0.99(0.00008)

*Inventory ratio = (pCi Pu/m² in component)/(total pCi Pu/m²). All plutonium values are ^{239,240}Pu except Mortandad Canyon which is ²³⁸Pu; parenthetic value is coefficient of variation (CV = standard deviation/mean).

The relative inventory of plutonium within all our study ecosystems is governed primarily by component mass relationships since differences in mass of the various ecosystem components are much greater than the differences in plutonium concentrations between the same components. The data in Table 9 demonstrate that mass inventory ratios for Mortandad Canyon provide a good approximation of the plutonium inventory ratio.

TABLE 9 Mass and Plutonium Inventory Ratios in Mortandad Canyon at Los Alamos

Component	Component mass, g/m ²	Mass inventory ratio	Plutonium inventory ratio*
Soil (0 to 15 cm)	2.3×10^5	0.999	0.999
Grass	20.0	9.0×10^{-5}	4.1×10^{-5}
Forb	10.0	4.4×10^{-5}	4.8×10^{-5}
Rodents	0.03	1.3×10^{-7}	1.5×10^{-9}

*Data from Table 8.

Plutonium Transport

Soils

Rainstorm runoff in the intermittent streams receiving wastes was identified over 30 yr ago in the environmental transport of plutonium (Kingsley, 1947). Additional studies were begun to determine the relationships of rainfall, runoff, and suspended sediments with radionuclide transport (Purtymun, 1974; Purtymun, Johnson, and John, 1966; Hakonson, Nyhan, and Purtymun, 1976).

Results of these studies demonstrate that runoff from snow melt and summer rainstorms serves as a radionuclide transport vector in Los Alamos intermittent streams and that the magnitude of this transport is highly dependent on the hydrologic

characteristics of the watershed and the intensity of runoff flow (Purtymun, 1974; Hakonson, Nyhan, and Purtymun, 1976). The dependency of concentrations of suspended sediments and plutonium in runoff on flow rate is indicated in Fig. 3 for one rainstorm runoff event in Mortandad Canyon. The nonlinearity in the curve is due to the relationship of flow rate with the particle size of resuspended material. At flows less than 0.25 m³/sec, only the silt-clay size materials were in suspension in the runoff. However, at flows greater than 0.25 m³/sec, coarser sands containing most of the sediment plutonium inventory (Table 5) were resuspended, which resulted in increased suspended sediment and radionuclide concentrations. High flow rates typically occur during the early phases of runoff events at Los Alamos owing to the intense nature and short duration of area rainstorms. We found that nearly 80% of the sediment and 70% of the radioactivity was transported within the first half of such events.

Additionally, there was a highly significant ($P < 0.01$) relationship between suspended sediment and radionuclide concentrations in runoff water. About 99% of the radioactivity in runoff was associated with suspended sediments greater than 0.45 μm in diameter, whereas only 1% of the radioactivity in the liquid phase was associated with sediments less than 0.45 μm in diameter.

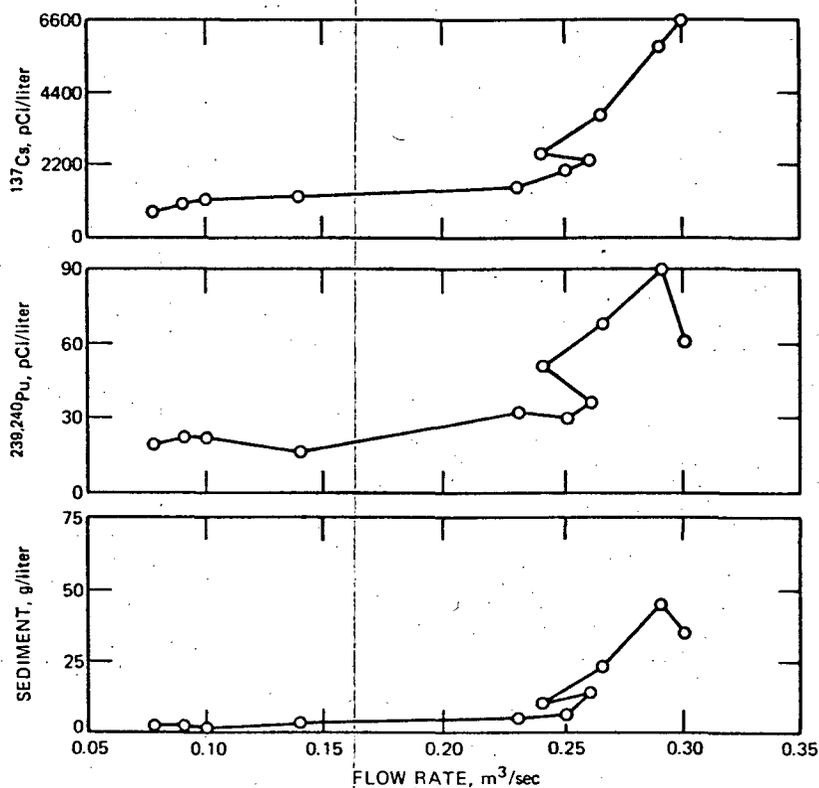


Fig. 3 Concentration of sediment and radioactivity in unfiltered runoff water from Mortandad Canyon as a function of runoff flow rate.

Area 21
 < 10⁻⁴ (0.76)
 < 10⁻⁵ (0.77)
 < 10⁻⁴ (0.81)
 < 10⁻⁹ (0.47)
 (0.00008)

²⁴⁰Pu except
 (n/mean).

governed
 by various
 concentrations
 inventory
 inventory

over 30 yr
 of studies
 sediments
 1, 1966;

summer
 streams
 hydrologic

Studies were recently begun on wind transport of plutonium in the Trinity fallout zone, where evidence of wind erosion of soil is readily apparent. Although these studies are not complete, several important observations have been made. First, soil flux by surface creep and saltation processes is highly seasonal and has peaked in the months of July and August for two consecutive years of observation. Second, soil particle size analyses on dust-collector samples show major differences in the amount of silt-clay material between study sites. About half the dust material at Area 21 is in the silt-clay size range, whereas less than 1% of collected dust at Area GZ is in the silt-clay size range. These differences become important when coupled with the plutonium concentrations in the various soil particle size fractions (from Table 5). For example, silt-clay material in dust collectors at Area 21 contains over 200 times as much plutonium as the silt-clay fraction of dust samples at Area GZ.

Summary and Conclusions

Despite differences in ecosystems and plutonium source, there are several similarities in plutonium distribution between the Los Alamos and Trinity study areas. First, the soils-sediment component contains virtually all the plutonium, with vegetation and rodents containing less than 0.1% of the total. Plutonium has penetrated to considerable soil depths at both locations, although it has occurred much more rapidly and to a greater degree in the alluvial soil at Los Alamos than in the arid terrestrial soils at Trinity. At both locations less than 50% of soil column plutonium inventories was found in the surface 2.5 cm.

The plutonium penetration depth appears to correspond to the moisture penetration depth in the Trinity fallout zone. This is probably the governing factor at Los Alamos, although storm runoff and accompanying turbulent mixing complicate the process. In Acid-Pueblo Canyon, the bulk of the soil column inventory lies in the lower profiles, an indication of the loss of plutonium from surface layers due to sediment transport.

The plutonium in most cases was associated with relatively coarse soil size fractions. The silt-clay (<53 μm) fraction contained relatively little (<15%) of the plutonium, a reflection of the small amounts of this size fraction in study area soils. An exception was in Area 21 at Trinity, where the <53- μm soil size fraction contained about 73% of soil plutonium inventories. The importance of these distributional differences stems from the fact that silt-clay soil particles can be transported farther and are more likely to adhere to biological surfaces than larger size fractions.

Concentrations in herbaceous ground vegetation were generally related to those in soils from all sites. Our data strongly indicate that external contamination of plant surfaces is the major soil-to-plant transport mechanism in these arid systems. The plutonium concentrations in pelt and GI tissues were related to corresponding soil concentrations at all sites. Over 95% of the plutonium body burden in rodents was associated with pelt and GI tract samples, an indication of the dominance of physical and/or biological processes as the contaminating mechanism.

Horizontal transport in both areas is dominated by wind- and water-driven processes. At Los Alamos surface runoff water governs the downstream transport of plutonium; indications are that wind is a relatively more important transport vector at Trinity,

although splash-up of soil by raindrops may be an important transport mechanism in these arid, sparsely vegetated study locations.

There was no evidence for a trophic-level increase of plutonium from soil to vegetation to rodents. We believe that rodents come into contact with environmental plutonium directly from the soil and to a lesser extent through a food-web intermediary.

Research Needs

The importance of the soils component as a receptor of plutonium released to the Los Alamos and Trinity Site study areas coupled with the direct role these soils play in contamination of biota emphasizes the importance of understanding soil formation and transport processes. Factors governing these processes will be instrumental in determining plutonium distribution and transport as a function of time. Hydrologic and wind transport processes discriminate against certain soil particle sizes; therefore studies on the relationship of plutonium to soil separates will be useful in evaluating the potential importance of a transport pathway and the resulting hazard. We know, for example, that wind transport of silt-clay material at Area GZ, Trinity Site, would represent a relatively smaller inhalation hazard than corresponding transport at Area 21 simply because the silt-clay fraction of Area GZ soil contains very little of the plutonium inventory.

Factors affecting migration of plutonium into the soil profile require understanding since depletion of plutonium from the soil surface will likely reduce the horizontal transport potential and may alter the availability of the element to vegetation.

Field studies should be conducted to quantify the relative importance of the root pathway for contaminating vegetation to serve as a basis for judging changes in physiological availability of environmental plutonium with time. As yet few field studies have been able to show conclusively the relative importance of the two contamination mechanisms.

In our opinion studies should be continued on the availability of plutonium to native animals in our study ecosystems; however, on the basis of present concentrations and the high variability associated with these measurements, we believe that the frequency of sampling should be drastically reduced. Perhaps sampling at intervals of 5 to 10 yr would be adequate to judge whether significant changes in plutonium availability have occurred.

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