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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

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.

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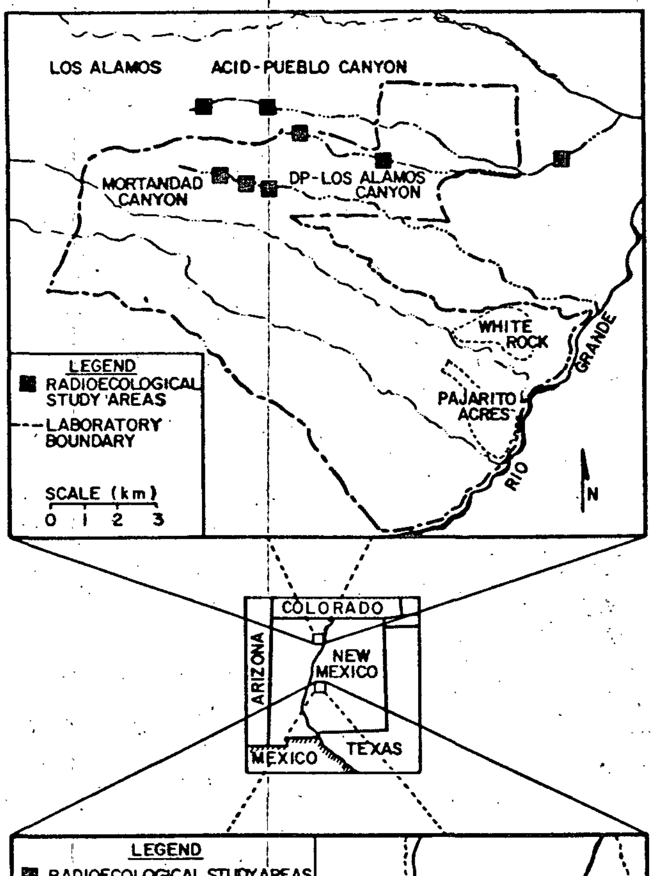

accounts for about 90% of the annual precipitation. The area supports a relatively dense vegetation cover, considering the region; total vegetative ground cover ranges from about 15 to 25% (Neher and Bailey, 1976).

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/m²	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/m²	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/m²	0.2–20	0.03–2

*Dry-weight concentrations.
†Includes ^{238}Pu and $^{239,240}Pu$.
‡Coefficient of variation (CV = standard deviation/mean).

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

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/m^2 in 1950 (Olafson, Nishita, and Larson, 1957) and 18 nCi/m^2 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}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

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 (Alldredge, 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

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

	Plutonium inventory ratio*							
	Los Alamos				Trinity			
Component*	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^{-5}(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^{-8}(1.7)	20	2.3×10^{-9}(0.47)
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

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 m^3/sec, only the silt–clay size materials were in suspension in the runoff. However, at flows greater than 0.25 m^3/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.

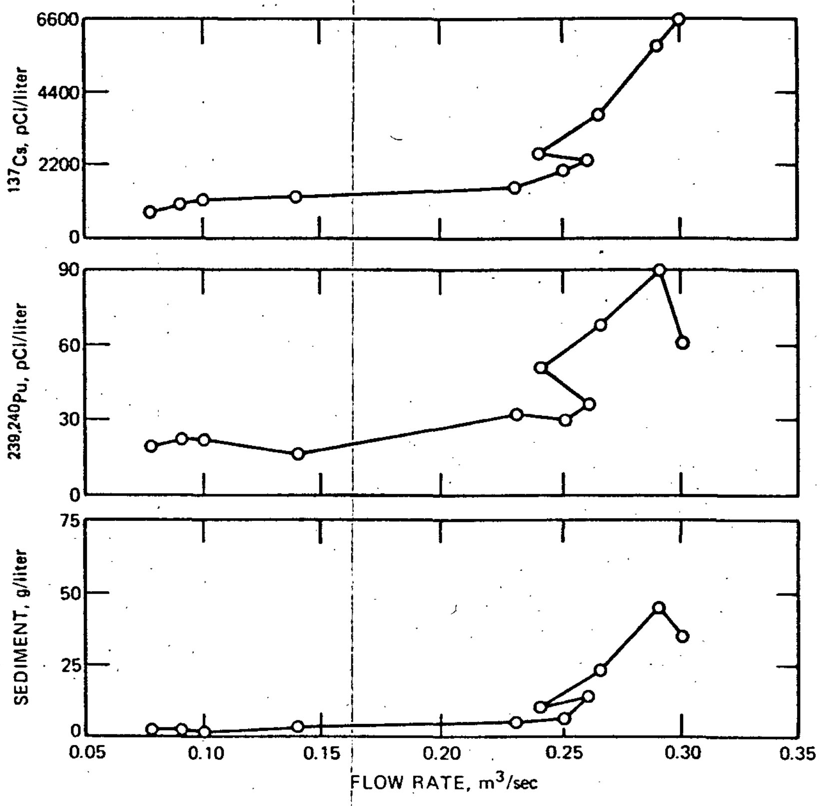

Fig. 3 Concentration of sediment and radioactivity in unfiltered runoff water from Mortandad Canyon as a function of runoff flow rate.

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