Plutonium in a Grassland Ecosystem

CRAIG A LITTLE

This chapter is primarily concerned with plutonium contamination of grassland at the U S Department of Energy Rocky Flats plant, which is located northwest of Denver Colo Major topics include the definition of major plutonium containing ecosystem compartments, the relative amounts in those compartments whether or not the predominant isotopes, ^{238}Pu and ^{239}Pu, behaved differently, and what mechanisms might have allowed for the observed patterns of contamination

Samples of soil, litter, vegetation, arthropods, and small mammals were collected for plutonium analysis and mass determination Small aliquots (5 g or less) were analyzed by a rapid scintillation technique and by alpha spectrometry

Of the compartments sampled, greater than 99% of the total plutonium was contained in the soil The concentrations of plutonium in soil were significantly inversely correlated with distance from the contamination source, depth of sample, and particle size of the sieved soil samples The soil data suggested that the distribution of contamination largely resulted from physical transport processes

Concentrations of plutonium in litter and vegetation were inversely correlated to distance from the source and directly correlated to soil concentrations at the same location. Comparatively high concentration ratios of vegetation to soil suggested wind resuspension of contamination as an important transport mechanism

Arthropod and small-mammal tissue samples were highly skewed, kurtotic, and quite variable Plutonium concentrations were lower in bone than in other tissues Hide, gastrointestinal tract, and lung were generally not higher in plutonium concentration than kidney, liver, and muscle. All data tended to indicate that physical transport processes were the most important

Median isotopic ratios of ^{239}Pu to ^{238}Pu by activity concentration in soil were 40 to 50 Litter and vegetation isotopic ratios were similar to those of soil Arthropod and small-mammal isotopic ratios were lower than those of soil, which implied that the two isotopes were differentially incorporated into the animal bodies and ^{238}Pu was taken up at a higher rate However, further investigations suggested that statistical bias may have spuriously contributed to the lower isotopic ratios in small animals

Most of the world's agriculture occurs on land that, before tilling, was once covered by stands of grasses An important untilled tract of land contaminated with plutonium* is the grassland immediately adjacent to and contained within the Rocky Flats plutonium processing plant and associated buffer zone about 12 km northwest of Denver, Colo , metropolitan area Because Rocky Flats is a prime example of plutonium-contaminated

*The word "plutonium" indicates ^{239,240}Pu in this chapter, unless otherwise noted

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grassland, this chapter will dwell primarily on data from environmental sampling at Rocky Flats

The Rocky Flats installation uses nearly 30 km^2 as a buffer zone to separate the public from plutonium-handling operations The climate at the installation is typified by occasional strong WNW winds exceeding 40 m/sec and moderate precipitation i e , 40 cm/yr average (Rocky Flats 1975 annual weather summary unpublished) The Rocky Flats grassland has been modified by the activities of humans and includes plant species typical of short-grass plains (*Bouteloua gracilis* and *Buchloe dactyloides*) as well as tall-grass prairie (*Agropyron* spp and *Andropogon* spp) and ponderosa pine (*Pinus ponderosa*) woodland (Weber, Kunkel, and Shultz, 1974) Mule deer (*Odocoileus hemionus*) are found on the site along with grassland species of reptiles rodents, and birds (Whicker, 1974)

Source of the Contamination

Investigations by Krey and Hardy (1970) of DOE's Environmental Measurements Laboratory (EML, formerly Health and Safety Laboratory) suggested that the most likely contamination source was a storage area of stacked 55-gal barrels that leaked plutonium-polluted oil Data supporting the conclusion of Krey and Hardy and a description of the nature of the stored oil—plutonium mixture are delineated

Air-sampling data from Rocky Flats link the barrel storage area to the east-southeast contamination pattern discovered by Krey and Hardy (1970) Air-sampling station S-8, one of many such stations maintained and sampled regularly by Rocky Flats personnel, is located about 75 m east and slightly south of the barrel storage area Except for a brief period during 1961, monthly averages of daily airborne contamination values have been kept since at least 1960 to the present (Fig 1)

The S-8 air-sampling data indicated that contamination peaks in the air were associated with dates of perturbation of the contaminated surface (Table 1 and Fig 1) Except for periods of disturbance, the gross alpha concentrations in ambient air were near 0 01 pCi/m^3 However, during excavation and paving of the barrel storage area the alpha concentration in air markedly increased (Table 1)

The plutonium-contaminated cutting oil about as viscous as lightweight motor oil but thinned by carbon tetrachloride, was stored in the 55-gal barrels for periods of up to 7 yr The interactions between the oil, air, CCl_4, and plutonium within the barrels were probably quite important in determining the eventual fate of the element

The oil was filtered through 2- to 3-μm filters before being placed in the barrels The discard limit at the time of storing was 1 x 10^{-2} g of plutonium per liter of oil If the limit and the filtering had been observed and performed faithfully, each of the approximately 3570 plutonium-containing barrels would have had no more than 2 1 g of plutonium (0 13 Ci) (M A Thompson, Environmental Sciences, Rocky Flats Plant and F J Miner, Chemical Resources, Rocky Flats Plant, personal communications)

It is difficult to assess what occurred once the oil was inside the barrels The presence of carbon tetrachloride in the drums allows the possibility that hydrochloric acid was formed, which, in turn may have reacted with the plutonium metal to form very low concentrations of plutonium chloride, a more-soluble form of the element (J M Cleveland, Environmental Studies, Rocky Flats Plant personal communication) This possibility is given credibility by the work of J Navratil of Rocky Flats Chemical Research Division, who has studied contaminated cutting oil in recent years

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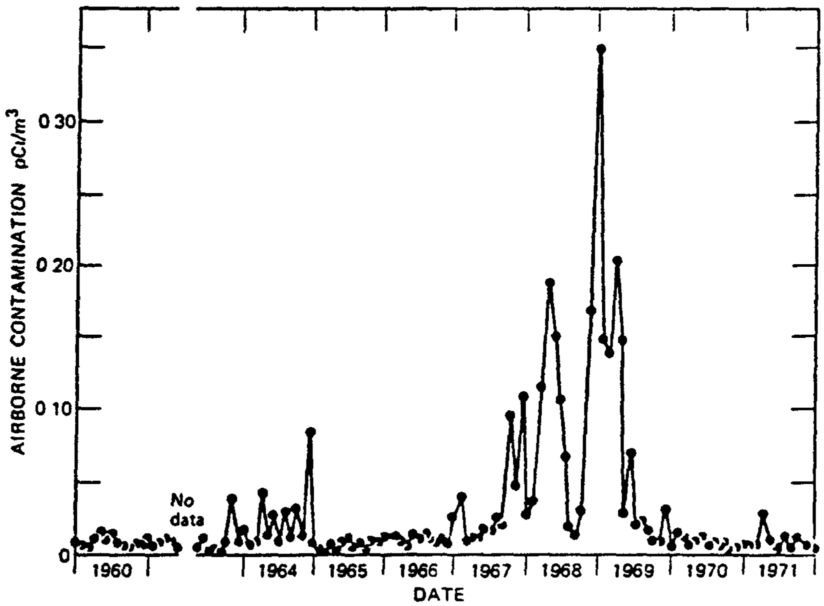

Fig. 1 Monthly means of daily gross alpha activity in ambient air at station S-8 (75 m east of the oil-barrel storage area) Aliquots of Gelman AE filter material were counted in a gas-flow proportional detector Data adapted from D C. Hunt, Environmental Sciences, Rockwell International, personal communication

TABLE 1 Total Monthly Gross Alpha Activity in Ambient Air at Station S-8 (75 m East of Oil-Barrel Storage Area) During Disturbances of the Storage-Area Surface*†

Dates	Event	Alpha activity, pCi/m³
7/59–9/63	No large-scale leaking	0 009
1/64–1/65	Large-scale leaking	0 025
1/65	Contaminated soil covered with fill	0 01
1/66	Small building added to filter contaminated oil from leaking to new drums	0 014
1/67	Drum-removal activity begun	0 038
6/68	Last drums removed but high winds spread some activity	0 188
2/69	Weeds burned and area graded for paving	0 34
9/69	Asphalt pad completed	0 013
11/69	Four sampling wells dug through pad	0 033
4/71	Drainage ditch dug on west side of asphalt pad	0 033

*Air-filter material was counted directly in a gas flow proportional counter

†Adapted from D C Hunt, Environmental Sciences, Rockwell International, personal communication.

Navratil and Baldwin (1976) found that filtering the contaminated oil through a 0 01-μm filter removed only about 50% of the plutonium This result strongly suggested that about half the plutonium was in a relatively large particulate form whereas the other half was associated with very small particles Fission-track analysis of the filtered oil confirmed that the remaining plutonium was monomeric It is doubtful that the barrels consistently held the above proportions of particulate and nonparticulate plutonium oxide, but each probably contained some plutonium chloride

J M Cleveland (personal communication) also suggested that the filtered 3-μm plutonium particles might combine to form larger aggregates of the metal Of course, the size and binding tenacity of these conglomerates are unknown

Methods

Two macroplots were chosen for intensive sampling of plutonium in soil, vegetation, and litter The locations of these macroplots relative to the supposed plutonium source, the barrel storage area, and the prevailing wind are shown in Fig 2 A sampling grid was superimposed over each macroplot The macroplot 1 grid was approximately 0 75 ha

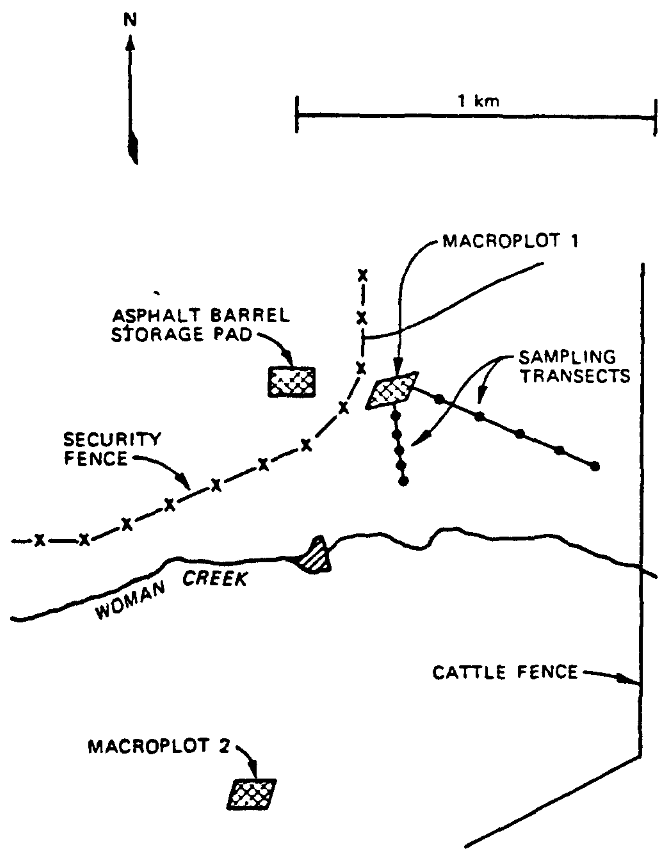

Fig. 2 Schematic representation of the southeast corner of the Rocky Flats Plant indicating the location of study macroplots and sampling transects. The wind rose indicates the relative magnitudes of wind velocities during 1974

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(1 ha = 2.5 acres) and contained 100 sampling markers. Plutonium data in this chapter are from macroplot 1 unless specifically noted otherwise.

Depth profile soil samples were taken by hand with a trowel. After vegetation and litter had been clipped and bagged separately, four 5 by 5 by 3-cm samples were removed and bagged separately for each of seven 3-cm-depth layers to 21 cm deep. If rocks precluded sampling at a given depth, the column was resumed below the blockage. Soil samples were transported to the laboratory and air-dried. Rocks or debris greater than 0.5 cm in diameter were removed from the sample. After oven-drying and weighing, samples were mechanically shaken on brass soil sieves. The accumulation on each sieve was weighed and placed in a small paper envelope, and the envelope was sealed with tape.

Litter and standing vegetation were sampled from 0.25 m^2 and 0.5 m^2 areas, respectively. Vegetation was clipped and bagged, and litter was gathered by hand and bagged. In the laboratory litter and vegetation samples were air-dried and weighed. Soil was separated from the litter samples by a flotation process (Little, 1976). The net vegetation or the litter dry weight was divided by the microplot size, 0.5 m^2 or 0.25 m^2 respectively, to calculate mass per unit area. For plutonium analysis vegetation and litter samples were ground on a Wiley mill with an 850 μm opening screen, and 5-g aliquots were taken.

Arthropods were sampled by a combination of sweep netting, pitfall trapping, and drop trapping at random sites on established grids. At the laboratory animals were separated into generic groups that were weighed separately. These generic totals were combined for an estimated weight per 0.5-m^2 microplot. Arthropods obtained by the drop-trap method were not analyzed for plutonium owing to fear of cross contamination from soil during the vacuuming process. Samples for plutonium analysis and a species inventory list resulted from the sweep netting and pitfall traps. For plutonium analysis a representative composite was analyzed for each sampling period. Arthropods were not cleaned prior to plutonium analysis.

Small mammal trapping grids were superimposed over each macroplot in a manner resembling that used by the U. S. International Biological Program Grassland Biome (Packard, 1971). Animals were trapped about six times yearly. Sherman live traps were used for cricetid and sciurid rodents. Geomyid rodents were trapped less regularly in homemade live traps placed in burrows systems. Approximately 15% of the estimated population was removed from each macroplot during each trapping session. These animals were collected by removing dead-in-trap individuals during the regular session and the remainder randomly in one extra trapping night. Small mammals were either dissected immediately or frozen for dissection later. Special precautions were taken during dissection to minimize cross contamination between tissues. Approximately 10 cm^2 of hide was used as an aliquot. Lungs, liver, and gastrointestinal (GI) tract were removed intact. Muscle samples were taken from the legs in most cases. Bone samples consisted of the whole skeleton, which had been cleaned of flesh by a dermestid beetle colony. All samples except bone were placed on tared, ashless filter papers, oven-dried at 50 to 60°C, and weighed. The sample was placed in a snap-cap vial for storage or transport to a commercial laboratory.

Some soil-sample aliquots (5 g) were analyzed for plutonium content by commercial laboratories (LFE, Richmond, Calif., and Eberline Instrument Corp., Albuquerque, N. Mex.). Most soil samples were analyzed in our laboratory, as were most litter and vegetation samples. Small-mammal tissues and arthropods were commercially analyzed. The LFE method used concentrated hydrofluoric acid to dissolve samples (Wessman

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et al 1971) Eberline modified a pyrosulfate fusion technique for the same purpose (Sill, 1969) Ion-exchange columns removed interfering elements and isolated plutonium from the samples before alpha spectrometry analysis Chemical recovery was measured by adding ^{236}Pu tracer to each sample Agreement between homogenized split samples sent to these laboratories was good (Little, 1976) In our laboratory a procedure was used that incorporated harsh digestion of the sample by nitric and hydrofluoric acids ion exchange organic extraction, and liquid scintillation spectrometry (Little, 1976) This method had an estimated minimum detectable activity of 0 42 pCi ($P < 0.05$) Plutonium data in this chapter are ^{239,240}Pu unless otherwise noted Plutonium 240 contributed about 20% on the average to the alpha activity of ^{239,240}Pu

Plutonium Compartmentalization

The inventories of plutonium in the principal compartments of the grassland ecosystem were calculated Compartments investigated were soil, in 3-cm increments from 0 to 21 cm, litter, standing vegetation, arthropods, and small mammals

The compartmental inventories of plutonium were calculated by multiplying the mean mass of each compartment (g/m^2) by the mean plutonium concentration of the compartment (μCi/g) A total ecosystem inventory was calculated by summing over all compartments The compartmental fraction (unitless) of the total plutonium inventory was calculated by dividing each compartmental inventory (μCi/m^2) by the total inventory (μCi/m^2)

The soil compartment had vastly the largest fraction of the total plutonium, 99 69% (Table 2) As expected, the fraction of the total plutonium contained within a soil layer decreased as the depth increased The litter compartment comprised less than 1% of the total plutonium (53 nCi/m^2) in the study areas, and the vegetation represented only about 0 01% of the total plutonium By virtue of representing both low biomasses and plutonium concentration, the animal compartments arthropods and small mammals, had extremely small fractions of the total plutonium 6.8×10^{-9} and 3.3×10^{-9}, respectively

In summary the compartmentalization data indicated that greater than 99% of the plutonium in the study area was located in the soil At the time of sampling, nearly one-half (49 7%) the total plutonium was in the top 3 cm of soil In decreasing order, smaller plutonium-inventory fractions were found in litter, vegetation, arthropods and small mammals The implication of these results is that, in the present state, transport of plutonium is closely linked to soil movement or erosion Therefore efforts to prevent plutonium transport off contaminated grasslands should be directed primarily at minimizing soil transport rather than mobilization by biota

Plutonium in Soil

Analysis of the soil plutonium data suggested that two primary generalizations about plutonium in soil could be stated First, the plutonium concentrations in the soil samples were highly variable Second, the plutonium concentration in a soil sample was a function of sample location sample depth, and the soil particle composition of the sample Rationales for both of these conclusions are examined in some detail

Frequency distributions of plutonium in soil samples were positively skewed ($P < 0.05$) with coefficients of variation (CV = standard deviation − mean) ranging to

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TABLE 2 Distribution of ^{239}Pu in Samples from the Rocky Flats Study Macroplot*

Compartment	Mean	n†	95% confidence interval‡
Plutonium concentrations, pCi/g			
Soil, 0–3 cm	835	72	554–1120
Soil, 3–21 cm	105	309	69–141
Litter	412	29	314–509
Vegetation	28 6	76	15 7–41 4
Arthropods§	5 48	23	3 13–7 83
Small mammals	6.50	304	2 38–10 6
Fraction of total plutonium			
Soil, 0–3 cm	$5\ 0 \times 10^{-1}$		$2\ 5 \times 10^{-1}$–$7\ 4 \times 10^{-1}$
Soil, 3–21 cm	$5\ 0 \times 10^{-1}$		$2\ 5 \times 10^{-1}$–$7\ 5 \times 10^{-1}$
Litter	$2\ 9 \times 10^{-3}$		$1\ 6 \times 10^{-3}$–$4\ 2 \times 10^{-3}$
Vegetation	$1\ 0 \times 10^{-4}$		$4\ 1 \times 10^{-5}$–$1\ 6 \times 10^{-4}$
Arthropods§	$1\ 2 \times 10^{-8}$		$4\ 6 \times 10^{-9}$–$2\ 0 \times 10^{-8}$
Small mammals	$3\ 3 \times 10^{-9}$		$6\ 6 \times 10^{-10}$–$6\ 0 \times 10^{-9}$

*Compartmental ^{239}Pu inventory (pCi/m^2) equals mean biomass [g(dry)/m^2] times mean concentration [pCi/g(dry)] Fraction of total equals mean compartmental inventory (pCi/m^2) divided by total inventory

†Number of samples for which the mean is calculated For arthropods and vegetation n is the number of groups of individuals analyzed, for small mammals n is the number of tissue samples, not individual animals.

‡95% confidence interval equals mean ± (1 96 standard error of the mean)

§Includes data from Bly (1977)

greater than 2 0 Although positive skewness is a characteristic of lognormal distributions, the natural-log transformation of soil data did not result in normal distributions (Kolmogorov–Smirnov one-sample test, P > 0 05) but did reduce the skewness for the seven depth groups tested

Three adjacent soil columns (5 by 5 cm) from a 5- by 15-cm area on macroplot 2 exhibited the extreme spatial variability that sometimes occurred in plutonium concentrations in the soil The mean plutonium concentrations in the 5-g aliquots from each column were 480 (column A), 5 4 (column B), and 0 57 pCi/g (column C) at the 0- to 3-cm depth Virtually all the plutonium in column A was in the top 3 cm, the other depths (in 3-cm increments to 21 cm) being at or near background In columns B and C, the majority of the plutonium was found at lower depths No other cases of such extreme spatial variation in soil plutonium concentrations were detected during the sampling at Rocky Flats

As expected, surface soil samples (0 to 3 cm) had a higher mean plutonium concentration than subsurface samples (Table 3) This result agreed with data from Rocky Flats soil sampling reported by Krey and Hardy (1970) Plutonium concentrations were also a function of the size range of soil practices comprising the aliquot (Tables 3 and 4)

TABLE 3 Mean Plutonium Concentrations of Soil Samples from Rocky Flats

Soil particle size range, μm	Plutonium concentration, pCi/g						
	0–3 cm	3–6 cm	6–9 cm	9–12 cm	12–15 cm	15–18 cm	18–21 cm
850–2000	740	140	88	27	13	5.4	1.8
425–850	460	120	100	36	29	7.0	5.5
250–425	440	130	89	30	30	13	5.7
180–250	460	120	130	39	30	14	8.9
150–180	770	130	100	35	140	25	6.5
75–150	870	210	100	68	56	44	11
45–75	1400	310	210	100	160	84	35
0–45	1500	180	810	190	220	85	27
0–2000	830	170	200	66	86	35	13

TABLE 4 Regression Parameters of Soil Plutonium Concentration (pCi/g) Adjusted for the Sample Location as a Function of Soil Particle Diameter at Various Depths*

Depth, cm	Intercept (b_0)	Slope (b_1)	Correlation coefficient (r)	Significant at α =	n
0–3	4.8	−0.336	−0.312	0.01	72
3–6	3.6	−0.270	−0.291	0.05	69
6–9	0.89	−0.753	−0.471	0.001	50
9–12	1.6	−0.544	−0.564	0.001	69
12–15	0.67	−0.799	−0.719	0.001	52
15–18	−0.21	−0.775	−0.706	0.001	47
18–21	−0.42	−0.572	−0.358		22

*The model used was $\ln Pu = b_0 + b_1 \ln D$

Least-squares regressions were calculated with linear exponential and power-function models of plutonium concentration in surface soil as functions of the distance east or south from the asphalt pad The power-function model gave the best fits of the data for both curves (Figs 3 and 4) A t-test indicated that the slope of the distance-south curve (Fig 4) was steeper ($P < 0.05$) than that of the distance-east curve (Fig 3) These results conform to the concept of wind-distributed plutonium the more effective, stronger winds were to the east, and hence the slope of that curve was smaller

Several multiple linear-regression models were calculated The model that accounted for the largest fraction of the total variance (0.868) had the following form

$$\ln Pu = 11.15 - 0.0535 \ln E - 1.628 \ln S$$

where Pu is the plutonium concentration (pCi/g), E is the distance east of the asphalt pad centerline (m), and S is the distance south of the asphalt pad centerline (m)

With this model plutonium concentrations of samples in the soil depth profile were adjusted to estimate the concentration expected at a common location The adjusted values were then regressed as a function of sample depth (Fig 5) As with the distance

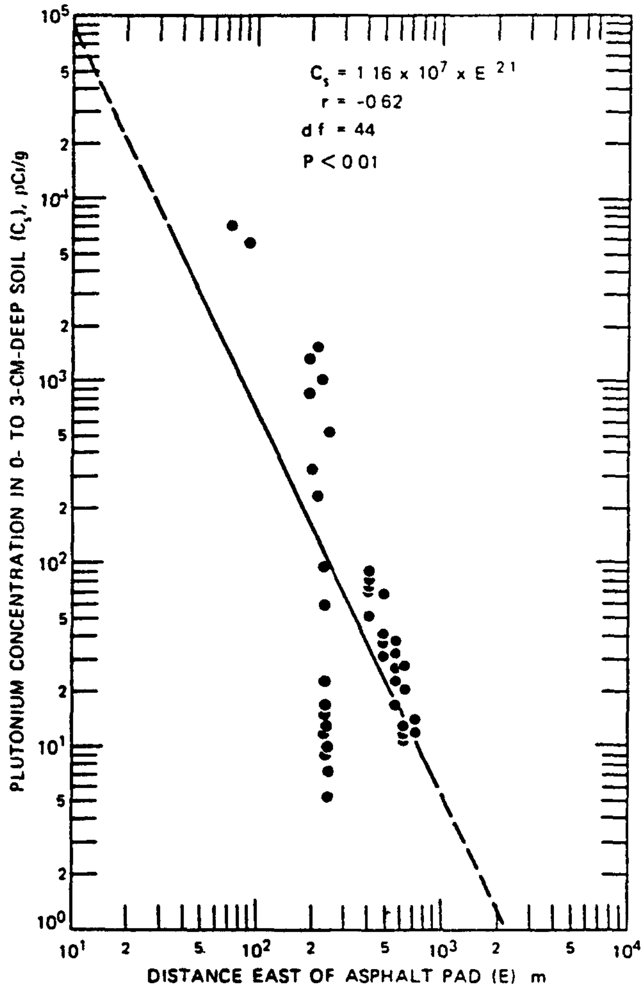

Fig 3 Plutonium concentration in 0- to 3-cm-deep Rocky Flats macroplot 1 soil as a function of distance east of the center of the asphalt oil-barrel storage pad.

relationships, a power-function regression model had the highest correlation of plutonium concentration with depth of the models attempted and was significant ($P < 0\ 01$)

The relationship of plutonium concentration in soil as a function of soil particle diameter (as represented by the opening of the final passage sieve) was examined for each depth layer (Table 4) The model resulted in a significant ($P < 0\ 05$) regression for each depth group except the 18- to 21-cm group The steepest slope (–0 799), at 12 to 15 cm, was significantly different ($P < 0\ 05$) from the flattest slope (–0 270), at the 3 to 6-cm level However, there was no obvious trend in slope vs soil particle-size curves with depth Because the amount of surface area represented by the soil particle spheres in a constant mass of soil is inversely related to soil particle diameter, it followed that the plutonium concentration in a soil sample should be inversely related to the surface area of the particles in the sample A tabulation of the fractions of the total soil sample mass

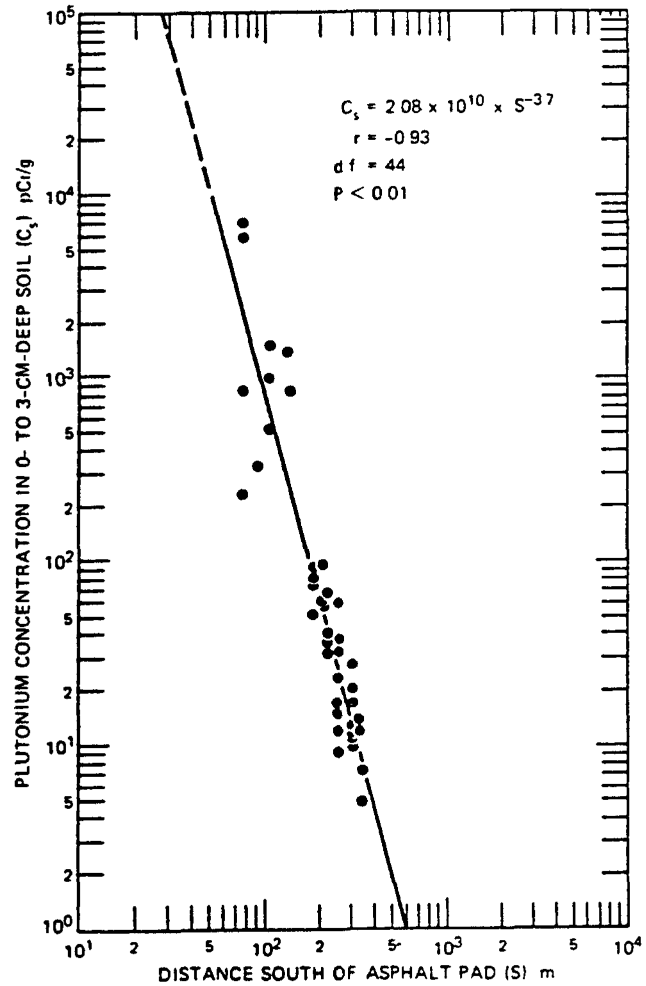

Fig. 4 Plutonium concentration in 0- to 3-cm-deep Rocky Flats macroplot 1 soil as a function of distance south of the asphalt oil-barrel storage pad

represented by each sieve size organized by depths did not produce any obvious patterns with either depth or particle size range Consequently regressions of the soil mass fraction per sample as a function of depth were not significant for most sieve fractions However, these last results would not preclude a surface attachment mechanism

The data on plutonium in soil at Rocky Flats can be summarized by several statements First the variance in the plutonium concentrations of the soil samples was large. CV s within groups of like samples (same depth and particle size) ranged to over 2 0 Frequency distributions for soil samples were positively skewed Spatial variation was also large in one instance the plutonium concentrations of aliquots taken less than 15 cm apart varied by nearly three orders of magnitude

Second, in spite of the large degree of variance in the data the plutonium concentrations in soil were significantly correlated with the location and soil particle

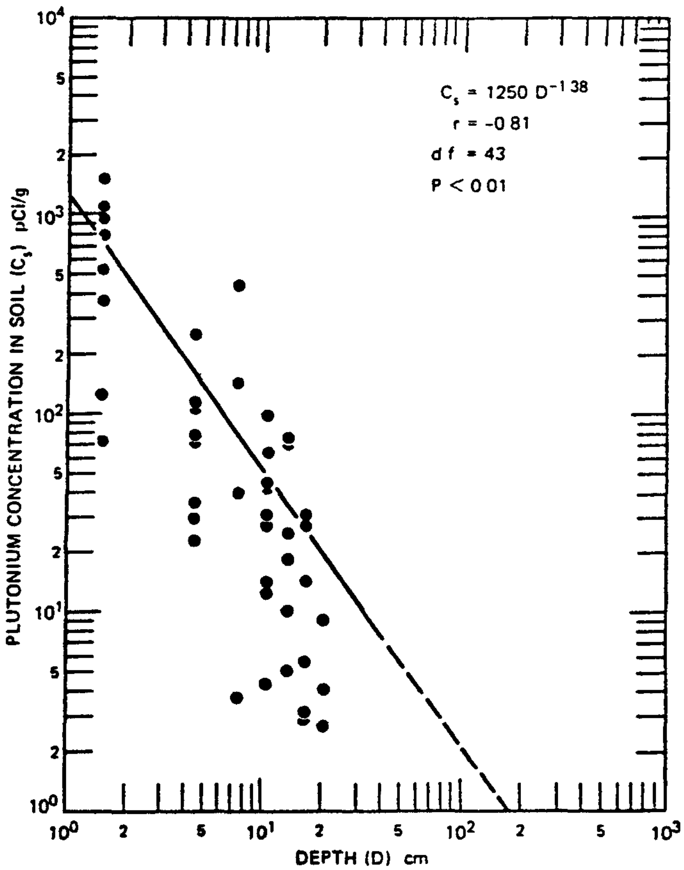

Fig. 5 Plutonium concentration in Rocky Flats macroplot 1 soil as a function of depth of sample Sample concentrations adjusted for distance east and south of center of asphalt oil-barrel storage pad.

composition of the soil sample The spatial distribution of plutonium (i e , more plutonium downwind than downslope) implicated wind as the prime mechanism of plutonium transport onto the studied areas Such factors as resuspension with or without added mechanical disturbances by humans or fauna undoubtedly contributed to the wind transport of plutonium but to a presently unknown degree The data also indicated that plutonium was found to a depth of 21 cm in most samples from downwind of the barrel storage area but that about two-thirds of the contamination was in the top 5 cm The relationship between plutonium concentration and soil particle size suggested a surface-attachment mechanism of plutonium attachment to soil particles However, the lack of any pattern of soil mass fraction with depth for the various particle sizes probably indicates that plutonium transport with depth is not simply a case of transport of the various plutonium–soil particles downward

Plutonium in Plant Compartments

The vegetation community of the study area was composed mostly of grasses and members of the sunflower family Members of the sedge, pea, and mustard families were

TABLE 5 Plutonium Concentrations in Rocky Flats Vegetation and Litter Samples

	Plutonium concentration, pCi/g		
	Mean	Coefficient of variation	n
Litter	412	0.65	29
Vegetation	28.6	2.02	76

also present but in much lower numbers of individuals. Rather than study numerous plant types individually, two plant-derived compartments were studied, litter and detritus and standing vegetation. Although these compartments accounted for only a small fraction of the total plutonium (about 0.2%), the study of those compartments helped derive some concepts of plutonium transport.

As with the soil, frequency distributions for vegetation samples were positively skewed. Further, the hypothesis that plutonium concentrations in vegetation were lognormally distributed could not be rejected ($P > 0.05$). Unexpectedly, the hypothesis that plutonium concentrations in litter were normally distributed could not be rejected ($P > 0.05$).

Mean plutonium concentrations in litter were higher than those in vegetation (Table 5). Concentrations of plutonium in litter and vegetation were each inversely correlated with distance east or south from the asphalt pad ($P < 0.01$).

The fact that litter had a higher mean concentration of plutonium than standing vegetation is not surprising. This result reinforces the suggestion made above that soil transport was the primary mechanism of plutonium transport.

Plutonium in Animal Compartments

Two animal compartments were studied, arthropods and small mammals. These compartments together contained about 2×10^{-8} of the total plutonium estimated to be in the studied areas. Nevertheless, the mobility of the animals makes them potential transporters of plutonium, albeit relatively small amounts, off the site.

As expected from the soil and vegetative sampling, the frequency distributions of plutonium concentrations in small mammals were positively skewed as indicated by the histogram in Fig. 6. Not only were there many samples that had plutonium concentrations below the detection limit but also much of the total activity was supplied by relatively few samples. Frequency distributions of plutonium concentrations in arthropods were also positively skewed (Bly, 1977). Bly (1977) further indicated that logarithmic transformations were useful in alleviating the skewness. Therefore the plutonium concentrations of the arthropod samples were probably lognormally distributed.

Concentrations of plutonium in 23 groups of individual arthropods and in small mammal tissues were of comparable magnitude (Table 6). The small mammal

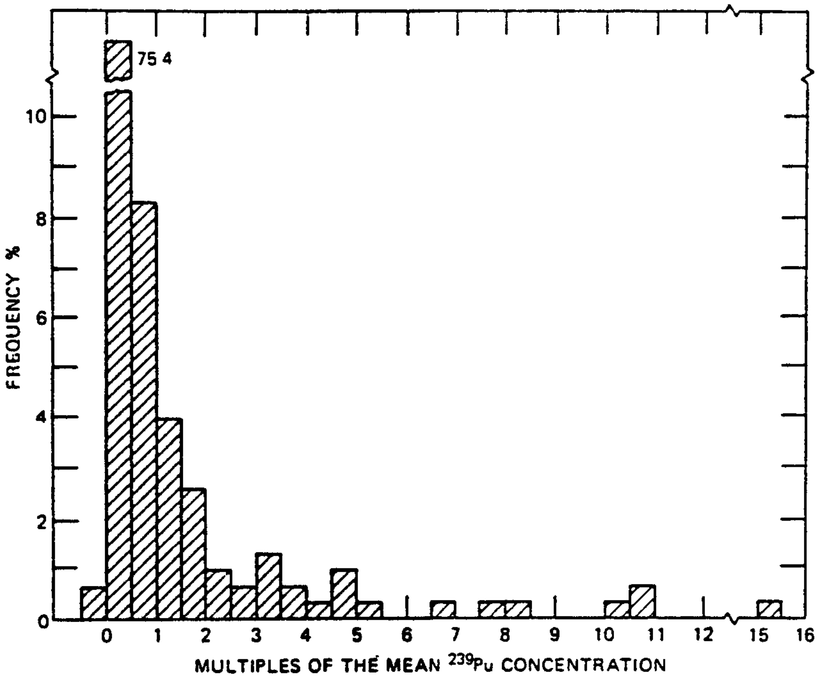

Fig. 6 Representative histogram of small-mammal tissue samples from Rocky Flats

TABLE 6 Mean Plutonium Concentrations of Arthropods* and Small Mammals Sampled from Rocky Flats Macroplot 1

Sample type	n	Plutonium concentration, pCi/g: Mean	Plutonium concentration, pCi/g: Coefficient of variation
Arthropods	23	5.48	1.05
Small mammals			
Bone	28	0.288	2.28
GI tract	40	7.03	2.50
Hide	47	1.51	1.84
Kidney	45	13.6	4.39
Liver	46	8.38	5.45
Lung	47	3.57	1.90
Muscle	50	8.92	5.85
External tissues	134	3.88	2.76
Internal tissues	169	8.59	5.58

*Includes data adapted from Bly (1977)

tissue-sample means ranged from 0.288 pCi/g for bone to 13.6 pCi/g for kidney, and the mean of whole arthropods was 5.48 pCi/g.

The patterns, or rather, lack of patterns, in the small-mammal data were puzzling. The tissues were arbitrarily classed either external or internal, depending on whether or not the tissue had a direct contact with the animal's environment. External tissues included GI tract, hide, and lung; internal tissues included bone, kidney, liver, and muscle. By virtue of the supposed low biological availability of plutonium and the proximity of the external tissues to the contaminated soil, external tissues were expected to have larger plutonium concentrations than internal tissues. Inexplicably, this was not the case. The three highest plutonium concentrations were found in internal tissues, i.e., kidney, muscle, and liver; hide and lung comprised two of the three lowest means. Additionally, the amount of variation in samples within a given tissue was quite high. The minimum tissue variation was in hide samples (CV = 1.84), and the maximum was in muscle (CV = 5.85).

Only two explanations for the high degree of variability are at hand. First, the possibility of cross contamination always exists no matter how carefully one removes tissues during dissection. Second, the extremely small sample mass of a few samples (a dry kidney may be as small as 0.05 g) may have had a tendency to magnify the relative plutonium concentrations. However, a plot of plutonium concentration in small mammals vs. sample mass indicated that about as many samples had large mass and small plutonium concentrations as had small mass and large concentrations. Beyond this, however, the tendency for small-mass samples to skew the distribution has not been investigated.

The nonparametric Kruskal–Wallis technique (Siegel, 1956) was used to test whether or not the seven tissue means were from the same population. The resulting chi-square value of about 44 indicated that the difference between the tissue groups was highly significant ($P < 0.001$). Although no test was performed, it was intuitively obvious that the mean plutonium concentration of the bone samples (0.29 pCi/g, n = 28) was lower than that of other tissues.

Plutonium Concentration Ratios

The concentration ratio (CR) is a potential indicator of plutonium redistribution by wind, water, or plant uptake. Concentration ratio is defined as the concentration in activity per unit mass or volume divided by the concentration of the same nuclide in the same units in another material. In this section the CR will have 0- to 3-cm-deep soil as the material in the denominator [e.g., CR of litter = (pCi Pu/g litter) ÷ (pCi Pu/g 0- to 3-cm-deep soil)].

The CR's of litter, vegetation, arthropods, and small mammals are listed in Table 7. Litter had the largest CR followed in descending order by vegetation, small mammals, and arthropods. Regressions of litter and vegetation CR's vs. distances east and south of the asphalt pad did not achieve significant correlation coefficients ($P > 0.05$).

The plutonium concentrations in litter and in vegetation were plotted vs. soil plutonium concentrations from the same locations. Only the litter curve is shown here (Fig. 7). The litter regression was interesting because of its high correlation (r = 0.975) and near-unit slope (1.001). Although the number of samples here was limited, the data comprising Fig. 7 suggested that litter may be an excellent estimator of soil plutonium concentration in the grassland. The regression of plutonium concentration in vegeta-

TABLE 7 Plutonium Concentration Ratios and 95% Confidence Intervals of Ecosystem Compartments in Rocky Flats Macroplot 1 with 0- to 3-cm-Deep Soil*

Compartment	Concentration ratio	95% confidence interval
Litter	4.9×10^{-1}	2.9×10^{-1}–7.0×10^{-1}
Vegetation	3.4×10^{-2}	1.5×10^{-2}–5.4×10^{-2}
Arthropods	6.8×10^{-3}	3.1×10^{-3}–1.1×10^{-2}
Small mammals	7.8×10^{-3}	2.1×10^{-3}–1.3×10^{-2}

*Mean plutonium concentration in 0- to 3-cm-deep soil equals 835 pCi/g. Concentration ratio equals mean pCi/g compartment divided by mean pCi/g in 0- to 3-cm-deep soil

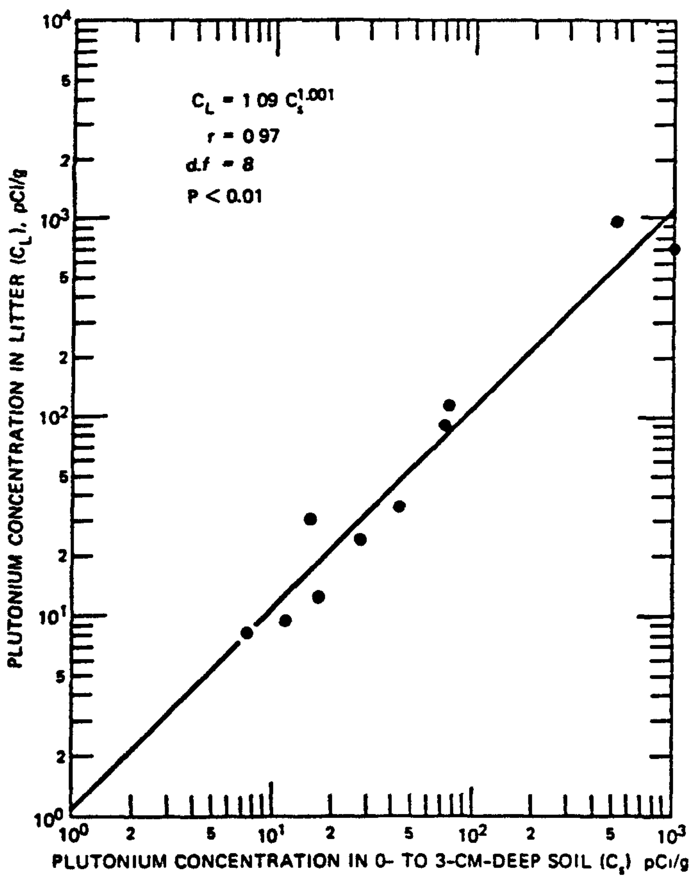

Fig. 7 Plutonium concentration in litter vs plutonium concentration in soil at the same sample location.

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tion vs plutonium concentration in underlying soil was also statistically significant ($P < 0.01$) but was less conclusive than the litter vs soil curve and is not shown here

The CR's of the vegetation were higher than those produced in greenhouse studies Typically, uptake of plutonium under laboratory conditions has been on the order of 10^{-6} to 10^{-4} of the soil concentrations (Newbould, 1963, Wilson and Cline, 1966, Romney, Mork, and Larson, 1970, Schulz, Tompkins, and Babcock, 1976) The Rocky Flats CR of 3.4×10^{-2} suggests either increased root uptake by grassland species or another method of contamination, such as aerial deposition of resuspended soil particles The high surface-to-volume ratio of grasses and the hairy nature of the leaves of many members of the sunflower family would be amenable to a high rate of impaction and attachment of small soil particles Given wind-redistributed plutonium at Rocky Flats, surficial attachment of contaminated soil particles to plants is the likely mechanism of contaminating the vegetation

Plutonium Isotopic Ratios

Ratios of plutonium isotopes or ratios of ^{239}Pu and ^{241}Am have been reported from several sites (Emery et al , 1976, Gilbert et al , 1975, Hakonson and Johnson, 1974, Markham, 1976) In the hope that the examination of the isotopic ratios of ^{239}Pu and ^{238}Pu in the grassland would give some insight into the relative ecological availability of these two nuclides, isotopic ratios were calculated for samples analyzed by alpha spectrometry [isotopic ratio (IR) = ^{239}Pu pCi/g of sample ÷ ^{238}Pu pCi/g of same sample] Ratios were not calculated for samples where either isotope was below the detection limit Ratios were tabulated according to sample type and tested for goodness of fit to a normal distribution The distribution of IR in the various soil depths was either lognormal or marginally normal Small-mammal tissues appeared to be lognormal with respect to the IR

As suggested by Doctor and Gilbert (1977), the concentration of ^{239}Pu was plotted vs ^{238}Pu for each of the seventeen sample types Seventy percent of these groups exhibit a zero intercept, based on a t-test These results implied that the ratios were constant within the tested sample groups and that $^{239}\overline{Pu}/^{238}\overline{Pu}$ would be an unbiased estimator However, because of the likelihood that the IR is lognormally distributed within most of the sample groups, median IR's are reported here (Table 8) The R_4 method of calculation discussed by Doctor and Gilbert (1977) was used to calculate these values

At first glance the median isotopic ratio in soil appeared to decrease as depth increased However, the overlapping 95% confidence intervals for the listed medians suggested that the ratio is relatively constant As expected, neither linear, exponential, nor power-function regressions of the raw IR data vs soil depth were statistically significant ($P > 0.05$)

Despite the limited number of litter and vegetation samples analyzed for both ^{239}Pu and ^{240}Pu, the median IR's of these two compartments were very similar to IR s of the soil These results tended to indicate that the litter and vegetation were closely linked to the soil

The IR's in the animal compartments raised some very interesting questions (Table 8) Only two sample types (GI tract and muscle) exhibited 95% confidence intervals that overlapped with soil IR confidence intervals Therefore, it appeared that the small mammal and arthropod compartments had lower IR s than soil A lower IR would imply

TABLE 8 Median Isotopic Ratios (^{239}Pu pCi/g – ^{238}Pu pCi/g) in Rocky Flats Environmental Samples*

Compartment	Isotopic ratio† Median	Isotopic ratio† 95% confidence interval	n
Soil depth, cm			
0–3	54 70	37 04–80 78	10
3–6	43 70	35 09–54 43	7
6–9	46 41	35 23–61 14	8
9–12	46 45	40 67–53 06	6
12–15	48 76	42 81–55 54	6
15–18	46 94	32 27–68 26	3
18–21	38 67	34 16–43 78	2
Litter	55 47	51 62–59 61	5
Vegetation	59 98	39 92–90 12	3
Arthropods	9 88	5 69–17 15	9
Small-mammal tissues			
Bone	7 49	2 99–18 71	9
GI tract	24 82	17 18–35 90	20
Hide	19 94	13 99–28 43	21
Kidney	11 07	3 98–30 80	7
Liver	17 55	11 62–26 50	12
Lung	7 42	3 97–13 87	10
Muscle	13 20	4 66–37 41	9

*Only data in which both ^{239}Pu and ^{238}Pu were above detectable limits were included

†The median and confidence limits were calculated by method R_4 of Doctor and Gilbert (1977)

relatively enhanced assimilation of ^{238}Pu, compared to ^{239}Pu, into these compartments than into soil

Obviously, there are some physical reasons for skepticism regarding data which suggest that two isotopes of the same element behave differently in biological systems The difference in mass between ^{238}Pu and ^{239}Pu is less than that between ^{235}U and ^{238}U, on which millions of dollars have been spent for enrichment Alpha-recoil energy from ^{238}Pu and ^{239}Pu could displace other atoms from near the surface of a particle of plutonium metal However, unless the particle is composed of either pure ^{238}Pu or pure ^{239}Pu, there would probably be no preferential displacement of either nuclide relative to their ratio in the original metal Rocky Flats plutonium metal probably did not contain either pure ^{238}Pu or ^{239}Pu particles

However, if a particle of pure ^{238}Pu were in some way introduced into an organism, autoradiolysis by this high-specific-activity nuclide might allow relatively fast biological transport compared to ^{239}Pu This idea is not unprecedented Rats that inhaled $^{238}PuO_2$ and $^{239}PuO_2$ of the same particle size and crystalline form translocated up to seven times as much ^{238}Pu as ^{239}Pu to systemic organs at times up to a year postinhalation (Stuart, 1970) Ballou et al (1973) allowed rats and beagle dogs to inhale PuO_2 aerosols of identical size and preparation According to these workers, "The much greater translocation of ^{238}Pu suggests that solubilization of the $^{238}PuO_2$ occurs to a significant degree within the dog "

The previous two paragraphs do little to help explain the animal IR data A possible explanation may be had in statistical bias that heretofore has gone undetected Basically, the bias has to do with the fact that both ^{239}Pu and ^{238}Pu are probably lognormally distributed in environmental compartments Therefore the ratio of ^{239}Pu to ^{238}Pu should also be lognormally distributed (Aitchison and Brown, 1969, p 11) Unfortunately, the distribution of both ^{238}Pu and ^{239}Pu was censored i e, some proportion of the data points was below a detectable limit (Aitchison and Brown, 1969) Shaeffer and Little (1978) have shown that both the mean ratio and the variance of the ratio of two censored lognormal variates will be decreased relative to ratios of uncensored variates if the denominator (^{238}Pu) has a lower magnitude than the numerator (^{239}Pu) The magnitude of the decrease in mean ratio and variance is influenced by the relative closeness to the detection limit of the two variates

This appears to be essentially the case with the IR data presented herein The soil, vegetation, and litter compartments had relatively high plutonium concentrations and also relatively large IR's As the plutonium concentration began to approach the detection limit, e g, in arthropods and small mammals, the IR also decreased Therefore, if the censoring is large, an estimate of the mean or median of the uncensored ratios will be in error because of the effect of censoring

A solution for the problem of ratios of two censored distributions is to try to estimate the population parameters for each distribution and then use method R_2, i e, mean ratio equals mean ^{239}Pu divided by mean ^{238}Pu, as suggested by Doctor and Gilbert (1977) Kushner (1976) discusses two methods of estimating such parameters

Lognormality was assumed, and the methods of Hald (1949) as modified by Kushner (1976) were used to calculate population parameters Then a method of Aitchison and Brown (1969 p 45) was used to calculate the "minimum variance unbiased estimator" of the arithmetic mean isotopic ratio for hide The mean ratio of hide by these methods was found to be 37 The median ratio published in this chapter was 20, and the mean ratio calculated by summing all hide ratios and dividing by the number of ratios (method R_3 in Doctor and Gilbert, 1977) was 29 Therefore, although no confidence interval was calculated the mean IR in hide calculated by Kushner's (1976) method would be little different from the mean IR in soil Unfortunately some of the small-mammal tissue data are censored to such a degree that some of the functional values are extreme enough that they were not tabulated by Hald (1949), one of Kushner's (1976) prime references Therefore the parameters of most of the censored small-mammal data cannot be estimated by the methods of Kushner (1976) and Hald (1949)

In summary, the median IR was constant in soil and vegetation compartments However, the median IR's also suggest that ^{238}Pu is preferentially mobile in animal compartments of the grassland relative to ^{239}Pu and soil There is reason to believe that the IR data are biased toward lower magnitudes as influenced by their nearness to the detection limit The mean IR for hide estimated with procedures of Kushner (1976) and Hald (1949) suggested that these data may be similar to soil IR's Other small mammal tissues were not compatible with these estimation procedures Further field sampling to eliminate the censoring difficulties is probably necessary if the question of differential concentration of ^{239}Pu and ^{238}Pu in small mammals is to be resolved

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Summary

The soil to a depth of 21 cm contained more than 99% of the plutonium estimated to be in the studied areas of the Rocky Flats grassland. Litter contained a larger fraction of the total plutonium ($\sim 10^{-3}$) than vegetation ($\sim 10^{-4}$), arthropods ($\sim 10^{-9}$), or small mammals ($\sim 10^{-9}$). These results implied that soil–plutonium relationships and soil-management practices are very important at contaminated sites.

Plutonium-concentration frequency distributions for soil samples were positively skewed and characterized by CV's that were generally greater than 100%. Plutonium concentrations in surface (0 to 3 cm) soil were inversely related to distance from the plutonium source, the former oil-barrel storage area. Soil–plutonium concentrations tended to decrease as depth increased and tended to increase as the soil particle size decreased. This latter result suggested that plutonium–soil interaction was a surface-attachment mechanism.

Mean concentrations of plutonium were higher in litter than in vegetation. Frequency distributions of plutonium concentration were normal in litter and lognormal in vegetation. In a manner similar to soil, plutonium concentration both in litter and in vegetation was also inversely related to distance from the barrel storage area. Plutonium concentrations in plant-derived compartment samples were also significantly correlated to plutonium concentration in surface soil at the same locations.

Plutonium frequency distributions in arthropods and small mammals were also positively skewed. Plutonium concentrations in bone samples were lower than those in the other tissues sampled, namely, GI tract, hide, kidney, liver, lung, and muscle.

Concentration ratios of litter, arthropods, and small mammals relative to soil indicated that litter had the highest value. The other compartments, in descending order, were vegetation (3.4×10^{-2}), small mammals (7.8×10^{-3}), and arthropods (6.8×10^{-3}). The relatively high CR's suggested that most of the contamination of vegetation resulted from surficially attached plutonium–soil particles as opposed to root uptake. All the above data strongly indicate that in the grassland soil is by far the most important compartment insofar as plutonium content and transport are concerned. The primary conclusion is that, if transport of plutonium is to be avoided, then transport of soil should be avoided. Therefore soil stabilization should be promoted by maximizing vegetative cover growth and minimizing mechanical disturbances.

Isotopic ratios of ^{239}Pu to ^{238}Pu were calculated for soil, litter, vegetation, arthropod, and small-mammal samples processed by commercial laboratories. The soil results indicated that the median ratio was about 50. Litter and vegetation IR's were similar to IR's in soil. The IR's of small-mammal tissues and arthropods were likely lower than those of soil.

The meaning of the lower IR's in animal compartments was clouded by the fact that the frequency distributions of the ^{239}Pu and ^{238}Pu concentrations, from which the ratios were formed, were censored. Further, the ^{238}Pu concentration distribution was censored to a much larger degree than was the ^{239}Pu distribution. This situation may have the effect of spuriously decreasing the mean or median ratio if the ratios are formed before the average is calculated. An estimation procedure was used to calculate the mean of both ^{239}Pu and ^{238}Pu by taking into account the degree of censorship. Although most small-mammal compartments may not be amenable to such a procedure, the ratio in hide was calculated to be about 37. This value was within the 95% confidence interval of most of the soil IR's. Without further analysis, the hide data suggested that the IR may not be changing between environmental plutonium compartments, as previously suggested (Little, 1976), but may indeed be constant

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