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Plutonium in a Grassland Ecosystem at Rocky Flats

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ABSTRACT

Portions of the grassland around the Rocky Flats installation of the U.S Department of Energy, located about 12 km northwest of the Denver metropolitan area, were contaminated with Pu between 1959 and 1964 This study was designed to elucidate the Pu distribution patterns in the terrestrial ecosystem immediately downwind of the contamination source. We concluded that wind erosion of contaminated soil was likely the chief mechanism of plutonium transport and that soil contained 99.7% of the plutonium in the system Plant and small animal compartments combined contained less than 0.3% of the total Pu inventory which averaged about 53 μCi/m² in the study area

Additional Index Words **plant uptake, concentration ratio, Pu-transport, radionuclides.**

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Although there have been numerous studies of plutonium in the environment, the study reported here was the first designed to quantify the distribution of Pu in compartments of a grassland ecosystem This paper reports results of a study begun in 1972 to describe the Pu deposition and distribution patterns in the grassland buffer zone downwind of a previously used barrel storage area at Rocky Flats

SITE AND METHODS

The Rocky Flats installation, about 12 km northwest of the Denver metropolitan area, utilized nearly 30 km² as a buffer zone which separates the public from production operations The climate is typified by occasional strong west-northwest winds >40 m/sec and moderate precipitation (40 cm/year average) (Rocky Flats 1975 annual weather summary, unpublished) The physiognomy of Rocky Flats is best described as modified grassland which includes species typical of shortgrass plains (*Bouteloua gracilis, Buchloe dactyloides*) as well as tall-grass prairie (*Agropyron* spp , *Andropogon* spp) and ponderosa pine (*Pinus ponderosa*) woodland (Weber et al , 1974) Mule deer (*Odocoileus hemionus*) and numerous species of reptiles, rodents, and birds reside in the area (Whicker, 1974)

Drums containing Pu-contaminated oil which leaked during 1959-1964 on the barrel storage area were the major source of Pu in the Rocky Flats environment (Krey and Hardy, 1970) Investigations by the U S Department of Energy Environmental Measurements Laboratory (EML, formerly Health and Safety Laboratory), Rocky Flats personnel, and others indicated that Pu concentration patterns in soil were consistent with the predominant wind directions (i e , Pu concentration isopleths in soil extended primarily east and southeast from the barrel storage area) (Krey and Hardy, 1970, Poet and Martell, 1972) The barrel storage area was asphalted in late 1969 and no barrels have been stored there since

We established two sampling areas in the buffer zone southeast (downwind) of the asphalt pad covering the former oil barrel storage area a 0 75-ha study macroplot about 200 m away, and a 500-m-long sampling transect extending from 270 to 770 m downwind (southeast) During 1972–1974, we collected samples of soil, litter, composited vegetation, and the most common arthropods (orders Arachnida Thysanura, Orthoptera, and Coleoptera) and small mammals (*Peromyscus maniculatus, Thomomys talpoides,* and *Spermophilus tridecemlineatus*) from the study macroplot for Pu analysis and biomass estimations Soil, litter, and vegetation were also sampled along the downwind transect Soil was sampled using a hand trowel to 21 cm in seven 3-cm depths, i e , 0–3, 3–6 6–9 cm, etc (Little and Whicker, 1978) Soil samples were dry-sieved Small mammals were dissected and individual tissues of bone, GI, hide, kidney, lung, liver, and muscle were analyzed Vegetation was clipped about 1.5 cm above the soil and was not washed prior to analysis for Pu content Arthropods were sampled using sweep nets and pitfall traps, and were analyzed whole for Pu content All samples were oven dried prior to analysis for Pu content Sample masses analyzed by liquid scintillation in our laboratory (Little, 1976) or by alpha spectrometry in a commercial laboratory were kept small (<5 g) to reduce analytical cost per sample Analytical error was relatively low, the mean fractional recovery of Pu from 29 aliquots of a homogenized Rocky Flats soil supplied by EML was 100% of the EML value and had a standard deviation that was 23% of the mean Nonparametric statistical tests were used in many cases because highly skewed sample frequency distributions were observed A more detailed description of the methods was given by Little (1976)

RESULTS AND DISCUSSION

Plutonium Compartmentalization

Plutonium concentrations[3] in surface soil (0–3 cm) were higher than Pu concentrations in soil from lower depths (Table 1) Litter had a higher Pu concentration than vegetation, but both of these had less Pu than surface soil Concentrations of Pu in small mammals and arthropods averaged about 100 times lower than those found in soil (Table 1) No significant differences ($p > 0.05$) were found between Pu concentrations of the seven tissue types The mean concentration of the 304 individual tissue samples was assumed to be representative of the entire small mammal biomass

Mass estimates and mean concentrations for each of the sampled compartments were used to calculate the fraction of the total Pu inventory in each compartment (Table 1) ' The compartmental estimates indicated that the top 21 cm of soil contained >99% of the total Pu inventory of the ecosystem, the 0–3 cm and 3–21 cm

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[3] Unless noted otherwise data in this paper refer to ^{239}Pu which includes aproximately 3% activity contribution from ^{238}Pu

Table 1—Distribution of ^{239}Pu in samples from the Rocky Flats study macroplot †

Compartment	Mean	n‡	95% confidence interval
	Plutonium concentration, dpm/g		
Soil, 0-3 cm	1 850	72	1,230-2 480
Soil, 3-21 cm	233	309	154-312
Litter	914	29	698-1 130
Vegetation	63 4	76	34 8-92 0
Arthropods§	12 6	23	7 19-18 0
Small mammals	14 4	304	5 29-23 5
	Fraction of total Pu		
Soil, 0-3 cm	5.0×10^{-1}		2.5×10^{-1}-7.4×10^{-1}
Soil, 3-21 cm	5.0×10^{-1}		2.5×10^{-1}-7.5×10^{-1}
Litter	2.9×10^{-3}		1.6×10^{-3}-4.2×10^{-3}
Vegetation	1.0×10^{-4}		4.1×10^{-5}-1.6×10^{-4}
Arthropods	1.2×10^{-6}		4.6×10^{-7}-2.0×10^{-6}
Small mammals	3.3×10^{-7}		6.6×10^{-10}-6.0×10^{-7}
	Concentration ratio¶		
Soil, 0-3 cm	1.0×10^{0}		-
Soil, 3-21 cm	1.3×10^{-1}		6.6×10^{-2}-1.9×10^{-1}
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}		2.2×10^{-3}-1.3×10^{-2}

† Compartmental ^{239}Pu inventory (dpm/m²) = mean biomass (g dry/m²) × mean concentration (dpm/g dry) Fraction of total = mean compartmental inventory (dpm/m²) ÷ total inventory (dpm/m²) Concentration ratio = mean concentration of compartment (dpm/g) ÷ mean concentration of 0-3 cm soil (dpm/g)

‡ n = 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, the number of tissue samples not individual animals.

§ Includes data collected by Bly (1977) unpublished manuscript., Dept. Radiology and Radiation Biology Colo. State Univ., Ft. Collins.

¶ Relative to 0-3 cm soil.

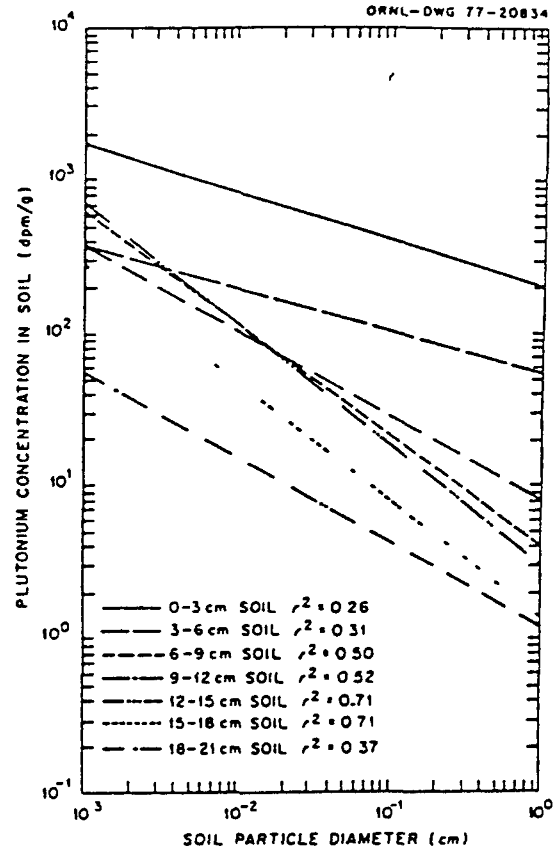

Fig 1—Plutonium concentration in soil from the Rocky Flats study macroplot plotted versus soil particle diameter The value of r^2 is the coefficient of determination

layers had about equal fractions (Table 1) As expected from the mass relationships of the ecosystem, the fractions of total Pu in the non-soil compartments were orders of magnitude lower than the soil Litter contained a higher fraction of the total Pu inventory than vegetation which in turn contained a larger fraction than the animal compartments studied These data imply that soil-Pu relationships and soil management practices are very important on contaminated areas and also that Pu quantities in biotic tissues are relatively insignificant

Plutonium in Soil

Because of the importance of soil in terms of the total Pu inventory, much of this study was devoted to describing the distribution of Pu in soil at the Rocky Flats study area These data have been reported in greater detail elsewhere (Little, 1976, Little and Whicker, 1978), but we will allude to them briefly here. The concentration of Pu in surface soil was a function of both distance east and distance south of the Pu source (Little and Whicker, 1978) Wind was implicated as the most effective Pu transport mechanism by two coincident facts First, winds at Rocky Flats are predominantly to the east Second, the slope of the curve of soil plutonium vs distance south of the source was more negative ($p < 0.05$) than the slope of the curve of soil Pu vs distance east This result implies that transport to the east (predominant wind direction) was more effective than transport to the south (predominantly downslope)

Soil Pu concentrations were also described by an inverse power function of sample depth (Little, 1976, Little and Whicker, 1978) The Pu concentration in soil at each depth was further described by an inverse power function of the soil particle size range (Fig 1) As indicated by the seven lines in Fig 1, there was no apparent change in the relationship between Pu concentration in soil and soil particle diameter as depth increased At all depths, the higher Pu concentrations were associated with the smaller soil particles or aggregates The fractional contributions to sample mass by each soil aggregate size were also investigated and found to exhibit no trends with depth Because the surface area of a given mass of soil increases as the soil particle diameter decreases, these data (Fig 1) are consistent with a mechanism of surface attachment of Pu particles to soil particles

The trends of Pu distribution in Rocky Flats soil profile found during this study basically agree with the few published soil profile results from other sites Both Hakonson and Johnson (1974) at the Trinity Site in New Mexico, and Essington et al (1975) at the Nevada Test Site found that in most profiles the majority of the Pu was within a few centimeters of the surface Essington et al (1975) further reported detectable plutonium concentrations at the deepest-sampled levels, 25 cm Some of the profiles closer than 24 km from ground zero reported by Hakonson and Johnson (1974) had relatively uniform Pu distributions in the top 7 5 cm and rapidly decreasing concentrations below that point We know

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of no soil profile sampling around facilities that have had Pu releases similar to those at Rocky Flats As stated above, however, the weapons-related releases of Pu and releases at Rocky Flats frequently resulted in most of the soil Pu residing near the surface

Distributions of Pu concentration in soil due to random spatial variation appeared log-normal with C V over 1.0. An example of the high degree of spatial variation was found at one sampling location about 1 km south of the asphalt pad where aliquots from each of three contiguous surface samples (5 cm by 5 cm by 3-cm deep) had mean Pu concentrations of 1,060, 11.9, and 1.3 dpm/g Plausible explanations for this large range in Pu concentration include (i) a single large particle of Pu oxide resulted in the detected activity or (ii) many small Pu particles agglomerated with larger soil particles to form aggregates. If a single large Pu particle contributed the activity, the 5,300 dpm in the most active 5-g aliquot mentioned above could have come from a 19-μm diam PuO_2 particle (Sill, 1971) However, fission track analysis of contaminated Rocky Flats soil yielded mean and maximum PuO_2 particle diameters of 0.3 and 3.0 μm, respectively (Hayden, 1973, McDowell and Whicker, 1978) The second explanation for the large degree of variability in Pu concentration may be the more likely. For example, 10 50-μm-diam soil spheres, each with a surface area of 7.85×10^3 μm², could provide host attachment to 8.73×10^5 PuO_2 spheres of 0.3 μm diam for a total activity of 1.7×10^4 dpm For the sake of simplicity, this calculation assumes the amount of soil particle surface area required to attach a Pu particle is the diameter of the Pu particle squared Therefore, 0.3 μm PuO_2 particles covering only about 30% of the soil surface area could contain 5,300 dpm Such a spherical particle aggregate would have a diameter <200 μm and, thus, would be highly erodible (Healy and Fuquay, 1958) After erosion and deposition, weathering could have separated the components of the composite particle This postulated process, although speculative, is compatible with the expressed variability, necessary qualities of erodibility, and observed particle size distributions in contaminated soils

Plutonium in Plant Compartments

Frequency distributions were normal for litter and log-normal for vegetation (Kolmogorov-Smirnov two-sample test, $p > 0.05$) Mean Pu concentrations were higher in litter than in vegetation (Table 1) Concentrations of Pu in litter and vegetation were also inversely correlated to downwind distance from the barrel storage area ($p < 0.01$) and positively correlated to Pu concentration in soil underneath ($p < 0.01$)

Concentration ratios of litter, arthropods, and small mammals relative to soil (Table 1) were similar to those found by investigators in other field studies (Hakonson et al , 1974, McLendon et al , 1976) However, concentration ratios of Pu for sampled Rocky Flats vegetation were several orders of magnitude larger than for experimentally grown vegetation Concentration ratios for vegetation grown to estimate Pu uptake by roots are commonly 10^{-6} to 10^{-4} (Newbould, 1963, Price, 1972, Schultz et al , 1976) The high concentration ratios of the Rocky Flats vegetation samples (3.4×10^{-2}, Table 1) suggested substantial surface deposition of Pu-containing dust or increased physiological availability of Pu through time A study by Arthur[4] (1977) corroborates the former conclusion Arthur found that, based on removal of activity b ultra-sound during washing, much of the Pu associated with Rocky Flats vegetation was attached to the plant surface

Plutonium in Animal Compartments

Frequency histograms of Pu concentrations in small mammal tissues were not normally distributed ($p < 0.05$), and the presence of many less-than-detectable values prohibited the use of logarithms for normalizing the data Mean Pu concentrations were raised somewhat by large concentration values in relatively few samples This phenomenon further contributed to the non-normality

As can be seen from Table 2, the variation about the mean for each tissue type was quite large Likewise, the apparent value of the mean varied considerably between tissue types To avoid the problem of non-normality, a nonparametric test, the Kruskal-Wallis (K-W) one-way ANOVA was used to test for significant effects between the seven tissue groups of each macroplot According to Siegel (1956), "the Kruskal-Wallis technique tests the null hypothesis that the k samples come from the same population or from identical populations with respect to averages "

For the 304 small mammal tissue samples of seven types, the calculated K-W chi-square value of about 44 indicated significant ($p < 0.001$) differences between the tissues Because of the non-normality of the distributions, a parametric test such as the Least Significant Difference test (L S D) was likely not valid to discern which tissue or tissues of the seven tested for each macroplot were different from the rest.

In the absence of such tests, the rank sums and mean ranks per sample for each tissue group were inspected (Table 2) Clearly, the bone tissues have much lower

[4] W J Arthur, III 1977 Plutonium intake by mule deer at Rocky Flats Colorado M S Thesis Colorado State Univ , Ft Collins Colorado

Table 2—Mean plutonium concentrations and Kruskal Wallis rank sums for small mammal tissues sampled from Rocky Flats

Tissue	n†	Pu concentration (dpm/g) Mean	C V ‡	Kruskal Wallis ranks§ Sum	Mean rank¶
Bone	29	0.64	2.3	2 200	75
GI	40	16	2.5	7 700	193
Hide	47	3.4	1.8	6 800	145
Kidney	45	30	4.4	7 800	174
Liver	46	19	5.5	6 500	142
Lung	47	7.9	1.9	8 700	186
Muscle	50	20	5.9	6 600	132
External	134	8.6	2.8	23 000	173
Internal	170	19	5.6	23 000	136

† n = number of samples for which the mean is calculated

‡ C V = standard deviation/mean.

§ All 304 samples were ranked in ascending order with rank value 1 assigned to the lowest valued sample and 304 to the highest valued sample The ranks for each tissue were summed and a chi squared calculated For a detailed discussion see Seigel 1956

¶ The mean rank per sample has no recognized statistical importance and is shown here for intuitive purposes only rank sums than any other tissues This difference, at least partially, results from the smaller number of samples of bone relative to other tissues Even so, when the rank sum for each tissue was divided by the number of samples to calculate the mean rank, the bone samples were still much lower While this result is not a testable quantity, it seems reasonable in light of the significant K-W results that the mean Pu concentration in bone is different from most, if not all, of the other tissue Pu means

The amount of Pu that might be biologically transported from the 0.75-ha study area or from the Rocky Flats plant site is not large The maximum small mammal biomass observed on the study area was 4.5×10^5 dry g (Little, 1976) Assuming that each individual animal was homogeneously contaminated to the mean Pu level of 14.4 dpm/g, all the small mammals combined contained <3 μCi. Given the large amount of Pu already transported off-site (5 8 Ci, Krey and Hardy, 1970), it seems doubtful that the small mammals could significantly increase that off-site inventory.

A study of Pu and soil movement by pocket gophers at Rocky Flats found that a population ranging from 6–16 individuals on a 2 6-ha site moved about 3,000 kg of below-surface soil with a mean Pu activity of 39 dpm/g over a 7-month period (Winsor and Whicker, in press) At that rate, the studied population of pocket gophers transported a total activity of 1.1×10^8 dpm (50 μCi) annually It should be noted that this activity was only transported from below surface to the surface and not off the study area

It also appears unlikely that Pu-transport by large mammals, namely deer, is large Arthur (1977)[4] estimated that a "standard weight deer (66 2 kg)" feeding on the study area at Rocky Flats would ingest 2.9×10^7 dpm (13 1 μCi) of Pu annually Based on Arthur's data, estimated deer population, and usage of the Rocky Flats study area, Hiatt (1977)[5] calculated probable transport of Pu by deer to be as much as 0 14 and 0 32 μCi to as far as 0.5 and 5 2 km, respectively, from the study area.

CONCLUSIONS

A significant conclusion of this study is that soil should be the compartment of principal concern with regard to transport of Pu (i.e , processes affecting soil transport will also influence Pu movement) Vegetation/soil concentration ratios further suggested that Pu associated with plant samples resulted mainly from Pu-containing soil attached to above ground plant surfaces as opposed to biological incorporation Therefore, to minimize Pu mobility after a contamination event, authorities should seek to minimize soil erosion

Although soil is the largest Pu compartment at Rocky Flats, modes of Pu mobilization other than soil erosion could prove to be important over very long time periods Soil movement by small mammal activities contributes to soil redistribution (Richens, 1966, Winsor and Whicker, in press) Mobile animals could transport Pu to other areas during migrations or wanderings, but the prospect for large-scale movement of Pu by animals seems small

Concern over short-term biological and ecological effects of Pu on the vegetation and small animals at Rocky Flats seems unwarranted at present Vegetation, arthropods, and small mammals had Pu concentrations much lower than those which produced biological effects in laboratory dogs and rodents (Park et al., 1972, Moskalev, 1972) Therefore, concern about ecological effects following an event resulting in Pu concentrations of the magnitudes found in the grassland studies at Rocky Flats should be centered more on the impacts resulting from cleanup or soil stabilization procedures than on Pu contamination directly

[5]G W Hiatt 1977 Plutonium dispersal by mule deer at Rocky Flats Colorado M S Thesis, Colorado State University, Ft Collins Colorado

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