**RADIOECOLOGY OF
NATURAL SYSTEMS**

MASTER

THREE YEAR SUMMARY REPORT

for the period
May 1, 1974 — July 31, 1977

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INTRODUCTION

This report is intended to provide a general summary of the progress and findings of work sponsored by ERDA under contract EY-76-S-02-1156 over the period May 1, 1974 through July 31, 1977. During this period, the overwhelming majority of effort was devoted to studies on the distribution, transport, characterization and ecological consequences of plutonium in the terrestrial environs of the Rocky Flats nuclear weapons plant near Denver, Colorado. During the period, however, additional studies were carried out on the geochemistry of stable lead in an alpine lake and its watershed, cesium kinetics in a montane lake, long-term patterns of fallout ^{137}Cs in trout and mule deer populations, and effects of chronic gamma irradiation on a shortgrass plains ecosystem. Since the studies on lead and cesium have been of comparatively low intensity and because the data generated have not been analyzed sufficiently for a substantive summary, they are not reported on here. Those interested in the lead and cesium studies should consult the recent annual progress reports. In 1975, the investigation of the effects of chronic irradiation on a shortgrass plains ecosystem was transferred to ERDA Contract EY-76-S-02-2743; therefore, it is not reported here either. This report provides a brief synthesis of progress on the plutonium studies.

Public and scientific interest in plutonium contamination of the environment has been keen in recent years. One focal point of concern is the grassland near the Rocky Flats installation of the U. S. Energy Research and Development Administration northwest of Denver, Colorado. Now operated for ERDA by Rockwell International, the Rocky Flats Plant handles large amounts of plutonium metal for defense purposes. The plant, approximately 12 km northwest of the Denver, Colorado, metropolitan area, utilizes nearly 30 km² as a buffer zone which separates the public from production operations. The climate of Rocky Flats is typified by occasional strong W-NW winds exceeding 40 m/s and moderate precipitation (40 cm/y average). The physiognomy of Rocky Flats is described as modified grassland which includes species typical of shortgrass plains (Bouteloua gracilis, Buchloe dactyloides, etc.) as well as tall-grass prairie (Agropyron spp., Andropogon spp.), and ponderosa pine woodland. Mule deer (Odocoileus

hemionus hemionus) reside alongside typically grassland species of arthropods, reptiles, rodents, and birds.

Drums containing plutonium-contaminated oil which leaked during 1959-1964 on the barrel storage area were the major source of plutonium contamination. Investigations by ERDA's Health and Safety Laboratory, Rocky Flats personnel, and others indicated that plutonium contamination patterns in soil were consistent with predominant wind directions (i.e., plutonium concentration isopleths in soil extended primarily east and southeast from the barrel storage area). Evidence supports the concept that soil particles near the base of leaking drums became heavily contaminated with plutonium and were subsequently resuspended by strong winds. Contaminated soil particles were deposited downwind at levels which decreased with distance. The plutonium has largely remained in soil, near the surface. However, measureable amounts have been found to depths of 30 cm in soil and in the biota. Public concern, lack of knowledge on the environmental behavior of plutonium, and the high radiotoxicity of the element all provided justification for the research program described in this document.

The findings described in this report were obtained by (alphabetically): staff members A. W. Alldredge, J. A. Bly, L. Fraley, Jr., and T. F. Winsor; and graduate students L. E. Alexander, W. J. Arthur, S. J. Baker, M. P. Carson, R. A. Geiger, G. S. Hiatt, C. A. Little, L. M. McDowell, S. L. Mecker, and M. L. Miller.

SUMMARY OF FINDINGS

Distribution of Plutonium in the Terrestrial Ecosystem

We established three sampling areas southeast (downwind) of the former oil barrel storage area: a 0.25 ha plot 50 m away, a 0.75 ha study macroplot about 200 m away, and a 500 m-long sampling transect running from 270 to 770 m downwind. Two other study plots were established, one 1400 m south of the former oil barrel storage area, and one in the northwest corner and upwind of the plant. During 1972-1976, we collected samples of soil, litter, vegetation, arthropods (orders Arachnida, Thysanura, Orthoptera, and Coleoptera), small mammals (Peromyscus maniculatus, Thomomys talpoides, and Spermophilus tridecemlineatus), mule deer, snakes (Coluber constrictor flaviventris, Pituophis melanoleucus sayi, and Crotalus viridis viridis), and mourning doves (Zenaidura macroura) from the study areas for plutonium analysis and biomass estimations. Samples were analyzed by liquid scintillation counting in our laboratory or alpha spectrometry by commercial laboratories.

Data from or representative of macroplot 1, the principal study plot 200 m downwind of the former barrel storage area, are given in Table 1. Mean concentrations, compartmental inventories, and concentration ratios are summarized. The data indicate that the top 21 cm of soil contained more than 99% of the total plutonium inventory of the ecosystem; the 0-3 cm and 3-21 cm layers had about equal fractions. The fractions of total plutonium in the non-soil compartments were orders of magnitude lower than the soil. Litter had a higher fraction of the total plutonium inventory than vegetation which in turn had a substantially larger fraction than the animal compartments. These data imply that because most of the plutonium is in soil, land management practices are very important on contaminated areas and also that plutonium transport by biota appears relatively insignificant, at least in the short term.

The values given in Table 1 for vegetation and small mammals need to be qualified in the light of recent findings. For example, vegetation from macroplot 1, when cleansed of surface dust using an ultrasonic bath, contains of the order of 1 to 2 dpm/g of ^{239}Pu . This is strong evidence

Table 1. Distribution of ^{239}Pu in samples from Rocky Flats study macroplot no. 1. Compartmental ^{239}Pu inventory (dpm/m^2) = mean biomass (g/m^2) x mean concentration (dpm/g). Fraction of total = mean compartmental inventory (dpm/m^2) $\div$ total inventory (dpm/m^2). Concentration ratio = mean concentration of compartment (dpm/g) $\div$ mean concentration of 0-3 cm soil (dpm/g).

Compartment	Mean	n*	95% confidence interval	
Plutonium concentrations (dpm/g)				
soil, 0-3 cm	1850	72	1230	- 2480
soil, 3-21 cm	233	309	154	- 312
litter	914	29	698	- 1130
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^{-8}		4.6×10^{-9}	- 2.0×10^{-8}
small mammals	3.3×10^{-9}		6.6×10^{-10}	- 6.0×10^{-9}
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}

*n = no. 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.

that the mean value of 63 dpm/g in Table 1 mainly reflects plutonium in the surficial dust. In addition, more recent data on small mammal samples which were pooled to obtain more activity per sample, gave lower mean values, on the order of 0.4 dpm/g.

Levels of ^{239}Pu in the tissues of mule deer, snakes, and mourning doves in most cases have been below the analytical detection limit. In no case has a sample of internal tissue from these species contained more than 1 dpm/g. These data collectively indicate that the plutonium in the environs of Rocky Flats is highly insoluble and is not moving into the biota to an unexpected extent. Of the animals sampled, arthropods clearly contain the highest concentrations of ^{239}Pu and also the largest fraction of the inventory associated with animals. As with vegetation, however, arthropods were not cleansed of surficial activity prior to assay and the ^{239}Pu concentrations of internal body parts is not known.

Plutonium concentrations in Rocky Flats soil varied inversely with distance from the original plutonium source, depth of the sample, and particle size of sieved soil samples. Coefficients of variation of plutonium in soil ranged to more than 300%; and frequency distributions were highly skewed with most samples lying below the mean and a few being much larger than the mean. The plutonium distribution patterns and known characteristics of the plutonium source indicated that the mechanisms of environmental dispersion may have involved: the attachment of plutonium oxide to soil particles; primary dissemination of the contaminant from the source by wind; and weathering, microdispersal, and penetration into soil of deposited particles. The high degree of spatial variability, in particular, suggested that the most common functional form of the contaminated soil during dissemination was probably an agglomerated particle containing many plutonium oxide and soil particles bound together.

In an effort to more fully understand the problem of sampling heterogeneity, soil samples from macroplot 1 were examined microscopically so that plutonium particle sizes and micro-distribution could be investigated. We wanted to explore the idea that occasional high-activity samples were possibly caused by "hot particles" of plutonium. An autoradiographic technique, utilizing nuclear emulsion plates, was used to obtain abundance of, and equivalent size distributions for, plutonium particles in Rocky

Flats surface soil. A total of 1700 particles were sized, for which mean $^{239}\text{PuO}_2$ equivalent diameters of 0.29 μm , 0.25 μm , and 0.20 μm were found for 7-, 14-, and 37-day exposures of soil to emulsion plates. A method to scan for particles greater than 1.50 μm equivalent diameter, utilizing Kodak AA Industrial X-Ray film, was also used. The largest particle sized with this procedure was 6.86 μm $^{239}\text{PuO}_2$ equivalent diameter. None of the particles sized was of sufficient magnitude alone to account for elevated plutonium activity observed previously in Rocky Flats surface soil samples. Variability in particle concentrations was observed, however, which suggested that heterogeneity in the spatial distribution of plutonium particles in the soil may partially explain observed variability in soil plutonium concentrations. While no "hot particles" (> 1000 dpm) were found, their presence cannot be ruled out because of the limited quantity of soil that can be feasibly examined by the autoradiographic technique.

Based upon data from samples taken across gradients of plutonium contamination, levels of plutonium in litter, vegetation and arthropods were significantly correlated to concentrations in soil. These correlations provided additional evidence that most of the plutonium associated with these samples was actually attached to surficial dust. This study corroborates the idea expressed by other investigators that plutonium moves in the environment principally by physical rather than physiological mechanisms.

Data on ^{239}Pu concentrations in vegetation, small mammals and arthropods were too variable to show statistically significant differences between species or taxonomic groups. We also have not been able to show significant differences between collection dates nor types of small mammal tissues. Statistical analyses have been exhaustive, in that data were log-transformed to reduce heterogenous variance and skewness of distributions, and normal as well as non-parametric procedures have been used.

Plutonium Transport Processes

In an effort to understand and possibly predict the behavior of plutonium in the environment, several studies dealing with transport processes have been undertaken. These include measurements of aerial

deposition rates, soil erosion, plutonium export resulting from biological processes, and plant uptake. Data from these studies have not been fully analyzed, but several noteworthy points have emerged.

Aerial deposition rates over the past two years have averaged about 20 dpm/m²-day in macroplot 1, based upon data from 21 wet pot collectors. This value is in good agreement with mean air concentrations over the area of about 10^{-15} μ Ci/cm³ and a reasonable deposition velocity figure of 5 cm/sec. This deposition rate would provide an increment to the soil inventory of only 0.006% per year. A deposition rate of 20 dpm/m²-day would theoretically lead to a ²³⁹Pu concentration in vegetation of about 2 dpm/g, assuming a mean biomass of 220 g/m² and an effective half-time of 14 days.

Since over 99% of the plutonium in the ecosystem is associated with the soil, erosional processes offer the potential of dispersing significant plutonium contamination. Such processes include water erosion, both gully and sheet forms, and resuspension by winds. These processes may be enhanced by soil disturbance from natural phenomena such as animal activity and needle ice formation. We attempted to measure soil erosion by tracing through time the location of soil particles tagged with ⁵⁹Fe and by recording beta counts from ⁹⁰Sr sources buried at fixed positions beneath the soil surface. These data gave no indication of measureable soil erosion over the duration of the study in undisturbed, well-vegetated areas. If surficial erosion were to occur, the fact that most of the soil plutonium is greater than a few mm deep lends assurance that significant quantities of the element would not move. Severe gully erosion could move substantial quantities of plutonium, but this has not been observed except in steep, disturbed areas.

The potential of mammals for dispersing plutonium has been investigated in considerable detail by quantifying certain activities of pocket gophers and mule deer. Burrowing and mound building by small mammals exposes contaminated soil to erosional forces. Pocket gophers at Rocky Flats form mounds and dig tunnels and burrows, and move large quantities of soil in the process. In a plutonium-contaminated area of 2.6 ha, gophers constructed about 4.5 mounds/day, representing about 5,000 kg of soil per year which contained about 85 μ Ci of ²³⁹Pu. Most of the

mound soil originated from the 10-15 cm horizon. The fate of plutonium in exposed mounds has not been studied, but observations suggest that most remains in the immediate area of the mound. Mule deer ingest plutonium associated with vegetation and soil and transport the material to the surrounding area, where most is excreted with fecal material. Measured soil ingestion rates of up to 30 g/day and ingestion of contaminated vegetation (1100-1400 g/day) lead to an estimated probable annual intake of 0.07 μCi per deer feeding around the contaminated areas at Rocky Flats. An estimated maximum possible annual intake per deer is 13 μCi if the animal were confined entirely to macroplot 1. Around 100 deer utilize the Rocky Flats buffer zone, but their activity is seldom concentrated in the more highly contaminated areas. An upper limit estimate of the proportion of total plutonium dispersed from macroplot 1 by mammalian activities is of the order of 0.1 to 1% per decade. A more probable estimate is 0.001 to 0.01% per decade.

We have attempted to measure the degree of plutonium uptake from soil to plants through root assimilation, and the comparative importance of root uptake and aerial deposition. Intact, contaminated soil blocks and columns from macroplot 1 were transplanted either to an uncontaminated area northwest of the Rocky Flats Plant or to a greenhouse at Colorado State University. Vegetation growing on these blocks and columns has been clipped at various time intervals and assayed for plutonium to get an estimate of root uptake. The values have run of the order of 0-4 dpm/g dry vegetation, which is similar to the values for ultrasonicated vegetation from macroplot 1, but substantially less than unwashed vegetation from macroplot 1 which averaged about 60 dpm/g. These facts strongly implicate aerial deposition as the major transport mechanism. However, other data do not support this concept. For example, uncontaminated soil blocks placed in contaminated macroplot 1 generated vegetation that was also of the order of 0-4 dpm/g. Also, aerial deposition rates as measured by wet pot collectors in macroplot 1 would only account for about 2 dpm/g, assuming a retention half-time of 14 days for plutonium on vegetation surfaces. Because of this discrepancy and our inability to explain it, we cannot yet draw a definite conclusion as to the relative importance of root uptake and aerial deposition.

Comparative Behavior of ^{238}Pu and ^{239}Pu

It has been consistently observed that the isotopic activity ratio ($\text{dpm } ^{239}\text{Pu/g} \div \text{dpm } ^{238}\text{Pu/g}$) is of the order of 65 in surface soil samples. However, the isotopic ratio (IR) has been observed to decrease with soil depth. Also, IRs for animal tissues have been substantially lower than for surface soil, running of the order 5-30. Our initial interpretation of this observation was that ^{238}Pu is more mobile than ^{239}Pu and penetrates more rapidly into the soil and through biological membranes more efficiently. Such isotopic discrimination has some theoretical basis because it has been demonstrated that particles of ^{238}Pu break down in aqueous systems much more rapidly than ^{239}Pu particles because of the higher specific activity of ^{238}Pu . The more rapid breakdown is consistent with the process of "radiolytic weathering."

An argument against the concept of differential rates of particle breakdown from radiolysis is the fact that we have no reason to suspect that ^{238}Pu particles occur distinct from ^{239}Pu particles. Plutonium particles in the environs of Rocky Flats likely contain both isotopes intimately mixed in a reasonably predictable ratio. Thus, the radiolytic effect of ^{238}Pu decay, manifested in recoil energy of the ^{234}U nucleus, should dislodge fragments containing ^{239}Pu as well as ^{238}Pu . Another argument is that much of the plutonium in soil at Rocky Flats appears monomeric and there is little if any reason to suspect isotopic discrimination for monomeric plutonium.

Recently, we have examined several data sets by plotting the IR as a function of total plutonium in the sample. Each data set has shown a similar pattern, with IR values below 50 nearly always associated with samples containing less than 10 dpm of plutonium. Above 10 dpm, the IR appears independent of the total plutonium in the sample. Therefore, it seems clear that the IR is biased by sample activity, the bias becoming more severe as the total Pu content decreases below 10 dpm. Unaccounted-for interference under the ^{238}Pu peak of the alpha spectrum could explain the bias observed, but this possibility has not been explored fully. Formulation of conclusions regarding the isotopic ratio variations therefore awaits further data analysis.

Ecological Consequences of Plutonium Contamination at Rocky Flats

The contamination of a portion of the terrestrial environs of Rocky Flats with substantial quantities of plutonium provided the opportunity to search for possible ecological effects resulting from the presence of the element. Gross population effects from the levels of plutonium present in the environment were not expected on the basis of predictions and extrapolations from laboratory studies. However, prior to our studies, a lack of population effects had not been demonstrated and furthermore, extrapolation from the laboratory to the natural environment is frequently not a valid practice, owing to complexities and interactions which occur in nature but not in the laboratory.

We conducted studies which permitted comparisons of various biological measurements and pathological data between ecologically similar study areas at Rocky Flats of widely varying plutonium levels. Soil in the principal study areas ranged from 100 to over 20,000 dpm $^{239}\text{Pu}/\text{g}$ in the upper 3 cm (2-400 $\mu\text{Ci}/\text{m}^2$). In addition, comparative data were obtained from control areas, containing only world-wide fallout plutonium of the order of 0.1 dpm/g (0.002 $\mu\text{Ci}/\text{m}^2$). Biological measurements such as vegetation community structure and biomass; litter mass; arthropod community structure and biomass; and small mammal species occurrence, population density, biomass, reproduction, and physical size of whole carcass and organs were made. In addition, pathological examinations of small mammals, including x-ray for skeletal sarcomas, microscopy for lung tumors, and necropsy for general pathology and parasite occurrence were carried out.

While minor differences in certain biological attributes between study areas were observed, none could be related to plutonium levels. Pathological conditions and parasites were found in some rodents, but occurrence frequencies between control and contaminated areas were similar. No evidence of cancers or other radiogenic diseases was found. These observations and measurements, combined with intensive field observations over a period of five years, leads us to conclude that plutonium contamination at Rocky Flats has not produced demonstrable ecological changes. Furthermore, the levels of plutonium observed in tissues of plants and animals in contaminated areas were insufficient to produce

the doses that would be required to produce obvious biological changes. Subtle biological changes, such as chromosome aberrations, cannot be ruled out at Rocky Flats. However, even if chromosome aberration frequencies were increased in the more highly contaminated areas, population-level changes would likely not persist because of the surrounding reservoir of normal genetic information.