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DISSERTATION

PLUTONIUM IN A GRASSLAND ECOSYSTEM

Submitted by

Craig Alan Little

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In partial fulfillment of the requirements

for the Degree of Doctor of Philosophy

Colorado State University

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COLORADO STATE UNIVERSITY

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WE HEREBY RECOMMEND THAT THE THESIS PREPARED UNDER OUR SUPERVISION
BY CRAIG ALAN LITTLE
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ABSTRACT OF DISSERTATION

PLUTONIUM IN A GRASSLAND ECOSYSTEM

This study was concerned with plutonium contamination of grassland at the U. S. Energy Research and Development Administration Rocky Flats plant northwest of Denver, Colorado. Of interest were: the definition of major plutonium-containing ecosystem compartments; the relative amounts in those compartments; how those values related to studies done in other geographical areas; 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 from each of two macroplots. Small aliquots (5 g or less) were analyzed by a rapid liquid 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 the sample, and particle size of the sieved soil samples. The soil data suggested that the distribution of contamination largely resulted from physical transport processes. A mechanism of agglomerated submicron plutonium oxide particles and larger (1-500 μm) host soil particles was proposed.

Concentrations of Pu 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 samples were highly skewed, kurtotic, and quite variable, having coefficients of variation (standard deviation/mean) as high as 600%. Bone Pu concentrations were lower than other tissues. Hide, GI, and lung were generally not higher in Pu than kidney, liver and muscle.

In all compartments sampled, the expressed variation relative to the mean was quite high; coefficients of variation (standard deviation/mean) of up to 600% were found. The variation and the presence of zero or less than detectable values required the use of nonparametric statistical tests in many cases.

Isotopic ratios of ^{239}Pu to ^{238}Pu by activity concentration in surface soil were about 65. Litter and vegetation isotopic ratios were similar to soil suggesting that the contamination was largely surficial. Arthropod and small mammal tissue isotopic ratios were much lower than soil which implies the possibility that the isotopes were incorporated into the animal bodies and ^{238}Pu taken up at a higher rate. The isotopic ratio of bone was lower than for other tissues further suggesting different uptake rates for the two isotopes.

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INTRODUCTION

The partial destruction of Nagasaki, Japan, on August 9, 1945, by an atomic weapon was the first public appearance of the element number 94. The use of plutonium for weaponry and reactor fuel has steadily increased since that date. Much of this growth resulted from the proliferation of nuclear weapons during the Cold War between the U.S. and the U.S.S.R. The multiplication of nuclear weapons and the necessity to maintain them led to the origination of the U. S. Atomic Energy Commission (AEC) Rocky Flats plant near Denver, Colorado, during 1950. Again, as a result of the prevailing political climate, secrecy was the order of daily operations at the installation. Following an expensive fire involving plutonium in March, 1969, public awareness of Rocky Flats began to rise.

The efforts of the Colorado Committee on Environmental Information, headed variously by H. P. Metzger and E. A. Martell, in sampling for plutonium in soil around Rocky Flats and publicizing the results created a public concern about the installation (Martell et al., 1970). A study at Rocky Flats by the AEC's Health and Safety Laboratory (HASL) of soil contamination suggested three "plausible sources for the off-site plutonium" other than the 1969 fire:

1. a less expensive fire of September, 1957;
2. leaking drums of contaminated cutting oil in a storage area in the southeast corner of the plant complex;
3. chronic, low-level plutonium stack effluent.

The 1957 fire and the leaking barrels were judged to be the most probable sources (Krey and Hardy, 1970). The drums were placed in outside storage beginning in 1958. Following detection of initial leakage, and sporadic removal of some barrels, all the barrels were removed in 1968.

In 1971 the AEC expressed a desire to understand the distribution of plutonium in the grassland surrounding the Rocky Flats installation and funded the research resulting in this report. The need of ecological research at Rocky Flats was demonstrated by several factors. First and foremost, there was and is great concern for the hazards of plutonium on human as well as animal populations. This concern, coupled with much uncertainty about the ecological behavior of plutonium, is potentially frightening. Further, the continuing encroachment of housing developments to within a few kilometers downwind of the highest levels of plutonium contamination at Rocky Flats magnified the concerns.

The objectives of this thesis, generally stated, are to describe spatially and ecologically the patterns of plutonium concentration in plots representative of the grassland surrounding the facility and to infer or postulate mechanisms of plutonium transport. Specific questions associated with these objectives are:

1. What are the principal ecological compartments for plutonium?
2. What fraction of the total plutonium in two study areas is within these major ecological compartments?
3. How do plutonium levels and concentration ratios in the various ecosystem compartments compare to reported values from other studies?

4. Do the two predominant plutonium isotopes, ^{238}Pu and ^{239}Pu , differ in ecological behavior; if so, what is the magnitude of that difference?
5. What mechanisms or processes of plutonium transport may be postulated to explain the observed data?

These questions were approached by sampling soil, vegetation, litter, arthropods, and small mammals from various locations on the Rocky Flats plant site to provide estimates of compartmental mass and plutonium concentration.

HISTORICAL PERSPECTIVE

The following section on the discovery of plutonium is included to provide important perspective on element 94 to uninformed readers.

Discovery of Plutonium

One can begin the story of the transuranium elements with the discovery by E. Fermi and his group in 1934 that the irradiation of uranium by neutrons produced a number of radioactive substances. According to Seaborg (1961), the Fermi group felt these substances to be transuranium elements "because they decayed by beta emission, a process that might result in elements of higher atomic number than uranium, and because they differed in chemical properties from all the known elements in the region of uranium."

Early in 1939, O. Hahn and F. Strassman found that at least some of the radioactive substances resulting from the bombardment of uranium with neutrons were isotopes of barium and lighter elements. Most of the radioactivity previously assigned to "transuranium elements" was later found to be fission products. Further investigation of the fission process and fission products resulted in the discovery of the first true transuranium element, neptunium, element number 93 (Seaborg, 1961).

In 1940, A. C. Wahl, a graduate student, with J. W. Kennedy and G. T. Seaborg in the Department of Chemistry at the University of

California, Berkeley, began experiments bombarding uranium oxide with 16 MeV deuterons from a 60-inch cyclotron. They found that an alpha-emitting radioactive substance gradually appeared in the element-93 fraction previously chemically separated from the other target material. It was in these experiments that the first positive identification of element number 94, later named plutonium, was accomplished. It was later understood that the production reactions of the growing radioactive substances were as follows:

Shortly thereafter with the added collaboration of E. Segre, the search for the isotope theorized to be of major importance, ${}^{239}\text{Pu}$, was successful (Seaborg, 1961).

On March 28, 1941, the value of ${}^{239}\text{Pu}$ as a nuclear energy source was established with the first demonstration that the nuclide underwent fission when bombarded by slow neutrons produced in the 37-inch cyclotron of the University of California. The knowledge that the ${}^{239}\text{Pu}$ underwent fission by thermal neutrons, while showing that all neutrons emitted in the process could initiate further fission, established the great value of this isotope. This discovery resulted in the creation of the wartime Plutonium Project for the production of plutonium on a large scale and the eventual use of plutonium in the weapon at Nagasaki (Seaborg, 1961).

Following the discovery and study of plutonium in trace quantities, the amount of plutonium in existence increased tremendously, primarily because of its use in nuclear weapons and reactors. Plutonium may be produced from ^{238}U in a reactor with any type of neutron spectrum: thermal, intermediate, or fast. Plutonium-239 is produced as a result of neutron capture by ^{238}U and subsequent beta decay of ^{239}U and ^{239}Np . The isotopes of mass 240, 241, 242, and 243 plutonium are formed from successive neutron captures (Leonard, 1967). The short halflife of ^{243}Pu essentially terminates the plutonium isotope production at ^{242}Pu . A complete listing of these isotopes is included later.

Properties of Plutonium¹

Plutonium is a silvery white metal which melts at 639.5°C . This reactive substance oxidizes readily in warm, moist air. Finely divided amounts of plutonium may be pyrophoric. At warm temperatures plutonium will react with oxygen, halogens, nitrogen or ammonia, hydrogen, and carbon monoxide to form oxides, halides, nitrides, hydrides, and carbides, respectively.

Plutonium can exhibit five oxidation states from plus three (III) to plus seven (VII). The divalent state may exist in certain solid compounds. The four common oxidation states correspond to the cations Pu^{3+} , Pu^{4+} , PuO_2^{+} and PuO_2^{2+} .

The heptavalent state has been prepared by oxidation of Pu(VI) by ozone in alkaline solution. The four lower oxidation states are

¹The information in this section is largely from Taylor (1973).

stable in solution, and it is possible for all four states from Pu(III) to Pu(VI) to co-exist in the same solution. Obviously, the possible existence of concurrent multiple oxidation states leads to many complexities in plutonium chemistry.

Plutonium dioxide, PuO_2 , is one of the most important of all plutonium compounds, particularly with regard to environmental contamination. PuO_2 is formed when plutonium metal is burned in oxygen or when oxygen-containing compounds such as plutonium oxalate or plutonium peroxide are heated in vacuo at 1000°C . The refractory nature of PuO_2 generally increases as the preparation temperature increases. Other plutonium oxides which exist include the sesquioxide, Pu_2O_3 , and a possible monoxide, PuO , with plutonium in the divalent state.

Plutonium forms nitrate, sulfate and phosphate compounds. The following compounds and oxidation states exist: Pu(IV) and Pu(VI) nitrates, Pu(III) and Pu(IV) sulfates and Pu(III), Pu(IV), Pu(VI) and probably Pu(V) phosphates. Little is known about most of these compounds, but the phosphates are used during some plutonium processing.

In aqueous solutions the positively charged plutonium ions are not free, but associated with water in the hydrated form as the hydronium ion. Due partly to their high ionic charges and relatively small radii, plutonium ions undergo extensive hydrolysis in aqueous solution. The tendency towards hydrolysis occurs in the following order: $\text{Pu(IV)} > \text{Pu(VI)} > \text{Pu(III)} > \text{Pu(V)}$. Hydrolytic reactions usually occur more readily at higher pH, and, with complex formation,

hydrolysis is very important in plutonium chemistry in biological systems.

Besides association with water, plutonium ions may associate with other ions, molecules or groups of ions to form complexes. This process is closely related to hydrolysis, and thus, also depends on ionic potential. Therefore, the small and highly charged Pu^{4+} ion plays an important role in the solution chemistry of Pu(IV). The oxidation states of plutonium form complexes in the following decreasing order: $\text{Pu(IV)} > \text{Pu(III)} > \text{Pu(VI)} > \text{Pu(V)}$.

Since the discovery of ^{238}Pu by Seaborg and his co-workers in 1940, fifteen further isotopes of plutonium have been synthesized. All of these are radioactive, most of them decaying by alpha particle emission with half lives ranging from 0.18 seconds to 76 million years. Table 1 contains a complete listing of these isotopes.

From the standpoint of power production and nuclear weapons ^{239}Pu is the most important of the isotopes of plutonium. However, ^{238}Pu may ultimately be more important in the environmental and biological sense because of its specific activity which is 280 times higher than ^{239}Pu .

Biological Research

Concern about the biological metabolism and effects of plutonium grew soon after the discovery of the element. In late 1942, a program was initiated at the University of California, Berkely, under the leadership of J. Hamilton, to determine the metabolic properties of plutonium in rats. The actinide elements ^{239}Np , ^{241}Am , ^{242}Cm , and the

Table 1. Properties of plutonium isotopes^a (adapted from Taylor, 1973, after Lederer *et al.*, 1967).

Isotope	Half-life	Specific alpha activity(Ci/gm)	Principal radiations(energy in MeV)
²³² Pu	36 m	4.50x10 ⁵	α 6.59(2%); EC(98%)
²³³ Pu	20 m	4.03x10 ⁵	α 6.31(0.1%); EC(99%)
²³⁴ Pu	9.0 h	8.94x10 ⁴	α 6.20(4%); 6.15(1.9%); EC(94%); γ(Np x-rays)
²³⁵ Pu	26 m	909	α 5.86(0.003%); EC(99+%); γ(Np x-rays)
²³⁶ Pu	2.85 y	532	α 5.77(69%), 5.72(31%); γ UL x-rays, 0.048(.31%), 0.109(0.012%); e ⁻ 0.028, 0.043
²³⁷ Pu	45.6 d	40.3	α 5.66, 5.37(3.3x10 ⁻⁴ %); γ Np x-rays 0.060(5%); e ⁻ 0.026, 0.032, 0.038, 0.042, 0.056
^{237m} Pu	0.18 s	--	γ Pu L x-rays 0.145(2%); e ⁻ 0.125(75%), 0.140(23%)
²³⁸ Pu	86.4 y	17.4	α 5.50(72%), 5.46(28%); UL x-rays 0.099(8x10 ⁻³ %), 0.150(10 ⁻³ %); 0.77(5x10 ⁻⁵ %); e ⁻ 0.024, 0.039
²³⁹ Pu	24,390 y	6.14x10 ⁻²	α 5.16(88%), 5.11(11%); γ 0.039(.007%), .052(.02%), 0.125(.005%), .374(.0012%), .414(.0012%), .65(8x10 ⁻⁵ %), .77 (2x10 ⁻⁵ %); e ⁻ .008, .019, .033, .047
²⁴⁰ Pu	6580 y	.227	α 5.17(76%), 5.12(24%); γ UL x-rays 0.65(2x10 ⁻⁵ %), e ⁻ 0.026, .040
²⁴¹ Pu	13.2 y	--	β ⁻ 0.021 max (99+%), α 4.90, 4.85 (2.3x10 ⁻³ %); γ UL x-rays 0.145 (1.6x10 ⁻⁴ %)
²⁴² Pu	3.79x10 ⁵ y	3.90x10 ⁻³	α 4.90(76%), 4.86(24%); γ UL x-rays
²⁴³ Pu	4.98 h	--	β ⁻ 0.58 max; e ⁻ 0.019, 0.036; γ Am L x-rays 0.085(21%), 0.381(0.7%)
²⁴⁴ Pu	7.6x10 ⁷ y	1.93x10 ⁻⁵	α 4.58
²⁴⁵ Pu	10.1 h	--	β ⁻
²⁴⁶ Pu	10.85 h	--	β ⁻ 0.33 max(10%), 0.15 max; e ⁻ 0.020, 0.038, 0.055, 0.156; γ Am L x-rays 0.044(30%), 0.180(10%), 0.224(25%)

^aEC = electron capture.

fission product ^{90}Sr were later included in these experiments. Some of the findings of this work included: carriers of the nuclide were important in the nuclide's biological fate; the actinide nuclides were not absorbed in significant quantities from the digestive tract; the actinide nuclides were found to migrate to the skeleton where they were surficially deposited and tightly bound; and liver deposition was important (Hamilton, 1948).

As the metabolic information increased, it became obvious that the metabolic distribution of plutonium was closely related to and determined by the following factors: the chemical form of the plutonium administered, the route of its administration, the species of animal exposed, and the age of the individual animal.

Comparative plutonium metabolism in man and experimental animals was studied. In April, 1945, the first 12 of 16 terminal human patients were given acute intravenous injections of plutonium (as $^{239}\text{Pu(IV)}$ citrate complex) as a joint effort of the Los Alamos Scientific Laboratory and the University of Rochester School of Medicine and Dentistry. These experiments helped determine that in man, as in experimental animals, the skeleton and liver were principal sites for deposition of plutonium. The amount in bone and liver decreased slowly in man (i.e., evidenced a long biological half-life) while plutonium in the rodent liver decreased more rapidly. Investigators found that less than 10% of the plutonium in human patients was excreted by those surviving during the five years after the injections (Stover and Stover, 1972). The earliest effects studies with mice showed deleterious effects of plutonium which included the

occurrence of osteosarcomas and gross skeletal damage (Lisco et al., 1947). The acute toxicity of plutonium in dogs was shown by the appearance of massive damage to the hematopoietic system (Painter et al., 1942).

Later studies at the University of Utah showed dose-dependent granular leukocyte depression with beagles injected with 0.016-2.8 $\mu\text{Ci/kg}$ of ^{239}Pu . The higher dose levels led to sustained lymphopenia and a transient early thrombocytopenia and anemia (Dougherty, 1972).

The same data indicated the toxicity of plutonium through induction of osteosarcomas. The incidence of sarcoma fatalities decreased to 33% at the lowest dosage (0.016 $\mu\text{Ci/kg}$). The post-dosage time to tumor appearance was inversely related to dosage level (Jee, 1972).

In comparison to radium-226, plutonium-239 is five to ten times more toxic relative to osteosarcoma induction in beagle dogs based on the same total energy delivered to bone. This difference results from plutonium deposition on bone surfaces (Mays and Dougherty, 1972). Two factors contribute to the opinion that plutonium is more toxic to humans than dogs with regard to osteosarcomas: a lower surface-to-volume ratio in human trabecular bone and a lower rate of surface deposit burial by new bone in humans (Lloyd and Marshall, 1972). Because of these two characteristics more ^{239}Pu energy is deposited at the bone surface near cells, whereas most of the ^{226}Ra alphas are deposited in bone mineral.

The acute and chronic effects of plutonium inhalation was studied in dogs. The earliest response in animals inhaling 0.2 to 1.0 μCi of

of PuO_2 was seen to be lymphopenia. Forty of the 65 dosed dogs died between 55 to 200 days after exposure because of plutonium-induced pulmonary insufficiency. Twenty-two dogs surviving more than 1600 days had malignant lung tumors (Park et al., 1972).

Studies with rats suggest lung cancer as the most sensitive response of PuO_2 inhalation. Osteosarcomas developed in rats which inhaled more soluble forms of plutonium (Moskalev, 1972).

In summary, much is known about plutonium metabolism and toxic effects. Plutonium metabolism is affected by the form of the element, its route of entry, and the age and species of the subject. The organs and tissues affected by plutonium also depend on the element's entry route and form. Injected plutonium in sufficient quantities may result in bone tumors; inhaled insoluble plutonium may produce lung tumors; inhaled soluble plutonium may lead to both lung and bone tumors. A more detailed summary of plutonium biomedical research is offered by Bair and Thompson (1974).

Sources of Environmental Contamination

Plutonium may be introduced into the biosphere in a number of ways: either a high-explosive detonation or fission of a nuclear explosive device, a routine or accidental release from a nuclear industrial facility, burn-up of a plutonium-fueled space power source during atmospheric reentry, or a transportation accident during shipment of plutonium (Stannard, 1973). Sites of note which have been contaminated by the above mechanisms are listed in Table 2.

Table 2. Land areas of note contaminated by plutonium.^a

Location	Facility	Contamination Mechanism
Southern New Mexico	Trinity Site	Nuclear detonation
Enewetok Atoll, South Pacific	Pacific Proving Ground	Nuclear detonation
Central Nevada	Nevada Test Site	Nuclear detonation
Nagasaki, Japan	--	Nuclear detonation
Palomares, Spain	--	Non-nuclear detonation
Thule, Greenland	--	Non-nuclear detonation (bomber crash)
Central Nevada	Nevada Test Site	Non-nuclear detonation
Northern New Mexico	Los Alamos Scientific Lab.	Accidental or routine releases
Eastern Washington	Hanford Reservation	Accidental or routine releases
Western South Carolina	Savannah River Plant	Accidental or routine releases
Southern Idaho	Idaho National Enginrg. Lab.	Accidental or routine releases
Central Colorado	Rocky Flats Plant	Accidental or routine releases
Southern Hemisphere	--	Ablated ²³⁸ Pu-powered generator (satellite)

^aInformation from Hanson (1975) and Stannard (1973).

After a land area has been contaminated, the forces of nature and/or man may cause the contaminated site to serve as a source of further contamination to adjacent, uncontaminated areas. Wind and water are prime influences in this regard. Mechanical disturbance by humans and other animals potentially serve as forces of further contamination.

Plutonium in Soil

Plutonium associated with soil has been studied by many investigators during the past three decades. Characteristics of the soil which influence plutonium adsorption onto soil particles include the pH of the soil, the amount and type of various organic substances, and the soil particle sizes available for attachment. Once the plutonium is attached to soil particles, it may be moved by agents including gravity, wind and water. Several studies in various biomes of contaminated soil areas have determined the profiles of plutonium downward through the soil and suggested causes for the results.

Some of the first work on plutonium movement in soil concerned possible plutonium contamination from subsurface waste disposal cribs at the Hanford Reservation (Rhodes, 1957a,b). In laboratory studies, adsorption of trace quantities of plutonium on soil was found to be pH-related in that 98-100% of the plutonium was adsorbed at a pH range of 2 to 8 while only 80% was adsorbed at a pH range of 8 to 12. Rhodes concluded that plutonium was adsorbed onto soil particles at $\text{pH} > 2$ as positively charged polymers of the $\text{Pu}(\text{OH})^{+n}$ type. The

lower adsorption at higher pH apparently resulted from change in the polymers' characteristics.

Rhodes (1957a,b) further found that high concentrations of inorganic salts appeared not to affect the plutonium adsorption on soil, but equivalent concentrations of organic salts, e.g. Ca and Ba acetate, reduced adsorption to nearly 60%. Rhodes concluded that a complex ion of plutonium acetate was formed. In their work concerning movement of plutonium through Los Alamos tuff, Christenson and Thomas (1962) mentioned the possibility that organic plutonium complexes were in some way responsible for transportation of plutonium to depths as great as 8.5 m.

Knoll (1969) studied the effects of organic materials on removal of radionuclides adsorbed on soil in low-level wastes. Significant quantities of ^{239}Pu , ^{85}Sr , and ^{241}Am were removed by two of the organic solutions tested (hydroxyacetic acid and a mixture of di-(2-ethylhexyl) phosphoric acid and tributylphosphate in a commercial mixture of straight chain hydrocarbons). Hydroxyacetic acid alone removed all of the above nuclides, but not ^{137}Cs . Removal was directly related to the hydroxyacetic acid concentration. More recently, Bondietti et al. (1975) explored the interaction of plutonium with complexing agents in soils. They found that reduction of Pu(IV)-Pu(VI) and Pu(IV)-Pu(V) couples by phenolic materials such as humic substances and reducing sugars such as uronides occurred readily. Since materials such as those noted are ubiquitous in the natural environment, plutonium oxidation states higher than Pu(IV) were not likely.

Two years following a detonation at the Nevada Test Site (NTS), Mork (1966) reported that the major portion of the plutonium was found associated with particles greater than $44\text{ }\mu\text{m}$ in diameter. Results of a study by Tamura (1975) may conflict with those of Mork (1966). Tamura used soil samples from three sites to study plutonium association with soil particle sizes. The three soil sampling sites were the Nevada Test Site (NTS), Mound Laboratory (ML), Miamisburg, Ohio, and Oak Ridge National Laboratory (ORNL), Oak Ridge, Tennessee. Tamura found that the plutonium in the samples from NTS and ML occurred predominantly in the silt ($2\text{--}53\text{ }\mu\text{m}$) and clay ($<2\text{ }\mu\text{m}$) size fractions, respectively. The clays of ML, however, exist as aggregates and behave as silt particles in the natural state. In the ORNL sample the plutonium was more evenly distributed in the silt and clay sizes, but the clay size was the higher contributor.

Tamura (1975) also examined the plutonium solubility in the various soil samples. Citrate extractions showed the highest plutonium solubility in the ML sample, followed by the ORNL sample, and least in the NTS sample. There was a direct relationship between citrate solubility and published vegetation/soil concentration ratios for the NTS and ORNL samples which suggests a useful technique for predicting potential uptake by plants.

Once plutonium is attached to soil particles, it may be mobilized by several natural forces, including gravity, water and wind.

As indicated by moisture data, rates of liquid flow, and visual inspection, Christenson and Thomas (1962) in their study at Los Alamos, felt that most of the plutonium movement occurred along

fissures in the tuff. Essington et al. (1975) suggested that it was possible that particles containing plutonium migrated downward from the surface into the fractures at the NTS.

Hajek (1966) studied plutonium mobility in the soil beneath the same subsurface storage cribs studied by Rhodes at Hanford. He predicted that movement of plutonium by contact diffusion would be less than 10 cm after 2.4×10^5 years. Hajek further concluded that more than 900 cm of infiltrating groundwater would be required to leach 0.1% of the plutonium from the surface layer. Based on an assumed, but to him unknown, fraction of the annual precipitation available for leaching, Hajek estimated several half lives of ^{239}Pu would occur before the nuclide reached groundwater reservoirs.

A study by Nyhan (1973, cited by Hanson, 1975) at the Trinity site revealed a direct correlation of water percolation rates with plutonium distributions. Nyhan found that light, porous soils with water intake rates of 192 mm/hr had nearly uniform plutonium throughout the upper 30 cm depth, contrasted with intake rates of 92 mm/hr in clay soils having 90% of the Pu concentrated in the upper 2.5 cm. Bondietti et al. (1975), however, felt that leaching losses of plutonium in natural systems would be insignificant compared to physical movement of contaminated soil particles.

Particularly with regard to nuclear detonations at the NTS, several investigators (Larson et al., 1951; Olafson et al., 1957; Olafson and Larson, 1963) reported the principal mode of horizontal or surface transport of plutonium to be resuspension of plutonium-contaminated particles by wind. Krey and Hardy (1970), studying the

contamination patterns of soil at Rocky Flats, felt that the strong westerly and northwesterly winds of the areas were the probable causes of most plutonium movement at the facility.

Profiles of plutonium concentration with depth have varied from site to site. Christenson and Thomas (1962) found Pu to depths as great as 8.5 m in the highly heterogeneous Los Alamos tuff. Two years following detonations at the NTS, Mork (1966) found little movement of plutonium downward through the soil.

Romney et al. (1970) found considerably more downward plutonium movement in soil 10 years following detonation at the NTS than was observed at the Trinity site after the same time interval. These workers believed that weathering processes contributed more to the downward migration than solution chemical processes. Most of the data from the NTS reported by Essington et al. (1975) evidenced reasonably smooth profiles with depth. Nearly one-half the plutonium profiles contained greater than 95% of the total profile plutonium in the top 5 cm. Even so, in almost all of the profiles reported, plutonium was found to a depth of 25 cm.

Studies by several investigators (Olafson et al., 1957; Olafson and Larson, 1963) revealed relatively immobile plutonium at the Trinity site. Hakonson and Johnson (1973) found relatively uniform Pu concentrations throughout the upper 7.5 cm of several soil cores obtained out to a distance of 24 km from ground zero (GZ) at the Trinity test site 27 years following the detonation. Further from GZ, however, the Pu was found to be increasingly concentrated in the upper 2.5 cm. Krey and Hardy (1970) in their often-cited report,

HASL-235, stated that 9-61% of the Pu in the top 20 cm was at a depth greater than 5 cm in samples from near the Rocky Flats installation. However, the two samples with the highest fraction of plutonium below the 5 cm depth were in city parks of Denver, Colorado, where the practice of liming and fertilizing was thought to enhance downward movement.

McLendon et al. (1975) sampled soil (0-15 cm depth) and biological components of the ecosystem surrounding the Savannah River Plant, Aiken, South Carolina. They stated that plutonium concentrations of the soil cores varied widely, but generally reflected the sampling distance from the plutonium sources at the Savannah River Plant. Plutonium concentrations of resuspendible materials showed the same relation to source-to-sample distance, but had generally higher concentrations than the soil.

In summary, several concepts of soil-plutonium interaction arise. Researchers have found that plutonium adsorption onto soil particles is influenced by the soil type, the amount and type of organic substances in the soil, and inversely related to pH and the soil particle sizes available for Pu attachment. Most workers believe that physical, rather than biological or chemical, forces have the greatest effect on plutonium redistribution throughout the environment. Of these physical forces, including gravity, water and wind, wind is probably the most effective at mobilizing the soil particles. In spite of the large effect of physical interactions, there is some potential for biological-chemical interactions to allow environmental redistribution.

Plutonium and Plants

With regard to plutonium associated with vegetation, the overriding impression is that uptake of plutonium via the roots is very low. The soil to plant uptake magnitudes of plutonium relative to other nuclides has the following arrangement (Nishita *et al.*, 1965):

Effects of various factors on the rate or magnitude of plutonium uptake by plants are: pH of the soil or growth medium and soil type gave various results; decrease in Pu concentration of the soil increased uptake; presence of chelating agents or organic material increased uptake; soluble forms of Pu were more easily uptaken; time increased uptake. Studies both in the lab and the field have determined uptake and/or contamination via aerial resuspension at several sites. For plants the term "concentration ratio" is defined as (activity/dry g plant) $\div$ (activity/dry g soil).

One of the earliest papers contained the observation by Jacobson and Overstreet (1948) that barley plants translocated very small portions of plutonium from clay suspensions to roots to leaves. Jacobson and Overstreet measured the percent plutonium translocated to roots and to leaves of 3-week-old barley plants from clay suspensions during a 24-hour period. The percentages of the original dose attached to the roots were 37.8, 24.6 and 20.3 for the PuO_2^{+2} , Pu^{+4} , and Pu^{+3} , respectively. The Pu fractions translocated to leaves were 0.010%, 0.0024%, and 0.00045% of the original dose for PuO_2^{+2} , Pu^{+4} , and Pu^{+3} , respectively. Of the nine radionuclides tested, ^{89}Sr had the highest percentage uptake, 1.6%, and plutonium the lowest.

Using the Neubauer seedling test to study the uptake of fission products by various plants, Rediske and Selders (1953) found that concentration ratios $[(\text{amount/g plant}) \div (\text{amount/ml solution})]$ ranged from 2×10^{-5} for Russian thistle leaves and stems to 3×10^{-4} for bean leaves for a nutrient solution of pH 6.0.

Newbould (1963) grew rye-grass in three different soils and found no significant difference in uptake even though the soils were of different pH. The uptake was on the order of 10^{-6} of the soil concentration. However, Rediske et al. (1955) found the concentration factor to increase by an order of magnitude as the nutrient culture pH changed from 7 to 4.

Wilson and Cline (1966) grew barley for 18 days after planting in 100 gm of soil containing $10 \mu\text{Ci } ^{239}\text{Pu/gm}$ soil as plutonium nitrate. The soil types used were: Ephrata fine sandy loam, a slightly alkaline soil; Milville silt loam, a calcareous soil; and Cinebar silt loam, a medium acid forest soil. The uptake of Pu varied from 6.0×10^{-7} to 2.2×10^{-6} of the total activity in the soil depending on the soil type used. Uptake was greatest from the acid, Cinebar soil.

Wildung and Garland (1974) grew barley using both a split-root soil-nutrient solution plant culture technique and a modified Neubauer technique to measure the effect of soil plutonium concentration (ranging from 7 to 550 pCi/g soil) on the uptake and distribution of the nuclide in the plants. They found that the "concentration factor" $[(\text{activity/g dry plant}) \div (\text{activity/g dry soil})]$ in the plants increased as the concentration of plutonium in soil decreased.

Either a concentration-dependent change in plutonium availability or a toxic effect of plutonium on the plant may have caused this inverse relationship. The authors felt that the latter explanation was most likely, but noted that microbial activity could potentially alter the plutonium form and availability in soil which could explain the observed effects.

In a recent paper, Romney et al. (1975) again analyzed data from the Nevada Test Site and found "vegetation/soil" ratios (same as concentration ratios, above) ranging from .024 for the TTR Double Track shot in 1963 to 0.44 for Area 13 contaminated in 1957. These workers felt that the plutonium concentration ratio in samples tended to increase as plutonium activity in soil decreased. Further, they felt this tendency might reflect the increasing proportion of finer particles of fallout material deposited at greater distances from ground zero.

Schulz et al. (1975) found uptake of plutonium and americium by wheat to be influenced by soil type. They used Panoche, Aiken and Yolo soils which are alkaline, acid and neutral, respectively. The plants grown in alkaline soil had 7- and 35-fold greater uptake of Pu than the neutral and acid soils, respectively.

Romney et al. (1970) found a five-fold increase in the plutonium concentration of ladino clover under glasshouse conditions and harvested annually during a five-year period. From soils with a concentration of 10^5 dpm/g plutonium concentration, the plutonium concentration increased from 3.1 dpm/g in 1958 to 22.6 dpm/g in 1962

harvested material. These workers felt the apparent increase might result from natural chelation of the residual plutonium by decaying roots inside the pots. Newbould (1963) also saw a time-related change in uptake. He grew rye-grass in three different soils and found somewhat greater uptake the second year than the first.

In support of the natural chelation idea, Romney et al. (1970) showed that plutonium uptake from some soils could be increased by the addition of chelating agents. They added 100 parts per million of DTPA (diethylenetriamine pentaacetic acid) and EDDHA (ethylene-diamine di-o-hydroxyphenylacetic acid) to soil of unknown plutonium concentration. Alfalfa grown in DTPA-treated soils showed a six-fold increase in uptake over the controls; uptake increase by the EDDHA-treated soils were insignificant.

Several studies have examined the influence of plutonium chemical form. Paine (1974) reported concentration ratios for barley grown in a complete growth medium ranging from 2.2×10^{-6} to 1.1×10^{-3} for plutonium in the medium as a soluble nitrate and 4×10^{-4} to 4×10^{-3} for plutonium as PuO_2 . In this case, the concentration ratio denominator had units of activity/ml growth medium. In a recent study Schulz et al. (1975) grew wheat on three soils previously contaminated by addition of plutonium in dilute chloride or nitrate solutions. Depending on the soil type used, the concentration ratio ranged from 4.4×10^{-8} to 3.8×10^{-6} . In alkaline Panoche soil, the nitrate form of the plutonium was taken up about 100 times greater than the chloride form.

Price (1972) grew tumbleweed (Salsola kali) and cheatgrass (Bromus tectorum) for two months under controlled conditions in pots containing Pu-spiked surface soil from near radioactive waste disposal sites at Hanford. For both plants, concentration ratios were significantly higher when the plutonium was added to the soil as complexes of organic acids (acetate, glycolate, oxalate, citrate), rather than a dilute nitric acid solution. Concentration ratios for tumbleweed ranged from 4.6×10^{-5} for the nitric acid to 3.1×10^{-4} for the citrate form. For cheatgrass the ratios ranged from 1.4×10^{-5} for the acetate to 5.1×10^{-5} for the citrate.

As mentioned above, the magnitude of uptake is generally quite low under laboratory or greenhouse conditions. However, under field conditions concentration ratios several orders of magnitude higher have been seen. Hakonson et al. (1973) reported plutonium concentration ratios from liquid waste disposal areas at the Los Alamos Scientific Laboratory of 10^{-2} to 10^{-1} for plants grown under field conditions on plutonium-containing sediments. McLendon et al. (1975) sampled ecosystem components from areas surrounding the Savannah River Plant, Aiken, South Carolina. McLendon et al. (1975) found concentration ratios for Camphor Weed (Heterotheca subaxillaris) of 0.218 on the plant site and 0.069 off-plant. For Honeysuckle (Lonicera japonica) the values were 0.93 on-plant and 0.077 off-plant.

The level of uptake of plutonium from soil by plants is best characterized as being low. Concentration ratios of 10^{-6} to 10^{-4} are most common. Concentration ratios from studies in the field are often higher than the levels expected from greenhouse studies. This

finding suggests that contamination of the plant surface, perhaps via windblown material, is important. This suggestion is compatible with the belief that physical forces are most important in redistribution of contamination.

Plutonium Uptake by Small Mammals and Arthropods

The biological behavior of plutonium in laboratory animals is well understood relative to what is known of these characteristics in native animals. Studies of plutonium translocation have been carried out at the University of Utah (Stover and Jee, 1972), Colorado State University (Bistline, 1973; Gomez, 1973; Schallberger, 1974) and at many ERDA laboratories. A number of previous animal studies were mentioned earlier in this chapter. Studies of small mammals contaminated by plutonium under field conditions have been limited in number. General conclusions reached include: low concentrations of Pu in small mammal tissues; low, but soil-dependent, concentration ratios; large amount of variation between individuals and between tissues within individuals; and resuspension, i.e., inhalation, as the most probable mechanism of small mammal contamination.

Romney et al. (1970) collected small mammals over a 10-year period in fallout areas at the Nevada Test Site. To obtain samples of sufficient size for ^{239}Pu analysis, tissues of kangaroo rats (Dipodomys microps and D. merriami) were pooled. Rat tissue ranged from a minimum of .02 dpm ^{239}Pu /sample to 61.28 dpm ^{239}Pu /sample. One sample of gastrointestinal tract (GI) contents had 2052 dpm ^{239}Pu .

Inter-tissue comparisons are difficult because of the author's practice of expressing contamination as dpm/sample. According to these workers, considerable variation in tissue burdens of adult jackrabbits (Lepus californicus) was found. Five individual jackrabbits sampled in an area of 20 dpm/g soil contained 0.27, 0.28, 0.57, 2.11 and 0.93 dpm ^{239}Pu per gram bone. Even greater variation was found in the gastrointestinal tracts (GI) of these individuals, reflecting their diet shortly prior to the collection. Only trace amounts of ^{239}Pu were found in muscle samples. Amounts found in lung were approximately the same as those found in the bone tissues. The data presentation of Romney et al. (1970) as dpm/sample made it impossible to calculate animal/soil values. However, these workers surmised that the relatively high levels of plutonium found in the GI indicated that "in addition to inhalation, these small mammals maintained contact with residual [soil] ^{239}Pu contamination through ingestion." The supposed source of the contamination was thought to be fallout particles ingested during eating, preening, digging, or taking dust baths.

Surveying the Trinity site 27 years following the detonation of the world's first atomic weapon, Hakonson and Johnson (1973) found considerable variability in rodent tissues. The pattern of concentration in rodent lungs, generally the highest of tissues sampled, was similar to that of soil and grass with respect to distance from ground zero of the shot. These workers felt that the lung deposition of plutonium suggested resuspension to be an important mechanism in the biological redistribution of the nuclide.

In similar work in the liquid waste disposal areas of the Los Alamos Scientific Laboratory, Hakonson et al. (1973) collected rodents for plutonium analysis. The plutonium concentration of the animal tissues collected at one sampling station in Mortandad Canyon had as much as three orders of magnitude variation. Lung and hide samples generally had the highest mean plutonium concentration. The investigators felt that these data justified speculation that inhalation of resuspended sediments was the main route of plutonium entry into the animals. The plutonium concentrations of liver and carcass samples were from 10^{-1} to 10^{-2} of the concentrations of hide and lungs.

McLendon et al. (1975) sampled grasshoppers and cotton rats (Sigmodon hispidus) around the Savannah River Plant, Aiken, South Carolina. Using data from the three grasshopper groups and eight adult rat samples, the calculated concentration ratio of animal plutonium to soil plutonium was about 10^{-2} . Plutonium concentration of juvenile and immature rat samples were similar to adult samples.

In summary, the few field studies of small mammal-plutonium interactions conducted to date have found concentration ratios relative to soil on the order of 10^{-2} to 10^{-1} , large amounts of variability in the data, and lung sample concentrations often higher than other tissues. Many workers agree that resuspension of plutonium in soil by wind is probably the most important mechanism of Pu association with wild animals. Relatively long lung (lower respiratory tract) retention half time estimates of 180-460 d in rats (Bair et al., 1961) are compatible with much of the Pu being found in the lungs of wild animals. Material cleared from the lung and swallowed during

the initial rapid clearance phase after lung deposition is absorbed from the GI in only small amounts, ranging from 5.8×10^{-7} to 1.9×10^{-2} , depending on the chemical form of the Pu (Bair, 1974). This finding agrees with the field studies which showed only small amounts of Pu in muscle tissues.

STUDY AREA AND PLUTONIUM SOURCE

The Rocky Flats installation, operated by Rockwell International for the United States Energy Research and Development Administration (ERDA) is located approximately 12 km northwest of the Denver, Colorado, metropolitan area at an elevation of about 1800 m (Figure 1). The location is almost equidistant from the cities of Boulder, 14 km to the north, and Golden, 15 km south.

The ERDA controlled area of the Rocky Flats plant is over 30 km². Of this, more than 95% is utilized as a buffer zone separating the public-at-large from plant production operations.

Rocky Flats is capped by glacial outwash 0.3 to 15 m thick and averaging 3 m thick (Malde, 1955). The outwash deposit is of Pleistocene age and consists largely of stony gravel and sand intermingled with clay (Scott, 1960). The gravel "is composed of about 60 percent quartzite, 20 percent schist and gneiss, 12 percent granite and pegmatite and 8 percent sandstone and siltstone" (Malde, 1955). Also according to Malde (1955), Rocky Flats soils are of pre-Wisconsin age.

The Soil Conservation Service (SCS) of the U. S. Department of Agriculture completed a soil survey of the Rocky Flats plant. The regions eventually chosen for macroplot locations were found to be similar. Both included the presence of Denver-Kutch-Midway soils with a 9 to 25% slope. According to Amen (1975), "Denver clay loams make up about 60 percent of the unit, Kutch clay loam about 25 percent and

Figure 1. Location of the Rocky Flats installation relative to nearby population centers.

Midway clay loam about 15 percent." The permeabilities of the Denver, Kutch and Midway soils are slow, slow and moderate, and the available water capacities are high, moderate and low, respectively. Most of the remainder of the macroplot 1 region was found to be moderately steep and steep gravelly land with 15 to 45% slopes, moderate to slow permeability, and moderate available water capacity. Most of the macroplot 2 remainder included Nederland cobbly gravelly sandy loam, sloping from 9 to 45%, with both moderate permeability and available water capacity (Amen, 1975).

Aliquots of Rocky Flats soil samples were analyzed by the CSU soil testing laboratory (Table 3). These data indicate the two macroplots to be similar. Macroplot 1 may be slightly more basic, with a slightly higher cation exchange capacity, and slightly higher amounts of exchangeable acetate and bicarbonate. These and other small differences obvious in the table probably are not significant in terms of plutonium-soil interactions.

Topographically, the land of the installation is characterized by five separate watercourses which generally run west to east. These stream channels have created a series of flat, wind-scoured plateaus divided by ravines in the northwest quadrant of the plant site. The characteristic channeling becomes less severe to the south and east sides of the plant site.

The climate of the Rocky Flats installation is typified by gusty winds and moderate rainfall. The 22-year average wind velocity is 3.7 meters per second (8.2 mph) with a record gust during those years of 47 m/s (106 mph) occurring January 18, 1959. The strong

Table 3. Characteristics of Rocky Flats soils; pooled aliquots of five independent samples of listed depths and macroplots.

characteristic	macroplot 1		macroplot 2	
	0-10 cm	10-20 cm	0-10 cm	10-20 cm
pH	7.7	8.0	7.5	7.8
conductivity, $\mu\text{mho}/\text{cm}$	0.7	0.8	0.5	0.6
Ca^{++} , meq/l ^a	7.0	5.5	2.0	4.0
Mg^{++} , meq/l	0.5	2.9	3.4	0.8
Na^{+} , meq/l	0.7	0.7	0.3	0.2
SO_4^{+} , meq/l	2.5	2.5	2.2	0.6
Cl^{-} , meq/l	0.6	0.8	0.4	0.7
$\text{CO}_3^{=}$, meq/l	0	0	0	0
HCO_3^{-} , meq/l	4.1	4.2	2.1	4.3
cation exchange capacity, meq/100g	25.0	25.8	21.6	21.2
Total N, %	0.17	0.12	0.23	0.17
$\text{NO}_3\text{-N}$, ppm	45	99	48	42
$\text{NO}_2\text{-N}$, ppm	<1	<1	<1	<1
$\text{NH}_4\text{-N}$, ppm	9	7	4	4
organic matter, %	2.6	3.5	3.1	5.7
bicarbonate extractable P, ppm	20	9	7	5
ammonium acetate extractable K, ppm	418	413	375	358
DTPA extractable Zn, ppm	1.9	6.6	3.4	1.8
DTPA extractable Fe, ppm	16.1	17.4	21.9	21.4
DTPA extractable Cu, ppm	2.4	23.6	1.9	2.3
DTPA extractable Mn, ppm	15.2	15.2	13.7	13.3
% sand	40	39	48	44
% silt	24	21	24	21
% clay	36	40	28	35

^a meq/l of water extracted from saturated soil sample.

winds are predominantly from the west and northwest. During 1974, 22% of the recorded winds were from the northwest. The 22-year mean of total annual precipitation is 40 cm (15.75 in) with a record of 63 cm (24.67 in) set in 1969. The mean temperature during the installation's existence has been 10°C (50°F) with a mean daily maximum of 25°C (77°F) and a mean daily minimum of -5.6°C (22°F) (Rocky Flats 1974 annual weather summary, unpublished).

The vegetation of the area may be described as disturbed grassland. However, this grassland is not necessarily typical of Colorado grassland, i.e. Rocky Flats land for the most part has not been grazed or otherwise systematically disturbed for over twenty years. The sunflower (Compositae) and grass (Gramineae) families are most abundant (Weber, 1975). Other families represented are the sedges (Cyperaceae), peas (Leguminosae), mustards (Cruciferae), figworts (Scrophulariaceae) and roses (Rosaceae) as well as 41 others. A few trees exist on the installation site; cottonwoods line several stream beds and scattered ponderosa pine are found on the western half of the site.

The SCS report mentioned above also classified the soils of Rocky Flats by their capability to sustain vegetative growth (Amen, 1975). Portions of eventual macroplots 1 and 2 were designated "clayey foothill good condition." Of the classification the report said, "Western wheatgrass, junegrass, green needlegrass, buffalograss, and Kentucky bluegrass dominate the site. A large number of forbs appear in small amounts. Production is around 800-900 pounds of air dry vegetation per acre per year." The remainder of the macroplot 2 region included

cobbly foothill soil in good condition which was dominated by needle and thread grass with little bluestem, sedge, sideoats grama, and blue grama prominent. Production estimates ranged from 1200-1500 lbs/acre/year. The same classification made up the rest of the macroplot 1 region, but only in fair condition. The fair condition designates more forbs and 40-50% less annual production.

The animals at Rocky Flats include arthropods, amphibians, birds, reptiles, and mammals. In terms of visual observation, the most prominent of the above groups are: grasshopper and wolf spider; leopard frog; killdeer, black-billed magpie, western meadow lark, and mourning dove; common prairie rattlesnake, common bull snake, and plains garter snake; white-footed deer mouse, northern pocket gopher, 13-lined ground squirrel and mule deer.

The location of the plutonium contamination source was important to the siting of the study macroplots. Investigations by Krey and Hardy (1970) of the Health and Safety Laboratory's (HASL) suggested that the barrel storage area was the most likely source of contamination.

More direct evidence linking the barrel storage area to the east-southeast contamination pattern comes from Rocky Flats air sampling data (Rocky Flats weather summary, unpublished data). Air sampling station S-8, one of many such stations maintained and sampled regularly by Rocky Flats personnel, is located just east of the barrel storage area. With a brief exception during 1961, monthly averages of daily airborne contamination values have been kept since at least 1960 to the present. These data strongly link airborne contamination

just east of the oil barrel storage area with leaks and disturbances of the storage surface.

The contamination peaks are virtually all associated with periods of perturbation of the contaminated surface (Table 4; Figure 2). For example, the S-8 air sample averages hovered at about $.01 \text{ pCi/m}^3$. After the asphalt pad was completed the ambient air concentration at S-8 was again $.01 \text{ pCi/m}^3$ except two periods when sample wells and a perimeter drainage ditch were dug. These data, although circumstantial, are strong evidence that the barrel storage area was the main contamination source.

The Pu-contaminated cutting oil, about as viscous as lightweight motor oil, but thinned by carbon tetrachloride, was stored in the 55 gallon barrels for periods of up to seven years. 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 2-3 μm filters before placement in the barrels. The discard limit at the time of storing was $1 \times 10^{-2} \text{ g Pu/l oil}$. If the limit and the filtering were observed and performed faithfully, each of the approximately 3570 plutonium-containing barrels would have no more than 2.1 g of Pu (1.3 Ci) (telephone conversations on 4 June 1976 with M. A. Thompson, Environmental Sciences, Rocky Flats Plant, and F. J. Miner, Chemical Resources, Rocky Flats Plant).

Once inside the barrels, it is difficult to assess what occurred. The presence of carbon tetrachloride in the drums allows the

Table 4. Approximate mean of total monthly long-lived alpha activity in ambient air at station S-8 (just east of oil barrel storage area) during perturbation events of the storage area surface.

dates	event	alpha activity, pCi/m ³
7/59-9/63	no large scale leaking	.009
1/64-1/65	large scale leaking	.025
1/65	contaminated soil covered with fill	.01
1/66	small building added to filter contaminated oil from leaking to new drums	.014
1/67	drum removal activity begun	.038
6/68	last drums removed, but high winds spread some activity	.188
2/69	weeds burned, area graded for paving	.34
9/69	asphalt pad completed	.013
11/69	four sampling wells dug through pad	.033
4/71	drainage ditch dug on west side of asphalt pad	.033

Figure 2. Monthly averages of daily airborne contamination values from air sampler S-8 located just east of the drum storage area.

possibility that hydrochloric acid was formed which in turn may have reacted with the Pu metal to form very low concentrations of plutonium chloride, a more soluble form of the element (telephone conversation on 4 June 1976 with J. M. Cleveland, Environmental Studies, Rocky Flats Plant). This possibility is given credibility by the work of J. Navratil of Rocky Flats Chemical Research Division who studied contaminated cutting oil in recent years.

Navratil found that filtering the contaminated oil through a .01 μm filter removed only about 50% of the plutonium. This result strongly suggests that about one-half of the Pu was in a large particulate form and the other one-half, very small particles. Fission track analysis of the filtered oil confirmed that the remaining plutonium was indeed monomeric (telephone conversation on 4 June 1976). It is doubtful that the barrels consistently held the above proportions of particulate and nonparticulate Pu oxide, but each probably contained some plutonium chloride.

Dr. Cleveland also suggested that the filtered 3- μm Pu particles might combine with each other to form larger aggregates of the metal. Of course, the size and binding tenacity of these conglomerates is unknown. Much of the above information concerning the plutonium-contaminated oil is based on speculation or memory of past events.

Assuming the barrel storage area to be the contamination source, two macroplots (study plots) for sampling and trapping (described in more detail later) were established within the Rocky Flats installation buffer zone (Figure 3). Macroplot 1 begins about 100 m southeast of the oil barrel storage pad (formerly the major plutonium source

Figure 3. Map of Rocky Flats installation with 1974 average wind rose.

term). Macroplot 1 was chosen because of its relatively high concentrations of plutonium in soil, its easy access, and its reasonably undisturbed vegetative community. Macroplot 2 is approximately 1 km south and slightly east of the oil barrel storage pad. While macroplot 2 and the oil barrel storage pad are at nearly the same elevation, they are separated by a watershed 55 m deep and 750 m wide, and macroplot 2 is on the leeward side of a hill. This location theoretically tends to shelter macroplot 2 from blown oil barrel storage area debris. Macroplot 2 was chosen because its plutonium concentration in soil was presumed to be much lower than that in macroplot 1, and because it is similar to macroplot 1 in vegetative community structure, exposure, and topography. Though less intensively than in macroplots 1 and 2, the plutonium concentrations in macroplot H, the hot spot, was investigated. Macroplot H is immediately adjacent to the oil barrel storage pad to the southeast, but differs from the pad in that macroplot H is on a sloping hillside and is not paved. This relatively small macroplot is topographically sheltered from strong westerly and north-westerly winds and was reasonably well vegetated during most of the study.

MATERIALS AND METHODS

Sample Collection and Preparation

Soil

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 Figure 3. A sampling grid was superimposed over each macroplot. The macroplot 1 grid was approximately 0.75 ha (1 ha = 2.5 acres) and contained 100 sampling markers. The macroplot 2 grid was smaller than macroplot 1 and contained 25 sampling markers, but the distance between markers was the same as macroplot 1.

The decision to make macroplot 2 smaller than macroplot 1 was based on a desire to sample the macroplot closer to the source more intensively. This was done, but more samples were ultimately analyzed from macroplot 2. The fact that a smaller area was chosen for sampling seems unimportant in the light of several considerations. Five-gram samples are an extremely small sample of the total mass available in each macroplot. Also, plutonium concentration does not vary significantly as a function of either north-south or east-west distance in macroplot 2. These facts seem to indicate that the smaller size of macroplot 2 did not bias the results significantly.

Soil samples from macroplots 1 and 2 were taken from 10 and 5 randomly chosen locations on each grid, respectively. Depth profile

soil samples were taken by first marking a 25 x 25 cm microplot. The standing vegetation was clipped and bagged, litter was collected and bagged. Next, soil of a 20 x 25 cm area was excavated to a depth of 3 cm using a hand trowel. This soil and subsequent 20 x 25 cm areas at lower depths were saved to refill the excavation after sampling. From the remaining 25 x 5 cm strip at the 0-3 cm depth, four 5 x 5 x 3 cm samples were removed and bagged separately. Following this, loose soil on the floor of the 3 cm deep 25 x 25 cm hole was removed to prepare for the next 3 cm removal. This process was repeated until all four columns of 5 x 5 x 3 cm blocks were sampled seven times to a depth of 21 cm. If rocks precluded sampling at a given depth, the column was resumed below the blockage. A disadvantage of this method is the potential for contaminating lower depths.

Soil samples were transported to the lab, air dried, and rocks or debris greater than 0.5 cm diameter removed from the sample. Following 24 hrs of oven drying at 80°C, samples were weighed. Samples were then mechanically shaken on 7.6 cm diameter brass soil sieves for 30 minutes. The accumulation on each sieve was weighed and placed in a small paper envelope which was sealed with tape.

Litter and Standing Vegetation

Litter and detritus and standing vegetation were taken approximately twice monthly during the growing season and monthly the remainder of the year from randomly chosen microplots, i.e., near randomly chosen grid markers. Areas of microplots sampled were 0.25 m² for litter and 0.5 m² for standing vegetation. Random numbers were used

with established grid markers in each macroplot. The vegetation frame was first dropped near the chosen marker and secured using large spikes and then the litter frame was secured inside the larger frame. Vegetation was clipped and bagged. Litter was "raked" using clippers, gathered by hand and bagged. Because these samples were to be analyzed for plutonium concentration, care was taken to prevent cross contamination.

In the laboratory, samples were air dried and weighed. To separate associated soil from the litter samples, the following procedure was followed as outlined by the U. S. International Biological Program: 1) samples were shaken on an American Society for Testing and Materials (ASTM) No. 10 sieve for five minutes, 2) the fines which passed the sieve were collected, 3) the fines were added to 1.0 l of water in a 2-qt container, 4) the floating fraction was removed and 5) reunited with the original sample which was 6) air dried and 7) reweighed, 8) the total weight was divided by the microplot size to compute mass of litter per unit area. In later cases the sieving was omitted due to poor success. The net vegetation weight was divided by the microplot size (0.5 m^2) to calculate mass of vegetation per unit area. For plutonium analysis vegetation and litter samples were ground on a Wiley mill with a $850 \text{ }\mu\text{m}$ -opening screen and 5 g aliquots were taken.

Arthropods

Arthropods were sampled using a combination of sweep netting, pitfall trapping, and drop trapping at random sites on established grids. Sweep netting and drop trapping were done approximately

monthly, and pitfall traps were left open continuously and checked when opportune. Sampling for biomass estimates was done exclusively by the drop trap method. The trap, containing 0.5 m², under a screened steel frame was randomly dropped and the enclosed vegetation and soil surface thoroughly vacuumed. Prior to dropping the trap the sampling vicinity was not disturbed, lest mobile arthropods escape detection. After transport to the laboratory, animals were separated into generic groups which were weighed separately. These totals were combined for an estimated weight per 0.5 m² microplot.

Animals obtained by the drop trap method were not analyzed for plutonium due 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. Collected arthropods were transported to the laboratory and killed. For plutonium analysis a representative composite was analyzed for each sampling period. Animals were not cleaned prior to plutonium analysis.

Small Mammals

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). Trapping was carried out about six times yearly using Sherman live traps for cricetid and sciurid rodents. The regimen followed during trapping efforts included two consecutive prebaiting days. The third day all traps were set, then operated for five consecutive days. Bait in all cases was an oatmeal-peanut butter mixture. Approximately 15% of the

estimated population was removed from each macroplot during each trapping session. These were collected by removing dead-in-trap individuals during the regular session and the remainder randomly in one extra trapping night.

Captured individuals were removed from traps and marked by toe-clipping. Species, sex, age class, reproductive condition, pelage, general condition, presence of ectoparasites, weight, and capture location were recorded for each animal. Animals were released where captured. The traps were then reset and rebaited.

Geomyid rodents were trapped less regularly using homemade live traps placed in burrow systems. Once captured, geomyids were handled in similar manner to the cricetids and sciurids.

Individuals were either dissected immediately or frozen for dissection at a later date. To prevent cross-contamination between tissues, special precautions were taken during dissection. The hide was removed totally prior to opening either the peritoneal or pleural cavities. A separate set of instruments was used for exterior and interior surfaces. Approximately 10 cm² of hide were used as an aliquot. Lung, liver and gastrointestinal tract were removed intact. Muscle samples were taken from the legs in most cases. Bone samples consisted of the whole skeleton cleaned of flesh by a dermestid beetle colony. All samples except bone were placed on tared, ashless filter papers, oven dried at 50-60°C, and weighed. The sample and filter paper, or bone sample alone, was placed in a 40-dram plastic snap-cap vial for storage or transport to a commercial laboratory.

Plutonium Analysis Procedure

Prepared samples of approximately 5 g each were sent to commercial laboratories or analyzed in our lab. Commercial labs used were LFE Environmental, Richmond, California, and Eberline Instrument Corp., Albuquerque, New Mexico. The basic LFE procedure is discussed by Wessman et al. (1971). The Eberline procedure is based on techniques developed by Sill (1969). These labs digested the samples using concentrated hydrofluoric acid when necessary; Eberline used a pyrosulfate fusion technique to dissolve soil. Ion exchange columns were utilized to remove interfering nuclides and to isolate plutonium from the samples prior to counting by alpha spectrometry. Chemical recovery was measured by addition of ^{236}Pu tracer to each sample.

The most satisfactory method of analysis in our laboratory has been harsh digestion, ion exchange, organic extraction and liquid scintillation counting. A 5 g sample was digested with concentrated nitric acid and ashed at 500-550°C for a minimum of eight hours. After ashing, concentrated hydrofluoric acid was applied for further digestion. The sample, in a 7.5 M nitric acid solution, was then passed through a one-cm (inner diameter) column of Amberlite CG400 100-200 mesh anion exchange resin. The plutonium was eventually removed from the column using 9 M hydrochloric acid which was later evaporated. Next, a glass liquid scintillation vial with plastic-lined cap was filled with the sample solution, nitric and boric acid solutions, and the scintillation cocktail. Solvent extraction was used within the vial to transfer the plutonium into the organic phase

for counting. Samples were counted on a Nuclear Chicago Mark II liquid scintillation system. Appendices A and B contain a more detailed description and a rationale for the analysis procedure, respectively. All sample Pu concentrations were expressed in dpm/g dry sample unless otherwise noted.

This procedure has an estimated minimum detectable activity (MDA) of 0.18 dpm/g, assuming a 5 g sample. The procedure has a cost per sample in the range of \$10-20 and a turn around time of about two weeks, both substantially less than for commercial laboratories. Very large or very small samples are not suited to analysis by the liquid scintillation method.

The plutonium data referred to herein are $^{239,240}\text{Pu}$ unless otherwise noted. Correction for ^{238}Pu contribution during the liquid scintillation procedure is noted in Appendix B.

Data Organization and Statistical Considerations

All data from the plutonium assay were punched onto computer cards, one sample per card. Pertinent information on each card included sample number, net weight in grams, count rate of sample (or total counts and counting time if from our lab), and codes denoting macroplot, microplot, sample type, soil depth, sieve size (if soil), tissue type, species and lab of analysis. Since data from commercial labs were reported as count rate concentration, the information was handled somewhat differently. However, all data, having been computed into dpm/g and corrected for blanks, isotopic ratio, etc., were punched by the computer onto cards in the same format.

Punching in this manner allowed combining all data for sorting into various groups for data analysis.

Virtually all statistical tests on the plutonium data, except some simple regressions, were accomplished using subprograms from the Statistical Package for the Social Sciences (SPSS) available at the CSU Computer Center (Nie et al., 1975). The SPSS program package is quite adaptable to needs of most disciplines and provides easy access and manipulation of relatively unwieldy procedures.

Calculation of simple regressions as well as the biomass data was accomplished using either of two Hewlett-Packard calculators: 9100 B desk-top programmable calculator or an HP-45 advanced scientific pocket calculator.

Nonparametric tests were substituted for more common tests on several occasions. Nonparametric tests are distribution free, i.e., do not require assumptions about distribution shape, and therefore are quite suitable to many of the non-normal data presented herein. Nonparametric tests, however, are less powerful (sensitive) than parametric methods. Siegel (1956) discusses the nonparametric tests used herein in detail.

RESULTS

Plutonium Compartmentalization in the Ecosystem

It can be expected that most of the plutonium in a given ecosystem will be in the soil with small portions found in other compartments. To test these expectations for the particular situations at Rocky Flats, compartmental inventories were calculated.

Mass estimates of litter, standing vegetation, arthropods and small mammals were made (Table 5). The tabulated values are means of data collected throughout several years. The plots of these data through time are incorporated into Appendix C. Not surprisingly, the litter and vegetation account for much more biomass than do either the arthropods or the small mammals. A t-test indicated that the arthropods comprise a larger biomass of tissue than do the small mammals ($p < .05$). Soil masses were estimated assuming densities of 1.2 g/cm^3 for 0-15 cm soil and 2.4 g/cm^3 for 15-21 cm soil (Krey and Hardy, 1970). Therefore, the top three centimeters of an area one meter square (1 m^2) contains $3.6 \times 10^4 \text{ g}$ of soil.

Next, the mean concentrations of plutonium were calculated for each of the categories of sample listed in Table 5, as well as the 0-3 and 3-21 cm soil groups (Table 6).

In macroplot 1, however, the litter had a higher concentration than the 3-21 cm soil, followed by vegetation, arthropods, and small mammals. In macroplot 2, the small mammals had the second highest mean concentration followed by 3-21 cm soil, litter, vegetation and

Table 5. Estimates of standing crop (vegetation and litter) and biomass (arthropods and small mammals) at Rocky Flats.

macroplot	compartment	mass (dry g/m ²)	
		mean	95% confidence interval
1	litter	4.3×10^2	$3.5 \times 10^{-2} - 5.0 \times 10^2$
	vegetation	2.2×10^2	$1.8 \times 10^2 - 2.5 \times 10^2$
	arthropods	1.3×10^{-1}	$9.2 \times 10^{-2} - 1.6 \times 10^{-1}$
	small mammals	3.1×10^{-2}	$1.9 \times 10^{-2} - 4.2 \times 10^{-2}$
2	litter	7.9×10^2	$5.4 \times 10^2 - 1.0 \times 10^3$
	vegetation	2.2×10^2	$1.7 \times 10^2 - 2.7 \times 10^2$
	arthropods	1.1×10^{-1}	$9.0 \times 10^{-2} - 1.3 \times 10^{-1}$
	small mammals	2.4×10^{-2}	$2.0 \times 10^{-2} - 2.8 \times 10^{-2}$

Table 6. Mean plutonium concentrations in selected ecosystem constituents from Rocky Flats macroplots.

macroplot	compartment	plutonium concentration (dpm/g)	
		mean	95% confidence interval
1	soil(0-3 cm)	1853	1221.7 - 2484.3
	(3-21 cm)	233	153.6 - 312.4
	litter	913.7	687.8 - 1139.6
	vegetation	63.4	34.2 - 92.6
	arthropods	7.03	1.60 - 12.46
	small mammals	14.4	5.11 - 23.7
2	soil(0-3 cm)	122.6	10.7 - 234.51
	(3-21 cm)	6.63	0.58 - 12.68
	litter	1.70	0.81 - 2.59
	vegetation	1.25	0.11 - 2.39
	arthropods	1.45	0.32 - 2.58
	small mammals	26.2	27.8 - 47.8

arthropods. The soil values listed in Table 6 were corrected for soil particle size distribution.

To calculate the compartmental fraction of plutonium for each compartment, the data in Table 5 (mass in g/m^2) were multiplied by the data in Table 6 (mean concentration in dpm/g). These data were then summed over compartments, including the two soil depth groups, to achieve a total activity for the average square meter of each macroplot. The compartmental inventories were then divided by this total to compute a fraction of the total for that compartment (Table 7). Confidence intervals (95%), assuming normality, for each appropriate value were calculated using standard formulae (Snedecor and Cochran, 1969; Arkin and Colton, 1970).

In both macroplots the soil compartment has the vast majority of the total plutonium (99.69% and 99.97%). In macroplot 1, the two soil groups are about evenly divided in terms of the fraction they contain. In macroplot 2, the surface soil group has about 50% more of the total activity than the 3-21 cm soil. Perhaps more important is the fact that the confidence intervals for both soil groups in macroplot 2 overlap zero in their ranges. This finding stresses the high degree of variation associated with the macroplot 2 soils.

In both macroplots the litter represents less than one percent of the total plutonium, but macroplot 1 has approximately 10 times as much plutonium in litter than macroplot 2. This is understandable considering the differences between macroplots in litter plutonium concentration and standing crop. The vegetation in both macroplots contains even less plutonium than the litter. Again, macroplot 1

Table 7. Compartmental distribution of plutonium in Rocky Flats macroplots.

macroplot	compartment	inventory (dpm/m ²)			fraction of total		
		mean	95% confidence interval		mean	95% confidence interval	
1	soil, 0-3 cm	6.671x10 ⁷	3.390x10 ⁷	- 9.952x10 ⁷	4.970x10 ⁻¹	2.101x10 ⁻¹	- 7.839x10 ⁻¹
	soil, 3-21 cm	6.710x10 ⁷	4.330x10 ⁷	- 9.090x10 ⁷	4.999x10 ⁻¹	2.671x10 ⁻¹	- 7.327x10 ⁻¹
	litter	3.892x10 ⁵	2.639x10 ⁵	- 5.145x10 ⁵	2.900x10 ⁻³	1.668x10 ⁻³	- 4.132x10 ⁻³
	vegetation	1.369x10 ⁴	6.913x10 ³	- 2.047x10 ⁴	1.020x10 ⁻⁴	4.231x10 ⁻⁵	- 1.617x10 ⁻⁴
	arthropods	9.139x10 ⁻¹	9.992x10 ⁻²	- 1.728x10 ⁰	6.809x10 ⁻⁹	7.496x10 ⁻¹⁰	- 1.287x10 ⁻⁸
	small mammals	4.464x10 ⁻¹	1.178x10 ⁻²	- 7.750x10 ⁻¹	3.326x10 ⁻⁹	6.758x10 ⁻¹⁰	- 5.976x10 ⁻⁹
	TOTAL	1.342x10 ⁸	9.408x10 ⁷	1.743x10 ⁸	1.000x10 ⁰	7.843x10 ⁻¹	- 1.216x10 ⁰
2	soil, 0-3 cm	4.414x10 ⁶	4.019x10 ⁵	- 8.426x10 ⁶	6.979x10 ⁻¹	-1.072x10 ⁻¹	- 1.503x10 ⁰
	soil, 3-21 cm	1.909x10 ⁶	1.559x10 ⁵	- 3.662x10 ⁶	3.018x10 ⁻¹	-4.628x10 ⁻²	- 6.499x10 ⁻¹
	litter	1.348x10 ³	5.489x10 ²	- 2.147x10 ³	2.131x10 ⁻⁴	1.560x10 ⁻⁵	- 4.106x10 ⁻⁴
	vegetation	2.775x10 ²	1.073x10 ²	- 4.477x10 ²	4.388x10 ⁻⁵	3.092x10 ⁻⁶	- 8.467x10 ⁻⁵
	arthropods	1.595x10 ⁻¹	4.440x10 ⁻²	- 2.746x10 ⁻¹	2.522x10 ⁻⁸	-2.050x10 ⁻⁹	- 5.249x10 ⁻⁸
	small mammals	6.288x10 ⁻¹	1.406x10 ⁻¹	- 1.117x10 ⁰	9.942x10 ⁻⁸	-4.580x10 ⁻⁹	- 2.034x10 ⁻⁷
	TOTAL	6.325x10 ⁶	1.909x10 ⁶	- 1.074x10 ⁷	1.000x10 ⁰	7.513x10 ⁻¹	- 1.249x10 ⁰

vegetation has a higher fraction than vegetation of macroplot 2. However, the pattern of higher fractions in macroplot 1 is reversed for the arthropods and small mammals. Both of these compartments in each macroplot have tiny fractions of the total plutonium. The macroplot 2 arthropod and small mammal fractions are suspect, however, because the confidence interval overlaps zero as did those of the soils.

In summary, the macroplot 1 compartmentalization data strongly suggest that greater than 99% of the plutonium in the macroplot is found in the soil. Far smaller fractions are found in the litter, vegetation, arthropods, and small mammals in decreasing order. In macroplot 2, the soil data again suggest that 99%+ of the total plutonium is found in the soil. However, these data have much more uncertainty associated with them. The other macroplot 2 compartments have much smaller fractions decreasing in the following order: litter, vegetation, small mammals, and arthropods.

Plutonium Concentrations in Ecosystem Constituents

Soil

Qualitative Observations: This investigation has shown soil to be the most important compartment in the Rocky Flats ecosystem in terms of concentrations and amounts of plutonium contained. This section describes the concentration and distribution of plutonium in the soil compartment of the ecosystem at Rocky Flats.

Table 8 categorizes plutonium concentration means in Rocky Flats soil samples by two macroplots, seven depth groups, and eight particle

Table 8. Mean plutonium concentrations of Rocky Flats soil samples.

macroplot	particle size range, μm	plutonium concentration, dpm/g [mean, CV(n)] of depth group aliquots							particle size mean
		0-3 cm	3-6 cm	6-9 cm	9-12 cm	12-15 cm	15-18 cm	18-21 cm	
1	2000-850	1640,2.02(7)	299,1.42(9)	196,1.83(6)	59.4,0.81(8)	29.6,1.60(7)	11.9,0.88(6)	3.99,0.63(3)	350,3.86(46)
	850-425	1030,0.82(9)	269,0.83(9)	228,1.70(6)	80.7,0.97(9)	65.3,0.72(7)	15.5,0.89(6)	12.3,0.45(3)	293,1.80(49)
	425-250	980,0.64(9)	293,0.83(9)	197,1.84(6)	65.7,0.51(8)	65.6,0.54(5)	29.2,0.80(6)	12.6,0.15(3)	297,1.58(46)
	250-180	1030,0.54(9)	277,0.57(8)	294,1.35(5)	85.6,0.90(9)	66.2,0.51(6)	31.2,1.05(6)	19.8,0.73(3)	313,1.47(46)
	180-150	1700,1.05(8)	278,0.64(8)	225,1.74(6)	77.1,0.63(8)	308,1.61(7)	56.6,0.74(6)	14.4,1.13(2)	448,2.12(46)
	150-75	1940,0.71(9)	463,0.79(9)	232,1.40(6)	151,0.85(9)	124,0.69(7)	96.6,1.29(6)	23.9,0.89(3)	528,1.73(49)
	75-45	3080,0.52(7)	698,0.45(6)	457,1.41(4)	221,0.60(8)	363,0.74(6)	187,1.38(4)	78.6,0.63(2)	876,1.48(37)
	45-0	3320,1.59(14)	390,1.07(10)	1800,1.82(11)	428,0.96(10)	498,0.98(7)	189,1.26(7)	60.1,0.31(3)	1280,2.38(62)
	depth mean	1910,1.46(72)	358,0.89(69)	592,2.79(50)	153,1.37(69)	194,1.58(52)	74.8,1.82(47)	26.5,1.06(22)	569,2.66(381)
2	2000-850	12.6,1.68(12)	2.11,0.89(9)	.800,1.59(8)	1.19,1.55(12)	.749,1.33(10)	.799,1.53(9)	1.27,0.80(6)	3.24,3.06(66)
	850-425	12.0,1.73(13)	1.45,0.91(9)	.983,1.51(13)	1.18,1.59(13)	.812,1.40(10)	.915,1.55(9)	2.67,0.61(6)	3.15,3.04(73)
	425-250	29.6,1.85(13)	4.49,1.05(9)	3.03,1.20(11)	3.42,1.56(13)	3.20,1.54(10)	3.42,1.46(9)	8.13,0.74(6)	8.67,2.92(71)
	250-180	58.6,2.26(13)	12.2,1.07(8)	5.06,1.51(11)	4.50,1.39(12)	8.65,1.41(10)	5.80,1.37(7)	9.51,1.05(6)	17.2,3.53(67)
	180-150	106,2.77(13)	21.5,2.00(8)	5.83,1.25(12)	7.01,0.94(12)	8.23,1.62(10)	8.92,1.08(8)	10.8,1.02(6)	28.0,4.69(69)
	150-75	150,3.28(13)	4.15,1.14(9)	3.87,1.43(13)	3.91,1.07(11)	4.70,1.61(10)	6.12,1.32(9)	7.93,0.76(6)	31.4,6.74(71)
	75-45	338,3.15(12)	12.7,1.02(10)	6.69,1.44(12)	6.02,1.15(10)	11.8,1.87(11)	28.6,1.68(7)	11.8,0.85(7)	68.7,6.49(69)
	45-0	29.2,1.29(11)	7.43,1.13(9)	8.96,1.36(11)	7.10,0.99(10)	8.03,1.63(9)	25.1,1.40(8)	10.9,0.50(6)	14.0,1.61(64)
	depth mean	91.6,4.61(100)	8.08,2.06(71)	4.47,1.64(91)	4.15,1.33(93)	5.82,2.04(80)	9.30,2.33(66)	7.96,0.98(49)	21.8,8.39(550)

size ranges. The coefficient of variation (CV, standard deviation/mean) and sample size of each category are also listed. One pertinent characteristic of this table is the order of magnitude difference ($p < .05$) between the grand mean of the soil concentration of the two macroplots. The 0-21 cm depth mean for macroplot 1 is 569 dpm/g and for macroplot 2 is about 22 dpm/g.

Several other aspects of Table 8 are worth discussing. First, in both macroplots there seems to be a rather pronounced depth gradient downward through the soil for virtually every particle size range. This tendency is also reflected in the depth means, but there appear to be several small peaks in the 6-9 and 12-15 cm depth groups. In macroplot 2, the data, as stated earlier, are more variable, resulting in a ragged depth gradient. Beginning with a value of about 8 dpm/g at 18-21 cm there seem to be depleted levels at the 6-9, 9-12 and 12-15 cm depth levels. Stated another way, the 15-18 and 18-21 cm groups may be higher than expected based on the mean concentrations in the top two depth groups. Nevertheless, there is good evidence of a strong depth gradient for plutonium concentrations in both macroplots.

These data (Table 8) also show a particle size effect for both macroplots. It appears that the smaller the particle the higher the plutonium concentration of the sample. In macroplot 1, the 2000-850 μm diameter particle group mean seems to be an exception to this statement by containing a higher concentration of Pu than the next three smaller size ranges. The same sort of phenomenon exists in macroplot 2 where the largest particle size group has a higher mean

concentration than the next smaller group. Another exception to the particle size gradient concept in macroplot 2 is found in the 45-0 μm diameter particles mean which is unexpectedly lower than the next four larger particle size groups. For both macroplots, regressions of each of the particle size fractions of the total mass as functions of the sample depth were not significant ($\alpha = .05$).

Three-way analyses of variance (ANOVA) of the raw and the logarithmically transformed soils data partitioned the variance into effects by the macroplots, depths, and particle sizes and two- and three-way interactions of these effects. These ANOVAs confirmed that there are significant macroplot, depth, and particle size effects ($\alpha = .001$). This result confirms the gradients suggested by the data in Table 8.

The variability seen in the soil samples is noteworthy. For instance, the coefficients of variation in the macroplot 1 cells range from 0.15 to 2.02. Generally, the macroplot 2 data are more variable, relative to the mean, with CVs ranging from 0.50 to 3.28. This observation is supported by the fact that the macroplot 2 overall CV (CV of the overall mean for all samples from a macroplot) is 8.39 compared to 2.66 for the macroplot 1 overall CV.

Table 9 gives an outstanding example of the large amount of spatial variation seen in macroplot 2 soil. Three adjacent columns (5x5 cm) from one microplot had 0-3 cm depth mean plutonium concentrations of 1062 (column A), 11.87 (column B), and 1.26 (column C) dpm/g.

Table 9. Plutonium concentrations in soil samples from three adjacent 5 x 5 cm columns of Rocky Flats macroplot 2 listed by depth and sieve numbers (concentrations corrected for background).

column	depth, cm	Plutonium concentration (dpm/g)								row means
		sieve #20	sieve #40	sieve #60	sieve #80	sieve #100	sieve #200	sieve #325	sieve pan	
A	0-3	76.9	75.8	204	490	1090	1790	3713	--	1062
	6-9	-.880	-.789	-.302	-.499	-.280	.072	.805	--	-0.268
	9-12	-.762	-.773	-.649	-.302	-.718	7.62	4.70	--	1.30
	12-15	-.351	-.752	-.653	-.695	-.553	-.534	-.266	--	-.543
	15-18	-.455	-.716	-.784	.304	-.113	-.111	.591	-	-0.183
	18-21	-.828	-.712	-.803	-.689	-.673	-.628	.157	--	-0.597
B	0-3	9.65	3.32	7.19	26.6	1.45	10.2	2.47	--	11.87
	3-6	-.446	-.732	-.580	-.529	127	-.027	2.02	--	18.1
	6-9	--	1.36	4.57	16.7	14.1	14.6	21.2	41.2	16.3
	9-12	3.99	6.42	11.6	12.0	14.3	6.13	--	19.4	10.55
	12-15	--	2.33	7.96	18.7	25.5	20.1	64.7	34.1	24.8
	15-18	.234	.675	.377	--	5.06	23.9	134	97.3	37.4
C	0-3	-.596	2.59	2.94	3.01	.726	.750	-.626	--	1.26
	3-6	1.99	1.56	9.51	9.61	5.43	10.8	32.1	-3.77	8.40
	6-9	3.79	5.22	4.42	3.81	7.72	10.3	.022	12.9	6.02
	9-12	5.72	3.49	11.1	13.3	18.5	6.95	--	13.7	10.4

The differences between the three columns extends to other depths as well, but the pattern shown in the 0-3 cm depths does not hold. Virtually all of the plutonium in column A is found in the top three cm, the other depths being at or near background. In column B, however, the majority of the plutonium is found at lower depths, with the highest concentration in the 15-18 cm level. Column C appears to have the same trend seen in column B; the deepest samples have the highest concentration of plutonium.

The data in both Tables 8 and 9 make the important point that there is a large amount of variation, with regard to depth, particle size distribution, and spatial dispersion, in the soils sampled from both macroplots. Further, the relative variability in macroplot 2 seems to be larger than macroplot 1.

Generally, radionuclide contamination of the environment results in log-normal distributions (Whicker et al., 1966; Cember, 1969). Following this pattern, the Pu data from soil sampling are highly skewed and kurtotic. In this study, the Kolomogorov-Smirnov (K-S) one-sample test of normality indicated the plutonium soil results are not normally distributed. The natural log transformation of these data, while not resulting in normal distributions, did greatly reduce the skewness and kurtosis for each of the seven depth groups from each macroplot tested. However, the results of the K-S normality tests on log-transformed data indicated that the macroplot 1 data seem more likely to be log-normally distributed than the macroplot 2 data.

Further transforming the data to $\ln(\ln \text{ Pu})$ generally increased the skewness and kurtosis. Consequently, these examinations confirmed that the natural log transform was the most useful of the three in "normalizing" the data.

Concentration and Source-to-Sample Distance: Directional gradients of Pu in soil relative to the source term and macroplot 1 grid were assessed. One tool for doing so was the use of regression of plutonium concentration as a function of various parameters of the sample. Two of the most promising measurements which were easily available were mentioned earlier; depth and particle size range of the sample. Another parameter considered was the distance of samples from the center of the asphalt pad.

Figure 4 illustrates two distance transects which were established. The downwind (DW) transect runs roughly east-southeast from the eastern boundary of macroplot 1 to the eastern boundary cattle fence. The transect name derives from the fact that the line direction approximates the predominant wind direction. If extrapolated westward, the transect would nearly hit the center of the asphalt pad. The other line, the downslope (DS) transect, follows the steepest slope from the southern edge of the macroplot 1 grid toward Woman Creek. Samples of soil, litter, and vegetation were taken at intervals of 100 m DW and 50 m DS, as marked on Figure 4. The DW distances used in later regressions were measured from the center of the asphalt pad. The DS distances were measured south of a line running east to west through the center of the asphalt pad.

Figure 4. Location of downwind and downslope sampling transects relative to macroplot 1, the asphalt pad, and the "hot spot."

Table 10. Calculation of a weighted average plutonium concentration of a given soil sample using the particle size distribution and plutonium concentration of each particle size fraction.

particle size range	net weight		Pu concentration in aliquot (dpm/g)		Pu dpm in group
2000-850	7.24	X	10.6	=	76.74
850-425	11.82	X	19.8	=	234.04
425-250	5.64	X	21.3	=	120.13
250-180	4.79	X	23.9	=	114.48
180-150	4.95	X	26.4	=	130.68
150-75	10.38	X	38.9	=	403.78
75-45	6.84	X	51.7	=	353.63
45-0	9.34	X	100.4	=	937.74
Total	61.0				2371.22
mean concentration = $2371.22/61 = 38.87$ dpm/g					

A regression of plutonium concentration in 0-3 cm soil versus radial downwind distance from the center of the asphalt pad (Figure 5) included samples from the downwind transect, macroplot 1 samples and "hot spot" samples. The dotted portion of the line which approximates the area covered by the asphalt pad is merely an extrapolation with supporting data.

A regression of plutonium concentration in 0-3 cm soil versus downslope distance which included samples from macroplot 1 and the downslope transect had the result shown in Figure 6. The distances in the equation are measured south from a line running east to west through the center of the asphalt pad. Although this equation does not fit its data as well as the downwind equation, only 14% of the variation is left unexplained.

Multiple linear regressions of the form $C = b_1 + b_2X + b_3Y$ where X is the distance east of the asphalt pad and Y is the distance south of the asphalt pad yielded very poor results. Perhaps multiple nonlinear regressions would result in better fits, but the expense and difficulty in assessing fits would have to be taken into account.

Concentration and Particle Size: An important relationship is the connection between plutonium concentration of soil and particle size of the sample. This was pursued using data adjusted by the downwind and downslope regressions discussed earlier. The soil plutonium concentration was regressed versus the attachment area for each depth group in both macroplots. Attachment area is an estimate of the amount of surface area available in a given volume of soil

Additional soil samples were included in the regressions; macroplot 1 and Hot Spot samples in the DW curve, and macroplot 1 samples in the DS curve. Since the macroplot 1 samples had been sieved into various particle size fractions and the DW, DS and Hot Spot samples were not, the macroplot 1 samples were reconstructed with regard to soil particle size distribution to their original state, i.e., the weighted average plutonium concentration was calculated.

The weighted average plutonium concentration of a given macroplot 1 soil sample was calculated by summing over all particle size groups the products of plutonium concentration of a given size group and the total mass of that as shown in Table 10. Even though small variations may result from aggregations of small particles into larger ones and variations in sieving time, this method effectively removes the variability of plutonium concentration associated with the particle size distribution.

For each relationship, a simple linear regression, a power function regression, and an exponential regression were calculated by linearizing the data; only the best fits (largest correlation coefficient) are reported herein. A simple linear regression takes the form $Y = a + bX$; a power function, $Y = aX^b$; and an exponential function, $Y = ae^{bx}$,

where Y = dependent variable

X = independent variable

a = intercept

b = slope.

Figure 5. Plutonium concentration in 0-3 cm soil versus distance downwind from center of asphalt pad.

Figure 6. Plutonium concentration of 0-3 soil versus distance downslope (see Figure 4).

uniformly filled with given sized soil spheres; the estimate is based on surface to volume ratio and the known particle size range from sieving data (Table 11). The same plutonium concentration data were plotted versus the soil particle diameter for each depth and macroplot. Linear, exponential, and power function models were attempted for each association.

Of the six curve types plotted, the power function model of Pu as a function of soil particle diameter gave the most consistently significant results (Table 12). The correlation coefficients for the macroplot 1 data are all significant ($p < .05$). The poor fits for the macroplot 2 data, significant only for the 6-9 through 15-18 cm depth groups, probably result from the relatively high degree of variability in that macroplot compared to macroplot 1.

Concentration and Depth: The plutonium concentration data from macroplot 1 were adjusted to a common point near the center of the macroplot, using the DW and DS regressions. These distance-adjusted data were regressed as a power function of depth resulting in the statistically significant ($p < .05$) equations in Figure 7. Macroplot 2 data (Figure 8), having no distance variation, were not corrected for distance.

The macroplot 2 curve has a much lower correlation coefficient than the macroplot 1 curve. The great deal of spatial variation evidenced in the macroplot 2 soils is a likely cause of the low correlation.

Multiple Linear Regression: A number of attempts were made to correlate plutonium concentration in soil depth, particle

Table 11. Relationship between sieve fractions and the attachment area function.

opening size, d (cm)		attachment area, cm ² /g ^a
accumulation sieve	passage sieve	
8.50 x 10 ⁻²	2.00 x 10 ⁻¹	25.0
4.25 x 10 ⁻²	8.50 x 10 ⁻²	58.8
2.50 x 10 ⁻²	4.25 x 10 ⁻²	118
1.80 x 10 ⁻²	2.50 x 10 ⁻²	200
1.50 x 10 ⁻²	1.80 x 10 ⁻²	278
7.50 x 10 ⁻³	1.50 x 10 ⁻²	334
4.50 x 10 ⁻³	7.50 x 10 ⁻³	667
0	4.50 x 10 ⁻³	1112

^aThe minimum value; assuming spheres of uniform diameter equal to the value of the passage sieve opening, a density of 1.2 g/cm³ for soil in the 0-15 cm depths, and that the total volume in any given sample is accounted for by the particles contained in the sample.

Table 12. Power function regression coefficients of soil plutonium concentration in dpm/g, corrected for the downwind and downslope transects, versus soil particle diameter in cm at various depths.

macroplot	depth	intercept(a)	slope(b)	correlation coefficient(r)	n
1	0-3	202.42	-.313	-.262 ^a	72
	3-6	54.67	-.281	-.311 ^a	69
	6-9	4.06	-.733	-.502 ^a	50
	9-12	8.31	-.550	-.524 ^a	69
	12-15	3.14	-.793	-.706 ^a	52
	15-18	1.36	-.768	-.710 ^a	47
	18-21	1.16	-.562	-.369 ^a	22
2	0-3	1.80	-.320	-.137	100
	3-6	0.34	-.390	-.176	71
	6-9	0.07	-.658	-.290 ^a	91
	9-12	0.05	-.835	-.446 ^a	93
	12-15	0.11	-.565	-.269 ^a	80
	15-18	0.03	-1.06	-.519 ^a	66
	18-21	0.24	-.596	-.287	49

^aSignificant at $\alpha = .05$.

Figure 8. Plutonium concentration in macroplot 2 soil versus depth of sample.

size, and distance using multiple regression techniques. One difficulty in using particle size range in such regressions is meaningfully relating particle size to plutonium concentration. This problem was approached by converting the particle size of the sample to the term known as attachment area (Table 11).

The minimum attachment area, which has units of cm^2/g , is a result of the surface-to-volume ratio of the soil particles, assumed to be perfect spheres, and the density of the soil, g/cm^3 . To achieve a value of the attachment area for a given soil particle distribution, e.g., 2000-850, 850-425 μm , etc., the diameter of the particle size distribution in question was taken to be equal to the opening of the passage, rather than the accumulation, sieve in cm. Consequently, in a range of particles from 2000 to 850 μm , the value of 0.2 cm was entered into the formula to calculate attachment area. This procedure prevents difficulties such as 0 cm diameter particles if accumulation sieve opening size was used or unequal interpolation values if intermediate values were used.

The overall effect on attachment area of using the largest possible diameter for each distribution is to decrease the available attachment area for each particle size group, i.e. a conservative estimate of attachment area. Even so, the 45-0 μm diameter particles yield minimum estimated value of 1112 cm^2/g soil. Assuming that the Pu oxide particles do attach to soil by some surface-dependent mechanism, this represents an immense number of potential sites for attachment.

The attempts to fit the soils data into significant multiple linear regressions using sample depth, attachment area, and distance were not satisfying (Tables 13 and 14). For the macroplot 1 data, the best equation of many attempts using several transforms of the independent variables explained only 55% of the total variation associated with these data. For macroplot 2, the best fit was only about 7% of the total variation. Several microplots had reasonably good fits, but there is a large amount of variation between microplots with the given model. The macroplot 2 data, as expected from the variation expressed earlier, had poorer fits in the multiple linear regression attempts.

One possible reason for the failure of the multiple regression attempts is the fact that the actual distribution of particle sizes in a given soil sample was not taken into account by the model. Therefore, a sample predominantly composed of smaller particles, with a higher overall concentration as a result of that composition, was not corrected for. To achieve a meaningful fit of any multiple regression, the distribution of particles within each sample must be described, i.e., the particle size variability must be adjusted in some manner.

Summary: Plutonium concentration in the soil at Rocky Flats is strongly related to the particle size distribution of the particular sample in question, the depth of the sample, and the sample location with regard to the predominant wind vector.

In terms of percent change over the soil volume in question (macroplot 1 to a depth of 21 cm), these relationships do not all have

Table 13. Coefficients of multiple linear regression for macroplot 1 soils ($\ln Pu = b_0 + b_1 \ln \text{DEPTH} + b_2 \text{AREA} + b_3 \text{DIST}$).

microplot(n)	ln of sample depth (cm)		attachment area (cm ² /g soil)		source-to-sample distance, m		b ₀	Total R ²	F	approximate ^a p
	b	R ²	b	R ²	b	R ²				
all (336)	-1.210	.436	1.34 x 10 ⁻³	.082	-1.15 x 10 ⁻²	.034	10.17	.552	135.7	10 ⁻⁵
01 (39)	-.8716	.513	8.76 x 10 ⁻⁴	.124	--	--	6.658	.637	30.72	10 ⁻⁴
13 (22)	-.155	.017	7.18 x 10 ⁻⁴	.116	--	--	6.208	.132	1.45	.259
20 (35)	-1.183	.636	7.12 x 10 ⁻⁴	.030	--	--	6.958	.666	31.86	10 ⁻⁴
29 (35)	-1.286	.217	3.23 x 10 ⁻³	.353	--	--	5.903	.570	21.18	10 ⁻⁴
34 (92)	-1.101	.443	9.14 x 10 ⁻⁴	.038	--	--	7.213	.481	41.35	10 ⁻⁴
39 (36)	-1.533	.736	1.43 x 10 ⁻³	.088	--	--	7.806	.823	76.95	10 ⁻⁴
53 (34)	-2.124	.779	1.21 x 10 ⁻³	.037	--	--	8.000	.816	68.55	10 ⁻⁴
72 (43)	-1.467	.482	1.53 x 10 ⁻³	.072	--	--	6.256	.554	24.88	10 ⁻⁴

^asignificance level of the equation

Table 14. Coefficients of multiple linear regression for macroplot 2 soils ($\ln Pu = b_0 + b_1 \ln \text{Area} + b_2 \ln \text{Depth}$).

Microplot(n)	Ln Area		Ln Depth		b_0	R^2	F	p^a
	b	ΔR^2	b	ΔR^2				
all (484)	.489	.044	-.474	.025	-1.17	.069	17.83	10^{-4}
15 (176)	.400	.025	.557	.026	-2.47	.051	4.623	.011
24 (94)	.656	.099	-1.50	.451	.137	.550	55.64	10^{-4}
32 (41)	.517	.216	.092	.005	-2.50	.221	5.383	.009
35 (32)	.271	.074	--	--	-1.17	.074	2.401	.132
53 (40)	.246	.017	-.797	.140	-.838	.157	3.455	.042
100 (37)	.739	.023	-4.06	.740	3.11	.763	54.47	10^{-4}
200 (38)	1.18	.179	.873	.076	-6.46	.255	6.004	.006
300 (26)	-.736	.094	1.24	.155	3.13	.249	3.818	.037

^a significance level of the equation

the same relative effect on plutonium concentration. Depending on the regression equation used (Table 12), particle size over the range of diameters considered, changes the calculated plutonium concentration from 70% to several orders of magnitude. As the depth increases from 1 cm to 21 cm, the Pu concentration decreases by almost 100%. Across the boundaries of macroplot 1, the DW regression effects a 74% decrease in Pu concentration in soil and the DS regression about an 82% decrease. Depth and particle size regressions for macroplot 2 had relative effects similar to the same relationships for macroplot 1. Much of the variation seen in soil aliquots from varying particle size groups can be removed by correcting for the particle size distribution.

Litter and Vegetation

The mean plutonium concentration for Rocky Flats litter and vegetation samples from macroplots 1 and 2 are listed in Table 15. The litter has a higher concentration of plutonium than vegetation in both macroplots ($p < .05$). Furthermore, macroplot 1 has a higher concentration of Pu in both litter and vegetation than macroplot 2 ($p < .05$). Interestingly, the coefficients of variation for vegetation are higher by about a factor of three than the litter CVs in both macroplots.

Generally, the CVs associated with the litter and vegetation data are similar to the soil CVs. A potential reason for the relatively low CVs in litter is the flotation method of removing loosely-bound soil from the litter, which may select for a relatively narrow distribution of small particles.

Table 15. Plutonium concentrations, dpm/g, in Rocky Flats vegetation and litter samples.

macroplot	plutonium concentration, dpm/g					
	litter			vegetation		
	mean	CV	n	mean	CV	n
1	913.7	0.65	29	63.4	2.02	76
2	1.70	1.41	30	1.25	4.05	78

Kolmogorov-Smirnov (K-S) one-sample tests of the four data groups listed in Table 15 indicate that the vegetation of both macroplots 1 and 2 are log normally distributed ($p < .05$). In agreement with this, the skewness and kurtosis associated with the untransformed vegetation data were greatly alleviated when the $\ln$ transform was applied. K-S tests of the litter showed these data to be normally distributed ($p < .05$).

Plutonium concentrations in macroplot 1 and vegetation were each regressed as a power function against downwind and downslope distances (Figures 9-12) including samples from the same transects previously mentioned for soil (Figure 4). The equations of the litter data fit better than the vegetation data for both the downwind and the downslope relationships. This fact may result from the method of preparation of litter, discussed earlier in this section, which might have lowered the variance. For both downwind curves (Figures 9 and 10), the dashed line indicates the area of the unsampled asphalt pad.

Comparisons of the slopes of downwind litter and vegetation are interesting. A t-test of the slope of litter (b_ℓ) versus the slope of vegetation (b_v), where $t = (b_\ell - b_v) / \text{standard error of } b_\ell$ (Draper and Smith, 1966), indicated no difference between the slopes in Figures 9 and 10 ($\alpha = .05$). This finding may provide a clue to the mechanism (or mechanisms) leading to the observed contamination levels in the litter and vegetation.

The downslope data curves (Figures 11 and 12) indicate about the same order of difference between compartment means as do the downwind

Figure 9. Plutonium concentration of litter versus distance downwind from center of asphalt pad.

Figure 10. Plutonium concentration of vegetation versus distance downwind from center of asphalt pad.

Figure 11. Plutonium concentration of litter versus downslope distance (see Figure 4).

γ

Figure 12. Plutonium concentration of vegetation versus downslope distance (see Figure 4).

data; the intercepts again are two orders of magnitude different. However, the downslope curves differ from the downwind curves by virtue of having significantly different slopes ($p < .05$); the slope of the vegetation curve having a higher absolute value.

In summary, the litter and vegetation data bear out the following points. Litter and vegetation from both macroplots evidence detectable plutonium contamination. Concentration means are higher in litter than in vegetation, and higher in macroplot 1 than in macroplot 2. Pu concentration in both litter and vegetation is strongly related to both downwind and downslope distance terms.

Arthropods and Small Mammals

Although relatively unimportant in terms of total amount of plutonium contained, the arthropods and small mammals are important constituents of the ecosystems. Study of these animals may yield information about mechanisms of plutonium mobility and distribution. Understanding the relationship between these animals and the source of contamination is difficult because of the organism's mobility. Further, arthropods were difficult to sample in quantities adequate for detection of contamination and small mammal data were sometimes confounded by small sample masses.

Arthropod samples from sweep netting or pitfall trapping were separated into similar classes, orders, or families for plutonium analysis as a group (Table 16). The mean plutonium concentration in macroplot 1, 7.03 dpm/g, appears to be about five times as large as the macroplot 2 mean, 1.45 dpm. A t-test of this difference was

Table 16. Plutonium concentrations in single samples of various arthropod classifications.

macroplot	class	order	family	Pu concentration, dpm/g (CV)
1	Arachnoidea	Arachnida ^a	mixed	4.26
	Insecta	Thysanura	Machillidae	7.84
	Insecta	Orthoptera	Acrididae	2.13
	Insecta	Orthoptera	Gryllidae	13.38
	Insecta	Orthoptera	mixed	26.1
	Insecta	Orthoptera	mixed	.628
	Insecta	Orthoptera	mixed	2.52
	Insecta	Coleoptera	Carabidae	2.85
	Insecta	Coleoptera	Scarabaeidae	7.79
	Insecta	Coleoptera	Tenebrionidae	2.83
	Mean			7.03 (1.09)
2	Arachnoidea	Arachnida ^a	mixed	1.68
	Insecta	Orthoptera	Acrididae	.15
	Insecta	Orthoptera	mixed	.202
	Insecta	Orthoptera	mixed	1.10
	Insecta	Hymenoptera	Formicidae	3.90
	Insecta	Coleoptera	Silphidae	1.41
	Insecta	Coleoptera	Tenebrionidae	1.69
	Mean			1.45 (0.87)

^aArachnida is actually a subclass, not an order.

not significant ($\alpha = .05$); but, a t-test of the difference between macroplot means using log transformed data was significant ($p < .05$). This latter result may indicate a skewed distribution.

The small mammal data offered some of the most interesting yet puzzling data in the project (Table 17).¹ The overall means for all tissues were 14.4 and 26.2 dpm/g, for macroplot 1 and 2, respectively. A tissue was designated external or internal depending on the presence or absence of direct contact with the animal's environment. External tissues included gastrointestinal tract (GI), hide and lung, while bone, kidney, liver and muscle were internals. Macroplot 1 internal tissues averaged 19.1 dpm/g while externals had a mean of 8.62. In macroplot 2, internal tissues averaged 16.1 dpm/g and external tissues 38.9 dpm/g. The bone seemed to have the lowest concentration in each macroplot with the highest mean being kidney in macroplot 1 and lung in macroplot 2.

There also appears to be a wide range of concentrations between tissues. The bone mean in macroplot 1 is nearly 50 times lower than the kidney mean concentration. In macroplot 2, the between tissues range is from .038 dpm/g in bone to 99.9 dpm/g in lung. The within tissue variation is also extreme. The minimum coefficient of variation (CV) is 1.59 in macroplot 2 liver, and the macroplot 1 muscle group has the highest CV of 5.85.

¹To allow expression of the low bone activities, data in this table were corrected for counting background, but not for the mean blank.

Table 17. Plutonium concentrations, dpm/g, in Rocky Flats small mammal tissue samples.

macroplot	tissue	n	²³⁹ Pu concentration dpm/g	
			mean	CV
1	externals - GI	40	15.6	2.50
	hide	47	3.35	1.84
	lung	47	7.93	1.90
	all	135	8.62	2.76
	internals - bone	28	.640	2.28
	kidney	45	30.2	4.39
	liver	46	18.6	5.45
	muscle	50	19.8	5.85
	all	169	19.1	5.58
	all tissues	304	14.4	5.62
	externals - GI	22	4.86	1.94
	hide	27	5.84	4.00
	lung	27	99.9	2.84
2	all	76	38.9	4.45
	internals - bone	18	.038	2.88
	kidney	25	36.1	3.92
	liver	27	2.91	1.59
	muscle	26	21.8	3.36
	all	96	16.1	5.07
	all tissues	172	26.2	4.98

Probably the most far-reaching characteristics of the data are the non-normal distributions. With the exception of the macroplot 2 bone samples, the Kolmogorov-Smirnov (K-S) goodness of fit test indicated very low probabilities that the various small mammal tissue groups were normally distributed (Table 18).

Histograms of the small mammal data including all tissues graphically illustrate the non-normality (Figures 13 and 14). For each macroplot histogram, the plotted values equal the plutonium concentration (dpm/g) of each tissue sample divided by the mean of that tissue group. The long tail and the piling up of cases below the mean in both figures is characteristic of skewness. For Figures 13 and 14, the skewness coefficients are 4.97 and 4.524, and the kurtosis coefficients are 29.280 and 22.952, respectively. All of these values are significant at the .01 level.

Perhaps because of non-normality and large coefficients of variation, a t-test indicated no significant difference between the two macroplot means for all tissues listed in Table 17 ($\alpha = .05$), while the Kolmogorov-Smirnov two-sample test found a difference ($p < .001$). The t-test indicated a difference between GI means between macroplots ($p < .05$). The K-S test further indicated differences between macroplots for internal tissues and between external and internal tissues for macroplot 1 ($p < .05$). The Mann-Whitney test found differences between macroplots for bone and between external and internal tissues for macroplot 1 ($p < .05$).

To avoid the problem of non-normality, a nonparametric test, the Kruskal-Wallis (K-W) oneway ANOVA was used to test for significant

Table 18. Kolmogorov-Smirnov goodness-of-fit test results of various small mammal data, plutonium concentration, dpm/g, versus a normal distribution having the same mean and standard deviation value as the sample group tested.

sample group tested		n	approximate ^a probability (α)
macroplot	tissue		
1	externals - GI	41	4.0×10^{-4}
	hide	47	1.7×10^{-3}
	lung	47	2.0×10^{-4}
	all	135	$< 10^{-5}$
	internals - bone	28	1.7×10^{-3}
	kidney	45	$< 10^{-4}$
	liver	46	$< 10^{-4}$
	muscle	50	$< 10^{-4}$
	all	169	$< 10^{-5}$
	all tissues	304	$< 10^{-5}$
	externals - GI	22	1.8×10^{-2}
	hide	27	$< 10^{-4}$
	lung	27	2.0×10^{-4}
2	all	76	$< 10^{-4}$
	internals - bone	18	9.9×10^{-1}
	kidney	25	3.0×10^{-4}
	liver	27	1.3×10^{-2}
	muscle	26	1.0×10^{-4}
	all	96	$< 10^{-5}$
	all tissues	172	$< 10^{-5}$

^atwo-tailed probability that fit between the sample distribution and the specified normal distribution could be achieved by chance variations alone.

Figure 13. Histogram of plutonium concentrations in Rocky Flats macroplot 1 small mammal tissues.

Figure 14. Histogram of plutonium concentrations in Rocky Flats macroplot 2 small mammal tissues.

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 macroplot 1, the K-W calculated a chi-square value of about 44. For macroplot 2, the chi-square was approximately 39. These statistics indicate significant ($p < .001$) differences between the tissues within a given macroplot. There was difficulty in determining the tissue or tissues of the seven tested for each macroplot which were different from the rest. Because of the non-normality of the distributions, a parametric test such as the Least Significant Difference test (LSD) was likely not valid.

In the absence of such tests, the rank sums and mean ranks per sample for each tissue group in each macroplot were inspected (Table 19). Clearly, in both macroplots, the bone tissues have much lower 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 are still much lower. While this result is not testable, as stated above, it seems reasonable in light of the significant K-W results that the bone mean is different from most, if not all, of the other tissue mean.

Table 20 summarizes the results of parametric and non-parametric tests of equality for the small mammal data. At the .05 level of significance, there is disagreement between test results more often

Table 19. Kruskal-Wallis oneway analysis of variance rank sums for small mammal tissue groups (corrected for ranking ties).^a

macroplot	tissue	n	rank sum	mean rank ^b per sample
1	bone	29	2163.5	74.6
	liver	46	6515.0	141.6
	muscle	50	6621.5	132.4
	hide	47	6790.5	144.5
	GI	40	7702.0	192.6
	kidney	45	7829.5	174.0
	lung	47	8738.0	185.9
2	bone	18	509.0	28.3
	GI	22	1937.0	88.0
	hide	27	2060.0	76.3
	liver	27	2311.5	85.6
	muscle	26	2325.5	89.4
	kidney	25	2534.0	101.4
	lung	27	3201.0	118.6

^aAll of the samples in a macroplot were ranked in ascending order with rank value 1 assigned to the lowest-valued sample and rank 304 (for macroplot 1) assigned to the highest-valued sample. The ranks for each tissue were summed and a chi-square calculated. For a detailed discussion, see Siegel (1956) pp. 184-194.

^bThe mean rank per sample has no recognized statistical value and is used here for intuitive purposes only.

Table 20. Results of parametric (t-test) and nonparametric (Kolmogorov-Smirnov two-sample and Mann-Whitney U) tests of equality of plutonium concentrations, dpm/g, in small mammal tissues.^a

test conducted ^c	approximate alpha level of resultant statistic ^b		
	t-test	K-S two-sample	M-W U
externals			
GI, MP1 vs. MP2	.217	.086	.045
hide, MP1 vs. MP2	.489	.053	.091
lung, MP1 vs. MP2	.029	.726	.429
all, MP1 vs. MP2	.046	.066	.136
internals			
bone, MP1 vs. MP2	.089	.074	.018
kidney, MP1 vs. MP2	.864	.768	.869
liver, MP1 vs. MP2	.426	.257	.715
muscle, MP1 vs. MP2	.934	.398	.669
all, MP1 vs. MP2	.816	.016	.232
all tissues, MP1 vs. MP2	.224	1.7×10^{-4}	.076
MP1, externals vs. internals	.267	1.0×10^{-4}	2.0×10^{-4}
MP2, externals vs. internals	.256	.084	.054

^aThe t-test is a test for equality of sample means, while the K-S two-sample and M-W U tests are measures of sameness of distribution, i.e., they indicate differences in central tendency, shape, etc.

^btwo-tailed probability

^cMP1 = macroplot 1; MP2 = macroplot 2

than agreement. For instance, when all of the tissues in macroplot 1 were tested against the tissues in macroplot 2, only the K-S test indicated a difference between the macroplots. The three tests agree that there are no differences between macroplots for hide, kidney, muscle or between internal and external tissues for macroplot 2. Further, the tests disagree on differences between macroplots for GI, lung, externals, bone, internals, and between macroplot 1 internals and externals.

The decision to label a test "significant" or "not significant" is arbitrary. For bone, if the level of significance was .10, the outcome of the three tests of significance listed (Table 20) agreed that there are indeed differences in the plutonium concentrations of bone between macroplots 1 and 2.

The light masses of the small mammal samples were a source of concern. In several cases, masses as low as 0.07 g were noted. A plot of plutonium concentration in dpm/g in macroplot 1 small mammal tissues suggests that small sample masses might be biased toward higher concentrations (Figure 15). For instance, 100 dpm in 1 g is 100 dpm/g, but a 100 dpm particle in .07 g equals 1429 dpm/g. Such samples would obviously add variation to the data unnecessarily. Fortunately, the number of small mass samples exhibiting extremely large concentrations is limited. The plotted digits in Figure 15 indicate up to nine data points in close proximity.

In attempts to lower the expressed variation and normalize the data, the possibility of transforming the small mammal plutonium concentrations data was examined. Some transforms, such as the

Figure 15. Plutonium concentration, dpm/g, in Rocky Flats macro-plot 1 small mammals versus sample mass, g.

logarithmic and square root transforms were not practical because of the presence of zero or less than zero data values. Expression of plutonium as quantity per sample (dpm/sample rather than dpm/g) was possible and was carried out.

Expressing the data as dpm/sample rather than dpm/g did little to lessen the variability or achieve normality. Most coefficients of variation were increased from their value when the dpm/g data were used. The K-S goodness-of-fit test indicated that none of the tissue groups were normally distributed. Therefore, expressing the data as dpm/sample rather than dpm/g was not a useful exercise for these analyses.

In summary, the plutonium concentrations in some small mammal tissues in microplot 2 appear higher than those in macroplot 1. Both macroplots have a large amount of variation associated with the mean values in each tissue and are not normally distributed. There is some indication that the internal and external tissues within each macroplot are from different distributions and that the mean values for internal and external tissues are different between macroplots.

Plutonium Concentration Ratios

A potential indicator of the extent of plutonium redistribution by wind, water, or plant uptake is the concentration ratio. Concentration ratio (CR) is defined as the concentration of a given nuclide in activity per unit of mass or volume of a material divided by the concentration of the same nuclide in activity per unit of mass or

volume of another material. In this section, the concentration ratio will always have 0-3 cm soil as the material in the denominator, e.g.,

$$\text{CR of litter} = (\text{dpm Pu/g litter}) \div (\text{dpm/g 0-3 cm soil}).$$

The concentration ratios for each macroplot of litter, vegetation, arthropods, and small mammals are given in Table 21. In macroplot 1, the litter has the highest ratio, followed in descending order by vegetation, small mammals, and arthropods. In macroplot 2, the small mammals are highest (suggesting laboratory bias) followed by litter, vegetation and arthropods.

Regressions of litter and vegetation concentration ratio as a function of downwind and downslope distances did not achieve significant correlation coefficients.

To examine further the relationship between soil and the litter and vegetation, the plutonium concentration in each of litter and vegetation versus the plutonium concentration of soil from the same locations was plotted (Figures 16 and 17). The litter curve is quite interesting because of its high correlation (.975), near-unit slope (1.001) and intercept (1.09). The vegetation data (Figure 17) are less conclusive than the litter data, but are still statistically significant ($p < .01$).

Plutonium Isotopic Ratios

Soil

This section describes the ratios of the two predominant plutonium isotopes, ^{238}Pu and ^{239}Pu , in Rocky Flats soil. The ratio, IR, is the ^{239}Pu content, dpm/sample (or concentration, dpm/g) divided

Table 21. Concentration ratios (CR) and 95% confidence intervals (CI) of litter, vegetation, arthropods, and small mammals with 0-3 cm soil from Rocky Flats; concentration ratio, litter = (mean dpm/g of litter) ÷ (mean dpm/g 0-3 cm soil).

compartment	macroplot 1		macroplot 2	
	CR	95% CI	CR	95% CI
litter	4.93×10^{-1}	$2.86 \times 10^{-1} - 7.00 \times 10^{-1}$	1.39×10^{-2}	$-7.94 \times 10^{-4} - 2.86 \times 10^{-2}$
vegetation	3.42×10^{-2}	$1.45 \times 10^{-2} - 5.39 \times 10^{-2}$	1.02×10^{-2}	$-3.03 \times 10^{-3} - 2.34 \times 10^{-2}$
arthropods	3.97×10^{-3}	$8.73 \times 10^{-3} - 6.71 \times 10^{-3}$	1.18×10^{-5}	$-1.59 \times 10^{-6} - 2.52 \times 10^{-5}$
small mammals	7.77×10^{-3}	$2.09 \times 10^{-3} - 1.34 \times 10^{-2}$	2.14×10^{-1}	$-4.16 \times 10^{-2} - 4.70 \times 10^{-1}$
mean Pu concentration (dpm/g) and (CV) in 0-3 cm soil:	1853 (1.46)		122.6 (4.61)

Figure 16.. Plutonium concentrations in litter versus plutonium concentration in soil from the same locations.

Figure 17. Plutonium concentration in vegetation versus plutonium concentration in soil at the same locations.

by the ^{238}Pu content or concentration, with the same units as the numerator, of a given sample. The ratio was calculated only if both isotopes were detectable in the sample. The number of samples available for isotopic ratios was relatively low for soils because most of the soil samples were processed in our own laboratory, i.e., by liquid scintillation counting. Plutonium-238 was not detectable in many of the samples processed in commercial labs.

Plutonium isotopic ratios in Rocky Flats soil are catalogued by macroplot and depth in Table 22. The mean ratio in macroplot 1 generally decreases from a high of 65 in the surface level to below 40 in the 18-21 cm level, with an overall mean of 51.1. Data from near the surface in macroplots 1 and 2 are similar, but the ratio decreases quickly to a value of less than 13 for the lower depth categories of macroplot 2.

The isotopic ratio data appear to be much less variable and more closely approximate a normal distribution than the Pu dpm/g data. Although the overall coefficient of variation (CV) for macroplot 2 isotopic ratios is still large, the below 3 cm groups show relatively low CVs in both macroplots. The values of IR, $\ln \text{IR}$, and $\ln(\ln \text{IR})$ were tested for goodness-of-fit against a normal distribution with the same mean and standard deviation as the tested groups. From these tests, both macroplots 1 and 2 seemed to be normal. The exception was the 0-3 cm depth group for each macroplot which, by the same criteria, seemed to be log normal.

The apparent inverse relationship between IR and the sample depth in cm did not result in significant regressions ($\alpha = .05$) for

Table 22. Plutonium isotopic ratios in Rocky Flats soil; all particle sizes included.

macroplot	depth, cm	^{239}Pu dpm/g/ ^{238}Pu dpm/g		n
		mean	CV	
1	0-3	65.4	.95	10
	3-6	44.6	.19	7
	6-9	48.4	.27	8
	9-12	46.8	.13	6
	12-15	49.1	.12	6
	15-18	47.3	.15	3
	18-21	38.7	.01	2
	all	51.1	.61	42
2	0-3	66.0	2.15	24
	3-6	25.6	.93	5
	6-9	7.27	.57	6
	9-12	6.45	.80	4
	12-15	8.74	.07	2
	15-18	12.9	.39	5
	18-21	7.92	0.40	5
	all	37.3	3.25	51

either macroplot. Kolmogorov-Smirnov (K-S) two-sample tests and t-tests of each depth group between macroplots indicated that macroplot 1 has a higher isotopic ratio for the overall mean and for every depth group mean except for the top two groups.

Outstanding trends in the soil isotopic ratio data are, perhaps, difficult to see due to the limited amount of data. Several facts stand clear, however. In macroplot 1 the ratio is near 50, but the ratio in macroplot 2 is about 25% lower. The surface levels are the same for each macroplot, but the ratio appears to decrease more rapidly with depth in macroplot 2.

Litter and Vegetation

The limited number of litter and vegetation samples analyzed by commercial laboratories are summarized in Table 23. No statistical comparisons can be made of the macroplot 2 data because of sample size. In macroplot 1 t-tests indicated no difference between the means of the litter and vegetation data. However, a t-test of the overall mean (litter and vegetation combined) of macroplot 1 versus the mean of macroplot 2 indicated a significant difference at the $\alpha = .05$ level. This finding is compatible with the soil results.

Arthropods and Small Mammals

The arthropod samples analyzed by commercial laboratories which resulted in detectable amounts of both ^{239}Pu and ^{238}Pu are listed in Table 24. Individual samples of each listed category were analyzed and the isotopic ratio calculated.

Table 23. Plutonium isotopic ratio means in litter and vegetation from Rocky Flats macroplots 1 and 2.

macroplot	plutonium isotopic ratio (^{239}Pu dpm/g/ ^{238}Pu dpm/g)					
	litter			vegetation		
	mean	CV	n	mean	CV	n
1 ^a	54.77	0.06	4	60.58	0.17	3
2	42.23	--	1	36.63	--	1

^a significant difference in overall means of macroplot 1 and macroplot 2 ($p < .05$).

Table 24. Plutonium isotopic ratios in single samples of various arthropod classifications.

macroplot	class	order	family	$\frac{^{239}\text{Pu dpm/g}}{^{238}\text{Pu dpm/g}}$ (CV)
1	Arachnoidea	Arachnida ^a	mixed	8.69
	Insecta	Thysanura	Machillidae	9.56
	Insecta	Orthoptera	Acrididae	9.26
	Insecta	Orthoptera	Gryllidae	12.9
	Insecta	Orthoptera	mixed	47.0
	Insecta	Orthoptera	mixed	3.75
	Insecta	Coleoptera	Carabidae	10.6
	Insecta	Coleoptera	Scarabaeidae	4.45
	Insecta	Coleoptera	Tenebrionidae	10.9
	Mean			13.01 (1.01)
2	Insecta	Orthoptera	mixed	.75
	Insecta	Hymenoptera	Formicidae	7.65
	Insecta	Coleoptera	Tenebrionidae	11.3
	Mean			6.57 (0.82)

^a Arachnida is actually a subclass, not an order.

The values in macroplot 1 range from 3.75 to 47.0 and in macroplot 2 from .75 to 11.3. The coefficients of variation are, therefore, large, and macroplot 1 seems to be more variable than macroplot 2. Using t-tests, the means of the two macroplots indicated no significant difference ($p < .05$). In both macroplots, the mean values are much lower than the mean values seen earlier for the upper soil layers and for litter and vegetation.

Even though a relatively large number of small mammal tissues were processed for plutonium by commercial labs a fairly small number of isotopic ratios resulted (Table 25). This fact results from several factors, including the laboratories' reporting practices and the low concentrations of ^{238}Pu in many of the samples.

T-tests and Kolmogorov-Smirnov two-sample tests indicate that for all tissues combined the macroplot 1 IR is larger than the macroplot 2 IR (Table 25). Further, these tests agree that the macroplot 1 bone, hide, and internal tissues all have higher values of IR relative to the corresponding tissue group in macroplot 2.

The coefficients of variation of these data appear to be much lower than previously seen in the small mammal concentrations data reported earlier. Also, variation from tissue type to tissue type was less than the concentrations data, ranging from a ratio of 2.65 in the macroplot 2 bone to 38.2 in the macroplot 2 lung. K-S goodness-of-fit tests of IR, $\ln \text{IR}$, and $\ln(\ln \text{IR})$ values indicated the distributions are likely to be log normal. Thereafter, all tests and calculations were performed on the log transformed isotopic ratios.

Table 25. Plutonium isotopic ratios (^{239}Pu dpm/g/ ^{238}Pu dpm/g) of Rocky Flats small mammal tissues.

macroplot	tissue	n	isotopic ratio	
			mean	CV
1	externals			
	GI	22	34.8	0.68
	hide	23	28.6	0.78
	lung	11	16.2	1.32
	all	56	28.6	
	internals			
	bone	9	11.8	0.81
	kidney	8	25.9	1.14
	liver	13	23.9	0.75
	muscle	11	34.4	1.32
	all	41	24.5	
	all tissues	97	26.9	0.96
2	externals			
	GI	9	32.1	1.14
	hide	10	16.8	0.99
	lung	8	38.2	1.10
	all	27	28.2	
	internals			
	bone	8	2.65	1.23
	kidney	7	25.4	1.59
	liver	9	25.4	1.39
	muscle	9	24.7	1.02
	all	33	19.7	
	all tissues	60	23.5	1.32

A three-way analysis of variance, two macroplots by seven tissues by two species (deer mouse and pocket gopher), of the ratios data showed a significant tissue effect at the $\alpha = .05$ level. No significant macroplot or species effects were shown. A one-way ANOVA of the seven tissue groups indicated significant tissue effects for both macroplots.

A least significant difference (LSD) multiple range test of $\ln IR$ of the tissues of macroplot 1 yielded the following homogeneous subsets ($\alpha = .05$): bone, lung, muscle, and kidney; lung, muscle, kidney, and liver; muscle, kidney, liver, hide, and gastrointestinal tract (GI). LSD results for the seven tissue groups in macroplots 2 are: bone and kidney; kidney, hide, GI, liver, muscle, and lung. These results seem to indicate that bone, and hide and GI are not equal in macroplot 1; in macroplot 2 bone seems to be separate of the other tissue groups.

In summary, there seems to be little difference between plutonium isotopic ratios in small mammal tissues in macroplot 1 and those in macroplot 2. The data seem to be log-normally distributed and are much less variable than the ^{239}Pu concentrations.

DISCUSSION

Plutonium Compartmentalization in the Ecosystem

The compartmental inventories were computed to assess the relative importance of the various ecosystem components as plutonium reservoirs. Although other investigations of Pu in the environment have been conducted, this study appears to be the only one in which such comparative inventories were measured. Knowledge of Pu compartmental inventories is important in conceiving a picture of distribution, assessing potential flows and hazards, and making land management decisions for contaminated areas.

The overwhelming fraction of the total ecosystem Pu is contained by the soil. The corollary of the above statement is that the other compartments sampled have very small proportions of the total Pu. Further, the animal compartment had much lower fractions of the total Pu than did the vegetation and litter compartments.

The implications of the fact that the soil has so much Pu are quite far-reaching. This fact strongly suggests most of the Pu mobility which occurred (or might be occurring) was also tied to soil behavior and mobility in some way. Therefore, to control large scale Pu movement one primarily needs to control soil movement. Because of this fact, soil conservation management techniques are very important.

Large scale plutonium mobilization by the botanic or zoologic members of the system has relatively low potential. This is not to say that biological members of the ecosystem should be ignored from

the standpoint of Pu movement. Knowledge of the migratory potential of animal populations and the individual's plutonium burden would allow prediction of Pu movement from a contaminated area.

In the generic sense, the compartmentalization data also clarify the relationships of the various compartments to the soil. It appears that as the relationship with soil became less intimate, the compartmental fraction of total Pu inventory decreased. For example, litter was more proximate with soil than arthropods in general and had a higher fraction of the total Pu. Though very crude, this concept might allow intuitive prediction of the relative importance of populations such as earthworms, reptiles, birds, or mule deer.

The compartmentalization results are important in future studies concerning Pu in the environment. Other researchers interested in modeling the flow of Pu at Rocky Flats would be spared the task of identifying the compartments which might be studied. Depending on the proposed sophistication and sensitivity of a future model, the compartmentalization results herein described might allow researchers to design their model more knowledgeably. These data indicate that researchers considering only Pu inventory of an area such as Rocky Flats are justified in sampling only soil, e.g., only soil was sampled in the Pu inventory of Rocky Flats by Krey and Hardy (1970). Future studies to measure potential of time in increasing Pu mobility could use the present data as a reference point. Finally, growing concern about possible increases in biological availability of actinides through time might also focus attention on these compartmental inventories.

The compartment inventories suggest that land management of Pu-contaminated areas should include soil conservation practices to prevent erosion and minimize soil disturbance. Further, large soil inventory fractions suggest the need for more study of soil processes; i.e., resuspension, migration, turnover, and fine Pu profile structure versus time. Soil ingestion by deer, cattle, etc., and denudation of soil by pocket gophers also appear important to quantify.

Plutonium in Soil

There are several salient points raised by the soil results. There was a significant difference in plutonium concentration in soil between macroplots. Similarly, the Pu concentration was significantly correlated to location relative to the source term. Further, the particle size distribution of the soil sample and its depth both influence the Pu concentration. Comparisons of the macroplots showed that the variability of data in macroplot 2 was higher than that in macroplot 1. Also, the effect of depth on the isotopic ratio apparently varies between macroplots.

These findings raise a number of questions concerning Pu mobility at Rocky Flats:

- 1) What is the likely mode(s) of transport of contamination from the source?
- 2) What causes the differences between macroplot means?
- 3) What is the significance of the particle size correlations?
- 4) What is the significance of the depth correlations?
- 5) Why is the relative variability higher in macroplot 2 than macroplot 1?

- 6) Why is the slope of Pu isotopic ratio versus depth steeper in macroplot 2 than macroplot 1?

To help answer some of the above questions a scenario of the contamination process will be described. The contents of the storage barrels and the likely interactions with the barrels were reported in the chapter, "Study Area and Plutonium Source". At various times the forces of nature, presumably internal condensation, wind, rain, frost and thaw, deteriorated some of the barrels to the point of leakage. The leakage was most likely not large or fast at first, but may have become so with time. The oil was deposited onto the ground surface and the plutonium contamination became bound by the soil, its eventual transport vector.

Depending on the condition of the soil first contacted and the form of the plutonium, the plutonium may or may not have attached immediately to a soil particle. The oil likely penetrated into the soil and stabilized it for some period. If the plutonium was deposited as a metal particle, it may have remained in position with little movement for some time. The plutonium metal bits likely oxidized slowly at normal temperatures in the presence of air. The resulting plutonium oxide was then relatively insoluble in water compared to so-called high-fired oxides but relatively insoluble in water compared to most metallic oxides. If plutonium chloride was deposited on the soil, it was likely hydrolyzed soon after first contact with water, and eventually became oxidized. The solubility of these compounds was probably again low relative to most compounds, but greater than the high-fired oxides (4 June 1976 telephone

conversation with J. M. Cleveland, Environmental Studies, Rocky Flats Plant). Eventually, however, it is highly probable that all PuO_2 particles, or molecules, became attached to soil particles.

Once the soil surrounding the barrel storage area became contaminated, the redistribution downwind of each plutonium leakage was probably a relatively short or erratic process occurring with disturbance and high winds. The data from the S-8 air sampler mentioned in an earlier chapter would seem to indicate that the redistribution past that sampling point, just adjacent to the eastern boundary of the oil barrel storage area, occurred over a period of about 24 years.

The S-8 air data, coupled with prevailing wind information (Figure 2) and the regressions of plutonium concentration versus downwind (DW) and downslope (DS) distance are strong evidence that wind was the primary mode of plutonium dispersion from the barrel storage area. Of course, there may have been some movement by water erosion downhill but this was probably minimal in and near the study plots because of dense vegetation cover. Tentative results of a study by the author using ^{59}Fe as a soil tracer (not described herein) gave little indication of intensive erosion by either wind or water on those study areas during a six-month period. Thus, wind was likely important in the initial distribution, but subsequent redistribution was probably minimal in stable areas.

The large differences in plutonium concentration in soil between macroplots 1 and 2 indicate that not only distance from the source term, but also direction with regard to the prevailing wind and the

presence of barriers are likely important. Macroplot 2 is almost due south of the barrel storage area, but has still received some contamination. However, except for a few relatively active samples, the mean activity of the macroplot would be much lower than found. A low mean activity was expected in macroplot 2 because of its location outside the prevailing wind pattern. Further, the distance between the source and the macroplot would tend to prevent contamination reaching macroplot 2.

Regarding the DW and DS regression, the slopes of the curves are notable. The slopes of both curves are relatively steep which probably means that some of the particles transported by the wind were rather large, i.e., the particles had a high terminal velocity (Healy and Fuquay, 1958). For instance, 8 μm diameter particles have a terminal velocity of 1.1×10^{-3} m/sec while 70 μm particles have a terminal velocity of .11 m/sec. If all the transported particles had been quite small, submicron or a few microns, the slopes of the DW and DS curves would probably have been much less steep.

The following exercise indicates the real possibility that erodible particles covered by the mean size Pu particles could account for the soil concentration at the source as estimated by the DW curve. Integrating the curve, $C_s = 1.09 \times 10^{11} X^{-3.321}$, from 1.0 to 82 m (approximately the eastern half of the barrel storage area) results in a mean Pu concentration in 0-3 cm soil of 5.80×10^8 dpm/g. To stay within the range of easily erodible particles, assume a soil particle diameter of 50 μm (Healy and Fuquay, 1958). Then assume that these 50 μm particles acted as host particles and became uniformly covered.

with 0.3 μ diameter Pu particles, likely near the mean oxide size found at Rocky Flats (telephone conversation, 14 June 1976, with J. Hayden, Environmental Studies, Rocky Flats plant). If the contaminated host particles are uniformly packed into a cubic centimeter of soil, an activity of 1.13×10^{10} dpm/g could result. This suggests that if five percent of that surface area contained by spherical 50 μ m particles was covered by 0.3 μ m Pu oxide particles, an activity of about 5.8×10^8 dpm/g could result. Thus, the host particle concept seem plausible.

In the sense that wind was an important causal factor in the contamination patterns as shown by the downwind and downslope regressions, the Rocky Flats data are similar to those seen by investigators at other sites (Christenson and Thompson, 1962; Romney et al., 1970; Hakonson and Johnson, 1973; Essington et al., 1975). These workers generally agree that plutonium contamination on their semi-arid to arid study areas was transported by wind or other physical factors rather than biological factors.

This is not to say that biological factors are totally uninfluential in contributing to transport. Burrowing by badgers or pocket gophers might allow previously sheltered particles to become subject to wind. Herbivory by mammals such as pocket gophers would remove shelter. Particles might be suspended above the soil surface by deer running or pocket mice engaging in dust baths. Ingestion of contaminated material followed by excretion at another location spreads some plutonium. Although these activities probably have

minimal impact in terms of total redistribution, single "hot" particles or agglomerates could be disturbed and moved.

At some point in time and space, the plutonium-contaminated soil particles were no longer redistributed significantly by wind; probably the particles were lodged in a lee, sheltered by larger particles, litter or vegetation. While lodged in the wind-protected position, the soil-plutonium particle may have been broken down by further weathering by the rain and freeze-thaw cycles. The presence of ^{238}Pu in the plutonium oxide might have contributed to particle deterioration through radiolysis. The eventual fate of the agglomerates probably depended on a number of factors including: the size and integrity of the original soil particles, the sizes and numbers of the attached Pu particles, the chemical form of the plutonium, and the isotopic ratio of the plutonium particles.

Prior to and during downward migration, the size distribution of the plutonium-soil particles was probably more complex than ever before; Pu particles might have detached from the soil particle and weathered or stayed attached and whole. A size range from submicron to ten microns is possible, but generally the Pu particles were probably smaller than at the source.

The mechanism of plutonium movement downward into the soil is difficult to assess. Since only apparently undisturbed soil was sampled, it seems that higher animal forces contributed little, if any, to the element's downward movement. The effects of earthworms, microbes, and vegetation root growth cannot be discounted. It seems

likely that physical processes and forces as well as biological forces were involved.

In terms of physical processes, the most plausible mechanism of soil penetration is a combination of gravitational force and water percolation. The likely submicron size of unattached Pu particles make passive transport of the particle by water feasible. Unfortunately, the available data provide little to assess this mechanism. There was no significant regression of the fraction of total mass with depth for most particle size ranges in either macroplot ($\alpha = .05$). If large amounts of small particles were transported downward by some mechanism, a significant regression would be expected. Regressions of Pu concentration as a function of soil particle size for each depth were generally significant ($p < .05$), but with one exception, the slopes within each macroplot were not significantly different from the mean slope of each macroplot. The implication of this is that there seems to be no detectable difference in the rate of movement of the large and small particles which presumably act as carriers of the plutonium contamination. While there are numerous possible mechanisms, their quantitative importance remains obscure.

The downward movement of Pu into the soil is important from the health aspect. Plutonium which has migrated a few centimeters into the soil is not imminently available to wind erosion. Therefore, some downward migration is desirable. Concern about movement of Pu into water tables seems unwarranted. The data indicate that most of the Pu is still near the surface after as much as 10 years since initial redistribution. The length of time required for the

the contamination to reach underground water supplies, if even possible, would appear to be very long.

The relationship between the plutonium concentration in soil and the particle size range of the sample is important; i.e., as the particle size range decreases, the plutonium concentration increases. Tamura (1975) found the highest proportion of plutonium in the silt fraction (2-53 μm) in soil samples from the Nevada Test Site and Mound Laboratory, Miamisburg, Ohio. These results would seem to agree with the Rocky Flats results. The tendency of Pu to preferentially associate with the smaller host particles tends to reduce re-suspension potential because monodispersed particles of less than 50 μm diameter are classified as difficultly erodible (Healy and Fuquay, 1958).

The results of plutonium concentration versus particle size give some insight into attachment mechanisms. Using surface to volume ratio of the soil particles, assumed to be spheres, it can be shown that the Pu concentration is positively correlated with the particulate surface area per gram of soil. This was expected from the Pu versus particle size regression since the surface to volume ratio of a particle increases as the diameter decreases. This lends support to the concept that plutonium contamination may be a surface attachment process.

The higher variability in macroplot 2 than macroplot 1 is difficult to explain. In particular, the data from three adjacent 5 x 5 cm columns of soil at macroplot 2 are exemplary of the high variability; the aliquots from the three samples varied by more than two orders

of magnitude within a maximum distance of 15 cm across the surface of the soil (Table 9). The maximum Pu concentration in these three samples was about 1100 dpm/g soil in a five gram sample, or a total of 5500 dpm.

There are several mechanisms of Pu attachment to soil which are possible. One is that the mean plutonium particle of 0.3 μm diameter suggested by J. Hayden of Rocky Flats (personal communication) would attach to the median plutonium bearing, or host, particle 10.3 μm (Nathans et al., 1971). Such a host particle could feasibly carry on its surface only about 70 dpm, and about 75 such particles would be required to independently find their way to the above contaminated column to give 5500 dpm. This prospect hardly seems likely.

Another remote possibility is that all of the 5500 dpm in the sample aliquot were supplied by one particle of pure Pu oxide. By the formula of Sill (1971), such a Pu particle would be about 20 μm in diameter. A 20 μm spherical particle of pure PuO_2 would be very difficult to erode by wind unless attached to a larger particle (Healy and Fuquay, 1958).

The most probable relationship of Pu to soil is the agglomerate particle, a combination of many Pu and soil particles into one functional unit. This idea is supported by Nathans et al. (1971) who state, "There is no correlation between the PuO_2 equivalent diameter and the [host] particle diameter. This lack of correlation supports the concept that the "hot" particles are agglomerates of small plutonium-containing particles and larger soil grains."

The range of activities which could have been contained by agglomerate particles is impressive. For instance, the most erodible host particles range from 50 to 500 μm diameter (Healy and Fuquay, 1958). The most probable PuO_2 particles range from 0.01 μm , the size of Navratil's filter (see "Study Area and Plutonium Source" in this study), to 3 μm , the largest particle supposedly allowed in the barrels. Assuming both the host soil and the Pu particles are spheres, that the host particle surface is maximumly utilized, and that the surface attachment requirement of the Pu particles is the diameter squared, the 50 μm host particles would contain from 56.6 dpm to 1.7×10^4 dpm for 0.01 to 3 μm diameter Pu particles, respectively. Similarly, a 500 μm host particle could have from 5660 dpm to 1.7×10^6 dpm adhered to its surface. Obviously, an even wider range of activities could result from the many different combinations from few .01 Pu particles to many 3.0 Pu particles; i.e., agglomerated particles of various Pu and soil particle sizes could result in myriad activities.

It appears that agglomerated particles are the likely form of the wind-redistributed contamination. This mechanism allows for particles which are readily wind erodible, is compatible with surface attachment, allows for particles which might weather after final deposition by wind, and might plausibly result in larger variances in macroplot 2 by the deposition of an occasional large agglomerate. Clearly, this mechanism seems more reasonable than presuming only large Pu particles or small Pu particles attached to a single soil particle.

A final important question raised by the soil data concerns isotopic ratios. In both macroplots the surface three centimeters have an isotopic ratio (IR) of about 65. As the depth increases in macroplot 1, the IR decreases to about 40 at the 18-21 cm level. In macroplot 2, the ratio decreases to 8 at the 18-21 cm level. The reason for this difference is unclear at present, but it may result from several causes. A probable cause was the bias caused by removal of some samples from consideration because the ^{238}Pu was below detection limits. This practice would effectively increase the mean value of ^{238}Pu in the samples and decrease the ratio artifactually.

It is also possible that the IR decreases more rapidly in macroplot 2 because of some of the Pu in the soil coming from a different source, perhaps the 1957 fire, or from different drums in the barrel storage area at different times.

Another interesting facet of the IR in soil is that the surface values in both macroplots are so similar. What makes this surprising is that the plutonium in the contaminated oil reportedly had an isotopic ratio of about 40 by activity (telephone conversation, 16 June 1976, with M. A. Thompson, Environmental Sciences and Waste Control, Rocky Flats Plant). The implication of the great difference in the isotopic ratio between the source and macroplots is that the ^{238}Pu was much more immobile with respect to redistribution by wind. Since the regressions of isotopic ratio with depth suggest that ^{238}Pu probably moved downward into the soil faster than ^{239}Pu , the ^{238}Pu was probably less susceptible to wind erosion from the barrel storage area.

In summary, the plutonium at Rocky Flats was most likely attached to soil in an agglomerate fashion and disseminated from the source by wind. The inverse Pu to particle size relationship suggested that the Pu was attached surficially to soil particles. Eventually, the agglomerates weathered and were probably carried downward into the soil by water along cracks in the ground, beside or inside decaying or live roots, through animal burrows, etc. Fortunately, the effect of the downward movement of Pu is to further reduce wind redistribution.

Plutonium in Litter and Vegetation

It is expected that the conclusions reached in examining the soil plutonium relationships have some bearing on the associations between litter and vegetation and soil. Plutonium concentrations in litter were shown to be very dependent on soil concentration of plutonium. A similar plot for vegetation was also significant, but more variable.

Further evidence of this conclusion is given by the significant regressions of Pu concentration in litter and vegetation as functions of both the downwind or downslope distance from the source. Since the contamination was likely dispersed as a function of soil movement by wind and the soil concentrations were significantly correlated with distance from the source, the litter and vegetation are circumstantially associated with the soil.

Relative importance of the modes of litter and vegetation contamination are unknown. Certainly, because of the different mean

concentrations, variances, and proximities to the contaminated soil, there must be different mechanisms of contamination at work or the same mechanism for different time periods.

In some respects, the mean concentration results are not what might be expected. In terms of root uptake alone, the vegetation should have been much lower. The greenhouse studies done during the past decades indicate that plutonium uptake via roots is on the order of 10^{-6} - 10^{-4} of soil concentrations (Jacobson and Overstreet, 1948; Rediske and Selders, 1953; Newbould, 1963; Wilson and Cline, 1966; Romney et al., 1970; Schulz et al., 1975). The Rocky Flats vegetation have a concentration ratio of about 10^{-3} - 10^{-2} . This fact suggests some other mechanism such as aerial deposition of resuspended soil particles.

Possible mechanisms of contaminating litter include impaction, gravitational settling, rainout and washout. Splashing of rain-dampened soil onto the overlying litter layer during rainstorms may be another important mechanism. During spring and fall needle ice formation with the attendant soil surface changes could contribute to contamination of litter.

One of the most plausible explanations of the higher litter concentrations regards the method of preparation in the laboratory. The technique of floating the litter to remove soil from the hand-collected samples may tend to increase selectively the mean Pu concentration. By floating the litter on water the large particles of soil are removed, but the particles small enough not to break the water's surface tension are probably left on the litter. Since the

plutonium concentration in soil is inversely related to the particle size of the soil sample, these unremoved small particles may evoke a higher concentration than is found in the environmental situation at Rocky Flats.

The vegetation is often composed of annual plants which would accumulate little surficial contamination during their short stand. Others, biennials or perennials, should accumulate more. A given mass of litter, on the other hand, may retain its integrity, and accumulate Pu for a number of years. If this concept is accurate, the litter might accumulate up to several times as much plutonium as the vegetation. The actual ratios of mean litter concentration to mean vegetation concentration were 14.4 and 1.36 in macroplots 1 and 2, respectively.

In summary, the mechanisms of contamination of litter and vegetation are not readily quantifiable from this study. It seems reasonable, however, that these mechanisms in some way involve physical interactions with the soil particles. Since laboratory studies with vegetation have suggested very low uptake of plutonium by various plants, it also seems reasonable that resuspended soil is in some way responsible for the comparatively high vegetation contamination levels. Further, it also seems reasonable that concentrations of Pu in litter are influenced by factors including wet and dry deposition, rain splashing, needle ice formation and the method of preparation in the laboratory.

Plutonium in Arthropods and Small Mammals

The arthropod and small mammal data are difficult to understand, probably because of animal mobility, several possible mechanisms of contamination, extreme variability in the data, and possible analytical laboratory bias. A number of questions concerning the animal results occur:

- 1) Are the animal contamination levels linked to the soil contamination?
- 2) How does the plutonium get into (onto) the individual tissues?
- 3) Do the results agree with research findings at other sites?
- 4) Why are the mean concentrations in small mammals not significantly different between the two macroplots?
- 5) Is there a difference in the biological interactions of plutonium -238 and -239?

Question number one is probably the most significant relative to the understanding of the contamination mechanisms. Because of the large fraction of total plutonium contained by the soil and animal proximity to and mobility in the soil, it is hard to imagine these animals not being contaminated by contact with the soil. However, Pu concentrations in the small mammal tissues cannot be directly correlated with the soil underneath by the present data, at least partly because of mobility.

There are several other bits of information which may help determine how strongly small mammal tissues are linked to soil. The data of McLendon et al. (1975) indicated that the concentration ratio of small mammal tissues to soil was independent of the soil Pu concentration, i.e., Pu levels in small mammals were highly dependent on

soil concentration of Pu. These workers sampled in an area of lower concentrations than Rocky Flats where little spatial variation occurred which might preclude the distance comparisons unavailable with these data. In contrast to the findings of McLendon et al. (1975), the concentration ratios presented herein vary between macroplots 1 and 2. This suggests that small mammals may not be closely tied to soil. However, there is some indication that laboratory error may play a role in the high macroplot 2 small mammal means and concentration ratios. This possibility is not clear cut and has yet to be tested.

The larger mean concentration of the log-transformed arthropod data in macroplot 1 seems quite reasonable. Since the whole animal was analyzed and the animal was sacrificed soon after capture, the arthropod difference probably reflects the dichotomy in soil concentrations in the two macroplots. It also seems reasonable that the arthropod's small size and high surface-to-volume ratio makes these individuals strong candidates for surface attachment mechanisms. However, the low ^{239}Pu to ^{238}Pu ratio relative to soil suggests considerable physiological incorporation.

There are presumably several ways in which Pu enters the animals. Because of their close proximity to the soil, it is reasonable to assume that the individuals inhale some soil. Some of this would probably find its way into the GI tract after being expelled from the trachea by mucus transport. In addition, soil would be ingested with the normal foodstuffs of the animals; plant parts for pocket gophers and insects for deer mice.

Unfortunately, the data do little to pinpoint the process of Pu entry into or onto the animal bodies; there are few significant differences between small mammal lung and GI or internal and external tissues. However, there seems to be little reason to suspect that assimilation of Pu by rodents at Rocky Flats occurs by methods different than reported by other investigators. Tissue concentration ratios were higher than those seen by some investigators (Hakonson et al., 1973) and quite comparable to those seen by others (Romney et al., 1970; McLendon et al., 1975). An interesting disagreement between the Rocky Flats data and those of Romney et al. (1970) is that those workers reported bone to have the highest concentration of any rodent tissue while bone was the lowest in this study. Also, the levels of variation seen in the data herein are comparable to those expressed by the above workers.

The fact that small mammal internal and external tissues are at about the same contamination level is intriguing. Several explanations are possible. Our methods of sometimes holding live captured animals for several days in the laboratory prior to sacrifice for analysis may explain the lack of high GI, hide and lung values.

The apparent equality of internal and external tissues might imply that there is a form of biologically-available Pu in small concentrations in the soil. Remembering the myriad weathering cycles to which some soil-plutonium particles were subjected, this possibility seems reasonable. Other possible explanations include: the potential of soil microbes, earthworms, or natural chelating agents from organic matter in the soil for solubilizing the Pu to make it

more biologically acceptable; the possibility of laboratory cross-contamination; the possibility that a few anomalously high tissue values led to a highly-biased mean.

The possible existence of a more available fraction of plutonium dispersed along with the insoluble and relatively large particulate contamination might help to explain the apparent lack of difference in small mammal means between macroplots. If the more available portion of contamination was spread uniformly across the plant site, the amounts of Pu seen in the individual tissue samples might reflect the maximum of equilibrium value of the more utilizable Pu distribution. One difficulty with this theory is that bone appears to have a lower concentration than the other tissues in each macroplot. If a "soluble" form of Pu existed in the animal's environment it would likely be found in bone to at least the same level as in the other internal tissues (Hamilton, 1948).

Concerning the question of dissimilarity in interaction of the two plutonium isotopes studies, there seemed to be some difference in the small mammal data. The ratios had much lower coefficients of variation than the ^{239}Pu data alone. This result would seem to indicate that the ^{238}Pu and ^{239}Pu varied together to some degree. The ratios for bone samples were generally lower than for other tissues. This result probably reflects bias resulting from lack of detectable ^{238}Pu concentrations in many samples or physiological differences between isotopes. The fact that the isotopic ratio in small mammal tissues is lower ($p < .05$) in each macroplot than the surface soil

indicates some physiological incorporation or difference in interaction between Pu isotopes.

In both macroplots, the mean ratios for arthropods are lower than the mean values seen earlier for the upper soil layers and for litter and vegetation ($p < .05$). This may indicate that ^{238}Pu is more available for attachment to or uptake by the arthropods. The potential mechanism for such a difference in availability is not clear at this point.

In summary, it is likely that the contamination of small mammals results from the soil Pu in some way, although some data contrary to this idea exist. The data on Pu concentrations in small mammals did not show any clear relationships to the soil contamination patterns, but the isotopic ratio data did suggest some physiological incorporation of plutonium. A postulated soluble or biologically-available plutonium compound or particle size distribution may have resulted in the similar internal and external small mammal tissue distributions within each macroplot and the agreement in mean activity per sample between macroplots. However, there is no direct information to support this contention in the present data. Contamination probably results from a combination of inhalation and ingestion of varied amounts of plutonium from the environment, leading to highly variable results between individuals and tissues. The facts that isotopic ratio in bone is apparently different from other tissues and that all tissues combined have a lower mean IR than soil would seem to indicate that the two isotopes, ^{238}Pu and ^{239}Pu , are being incorporated into tissues and at different rates. The same concept

applies to the arthropods, but only in macroplot 1 is the arthropod-soil difference significant ($p < .05$).

Detection of true differences in plutonium concentration within the small mammal populations would likely require kill trapping, large sample size, and perhaps classification of samples by age, species, sex, or location. It is questionable whether the benefits of such a program would justify the expense of such a project.

SUMMARY AND CONCLUSIONS

This study was initiated to elucidate and understand the patterns of plutonium contamination of the terrestrial ecosystem at ERDA's Rocky Flats plant northwest of Denver, Colorado. Of interest were the definition of ecosystem compartments which contain Pu, the patterns and relative amounts of Pu in those compartments, how those values compared to findings in other geographical areas, comparative behavior of ^{238}Pu and ^{239}Pu , and mechanisms which might explain the observed patterns of contamination.

Samples of soil, litter, vegetation, arthropods, and small mammals were collected for plutonium analysis from two macroplots on the Rocky Flats plant site. Sample masses analyzed were intentionally small (5 g or less) to permit population variability to be expressed. For instance, single rodent tissues, rather than pooled samples, were analyzed for Pu. Zero or less than detectable samples and highly skewed, non-normal distribution occurred, forcing the use of non-parametric statistical tests in many cases. In addition, estimates of mass were made for each of these ecosystem compartments. These data were used for calculation of compartmental fractions of the total plutonium in each macroplot.

Results of the compartmentalization calculations indicated that soil layers from 0-21 cm included greater than 99% of the plutonium in the sampled compartments from each of the study macroplots. The other compartments had fractions at least several orders of magnitude

lower than the soil. Litter had a higher fraction of the total Pu inventory measured than vegetation which was higher than the animal compartments studies. These data imply that soil-Pu relationships and soil management practices are very important on contaminated areas. Soil in macroplot 1, nearest the contamination source, the oil barrel storage area, had a mean concentration about thirty times that of the concentration in macroplot 2. Sampling distributions from both macroplots were characterized by coefficients of variation generally above 1.00 (100%) and extending to several times higher.

Concentrations of Pu in the 0-3 cm soil layer in macroplot 1 were found to be correlated with distance from the Pu source. Regressions along the downwind transect, running approximately in the predominant wind direction and the downslope transect running in a southerly direction from macroplot 1 had correlation coefficients of greater than 90%. The slopes of these curves suggest that the contaminated particulate material initially deposited was probably larger than submicron size.

Soil Pu concentrations were inversely correlated with soil particle size range. As the particle size decreased, the concentration of the sample increased. This result strengthened the belief that the plutonium-soil interaction is a surface attraction phenomenon. Also, the attachment of Pu to small diameter host particles strengthens the belief that present redistribution is minimal.

Concentration of Pu in soil was also correlated with sample depth. As the sample depth increased, the plutonium concentration of the sample decreased. This relationship took the form of a power

function in both macroplots. The downward movement of Pu was probably caused by gravity and water percolation, aided by biological processes and likely occurred following particle breakdown by weathering.

Multiple linear regression techniques were not suited to prediction of concentrations from distance, depth, and particle size of the soil sample. The failure of these computer-based techniques to give significant results probably stemmed from the various non-linear relationships achieved in the single regressions described earlier, and the failure to correct adequately for the distribution of fraction of total mass represented by various soil particle size ranges.

The data on Pu in soil led to the impression that the Pu-soil-wind interactions were dominant in the creation of the contamination patterns seen at Rocky Flats. The hypothesis that best seems to explain the data is that Pu became attached to the soil by way of agglomerated combinations of mostly submicron Pu particles and larger (1-500 μm) soil particles. These agglomerates were then dispersed by environmental forces as single particles larger in size than the components. Such particles were apparently deposited on the study plots in rather heterogeneous patterns and probably low densities to cause substantial sampling variability.

Generally, the mean concentrations were higher in litter than in vegetation, and higher in macroplot 1 than in macroplot 2. Sample distributions of litter were normal while vegetation was log-normal. Plutonium concentration in litter and vegetation was also

significantly correlated to both downwind and downslope distances. Vegetation and litter concentrations were also significantly correlated with soil underneath. Therefore, it seems likely that Pu concentrations in both vegetation and litter were strongly linked to soil.

Plutonium in arthropod samples appeared to be log-normally distributed. Using log-transformed data, the Pu concentrations in macroplot 1 arthropods were significantly higher than in macroplot 2 arthropods ($p < .05$).

Tissue samples from small mammals, however, showed no significant differences between macroplots if all tissues were considered as a group. There was evidence from several tests that the bone tissues were lower in both macroplots than the other tissues. Non-parametric statistical tests indicated the puzzling result that the external tissues in macroplot 1 were lower in Pu concentration than the internal tissues.

Histograms of the small mammal tissues from each macroplot were highly skewed and kurtotic and had the visual appearance of being log-normal. The presence of many zero and negative values prohibited the use of logarithms for transforming the data. Kolmogorov-Smirnov goodness-of-fit tests indicated that the distributions were not normal. The nonnormality of the sampling distribution was greatly enhanced by the fact that relatively few samples contributed much of the total activity and raised the means.

Concentration ratios of litter, arthropods, and small mammals relative to soil indicated that litter had the highest value in

macroplot 1 (4.9×10^{-1}). The other macroplot 1 compartments in descending order were vegetation (3.4×10^{-2}), small mammals (7.8×10^{-3}), and arthropods (3.8×10^{-3}). In macroplot 2, the descending order was as follows: small mammals (2.1×10^{-1}), litter (1.4×10^{-2}), vegetation (1.0×10^{-2}), and arthropods (1.2×10^{-5}). These results are similar to those found by other investigators in field studies with the possible exception of small mammals in macroplot 2. The high concentration ratios indicated that most of the contamination of vegetation probably results from surficial plutonium.

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 mean ratio near the surface was about 65 in macroplot 1 and about 25% lower in macroplot 2. In macroplot 1, the ratio decreased to about 40 in the 18-21 cm level. In macroplot 2, the ratio decreased to about 8 in the 18-21 cm depths. Neither curve of ratio versus depth was significant ($\alpha = .05$). Few litter and vegetation samples were analyzed by commercial laboratories and, therefore, the litter and vegetation ratios were limited in number. However, the macroplot 1 means of litter and vegetation combined were found to be significantly larger than the macroplot 2 means. Also, the litter and vegetation isotopic ratios were about the same as soil ratios which probably indicates surficial rather than internal contamination.

Isotopic ratios of arthropods showed that macroplot 1 ratios were significantly larger than ratios for macroplot 2 ($p < .05$).

Likewise, when all small mammal tissue samples were combined for each macroplot, macroplot 1 had a significantly larger ratio than macroplot 2 samples ($p < .05$). Only bone, hide and internal tissues from macroplot 1 were found to be significantly different (larger) than the same groups in macroplot 2. There seemed to be little difference in isotopic ratio between most tissues within macroplots; but, there was some evidence that bone was different from the other tissues in each macroplot. On the whole, since the isotopic ratios of small mammals were lower than soil, it appears that the plutonium-238 is being incorporated into tissues at a faster rate than the plutonium-239.

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APPENDICES

APPENDIX A

Plutonium Assay in Soil or Biological
Materials by Liquid Scintillation

Reagents and Supplies

1. Concentrated nitric acid (reagent grade, 16 molar).
2. Concentrated hydrofluoric acid (reagent grade, 52%).
3. 8 m nitric acid (500 ml conc. HNO_3 diluted to 1 liter).
4. 7.5 m nitric acid (469 ml conc. HNO_3 diluted to 1 liter).
5. 9 m hydrochloric acid (750 ml conc. HCl , 12 molar, diluted to 1 liter).
6. Hydroxylamine hydrochloride crystals (reagent grade).
7. 9 m hydrochloric acid - 5% ammonium iodide solution (5 g HN_4I in 100 ml 9 m HCl).
8. Anion exchange resin 50% slurry. [By volume combine equal parts of Amberlite (CG400 100-200 mesh) resin and deionized water. Wash resin by shaking the slurry, allowing resin to settle, and decanting the now cloudy water. Repeat wash procedure until water is clear. Finally, add one part water to the clean resin and store.]
9. 2 m Nitric Acid - 0.2 m Boric Acid solution (125 ml conc. HNO_3 + 12 grams H_3BO_3 diluted to 1 liter).
10. 4% Urea (4 grams urea per 100 ml water).
11. Keough & Powers Liquid Scintillation Cocktail [Pass approximately 200 ml of Bis(2-ethylhexyl) hydrogen phosphate (D_2EHPA) through a column of charcoal to remove color impurities if technical grade is used. Dissolve 200 ml of the D_2EHPA in 800 ml of toluene containing 5 grams of p-terphenyl and 0.05 grams of 1,4-bis-2 (5 phenyloxazole)

benzene (POPOP). This solution is stable for six months under refrigeration.]

12. BIORAD glass ion exchange columns, 0.7 x 15 cm, with polypropylene top and bottom fittings.
13. NALGENE or glass powder funnel, 7.7 cm inner diameter at top of cone, 2.8 cm x 1.7 cm outer diameter stem.
14. WHATMAN 54 filter paper, hardened, low ash.
15. 250 ml TEFLON beakers.

Digestion

1. Weigh approximately 5 g of sample, if available, into a 250 ml PYREX beaker.
2. Add enough concentrated nitric acid to cover sample in beaker using a minimum of 20 ml and allow to evaporate to dryness on a hot plate. Temperatures for all digestions and evaporations in this procedure should range from 150°-175°. High organic matter samples should be heated slowly or let stand in acid without heat overnight to avoid foaming.
3. Place dry sample beakers in a muffle furnace and ash at 600°C for a minimum of 8 hours. Cool before adding any liquid to beakers after removal from furnace.
4. Repeat step #2 without overnight predigestion.
5. Repeat step #3.
6. Place partially cooled beakers on a hot plate. Dissolve sample residue in concentrated nitric acid. Transfer to a TEFLON beaker using a NALGENE policeman along with a

concentrated nitric acid rinse to insure quantitative transfer. Evaporate to dryness on hot plate.

7. Add 20 ml of concentrated hydrofluoric acid to sample and evaporate to dryness on a hot plate.
8. Repeat step #7.
9. Add 20 ml of 8 m nitric acid to sample and evaporate to dryness on a hot plate. Sample is now ready for ion exchange.

Ion Exchange

1. Prepare column by removing top plug and pressing powder funnel into place atop polypropylene fitting. Leave polypropylene bottom cap in place.
2. Place a 150 ml beaker beneath the column as a catch-beaker. Fill column with deionized water by pouring down side of funnel to prevent air locking. Check for water leaks. Tap the column bottom to dislodge any trapped air bubbles. Shake slurry, pour into column, and allow to precipitate about 30 min. Final resin level should be slightly above the glass barrel top of the column. Do not allow resin to become dry at any point during procedure.
3. Place a small piece of glass wool on top of the resin bed. Tap down to allow any air trapped between the wool and the resin to escape.
4. Drain column by removing bottom cap.
5. Place quarter-folded Whatman 54 filter paper in funnel of the column. Wet the filter paper around the edges with

deionized water to prevent sample leakage around the paper.

6. Drain the column. Columns may be stored filled and moist until needed by replacing bottom cap. Take care to replace water lost to evaporation if columns stored for an extended period. Resin must be kept moist.
7. To condition the column pass 20 ml 7.5 HNO_3 through the column. Take care to detect air-locking between filter paper and resin column by observing for slowing or ceasing of flow. Air bubbles may be broken by slipping a clean glass rod between funnel and filter paper to top of resin bed.
8. Dissolve sample residue in 20 ml 7.5 m nitric acid; cover and allow to stand overnight. Pour into column.
9. Rinse beaker with 7.5 m nitric acid using 20 ml for each of two rinses. Take care to completely transfer all beaker contents to the column, using a glass rod or policeman if necessary.
10. Remove filter paper cone from column funnel after sample and rinses have drained through.
11. Pass 7.5 m nitric acid through column until iron complex is gone, approximately 20 ml. Iron complex is removed when column eluate is no longer yellow.
12. Discard nitric acid washes from the catch-beaker and replace beaker.

13. To remove the nitric acid, pass 20 ml 9 m HCl through the column. The eluate will be yellow, but this is not due to iron complexes.
14. Add about 1/4 teaspoonful hydroxylamine hydrochloride crystals to cover top of the resin bed.
15. To elute Pu, while leaving U on the column, pass 40 ml 9 m HCl - 5% NH_4I solution through the column. Drain the column.
16. Pass 20 ml 9 m HCl through the column.

Preparation for Counting

1. Add a few drops of concentrated nitric acid to eluate and evaporate to dryness, watching sample initially for boil over.
2. Add 20 ml concentrated nitric acid and evaporate to dryness on a hot plate.
3. Add 20 ml 8 m nitric acid and evaporate to dryness on a hot plate.
4. Dissolve residue in 5 ml of 2 m nitric acid - 0.2 m boric acid mixture and transfer to scintillation vial. Rinse beaker twice with the nitric-boric mixture and transfer to same vial. Total volume should not exceed 15 ml.
5. Add 1 drop 4% urea and 4 ml "Keough & Powers" cocktail.
6. Cap vials with polyethylene lined caps.
7. Place samples in shaker box and shake for 30 seconds at 5 minute intervals 6 times.
8. Count.

Counter Preparation

1. Ascertain optimal counting windows in Channels A and B using a plutonium standard. Most likely, a low gain setting is best. Generally, Channel A includes region from first steep increase in count rate to several energy channels above point where count rate drops off. Channel B has same upper limit, but is several energy channels wider at lower end. For example, if channel A was 10-21, Channel B would be 7-21.
2. Obtain and count an unquenched Pu standard.
3. Make a ferric chloride quenching solution using 6 g FeCl_3 , 6 H_2O dissolved in 100 ml deionized water.
4. Add ferric chloride solution to standard, one or two drops at a time, shake briefly, let the mixture stand for two minutes and count on above settings for one minute.
5. Record the cumulative number of drops and counts in both channels A and B.
6. Repeat #4 and #5 until the count rate in Channels A and B are very low, i.e., only a few counts per minute.
7. To determine channels ratio for each count divide the number of counts in Channel A by those in Channel B.
8. To determine the efficiency for each count, divide counts per minute in Channel B by the known activity of the unquenched standard.
9. Plot and calculate a linear regression of efficiency versus channels ratio. This will allow estimation of counting

yield (efficiency) for future channels ratios using the same cocktail.

10. The counts per minute in each sample should be corrected for counting yield as well as chemical yield which must be estimated by analyzing a known standard by the complete procedure, including digestion.

Additional Information

1. Safety glasses, clothing protection, and acid resistant gloves should be worn during all steps involving use of dilute or concentrated acids.
2. Samples should always be evaporated to dryness before placing them in the muffle furnaces after each nitric acid digestion. Moisture in the beaker will evaporate by popping and create the possibility of sample loss or cross-contamination.

APPENDIX B

Analytical Procedure Rationale
and Justification¹

¹This section is partially an adaptation of Whicker (1975).

The first two years of this study, commercial laboratories, chiefly Eberline Instrument Corp., Santa Fe, New Mexico, and LFE Environmental, Richmond, California, processed most of our samples for plutonium. The advantages of commercial laboratories include their use of widely accepted methods of total dissolution, isotopic spiking to measure chemical recovery, alpha spectrometry, their general reliability, and the fact that the purchasing institution is relieved of much tedious work. The disadvantages of outside labs are also noteworthy: high cost per sample (\$50-75), 6-10 weeks turnaround time, and lack of control of the laboratory by the purchaser. The cumulative inconveniences of the private labs led us to consider a simplified procedure that could be applied to samples expected to contain relatively high plutonium concentrations. This procedure, which employs an extraction procedure developed by Keough and Powers (1970) and involves liquid scintillation counting, was readily handled in our laboratory.

The benefits of the liquid scintillation (LS) analysis for Pu are lower cost per sample relative to commercial labs (approximately \$20 currently), about 2 weeks turnaround time, and total control of the analytical laboratory. The undesirable qualities are ignorance of chemical recovery on individual samples and isotopic composition, a higher minimum detectable activity than alpha spectrometry, large amounts of first-hand work, and the necessity to constantly cross check results with other labs by splitting aliquots.

During the past four years we have addressed the LS method's criticisms and its applicability to Rocky Flats plutonium samples in

soil, vegetation, litter, and animals. Since recovery and isotopic composition are not known, the capability of the HNO_3 -HF treatment to solubilize plutonium is of utmost importance. This digestion procedure is based on the assumption that air-oxidized plutonium is soluble in HNO_3 . Hydrofluoric acid is added to dissolve refractory plutonium oxides formed under oxidation at high temperature. If the HNO_3 -HF digestion procedure is not efficient poor chemical recovery causes calculated Pu activities to be low relative to more severe digestion methods.

Some capabilities of the LS method were assessed by Paine (1974) before our adoption of the method. He spiked sediment, water, vegetation, soft animal tissues and bone samples with known quantities of the soluble $\text{Pu}(\text{NO}_3)_4$. The method recovered 89-92% (mean of 90%) of the Pu. Next, Paine analyzed a soil sample provided by Dr. Claude Sill of the Idaho National Engineering Laboratory (INEL). The sample reportedly contained 33.56 dpm/g of high fired plutonium oxide and five aliquots analyzed by Paine ranged from 27.6-30.5 dpm/g with a mean of 29.6 dpm/g for 88% recovery. Finally, Paine sent aliquots of 14 sediment and vegetation samples to LFE Environmental for analysis using total dissolution, ^{236}Pu tracer addition, and alpha spectrometry. Results of the CSU LS method and LFE's more rigorous procedure agree satisfactorily.

Subsequently, we conducted inter-laboratory comparisons of plutonium analyses of homogenized soil sample aliquots. Results are shown in Table B1. A Rocky Flats soil sample (S-1835) was supplied

Table B1. Interlaboratory comparisons, aliquots of HASL soil sample S-1835 collected from Rocky Flats near CSU macroplot 1 and L. Roxy Ann soil sample collected in the Park Range of northern Colorado.

HASL S-1835 soil					
Analytical Laboratory	Digestion method	Concentration Pu(dpm/g)		CV	No. of Samples
		Mean	Range		
CSU	HNO ₃ + HF	1316	948-2178	0.23	29
CSU	HNO ₃ only	1492	1336-1635	0.10	3
Eberline	Total dissolution (Pyrosulfate fusion)	1418	984-2520	0.33	8
HASL	Acid leach	910	---	---	1
LFE	HF digestion	1300	---	---	1
L. Roxy Ann soil					
CSU	HNO ₃ + HF	7.2	5.5-9.1	0.17	8
Eberline	Total dissolution (Pyrosulfate fusion)	5.7	5.2-6.2	0.07	5
HASL	Acid leach	5.9	5.5-6.1	0.03	5
INEL(S111)	Pyrosulfate fusion	7.0	---	---	1
LFE	HF digestion	7.2	5.3-12.3	0.40	5

by ERDA's Health and Safety Laboratory (HASL), New York City and a naturally concentrated soil sample of plutonium fallout was taken from a high mountain lake (ca. 3300 m) in northern Colorado. In both cases agreement between laboratory results is favorable, even though different methods of analysis were utilized. T-tests comparing mean results for the HASL soil of both CSU methods listed in Table B1 and Eberline's results were not significant. A oneway analysis of variance of the L. Roxy Ann soil data showed no significant difference between laboratories ($\alpha = .05$).

To test the effectiveness of the HNO_3 -HF digestion, we analyzed five gram aliquots of HASL soil sample S-1835 using varied quantities of HF (Table B2). Sample residues were analyzed for Pu content using the usual HNO_3 -HF digestion. We repeated these steps for the residues of the residues. The Pu content of the two residues plus the original run activity were assumed to equal the total activity of the sample which in turn was used to calculate the fractional recovery for each HF treatment level. The overall mean recovery of .89 conforms with Paine's (1974) value of .90. The precision of the LS method indicated by the overall coefficient of variation (CV) is a heartening 0.06 with a maximum of 0.072 for the 50 ml HF treatment level.

The effect of varied amounts of HF listed in Table B2 is noteworthy. Silica volatilization by only 20 ml of HF results in an adequately small residue mass. The possible formation of CaF_2 precipitate and subsequent occlusion of Pu from the solution may explain

Table B2. Recovery of plutonium from 5g soil samples from HASL S-1835 versus the volume of HF used in the initial digestion. Recovery based upon total activity in initial sample run plus 2 successive runs on the sample residues. Three aliquots were run for each HF volume. Residues were digested using the normal HNO_3 + HF procedure.

HF volume (ml/digestion)	Plutonium concentration (dpm/g)		Mass of Residues (g)	Fractional Recovery Mean	CV
	Original run Mean	Original + 2 residues Mean			
0	1383	1492	4.05	0.93	.016
10	1140	1373	1.05	0.83	.036
20	1235	1422	0.36	0.87	.023
35	1221	1444	0.36	0.85	.054
50	1293	1514	0.36	0.85	.072
70	1275	1390	0.33	0.92	.006
100	1395	1512	0.31	0.92	.039
150	1665	1763	0.33	0.94	.012
Overall mean	1326	1489	0.89	0.89	--
CV	0.18	0.14		0.06	--

the reduced recoveries by moderate HF usage. These data suggest that most of the Pu is soluble in HNO_3 alone and, therefore, use of HF during digestion might be unnecessary.

Since recoveries in Table B2 were determined from assay of residues by LS, we decided to submit residues to LFE for an independent assessment. We combined residues from several samples to compose a residue batch. The group of residue batches in question is from sample series 626-739 in Table B3. The mean plutonium recovery during the initial run fell from 0.93 (expected from Table B2) to only 0.58. Later sample residue batches from non-HF analyses led to mean recovery values of 0.79 and 0.77 ranging from 0.57 to 0.90. Although the HF analyses do not appear to achieve greatly higher recoveries, the variation in the sample recovery seemed to be lower when HF was used. For this reason, as well as slightly higher recoveries, the HF digestion step is now used routinely.

To determine the residue type containing the majority of unrecovered plutonium, we saved and analyzed each type during another experiment. Using the normal HNO_3 -HF digestion LS procedure, we analyzed ten-gram aliquots of our Rocky Flats Macroplot H (immediately southeast of the asphalt pad) soil containing relatively high Pu concentrations. Some of each residue type (acid washes, anion exchange resin, or undissolved solids) were analyzed by our procedure and others by Eberline. These data and calculated recoveries are listed in Table B4. The relatively low mean recovery of 61% may be explained by poor digestion of the larger-than-normal 10 gm aliquots. By tallying the totals found in each residue type, we concluded that the

Table B3. Plutonium recoveries in 5g Rocky Flats soil samples using the CSU liquid scintillation method.

Sample Series	Basis of recovery value	HF used ?	Recovery Fractions		
			mean	S	range
626-739	LFE assay of 5 residue batches	no	0.58	0.15	0.44-0.79
740-811	LFE assay of 3 residue batches	no	0.79	0.07	0.77-0.87
733-809	LFE assay of 6 individual residues	no	0.77	0.11	0.57-0.90
1015-1074	LFE assay of 15 individual residues	yes	0.79	0.07	0.67-0.94
1218	assay of C. Sill's standard 29.8 dpm/g soil sample	yes	0.70	--	--
overall (n=30)			0.75	0.12	0.44-0.94
NO HF mean (n=14) = 0.71					
HF mean (n=16) = 0.78					

Table B4. Analyses of 10 gram soil samples from Rocky Flats Macroplot H and residues from the CSU liquid scintillation procedure.

Microplot	Sample depth (cm)	dpm/total sample					Recovery (%)
		Original sample	Original Sample Residues				
			Washes	Resin	Solids	Total	
1	0-3	138,727	250*	475*	10,190*	10,915*	93
	3-6	30,123	55	65	39,614	39,734	43
	6-9	20,289	22	46	12,915	12,983	61
	9-12	4,091	106*	32*	2,070*	2,208*	64
	12-15	7,990	14	27	5,318	5,359	60
	15-18	6,038	99*	36*	3,640*	3,775*	62
	18-21	1,569	5	2	904	911	63
2	0-3	115,016	244	234	55,545	56,023	67
	3-6	12,547	14	42	28,546	28,602	30
	6-9	11,126	73*	798*	4,040*	4,911*	69
	9-12	3,957	15	18	1,488	1,521	72
	12-15	2,508	4	9	1,900	1,913	57
	18-21	2,185	1	4	904	909	57

Mean fraction of total residue in:

-washes 0.01

-resin 0.02

-solids 0.97

*Residues analyzed by Eberline using total dissolution

vast majority of unrecovered plutonium is lost via the undissolved solid residues. This finding is compatible with possible CaF_2 precipitate formation during HF digestion.

Concern may be raised about the inability of the LS method to distinguish Pu isotopic composition. Data from samples processed by commercial labs are sufficient to calculate a common $^{239,240}\text{Pu}/^{238}\text{Pu}$ of about 50, i.e., 98% of the plutonium is ^{239}Pu . The variations of isotopic ratio with sample type are not large or sporadic enough to prevent routine isotopic corrections during activity calculations.

For vegetation, litter and soil samples the minimum detectable activity (MDA) of the liquid scintillation procedure is generally acceptable. The calculation is as follows:

$$\frac{\text{MDA (2)}}{\text{sample}} = \frac{2\sqrt{2 (R_b/t_b)}}{Y \cdot R}$$

where MDA = minimum detectable activity (disintegrations, d)

R_b = background count rate (cpm)

t_b = length of background count (m)

Y = counting yield (count/disintegration)

R = chemical recovery.

Using a theoretical sample mass of 5 g, R_b of 5 cpm, t_b of 100 minutes, Y of .90, and R of 0.78, the MDA equals 0.18 dpm/g. Most non-animal samples from Rocky Flats have a plutonium concentration of at least several dpm/g. Consequently, for all but small mass animal samples the LS procedure should be adequately sensitive. Pooling of animal tissues would allow analysis by LS. Not desiring to pool

Table B5. Plutonium values in aliquots from blended soil samples from different collection sites. Assay performed with the CSU liquid scintillation method.

Soil Type	Ref. No.	Pu alpha dpm/g	$\bar{X}$	CV
Rocky Flats (0-3 cm)	L-1	75	60	0.19
	L-2	64		
	L-3	43		
	L-4	59		
	L-5	59		
Rocky Flats (5-7 cm)	L-6	20	24	0.10
	L-7	23		
	L-8	24		
	L-9	26		
	L-10	25		
Rocky Flats (HASL S-1835)	L-204	1015	1012	0.07
	L-205	914		
	L-206	1041		
	L-207	1135		
	L-195	1092		
	L-197	993		
	L-198	948		
Roxy Ann (High mountain accumulation site, 0-3 cm)	L-18	2.5	4.0	0.25
	L-19	4.7		
	L-20	3.5		
	L-21	4.7		
	L-22	4.7		
Pawnee (shortgrass plains NE of Ft. Collins, 0-3 cm)	L-11	0.59	0.54	0.18
	L-12	0.64		
	L-13	0.61		
	L-14	0.43		
	L-15	0.56		
	L-16	0.40		

samples, we sent individual animal tissues to commercial labs (with procedures) capable of lower MDAs.

As expressed by the data in Tables B1, B2 and B5, the relative precision of the CSU LS method of Pu analysis is satisfactory. Between aliquots variation is the sum of true sample inhomogeneity, variation in the chemical and physical aspects of the sample preparation (analytical variation) and counting error. The counting error varies from a few percent to 58% of the total between aliquots variation. The contribution of spatial distribution variation, i.e., true sample inhomogeneity, is often large. These two facts coupled with the maximum observed CV of 0.25 in Table B5 indicate low sample preparation error.

APPENDIX C

Biomass Measurements¹

¹All the following figures are composites of sample data collected during several years. All sample values collected during a calendar month were averaged and plotted at the monthly midpoint.

Figure C1. Rocky Flats litter standing crop (1973-1975 combined).

Figure C2. Rocky Flats vegetation standing crop (1973-1975 combined).

Figure C3. Rocky Flats arthropod biomass estimates (1973-1974 combined).

Figure C4. Rocky Flats macroplot 1 small mammal biomass estimates (1973-1975 combined).

Figure C5. Rocky Flats macroplot 2 small mammal biomass estimates (1973-1975 combined).