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PLUTONIUM INVENTORIES IN TWO OLD-FIELD ECOSYSTEMS  
IN THE VICINITY OF A NUCLEAR-FUEL REPROCESSING FACILITY

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## ABSTRACT

Plutonium inventories were determined in two old-field ecosystems, Fields 1 and 2, located near a nuclear-fuel reprocessing facility that had released approximately 640 mCi  $^{238}\text{Pu}$  and 570 mCi  $^{239,240}\text{Pu}$  to the atmosphere in the 20 years preceding sampling. Field 1 was 230 m from the point of release (a 62 m stack) and contained 57 nCi  $^{238}\text{Pu}\cdot\text{m}^{-2}$  and 255 nCi  $^{239,240}\text{Pu}\cdot\text{m}^{-2}$ . Field 2 was 420 m from the stack and contained 15 nCi  $^{238}\text{Pu}\cdot\text{m}^{-2}$  and 79 nCi  $^{239,240}\text{Pu}\cdot\text{m}^{-2}$ . The distribution of Pu between soil and vegetation components was similar in the two fields with only 1.3% of the  $^{238}\text{Pu}$  and 0.2% of the  $^{239,240}\text{Pu}$  occurring in pines, herbaceous vegetation and litter. More of the Pu could be resuspended into the atmosphere by a  $6\text{ m}\cdot\text{sec}^{-1}$  wind on Field 1 than on Field 2 due to the lower biomass of herbaceous vegetation and litter on Field 1. Approximately 2% of the  $^{238}\text{Pu}$  and 0.5% of the  $^{239,240}\text{Pu}$  could be resuspended on Field 1, whereas only 0.5% of the  $^{238}\text{Pu}$  and 0.1% of the  $^{239,240}\text{Pu}$  could be resuspended on Field 2. Plutonium contamination of the vegetation occurred primarily by (1) direct deposition of recently released Pu onto vegetation and (2) resuspension of Pu from the soil to vegetation surfaces. Root uptake apparently made negligible contributions to the Pu contents of above-ground vegetation.

Key Words: Plutonium; inventory; concentration-ratio; old field; nuclear-fuel cycle; resuspension.

## INTRODUCTION

In man's history new technologies have become established based solely on economic considerations. Now we realize that technologies must be not only economically feasible, but that their impacts on the quality of life and the quality of the environment must also be considered. Nowhere is this more evident than in the development of new energy technologies. Yet, there is a dearth of basic data upon which to make sound management decisions concerning alternative energy technologies.

Breeder reactors are a potentially important new energy technology, but their implementation involves possible social and environmental problems because of the production and distribution of plutonium. Plutonium is a toxic element with several long-lived radioactive isotopes whose decay result in the release of alpha particles. If implemented in the United States, breeder reactors could lead to the production of over 1000 megacuries of Pu isotopes by early in the 21st century (Jacobs and Gera 1969). Similar breeder-reactor programs are being developed in Europe.

Barr (1976) estimates that approximately  $10^{-9}$  of the Pu involved in the breeder-reactor fuel cycle would be released to the environment. Most of this release would occur as atmospheric emissions of small (< 10 micron diameter) Pu-containing particles from nuclear-fuel reprocessing facilities. At present, there is very little information concerning the transport and fate of Pu released into the environment from reprocessing facilities. The majority of the data on Pu behavior in the environment are from studies of weapons testing or Pu released in aqueous discharges (Dahlman et al. in press, Hakonson and Nyhan in press, Hakonson et al.

1976, Hetherington et al. 1976, Martin and Bloom 1976, Nyhan et al. 1976, Romney et al. 1976).

This study is the first in a series on the transport and fate of Pu released from nuclear-fuel reprocessing facilities in natural and agricultural ecosystems. The objectives of this study were (1) to inventory the Pu contents of various components of an old-field ecosystem and (2) to draw inferences from these contents about the behavior of Pu in the ecosystem. For many elements, our understanding of cycling phenomena are such that inventories of amounts in ecosystem components are less informative than analyses of fluxes, transfer coefficients and rate controlling processes; however, this is not the case for Pu because its behavior in ecological systems is poorly known. The data collected in this study represent the first inventory of Pu released from a fuel reprocessing facility. Thus, these data were required before the important fluxes and transfers could be identified and simple models of Pu cycling constructed.

#### DESCRIPTION AND HISTORY OF THE STUDY AREA

We studied two fields (Fields 1 and 2) located adjacent to the H-area nuclear-fuel reprocessing facility on the U. S. Department of Energy's Savannah River Plant near Aiken, S. C. Low-level atmospheric releases of Pu have occurred from H-area since the start of operations in July 1955. Approximately 440 mCi of Pu were released from H-area before the installation of high-efficiency filters on the exhaust-air systems in December 1955. These releases contained  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  (which can only be differentiated using mass spectrometry and will hereafter be combined and symbolized as  $^{239,240}\text{Pu}$ ). From 1956 through

1966,  $^{239,240}\text{Pu}$  releases averaged  $4 \text{ mCi}\cdot\text{yr}^{-1}$ . From 1967 through 1974, normal releases averaged  $12 \text{ mCi } ^{238}\text{Pu}\cdot\text{yr}^{-1}$  and  $4 \text{ mCi } ^{239,240}\text{Pu}\cdot\text{yr}^{-1}$ . An accidental failure of the filtering mechanism in 1969 released an additional  $560 \text{ mCi } ^{238}\text{Pu}$  and  $58 \text{ mCi } ^{239,240}\text{Pu}$ . Total releases through 1974 were  $640 \text{ mCi } ^{238}\text{Pu}$  and  $570 \text{ mCi } ^{239,240}\text{Pu}$ .

Field 1 was  $145 \times 30 \text{ m}$  with its long axis oriented to the northwest away from the point of release, a  $62 \text{ m}$  stack. Field 2 shared the same long axis as Field 1 but was smaller ( $105 \times 30 \text{ m}$ ). The centers of Fields 1 and 2 were approximately  $230$  and  $420 \text{ m}$  from the stack, respectively. The A soil horizon and parts of the B soil horizon had been removed from Field 1 during construction of the reprocessing facility. In addition, some fill dirt had been deposited on Field 1. Field 2 had been disturbed less and had a soil profile that was typical for the area. Both fields supported a herbaceous plant community dominated by Andropogon spp., Lespedeza cuneata (Dumont) G. Don, Panicum spp. and Smilax spp. with scattered loblolly pines (Pinus taeda L.). The plant communities were typical of those occurring on abandoned fields of similar soil types in the southeastern United States. Pines were more abundant on Field 1 than on Field 2. Herbaceous vegetation was sparse in some areas of Field 1 due to exposed subsoil. A  $5 \text{ m}$  wide strip along the southwestern margin of each field had been mowed regularly for several years.

In the following analyses we compare Fields 1 and 2 to determine the effects of differences in soil, vegetation and the distance from the point of release on the distribution of Pu in various ecosystem components.

## METHODS

In October 1974 samples were collected from both fields to determine the amounts of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  in soil, pine trees, herbaceous vegetation, and litter (i.e., dead plant material lying on the soil surface). We also determined the amount of Pu which could be resuspended into the atmosphere from the litter and soil by a  $6 \text{ m} \cdot \text{sec}^{-1}$  wind (M<sup>i</sup>ham et al. 1976). From 27 February 1975 to 8 July 1975 we measured the rates of dryfall deposition of Pu on each field.

The location and diameter at breast height (dbh) of all loblolly pines taller than 2 m were determined. Six of these trees were randomly selected for determination of weights and Pu concentrations. Each of these trees was felled onto plastic sheets to prevent cross contamination from the soil, and samples of live and dead branches and leaves were removed for determination of Pu concentrations. The remaining leaves and branches were divided into live and dead materials and weighed after drying at  $65^{\circ} \text{C}$  for 72 h. A subsample of trunk was removed and divided into bark and bole wood components. These components were weighed before and after drying at  $65^{\circ} \text{C}$  for 400 h. The wet weight of the remaining trunk was determined, and the oven-dried weights of bark and bole wood were computed using wet weight:dry weight conversion factors obtained from the trunk subsample. Plutonium concentrations were determined in the bark and bole wood subsamples. Branch material was not divided into bark and wood fractions. Trees less than 2 m tall were considered to be herbaceous vegetation. The weights of components in unsampled trees were predicted by regressions of the logarithm of component oven-dried weight upon the logarithm of dbh (Beauchamp and Olson 1973).

The Pu concentrations of pine tree components in unsampled trees were predicted by regressions of the logarithm of concentration upon the logarithm of distance from the point of Pu release.

Samples of herbaceous vegetation, litter, resuspendible materials and soils were collected at 18 grid points in Field 1 and at 12 grid points in Field 2. Grids were composed of sampling blocks placed at 3, 15 and 27 m from the southwestern margin of the fields on transects across the short axis of the fields. These transects originated at 30.4 m (100 ft) intervals on the long axis of each field. Each sampling block was 3 x 10 m and contained ten 1 x 3 m plots. A randomly chosen plot was sampled in each block. Sampling blocks were marked for use in future studies.

Within each 1 x 3 m sampling plot, the herbaceous vegetation was clipped at ground level and placed in paper bags. Care was taken to prevent contamination of vegetation by soil. No distinction was made between live and dead vegetation at the time of sampling, but subsamples of the vegetation were later analyzed to determine the proportions of live and dead material. The biomass of the vegetation was determined after drying at 65°C for 72 h. Plutonium concentrations were determined in a  $\geq 200$  g subsample of the vegetation.

After the vegetation was clipped, the litter was removed from the center 1 x 1 m section of the plot by brushing the soil surface with a wisk broom. Litter was dried at 65°C for 72 h, sieved over a 1 mm mesh for 1 minute using a Cenco Sieve Shaker (Cat. No. 18480), redried, and weighed. A  $\geq 200$  g subsample of litter was used for determination of Pu concentrations.

Soil cores of 3.8 cm diameter were taken in each sampling plot using a split-barrel sampler and were divided into 0-5, 5-15, and 15-30 cm fractions. For unexplained reasons, the Pu concentrations in the 0-5 cm soil fraction were 50% lower than subsequent measures and could not be reproduced. Because of this discrepancy, we subsequently report Pu concentrations determined from 0-5 cm samples collected in November 1974 using a soil auger (Pinder and Paine in press) in the following discussions.

The amount of Pu in the litter and on the soil surface which could be resuspended into the atmosphere by a  $6 \text{ m} \cdot \text{sec}^{-1}$  wind was determined at 4 locations within each sampling plot by drawing a nearly laminar flow of air across a  $232 \text{ cm}^2$  (6 x 6 in) area under a 1 cm tall stainless-steel hood and collecting the resuspended materials in the paper vacuum-cleaner bag of a small, electrical vacuum cleaner (Sears Hand-Vac, Model 208.61110). A wipe of the interior of the plastic compartment holding the paper bag was taken to collect materials passing through the bag and was included in the sample. The oven-dried weight of the resuspended material was determined. The resuspendible samples were drawn from areas previously clipped of above-ground vegetation. The resuspended Pu does not represent a unique ecosystem component, but includes Pu from both the litter and soil. Consequently, resuspended Pu was not included in the computation of total Pu inventories in the ecosystem.

Dryfall deposition was determined using  $15.2 \times 15.2 \text{ cm}$  sticky paper collectors suspended 7.6 m above the ground near the center of each field. Eighteen determinations of dryfall input were made in each field with each sample being exposed for approximately 7 days.

All samples were ashed prior to determination of Pu concentrations. Vegetation, resuspension and deposition samples were ashed at 550°C. Soil samples were ground to a particle size  $\leq 500 \mu\text{m}$ , and ashed at 500°C.

Actinide elements were leached from a  $\leq 10\text{g}$  aliquot of the sample ash with hot 8M HCl, and valences were adjusted to insure formation of  $\text{Pu}(\text{IV})$ ,  $\text{Np}(\text{IV})$  and  $\text{U}(\text{VI})$ . Plutonium, Np and U were extracted into 10% triisooctylamine in xylene, and the Pu was separated from the Np and U by reducing  $\text{Pu}(\text{IV})$  to  $\text{Pu}(\text{III})$  with  $\text{NH}_4\text{I}$  and extracting into 8M HCl. This solution was evaporated to dryness and oxidized to destroy residual organic matter. The Pu was taken up in 8M  $\text{HNO}_3$ , and the valence was adjusted to  $\text{Pu}(\text{IV})$ . Final purification was accomplished by adsorbing the  $\text{Pu}(\text{IV})$  onto an anion exchange column and removing any residual Fe, U or other contaminants from the column with 4M  $\text{HNO}_3$ . The  $\text{Pu}(\text{IV})$  is reduced to  $\text{Pu}(\text{III})$  and eluted from the column with  $\text{H}_2\text{SO}_3$ . Following purification, the Pu was electrodeposited upon platinum plates, and the amounts of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  determined by alpha spectrometry using low-background high-resolution surface-barrier detectors. Counting times ranged from 2 to 7 days depending upon the Pu concentration of the sample. An internal standard of  $^{236}\text{Pu}$  was used to determine recovery efficiencies.

After determining the concentrations of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  in a sample, we computed the ratio of the  $^{238}\text{Pu}$  concentration to the  $^{239,240}\text{Pu}$  concentration and termed this the isotopic ratio (IR) of the sample. In subsequent discussions, we use the isotopic ratios to evaluate the relative importance of different pathways of Pu movement by assuming (1) that all isotopes of Pu are equally mobile and (2) that

similarity in IR between potential donor and acceptor components of the ecosystem indicates transfer of Pu between these components. We studied the H-area facility because we (1) expected the change in releases from  $^{239,240}\text{Pu}$  to  $^{238}\text{Pu}$  to have produced different IR in current deposition, resuspendibles and soils and (2) hoped to use these different IR to estimate the relative importances of various mechanisms resulting in Pu contamination of the vegetation.

Mean standing crops of Pu ( $\text{nCi} \cdot \text{m}^{-2}$ ) in herbaceous vegetation, litter and resuspendible materials were obtained by multiplying the component's weight at each sampling location by its concentration and averaging across sampling locations within each field. Standing crops of Pu in soil fractions were computed by multiplying average concentrations in each field by the weight of soil in that fraction where weights were computed using a bulk density of  $1.3 \text{ g} \cdot \text{cm}^{-3}$ . Bulk density determinations were made by D. C. Adriano of the Savannah River Ecology Laboratory. Plutonium standing crops in the bark, live branch and live leaf components of pine trees were obtained by summing the product of the predicted weights and concentrations within a field and dividing by the area of the field. Because statistically significant regression of the logarithms of concentrations on the logarithms of distance from the point of release were not obtained for dead branches and dead leaves, Pu standing crops in these components were estimated by multiplying the sum of the predicted weights for that component by the mean concentration and dividing by the area of the field.

Concentration data are expressed in units of  $\text{pCi Pu} \cdot \text{g}^{-1}$  and standing crops are expressed in units of  $\text{nCi Pu} \cdot \text{m}^{-2}$ . Although we would have preferred to use units of  $\text{g Pu} \cdot \text{g}^{-1}$  and  $\text{g Pu} \cdot \text{m}^{-2}$ , we could

not distinguish between  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  by alpha-spectrometry and, therefore, had to combine these isotopes. Statistical analyses were performed using the Statistical Analysis System (Barr et al. 1976). One sampling plot in Field 1 was omitted from the data analysis because of errors in processing.

## RESULTS AND DISCUSSION

### Aerial Deposition of Plutonium

The deposition rates of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  were usually less than  $0.2 \text{ pCi} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ , but greater rates were occasionally observed (Fig. 1). Mean deposition rates were  $0.33 \text{ pCi } ^{238}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  and  $0.14 \text{ pCi } ^{239,240}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  in Field 1 and  $0.31 \text{ pCi } ^{238}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  and  $0.37 \text{ pCi } ^{239,240}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  in Field 2. Differences in mean deposition rates between the fields were not statistically significant ( $P \geq 0.10$ ). We expected deposition rates to be greater in Field 1 because of its proximity to the stack and cannot explain why the fields had similar deposition rates when subsequent data showed total amounts of both isotopes to be greater on Field 1. Combining the data from both collectors yielded mean deposition rates of  $0.32 \text{ pCi } ^{238}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  and  $0.25 \text{ pCi } ^{239,240}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  and a mean IR of 1.90. These combined estimates will be used in all further discussions.

In computing the deposition rates in the preceding paragraph we omitted a 7-day period when deposition rates of  $4.52 \text{ pCi } ^{238}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  and  $158 \text{ pCi } ^{239,240}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  occurred on Field 1 and  $0.93 \text{ pCi} \cdot ^{238}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  and  $1.89 \text{ pCi } ^{239,240}\text{Pu} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  occurred on Field 2. Inclusion of these observations would have increased deposition rates and made the means on Field 1 greater than those on Field

0.32  
IR = 1.90

2; however, with only 18 sampling periods we cannot accurately determine the frequency of such high observations. Days of exceedingly high deposition could occur several times per year, or they may occur only once in 10 yr. An evaluation of the significance of these large depositions awaits further measurements of deposition rates and the results of in-progress studies of the size and chemical character of the released Pu particles.

#### Plutonium Concentrations

The concentrations of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  and the isotopic ratios in herbaceous vegetation, litter, resuspendible materials and soils are summarized in Table 1. The mean concentrations of both  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  were greater in Field 1 than in Field 2. In most cases the differences in mean concentrations were statistically significant. Only the mean  $^{239,240}\text{Pu}$  concentration in the 5-15 cm fraction of the soil was greater on Field 2. Plutonium concentrations in the lower soil depths were highly variable, and the greater mean  $^{239,240}\text{Pu}$  concentration in the 5-15 cm depth on Field 2 was due to a single large value. Plutonium concentrations in the 0-5 cm soil fraction were greater on Field 1 than Field 2, but the differences were not statistically significant because the sampling design was established to look at small scale variations (Pinder and Paine in press). Subsequent sampling has verified that soil Pu concentrations in the 0-5 cm depth are greater in Field 1.

Although concentrations differed between fields, the concentrations in the various ecosystem components showed similar patterns for both isotopes in both fields. The largest concentrations were usually observed in the resuspendible materials with the next largest concentra-

tions occurring in litter. The smallest concentrations were usually observed in the deeper soil samples.

Isotopic ratios were similar on the two fields even though concentrations differed. The largest IR were observed in the vegetation and litter. The smallest IR occurred in the soils. Isotopic ratios for the resuspendibles were intermediate between vegetation and soil. The IR for vegetation and litter compartments were similar to that observed in the deposition collectors and suggested that the majority of the Pu in vegetation was recently released Pu. The lower IR for soils reflects the earlier releases that were primarily  $^{239,240}\text{Pu}$ .

The intermediate IR observed in resuspendible materials indicated that both soil Pu and Pu-containing particles of more recent release had been resuspended. A  $6 \text{ m} \cdot \text{sec}^{-1}$  wind velocity is unusually fast for this region, and more typical, slower winds might resuspend only lighter particles and result in different Pu concentrations and isotopic ratios. Isotopic ratios in resuspendible materials in slower wind velocities could be greater because the heavier soil particles might not be resuspended.

Plutonium contamination of the vegetation can occur by three mechanisms: (1) root uptake and translocation to above-ground plant parts; (2) direct deposition upon the vegetation of Pu-containing particles released from the stack; and (3) impaction of resuspended Pu-containing particles on plant surfaces. The ratios of Pu concentrations in vegetation to those in the upper 5 cm of soil ranged from 0.064 for  $^{239,240}\text{Pu}$  in Field 2 to 0.86 for  $^{238}\text{Pu}$  in Field 1. These concentration ratios were substantially larger than the  $10^{-3}$  to  $10^{-5}$  ratios resulting from root uptake of Pu (Adriano et al. in press, Bennett 1976, Francis 1973,

Hanson 1975, Schulz et al. 1976) and indicated that direct deposition and resuspension were the important processes causing Pu contamination of the vegetation. If uptake had been the only process operating in the system, we would probably have observed concentrations of  $< 0.001 \text{ pCi } ^{238}\text{Pu} \cdot \text{g}^{-1}$  and  $< 0.003 \text{ pCi } ^{239,240}\text{Pu} \cdot \text{g}^{-1}$  in the vegetation on Field 1 and even lower concentrations on Field 2. Because observed concentrations were 100x these expected values, root uptake apparently contributed little to Pu concentrations in the vegetation.

Estimates of the relative importances of direct deposition and resuspension can be obtained from the isotopic ratios. If we assume that 1.9, 1.7 and 1.1 (weighted averages from Fig. 1 and Table 1) are representative IR for deposition, vegetation and resuspendible materials, respectively, then approximately 80% of the contamination in the vegetation resulted from direct deposition and 20% resulted from resuspension. This estimate was based upon the assumption of similar behavior for each isotope and was determined by solving the simultaneous equations,  $x + y = 1$  and  $ax + by = c$ , where  $x =$  the fraction of Pu in the vegetation due to deposition,  $y =$  the fraction of Pu in the vegetation due to resuspension,  $a =$  proportion of  $^{238}\text{Pu}$  in deposition  $= \text{IR}/(1 + \text{IR}) = 0.66$ ,  $b =$  proportion of  $^{238}\text{Pu}$  in resuspendible materials, and  $c =$  proportion of  $^{238}\text{Pu}$  in the vegetation. If lower wind velocities produce larger IR in resuspendibles, we have underestimated the importance of resuspension. Because of the importance of direct deposition and resuspension, most of the Pu contamination of the vegetation was probably surface contamination. Less than  $10^{-3}$  of this surface deposited Pu is translocated to internal plant tissues (Cataldo et al. 1976).

The concentrations of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  in loblolly pines (Table 2) were less than that in herbaceous vegetation and litter. The IR for

pine components were greater than 1.9 and indicated that the principle mode of contamination was direct deposition. Thus, most of the contamination was probably on the surface of the vegetation. The lower concentrations in pines may be due to different surface-to-volume ratios or to the apparent absence of resuspension contamination in pines. The concentrations of Pu in bole wood were not included in Table 2 because of cross-contamination from bark to bole wood during sample preparation. Although we could not estimate Pu concentrations in bole wood, our data suggested that these concentrations were less than  $0.001 \text{ pCi} \cdot \text{g}^{-1}$ .

Least-square regressions of the natural logarithm of concentration on the natural logarithm of distance from the stack indicated that  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  concentrations in leaves, branches and bark of loblolly pines decreased significantly ( $P \leq 0.10$ ) with increasing distance from the point of release. Similar decreases were observed in the Pu concentrations of dead leaves and dead branches, but the regressions were not statistically significant. Concentrations predicted from the regression equations (Beauchamp and Olsen 1973) for hypothetical trees located in the centers of Fields 1 and 2 were similar to the mean concentrations reported for these fields in Table 2.

#### Inventory of Plutonium Standing Crops

The total inventories of Pu (Fig. 2) in Field 1 were approximately  $57 \text{ nCi } ^{238}\text{Pu} \cdot \text{m}^{-2}$  and  $255 \text{ nCi } ^{239,240}\text{Pu} \cdot \text{m}^{-2}$ . In Field 2 the total inventories were approximately  $15 \text{ nCi } ^{238}\text{Pu} \cdot \text{m}^{-2}$  and  $79 \text{ nCi } ^{239,240}\text{Pu} \cdot \text{m}^{-2}$ . The inventories for  $^{238}\text{Pu}$  were only 20% of those for  $^{239,240}\text{Pu}$  although more  $^{238}\text{Pu}$  than  $^{239,240}\text{Pu}$  has been released from H-area. Most of the  $^{238}\text{Pu}$  release occurred during the accidental failure of the

filtering mechanism in 1969, and most (60%) of this release occurred during a single 24 h period. Presumably, the weather conditions during this period were such that little of the  $^{238}\text{Pu}$  was deposited upon our study areas.

Because the relative amounts of each isotope in pines, herbaceous vegetation, litter and soil were similar on the two fields, the data were combined to estimate the fraction of the total inventories contained in each of these components (Table 3). For both nuclides in both fields, the majority of the Pu was contained in the upper 5 cm of the soil. Approximately 1.3% of the  $^{238}\text{Pu}$  but only 0.2% of the  $^{239,240}\text{Pu}$  occurred in the pines, litter and herbaceous vegetation. The greater occurrence of  $^{238}\text{Pu}$  in the vegetation components probably reflects its more recent release from the H-area facility. Although the pines were less important than the herbaceous vegetation in this inventory, a more extensive canopy of pines could greatly alter the relative importance of pines and herbaceous vegetation. Both loblolly pines and hardwood trees can accumulate Pu under their canopies due to stemflow and leaf fall (Adriano and Pinder 1977). Including the bole wood component with assumed  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  concentrations of  $0.0005 \text{ pCi} \cdot \text{g}^{-1}$  does not measurably change the importance of pines.

There were more pines on Field 1 (28) than on Field 2 (7), but neither the greater abundance of pines nor the differences in amounts of herbaceous vegetation (Fig. 2) affected the proportion of either  $^{238}\text{Pu}$  or  $^{239,240}\text{Pu}$  occurring in either vegetation or litter. Minor structural differences in the plant community apparently have negligible effects on the distribution of Pu between soil and vegetation.

An important difference occurred between fields in the proportion of the Pu that could be resuspended. In Field 1, approximately 2% of the  $^{238}\text{Pu}$  and 0.5% of the  $^{239,240}\text{Pu}$  could be resuspended by a  $6 \text{ m} \cdot \text{sec}^{-1}$  wind, whereas in Field 2 only 0.5% of the  $^{238}\text{Pu}$  and 0.1% of the  $^{239,240}\text{Pu}$  could be resuspended. The mass of resuspended material averaged  $198 \text{ g} \cdot \text{m}^{-2}$  in Field 1 and  $81 \text{ g} \cdot \text{m}^{-2}$  in Field 2. These means were significantly ( $P < 0.05$ ) different. Differences in resuspendibility were probably caused by differences in the structure of the herbaceous plant community. The amounts of herbaceous vegetation and litter were greater on Field 2 than on Field 1. There was also a greater variation in litter weight in Field 1. Litter weights ranged from 7.5 to  $786 \text{ g} \cdot \text{m}^{-2}$  on Field 1. The range on Field 2 was only 91 to  $544 \text{ g} \cdot \text{m}^{-2}$ . The greater range on Field 1 resulted from poor plant growth on some very clayey areas where subsoils had been exposed. A significant negative correlation existed between the weight of resuspendibles and the weight of litter on Field 1 ( $r = -0.51$ ;  $df = 15$ ;  $P < 0.05$ ). The lower resuspension on Field 2 and the negative correlation between resuspension and litter weight in Field 1 suggest that litter cover can reduce resuspension of Pu.

At current deposition rates, the amounts of resuspendible  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  on Field 1 represent 9.9 and 13.0 yr of input, respectively. On Field 2, the resuspendible amounts represent only 0.6 and 0.8 yr of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  input, respectively. Thus, where resuspension is not retarded by vegetation and litter cover, Pu may remain on the soil surface in a resuspendible state for several years.

If, as indicated by the isotopic ratios, deposition was the major cause of Pu contamination of the vegetation, the deposition rates in Fig. 1 should be large enough to account for the Pu standing crops in

vegetation. Most of the herbaceous species on these fields produce above-ground vegetation from April through late September. The plants then die back to ground level and subsist through the winter as rosettes or under-ground propagules. The dead, above-ground material may remain standing for well over a year. Thus at the time of sampling, most of the live portion of the vegetation had been standing approximately 3 months, and most of the dead portion had been standing approximately 15 months. Approximately 55 and 46% of the herbaceous vegetation on Fields 1 and 2, respectively, was live vegetation. Thus, deposition could easily have accounted for the observed Pu standing crops in Field 2 because: (1) the  $^{238}\text{Pu}$  content of the vegetation represented only 82 days of input; (2) the  $^{239,240}\text{Pu}$  content represented only 35 days of input; and (3) once Pu-containing particles are deposited on vegetation surfaces they are not readily removed (Cataldo et al. 1976, Cataldo and Vaughan 1977). In Field 1, however, the deposition rates were not sufficient to explain Pu standing crops in the vegetation. Nearly all the Pu deposition in the past year in Field 1 would have had to impact on the vegetation to explain the observed standing crops. Deposition would be sufficient to explain the Pu standing crops if the exceptionally high deposition rates discussed previously occurred several times each year. Or, it may be that resuspension was more important on Field 1 than on Field 2. Mean isotopic ratios in the vegetation were lower on Field 1, but this difference was not statistically significant.

A greater proportion of the Pu in the soil apparently occurred at greater depths in Field 1. This difference probably reflects different histories of disturbance. Field 1 was close to old building sites and areas once used to support construction activities. Many past soil

disturbances were apparent in Field 1. Soil disturbances were also evident in Field 2. Because of these disturbances, the data were not used to estimate the mobility of Pu nuclides through the soil profile.

Two potentially important ecosystem components, plant roots and consumers, were not included in our inventory. Because plutonium concentrations in consumer organisms on the Savannah River Plant are approximately one-tenth the concentrations in plants (McLendon et al. 1976), their omission should have a negligible effect on the total inventory. The small roots of the herbaceous vegetation were not removed from the soil samples, and their Pu contents are contained in the soil components. Pine roots, however, were not sampled. Their omission should not greatly affect the inventory unless their concentrations were approximately 100 x those of bole wood. We omitted detailed studies of roots from our inventory because the adherence of even a few small soil particles to root surfaces can greatly bias determinations of Pu concentrations in roots.

#### SUMMARY AND CONCLUSIONS

The majority of both  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  in the two old-field ecosystems was contained in the upper 5 cm of the soil. Most of the remaining Pu was contained in the lower soil depths with less than 0.3% of the  $^{238}\text{Pu}$  and less than 0.1% of the  $^{239,240}\text{Pu}$  occurring in the vegetation. This distribution of Pu among soil and vegetation components is similar to that observed by Dahlman et al. (in press) in a forest ecosystem on old lake-bed sediments contaminated with  $^{239}\text{Pu}$ . The Pu distribution is also similar to those reported by Hakonson and Nyhan (in

press) for savannah-forest communities on the floodplains of contaminated streams and grass and forb communities on a former nuclear-test site in New Mexico and is similar to that reported by Romney et al. (1976) for nuclear test sites in Nevada. A maximum of 0.1% of the Pu at these sites was associated with the vegetation. Our distributions were similar although the amounts of Pu per  $m^2$  in our study areas were smaller than that at the other sites (Dahlman et al. in press, Hakonson and Nyhan in press, Nyhan et al. 1976, Romney et al. 1976).

Although most of the Pu was contained in the soil, from 0.1 to 2% of an isotope's total inventory was near the soil surface and could be resuspended into the atmosphere by a  $6 m \cdot sec^{-1}$  wind. The amounts of Pu that could be resuspended on Field 1 represented at least 9 yr of input at current deposition rates. Thus, Pu may remain at the soil surface for several years after deposition and may potentially be resuspended onto vegetation surfaces, lost from the system as airborne dust or resuspended during agricultural practices (Milham et al. 1976).

Total Pu inventories ( $^{238}Pu + ^{239,240}Pu$ ) were  $312 nCi \cdot m^{-2}$  on Field 1 and  $94 nCi \cdot m^{-2}$  on Field 2. The rapid decrease in inventories from Field 1 to Field 2 probably continues for several more km because most of the Pu released from reprocessing facilities is deposited within 3 km from the point of release (McLendon 1975, McLendon et al. 1976). Beyond the detectable effects of releases from the reprocessing facility, global fallout has produced an inventory of approximately  $2 nCi \cdot m^{-2}$  (McLendon 1975). Although total inventories differed between fields, the relative amounts in vegetation, litter and soil were similar. The only major difference between fields was that a greater percentage of both isotopes could be resuspended on Field 1. This

difference was apparently the result of sparser vegetation and litter cover on Field 1.

Our data indicated that two physical processes, direct deposition of stack released Pu onto plant surfaces and resuspension of Pu from the soil surface, were the important modes of transport to the vegetation. The amount of Pu reaching above-ground vegetation via root uptake was apparently negligible. Direct deposition appeared to be responsible for approximately 80% of the Pu in the vegetation, whereas resuspension was responsible for the remaining 20%. Although resuspension is the major cause of contamination of the vegetation in several arid ecosystems (Hakonson and Nyhan in press, Romney et al. 1976, Romney and Wallace 1977), our studies (see also McLendon et al. 1976) are the first to indicate the importance of resuspension in more humid ecosystems. Direct deposition has not been an important mechanism of plant contamination in other studies of Pu because of low deposition rates. The indicated importance of direct deposition demonstrates the problems involved in using results of Pu studies on weapons testing sites or contaminated floodplains to predict the behavior of Pu released from reprocessing facilities. Moreover, the importance of direct deposition suggests that the impact of a reprocessing facility on an agricultural crop may depend more upon the Pu releases during the crop's growing season than on the past history of Pu releases.

The inventories observed here cannot be taken as estimates of the inventories to be expected at future reprocessing sites. Advances in air-filtering technologies should result in a smaller fraction of the Pu handled by future facilities being released to the atmosphere (Barr 1976); however, these facilities may be much larger than H-area and

handle far more Pu. Most of the Pu was released from H-area before efficient filtration and during a single accidental release. If large accidental releases do not occur at future sites, their total inventories should be less than that observed at H-area. The Pu standing crops of the vegetation, however, could be greater at future sites if their average emission rates exceeded those of H-area.

The results of this study suggest five main conclusions concerning mobility of Pu released from nuclear-fuel reprocessing facilities: (1) Pu is retained in the upper regions of the soil profile; (2) Pu may accumulate on the soil surface and remain resuspendible for several years; (3) Pu contamination of vegetation will be primarily surface contamination resulting from physical rather than biological processes; (4) Pu releases occurring during a plant's growing period will have a considerable, if not the major, impact on its Pu concentration even though substantial amounts of resuspendible Pu are present; and (5) the impact of the biotic community on the distribution of Pu between vegetation and soil appears to be small, but the structure of the vegetation apparently affects the resuspension of Pu from the soil to the atmosphere.

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TABLE 1. Concentrations of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  and isotopic ratios ( $^{238}\text{Pu}/^{239,240}\text{Pu}$ ) in herbaceous vegetation, litter, surface resuspendibles and soil on Fields 1 and 2. Units are  $\text{pCi} \cdot \text{g}^{-1}$ .

Ecosystem Component	Field 1			Field 2			F <sup>a</sup>	df
	n	Mean	Standard deviation	n	Mean	Standard deviation		
-- $^{238}\text{Pu}$ --								
Vegetation	17	0.582	0.241	12	0.0833	0.0447	49.9**	1,27
Litter	17	2.50	1.71	12	0.913	0.575	9.42**	1,27
Resuspendibles <sub>b</sub>	17	6.82	7.96	12	0.771	0.373	6.84*	1,27
Soil 0-5 cm <sub>b</sub>	18	0.675	0.336	12	0.191	0.193	4.30	1,3
5-15 cm	17	0.0318	0.0308	12	0.0202	0.0165	1.42	1,27
15-30 cm	17	0.0419	0.0445	12	0.00975	0.00589	6.14*	1,27
-- $^{239,240}\text{Pu}$ --								
Vegetation	17	0.392	0.205	12	0.0445	0.0254	33.7**	1,27
Litter	17	2.00	1.27	12	0.422	0.195	17.8**	1,27
Resuspendibles <sub>b</sub>	17	6.66	8.69	12	0.835	0.557	5.33*	1,27
Soil 0-5 cm <sub>b</sub>	18	3.26	4.37	12	0.695	0.250	0.74	1,3
5-15 cm	17	0.0899	0.0975	12	0.223	0.509	1.12	1,27
15-30 cm	17	0.159	0.218	12	0.0248	0.0205	4.45*	1,27

TABLE 1. Continued

Ecosystem Component	Field 1			Field 2			F <sup>a</sup>	df
	n	Mean	Standard deviation	n	Mean	Standard deviation		
-- Isotopic Ratios --								
Vegetation	17	1.59	0.394	12	1.86	0.656	1.92	1,27
Litter	17	1.41	0.528	12	2.11	0.884	7.22*	1,27
Resuspendibles	17	1.14	0.468	12	1.03	0.358	0.44	1,27
Soil 0-5 <sup>b</sup>	18	0.558	0.496	12	0.308	0.135	0.49	1,3
5-15	17	0.461	0.215	12	0.241	0.148	9.40**	1,27
15-30	17	0.427	0.256	12	0.443	0.175	0.03	1,27

<sup>a</sup>F from analysis of variance testing the statistical null hypothesis of equal mean concentrations on Fields 1 and 2.

\*\* = (P ≤ 0.01); \* = (P ≤ 0.05).

<sup>b</sup>Data from Pinder and Paine, in press.

TABLE 2. Concentrations of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  and isotopic ratios ( $^{238}\text{Pu}/^{239,240}\text{Pu}$ ) in leaves, branches, bark, dead leaves and dead branches of loblolly pines on Fields 1 and 2. Units are  $\text{pCi} \cdot \text{g}^{-1}$ .

Component	Field 1			Field 2	
	n	Mean	Standard deviation	n	Mean
-- $^{238}\text{Pu}$ --					
Leaves	5	0.100	0.547	1	0.036
Branches	5	0.137	0.039	1	0.029
Bark	5	0.178	0.551	1	0.002
Dead Branches	3	0.220	0.164	1	0.119
-- $^{239,240}\text{Pu}$ --					
Leaves	5	0.050	0.032	1	0.013
Branches	5	0.059	0.020	1	0.012
Bark	5	0.068	0.024	1	0.001
Dead Branches	3	0.084	0.057	1	0.035
-- Isotopic Ratios --					
Leaves	5	2.09	0.320	1	2.84
Branches	5	2.38	0.411	1	2.46
Bark	5	2.74	0.688	1	3.26
Dead Leaves	4	1.92	1.18	1	3.40
Dead Branches	3	2.56	0.250	1	3.45

TABLE 3. The relative amounts of mass,  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  in pines, herbaceous vegetation, litter and soil.

COMPONENT	PERCENT OF TOTAL		
	Mass	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$
Pines	0.09	0.05	0.01
Herbaceous vegetation	0.07	0.18	0.03
Litter	0.05	1.07	0.15
Soil	>99.8	>98.6	>99.8

FIG. 1. Frequency distributions of deposition rates of Pu isotopes on Fields 1 and 2 located 230 and 420m, respectively, from the point of release, a 62 m stack. Sample size = 17.

FIG. 2. Inventories of biomass and Pu isotopes in Fields 1 and 2 shown on a logarithmic scale. The Pu contents of bole wood were estimated using concentrations of  $0.0005 \text{ pCi} \cdot \text{g}^{-1}$  and were not included in the total inventories.

FIELD 1

<sup>238</sup>Pu

<sup>239,240</sup>Pu

FREQUENCY

FIELD 2

pCi · m<sup>-2</sup> · day<sup>-1</sup>



