ATMOSPHERIC DEPOSITION, RESUSPENSION AND ROOT UPTAKE OF PLUTONIUM IN CORN AND OTHER GRAIN-PRODUCING AGROECOSYSTEMS NEAR A NUCLEAR FUEL FACILITY

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ABSTRACT-Plutonium released to the environment may contribute to dose to humans through inhalation or ingestion of contaminated foodstuffs. Plutonium contamination of agricultural plants may result from interception and retention of atmospheric deposition, resuspension of Pu-bearing soil particles to plant surfaces, and root uptake and translocation to grain. Plutonium on vegetation surfaces may be transferred to grain surfaces during mechanical harvesting. Data obtained from corn grown near the U. S. Department of Energy's H-Area nuclear fuel chemical separations facility on the Savannah River Site was used to estimate parameters of a simple model of Pu transport in agroecosystems. The parameter estimates for corn were compared to those previously obtained for wheat and soybeans. Despite some differences in parameter estimates among crops, the relative importances of atmospheric deposition, resuspension and root uptake were similar among crops. For even small depostion rates, the relative importances of processes for Pu contamination of corn grain should be: transfer of atmospheric deposition from vegetation surfaces to grain surfaces during combining > resuspension of soil to grain surfaces > root uptake. Approximately 3.9×10^{-5} of a year's atmospheric deposition is transferred to grain. Approximately 6.2×10^{-9} of the Pu inventory in the soil is resuspended to corn grain, and a further 7.3×10^{-10} of the soil inventory is absorbed by roots and translocated to grains.

INTRODUCTION

Plutonium isotopes may be released to the environment from nuclear weapons technology (Facer 1980; Perkins and Thomas 1980; Corey 1982), nuclear power technology (Barr 1976; Vaughan et al. 1976; Kreiter et al. 1980; Boone et al. 1981), and the use of ^{238}Pu in spacecraft power systems (Bennett 1981; Interagency Nuclear Safety Review Panel 1989). Once released to the terrestrial environment, Pu isotopes can contribute to dose to man through inhalation and ingestion. Plutonium contamination of food stuffs may occur as a result of: 1) root uptake and translocation of Pu to edible tissues; 2) interception and retention by plant surfaces of Pu-bearing particles in atmospheric deposition; and 3) resuspension of Pu-bearing particles from the soil surface to plant surfaces. Resuspension may occur due to the effects of wind (Sehmel 1980 and 1987; Shinn et al. 1983), raindrop splash (Dreicer et al. 1984), mechanical disturbance (Milham et al. 1976; Sehmel 1980), and animal activity (Sumerling et al. 1984). Particles present on leaf and stem surfaces due to atmospheric deposition and resuspension may be transferred to the surfaces of edible plant tissues during mechanical harvesting (McLeod et al. 1980; Adriano et al. 1982).

Although many studies of Pu transport in terrestrial systems have occurred [see reviews by Watters et al. (1980) and Watters (1987)], most studies have considered only a single transport process (e.g., root uptake) or have occurred several years after the release of Pu when atmospheric deposition was no longer occurring. Relatively few studies have determined the relative importance of root uptake, atmospheric deposition and resuspension for on-going instances of Pu release. Studies that compare the relative importances of these processes have occurred for wheat and soybean systems (McLeod et al. 1980; Adriano et al. 1982) for crops grown near nuclear fuel chemical separations plants.

The principal objectives of the study reported herein were 1) to evaluate the relative importances of root uptake, atmospheric deposition and resuspension in determining the Pu contents of vegetative and grain biomass for a corn agroecosystem and

2) to compare and contrast the the results for corn to those for wheat and soybeans. Corn systems have taller and more structured plant canopies than either wheat or soybeans, and the greater canopy structure is likely to affect atmospheric deposition and resuspension. The results for corn will be compared to those for wheat and soybeans using the estimates of transfer parameters of a model for Pu behavior in agroecosystems initially developed by McLeod et al. (1980). This model is described in the following section.

A SIMPLE MODEL OF PLUTONIUM BEHAVIOR IN AGROECOSYSTEMS

McLeod et al. (1980) and Adriano et al. (1982) developed and used a model of Pu transfers in agroecosystems to summarize their data. The model is simple, largely empirical and based primarily on data collected in the vicinity of the H-Area nuclear fuel chemical separations facility on the U. S. Department of Energy's Savannah River Site (hereafter SRS). The model estimates the transport of Pu in agroecosystems due to the interception and retention of atmospheric deposition, the resuspension of Pu from soils to plant surfaces, and the root uptake and translocation of Pu to grains (Figure 1). Plutonium transport is modeled for processes which occur in the field during the growth of the crop and those which occur in the combine during the mechanical harvesting of the grains. Processes occurring in the field include root uptake and translocation to grain and the interception and retention of deposition. Processes occurring in the combine include the transfer of Pu from vegetation surfaces to grain surfaces.

Although simple and empirical, the model reduces Pu behavior in agroecosystems to a set of four parameters that may be easily estimated and compared among systems. These four parameters are: 1) I_v, the fraction of the yearly atmospheric deposition of Pu that is retained on the surface of the vegetation at harvest; 2) T_{vg}, the fraction of the Pu on vegetation from deposition that is transferred to grain; 3) U_{sg}, the fraction of the soil inventory absorbed by roots and translocated to grain; and 4) R_{sg}, the fraction of the soil inventory of Pu that is transferred to grain surfaces by resuspension. Plutonium on

the surface of vegetation that enters the combine but is not transferred to grain is assumed to be deposited on the soil with the refuse vegetation.

The model depicted in Figure 1 contains two simplifications of the model originally presented in McLeod et al. (1980) and Adriano et al. (1982). The first simplification involves a reduction in the number of parameters used to express resuspension to grain. The original model contained two resuspension parameters associated with different processes. Resuspension in the original model involved 1) resuspension of Pu-bearing soil particles into the air during combining with subsequent intake into the combine and deposition onto grain and 2) the resuspension of Pu-bearing soil particles to vegetation surfaces with subsequent transfer to grain surfaces during combining. Although both of these processes probably occur, the estimation of parameter values for these processes was somewhat arbitrary because it was based on an assumption of equal transfer of Pu from vegetation to grain surfaces in the combine for Pu from both deposition and resuspension (i. e., equal T_{vg}). The current model does not separate these processes and uses a single parameter, R_{sg}, to account for the total movement of Pu from soil to grains by both processes.

Another simplification to the model originally presented by McLeod et al. (1980) and Adriano et al. (1982) is the omission of Pu movement to grain via foliar absorption. This process involves absorption at the leaf surface of atmospherically-deposited substances with subsequent translocation to grains and other plant tissues. Foliar absorption has been demonstrated for Pu (Cataldo et al. 1980). It was included in the original model but has been omitted here due to a lack of data comparing foliar absorption of Pu among different plant species. The omission of foliar uptake will be discussed in greater detail in a later section.

Atmospheric deposition is expressed as Bq m^{-2} d^{-1}, and the soil inventory and quantity of Pu harvest are expressed as Bq m^{-2}. The soil inventory is assumed to be uniformly mixed in the upper 0.15 m of soil by plowing, discing and other agricultural

operations. Because most releases from nuclear facilities will be ^{239}Pu, the effects of radioactive decay are negligible and are omitted from the model. Procedures for estimating the parameters are discussed by McLeod et al. (1980) and Adriano et al. (1982). Some of the original parameter estimates of McLeod et al. (1980) and Adriano et al. (1982) have been recalculated (Pinder et al. 1988b).

METHODS AND MATERIALS

Corn (*Zea mays* L., var. Coker-71) was grown in three agricultural fields > 0.5 ha in size near the H-Area nuclear fuels chemical separations facility on the SRS in Barnwell County, South Carolina. The atmospheric deposition rates and soil contents of ^{238}Pu and $^{239,240}Pu$ near H-Area have been increased due to deposition of Pu isotopes released to the atmosphere (see Pinder et al., 1979 for description and history of Pu releases). The fields were located 0.2, 0.4 and 0.7 km from the point of ^{238}Pu release, a 62-m tall stack at the H-Area facility. They will hereafter be referred to as Fields 1, 2 and 3.

The corn was planted on or about 6 April, 1976 after treating the fields with lime and 5-10-15 fertilizer. The crop was side-dressed with NH_4NO_3 in mid-May and harvested on 4 November, 1976 using a mechanical combine. Whenever possible, the procedures employed were chosen to simulate the size and complexity of full-scale agricultural operations.

The corn crops were sampled to determine the relative importances of root uptake, atmospheric deposition and resuspension and to estimate the parameters of the Pu transport model. Samples were obtained to determine crop biomass on or about 15 June, 1 August, 1 September and 27 October, 1976. For the 1 September and 27 October periods, vegetative and ear biomass were sampled separately. The concentrations of ^{238}Pu and $^{239,240}Pu$ were measured for all sampling periods from Fields 1 and 2. Separate determinations were performed for vegetative and ear biomass. For Field 3, plutonium concentrations were determined for only the 1 September and 27 October periods.

Sampling Procedures for Measuring Crop Biomass and Plutonium Concentrations

Corn vegetation was sampled using 10 randomly located 1 X 3 m plots arranged to contain 1 m long sections of three adjacent rows of corn plants. Corn plants were sampled in height increments to determine the distribution of biomass and Pu as a function of height above ground. For the 15 June and 1 August samples, the plants were sampled in 0.25 m height increments but combined into 1 m increments for Pu analyses. For the 1

September samples, the plants were sampled in height increments of 0.25 m from 0 to 2 m but were combined into 0.5 m increments for Pu determinations. Biomass above 2 m was treated as a single sample and was not subdivided into height increments.

The leaf morphology of corn plants changed with age and height above ground and necessitated different sampling procedures for the top, middle and bottom of the plant. The leaves at the top of the plant were nearly erect and were cut in 0.25 m increments. At the middle and bottom of the plant, the leaves were perpendicular to the stem, and the stem was cut in 0.25 m sections with all leaves attached to that section of stem being included in the sample. Leaf tips at the bottom of the plant tended to droop such that some portions of the leaves extended into the next lowest height increment. Thus, some of the biomass and Pu contents reported for increments of < 1-m tall were contained in lower increments. The tips of leaves for the lowest increments were sometimes in contact with the ground. These tips were removed before sampling.

The plants from the 27 October period were sampled and analyzed in height increments of 0 - 1 m and > 1 m. The larger increments were used because of deterioration in the structure of the corn canopy. As the plants entered senescence, the leaves dried and fell against the stem making sampling in resolutions of less than 1 m unnecessary.

Plant samples were dried at 60°C for 7 days, weighed to the nearest 0.1 g, and ashed at 550°C for 7 days. The concentrations of ^{238}Pu and $^{239,240}Pu$ for corn vegetation and grain were measured by the Savannah River Laboratory using methods described by Pinder et al. (1979). The inventories of Pu (i. e., mBq m^{-2}) in the corn canopy were computed by summing the product of plant biomass and Pu concentrations across height increments.

Determining the Distribution of Plutonium in Corn Ears

To determine the distribution of biomass and Pu among various components of corn ears, four random samples of from 5 to 25 ears each were removed from the plants in Fields 1, 2 and 3 during the 27 October sampling. Some of the ears were oven-dried at 60°C for 7

days, weighed to the nearest 0.1 g to determine average ear mass, ashed at 550 ^{o}C for 7 days, and submitted for Pu analyses. The remaining ears were separated by hand into outer leaves (i. e., those exposed directly to the atmosphere), inner leaves (i.e., those covered by outer leaves), cob and grain components. The masses of all tissues and the Pu concentrations for grain were determined for Fields 1, 2 and 3. In addition, the Pu concentrations in outer leaves, inner leaves and cobs were determined for the samples from Field 2.

These procedures provided data on the Pu concentrations of grains before combining and the ratio of grain mass to ear mass. The grain mass to ear mass ratios were used to compute the yield of grain (i.e., g of grain mass per m^2) from the estimates of ear mass per m^2 for the 27 October sampling.

Measuring the Plutonium Contents of Mechanically-Harvested Grains

Five random samples of grain were obtained from the combine for each field. The samples were sieved over a 4 mm mesh to remove vegetative plant parts, oven-dried at 60^{o}C for 7 days, ashed at 550^{o}C for 7 days, and submitted for Pu analyses.

Determining Plutonium Concentrations in Soils

To determine the Pu concentrations in soils, soil cores were taken from the plots used in the 27 October sampling. Cores were obtained using a split-barrel corer with a cross-sectional area of 0.001 m^2. The soil was sampled to a depth of 300 mm, divided into 0 - 50, 50 - 150 and 150 - 300 mm sections, and aliquots were removed from each section for Pu analyses using procedures described by Pinder et al. (1979).

Plutonium Deposition Rates

Deposition rates of ^{238}Pu and ^{239,240}Pu for the fields were predicted from regression equations that related deposition rate to the distance from the release stack (Corey et

al., 1982). Predicted deposition rates for ^{238}Pu were 28, 13 and 9 mBq m^{-2} d^{-1}, respectively for Fields 1, 2 and 3. Predicted rates for $^{239,240}Pu$ deposition for Fields 1, 2 and 3 were 13, 7.4 and 5 mBq m^{-2} d^{-1}, respectively. The predicted rates were verified against data obtained during the growing periods at Fields 1, 2, 4 and 6. Although some controversy exists concerning the appropriateness of tacky papers and other surrogate surfaces for measuring deposition (Hicks et al. 1980), the deposition rates measured by tacky papers at H-Area have been shown to be consistent with the accumulated inventories of Pu in plant canopies (McLeod et al., 1980; Adriano et al., 1982; Pinder et al. 1987, Pinder et al., 1988a). The Pu deposition near the chemical separations facilities involves particles of Pu metal or oxide (Saunders and Boni, 1980) of approximately 1 μm in size (Gay and Watts, 1981).

Measuring Root Uptake of Plutonium

To measure root uptake of Pu, plants were grown under greenhouse conditions in soils collected from the upper 50 mm of the soil profile from an area near H-Area where $^{239,240}Pu$ concentrations were known to be greater than those in Fields 1 and 2 (McLendon 1975). The greater $^{239,240}Pu$ concentrations were required to ensure measurable quantities of $^{239,240}Pu$ in plant tissues. Corn was grown under greenhouse conditions in plastic pots containing 12 kg of soil. Resuspension of soil to corn surfaces was prevented by a 10-mm thick fiberglass mat covering the soil surface. The plants grew through small openings cut in the center of the mat (see McLeod et al. 1981 for details). The Pu concentrations in leaves and grain were measured at maturity.

Determining the Relative Importances of Atmospheric Deposition and Soil Resuspension

Because the contributions of root uptake to the Pu content of plants grown in the fields near H-Area are usually negligible compared to surface contamination due to deposition and resuspension (McLeod et al. 1980; Adriano et al. 1982), the Pu content of

field grown crops and mechanically-harvested grains may be partitioned into components due to atmospheric deposition and resuspension using the isotopic ratio (ISR) of ^{238}Pu to $^{239,240}Pu$. There are distinct differences in the isotopic ratios for atmospheric deposition and soil resuspension at H-Area. The ISR for deposition onto Fields 1, 2 and 3 are 2.16, 1.80, and 1.71, respectively. These ratios were computed from the predicted rates of deposition and were verified against ratios observed in deposition samples during the growing season for Fields 1 and 2. The isotopic ratios for resuspension are 0.40 (Pinder et al., 1985) as estimated from samples of resuspendible materials collected from Fields 1 and 2. Resuspendible materials are defined as those particles on the soil surface that can be suspended into the atmosphere by a 6 m s^{-1} wind velocity. Numerous measures of this concentration have been obtained by passing an air current under a 0.15 m x 0.15 m X 10 mm tall metal hood and collecting particles from the outflow airstream (see Pinder et al., 1979 for a detailed description). It is believed that this measure is an accurate estimate of the Pu contents of the loose particles on the soil surface that can be transported by wind or raindrop splash. The Pu concentrations and ISR in resuspendible materials are also similar to those in soil particles < 125 μm (Pinder and McLeod 1988) which have been shown to be preferentially resuspended to and retained upon plant surfaces (Wallwork-Barber and Hakonson 1981; Dreicer et al. 1984). In partitioning the total Pu inventories for Field 3, the ISR of 0.40 for resuspendible materials on Fields 1 and 2 was assumed to be applicable to Field 3.

The total ^{238}Pu content of a vegetation sample can be partitioned into deposition and resuspension components by solving the simultaneous equations, $x + y = 1$ and $a\,x + b\,y = c$, where x = the fraction of ^{238}Pu in the sample due to deposition and y = the fraction of ^{238}Pu due to soil retention. The terms a, b, and c are the proportions (= ISR/(ISR + 1)) of ^{238}Pu in deposition, suspendible materials and the samples, respectively. After solving for x and y, the fraction of the ^{238}Pu content of the sample due to atmospheric deposition is estimated as $(a\,x)/(a\,x + b\,y)$. This procedure assumes negligible root

uptake. If root uptake represents an appreciable fraction of the Pu content of the plants, it will likely be confused as resuspension because of the similar ISR in soils and resuspendible materials. A further description of the methods for partitioning contributions due to deposition and resuspension is given by McLeod et al. (1980).

To verify the accuracy of the partitioning computations for vegetation, corn plants were grown in enclosures designed to permit atmospheric deposition but restrict resuspension. These enclosures were 1.2 m X 2.4 m X 1.2 m tall wood frames covered with polyethylene and laid on polyethylene sheets to prevent resuspension. They were open at the top to permit deposition. Corn plants were grown in the enclosures in plastic pots containing 12 kg of uncontaminated soil, harvested on 1 September, and analyzed for Pu contents. There were three enclosures located at distances of approximately 0.15, 0.35 and 0.50 km from the release stack.

RESULTS AND ESTIMATION OF MODEL PARAMETERS

Plutonium Concentrations in Soils

Mean Pu concentrations in soils for Fields 1, 2 and 3 are summarized in Table 1. The Pu concentrations in the 150 - 300 mm section were omitted from Table 1 because these concentrations were near or below detection limits for all fields. The Pu concentrations in soils decreased with increasing distance from the H-Area stack in a manner similar to that reported by McLendon (1975), McLendon et al. (1976) and McLeod et al. (1980). Mean ^{238}Pu concentrations (± S. E.) for the 0 - 150 mm section of the soils, which represents the principal rooting depth of the corn, were 7.6 ± 0.9, 2.1 ± 0.2 and 1.2 ± 0.1 mBq g^{-1}, respectively for Fields 1, 2 and 3. Mean $^{239,240}Pu$ concentrations for these soils were 22 ± 4, 6.0 ± 0.7 and 4.4 ± 0.5 mBq g^{-1}, respectively.

The Pu concentrations in the 0 - 50 mm and 50 - 150 mm soil sections usually differed by less than a factor of 2 X. This similarity reflects the mixing of the soil by agricultural operations. Before the soils were plowed and disced for crops, the Pu concentrations in the 0 - 50 mm sections were 4 - 30 X greater than those in the 50 - 150 mm sections (Pinder et al. 1979).

Plutonium Uptake by Corn Plants

The concentration ratios for root uptake by corn grown under greenhouse conditions were similar to those observed in other studies. The mean $^{239,240}Pu$ concentration (± S. E. for n = 6 samples) for the soils used in the uptake study was 63 ± 10 mBq g^{-1}, which was 3 - 50 X greater the means observed for the rooting depth of soils in Fields 1, 2 or 3. The mean $^{239,240}Pu$ concentration for corn leaves (± S. E. for n = 6 samples) was 18 ± 4 μBq per g dry mass which indicates a concentration ratio of approximately 2.9 X 10^{-4}. This is similar to the ratios observed for ^{238}Pu and ^{239}Pu uptake by corn in other studies using SRS soils (Hersloff and Corey 1978; McLeod et al. 1981; Adriano et al.

1986) and is similar to the ratio observed by Pimpl and Schmidt (1988) for corn uptake of Pu from a granitic soil.

The $^{239,240}Pu$ concentrations in grain samples were below the detection limits of approximately 3 $\mu Bq\ g^{-1}$ and suggested a concentration ratio for uptake of $< 10^{-5}$. Plutonium concentration ratios for grains are usually $\leq 10^{-6}$ and are less than those for leaves and stems (Romney et al. 1981), but there appear to be no published concentration ratios for corn grain. Hersloff and Corey (1978), McLeod et al. (1981), Adriano et al. (1986) and Pimpl and Schmidt (1988), who report ratios for vegetative tissues, do not report ratios for grains. In subsequent computations a concentration ratio for Pu in corn grains of 10^{-6} has been assumed.

Growth and Development of the Corn Crop

Corn biomass (Table 2) increased rapidly from planting and first emergence in late April to mid-June due to above average precipitation. From late June to early September precipitation was below average, and the biomass of the crop declined. The ears were formed and had largely filled by 1 September. After 1 September the plants entered senescence, and crop biomass declined as leaf biomass was lost and some ears were aborted. Although Fields 1, 2 and 3 received similar precipitation, differences in soils produced differences in moisture stress and ear production. The drought stress was especially pronounced on the more clayey soil of Field 1 (see McLeod et al. 1981 for soil descriptions) where many plants were < 1 m tall and did not produce ears.

Plutonium Concentrations of Field Grown Crops

The Pu concentrations for field grown corn varied among fields, among tissues and with height above ground (Table 3). Concentrations generally declined with increasing distance from the stack. The concentrations for leaf and stem samples averaged approximately 1.2 mBq ^{238}Pu and 0.6 mBq $^{239,240}Pu$ for Field 1, 0.5 mBq ^{238}Pu and 0.3 mBq

$^{239,240}Pu$ for Field 2, and 0.5 mBq ^{238}Pu and 0.3 mBq $^{239,240}Pu$ for Field 3. The concentrations for ears were usually < 0.1 mBq g^{-1} for both ^{238}Pu and $^{239,240}Pu$. Concentrations of ^{238}Pu were generally greater for the upper portions of plants, whereas the $^{239,240}Pu$ concentrations showed little apparent change with height increments. The number of replicates for determinations of concentrations were sometimes less than the number of plots sampled for biomass. This occurred for two reasons. First, the plot did not contain sufficient biomass for Pu analyses because of the effects of the drought. This was especially the case for samples of ear biomass. Second, some samples were destroyed in a small fire in the sample storage area.

The Pu concentrations for leaf and stem samples were greater than those expected to occur as a result of root uptake. For a concentration ratio of 2.9 X 10^{-4}, the expected concentrations for root uptake of $^{239,240}Pu$ from Fields 1, 2 and 3 are 7, 2 and 1 μBq g^{-1}, respectively. The expected ^{238}Pu concentrations are 2, 0.6 and 0.3 μBq g^{-1}, respectively. The expected concentrations were computed as the product of the concentration ratio and the Pu concentrations in the 0 - 150 mm section of the soil. The observed concentrations are approximately 100 X these expected values which indicates the far greater importance of surface contamination mechanisms. The contributions from root uptake are negligible compared to the surface Pu.

Tissue Masses and Plutonium Concentrations for Ears and Estimation of U_{sg}

There were considerable differences in Pu concentrations among the tissues of corn ears sampled from Field 2 (Table 4). The Pu analyses on ear tissues had greater accuracies and lower detection limits than those on leaf and stem samples due to the greater sample mass. The outer leaves of the ears, which were exposed to the atmosphere, had Pu concentrations similar to those observed for leaf and stem samples at the 27 October sampling. The inner leaves and other tissues had concentrations that were 10 - 100 X less than those for outer leaves. The $^{239,240}Pu$ concentrations in cob and grain

samples were below detection limits. Two of the four grain samples had detectable ^{238}Pu concentrations of approximately 1 $\mu Bq\ g^{-1}$. The ISR for outer and inner leaves suggested that atmospheric deposition was the principal source of the Pu.

Although care was taken to prevent cross-contamination from outer leaves to inner tissues, some movement of material among tissues should be expected. The outer leaves contained 94 % of the ^{238}Pu. Inner leaves contained 4 %, and the grain and cob contained approximately 1 % each. Because of the large difference in inventories between the outer leaves and grain, the possibility exists that much of the Pu content of the grain was the result of cross-contamination.

Fewer ears and less grain were available for Fields 1 and 3 which resulted in fewer determinations of Pu concentrations above detectable limits. For Field 1, only one sample had detectable ^{238}Pu concentrations of 0.6 $\mu Bq\ g^{-1}$. For Field 3, two of the four samples of grain had detectable ^{238}Pu concentrations of approximately 1 $\mu Bq\ g^{-1}$.

Grain accounted for the majority of ear mass on all three fields. The mean percent contributions of grain to ear mass ($\pm$ S. E.) were 69$\pm$ 2, 71 $\pm$ 1 and 69 $\pm$ 1 % for Fields 1, 2 and 3, respectively. These percentages and the biomass of corn ears at harvest (Table 2) indicate grain yields of 1, 222 and 63 g m^{-2} for Fields 1, 2 and 3. The small yield for Field 1 reflects the effects of drought. Assuming a concentration ratio of 10^{-6} between grain and soil, these yields result in estimates of U_{sg} (i. e., the fraction of the soil inventory transferred to grain by root uptake of 7.1 X 10^{-12}, 1.1 X 10^{-9} and 3.2 X 10^{-10} for Fields 1, 2 and 3.

The Plutonium Contents of Corn Due to Atmospheric Deposition

The inventories of ^{238}Pu (i.e., mBq m^{-2}) attributed to atmospheric deposition and resuspension are given in Table 5. The ^{238}Pu inventories due to deposition were fairly constant, and changes in inventories generally reflected changes in crop biomass. An exception to this pattern is the small inventory for Field 2 for the 15 June sampling

period. The ^{238}Pu concentrations for the vegetation on Field 2 were unusually small at this time period (Table 3). Unusually small concentrations were not observed for Field 1. The reason for the small concentrations is unknown.

The inventories attributed to atmospheric deposition for corn were greater than those for wheat and soybean crops grown on the same fields. Inventories for wheat were approximately 230 and 100 mBq ^{238}Pu m^{-2} for Fields 1 and 2 (McLeod et al. 1980). The inventories for soybeans were approximately 100 mBq ^{238}Pu m^{-2} for both Fields 1 and 2 (Adriano et al. 1982). The greater inventories for corn were not due to greater deposition rates of Pu. Measured deposition rates were consistent with predicted rates and similar to those observed for wheat and soybeans. The greater inventories for corn probably resulted from the greater biomass for corn. The biomass for wheat and soybeans were usually < 300 g dry mass per m^2.

At the time of harvest, the corn biomass and ^{238}Pu inventories attributed to atmospheric deposition were similar to those for wheat and soybeans due to leaffall associated with the senescence and drying of the plants. Estimates of I_v, the fraction of the yearly deposition retained on vegetation at harvest, for Fields 1, 2 and 3 were 0.019, 0.048 and 0.027, respectively.

The ^{238}Pu attributed to atmospheric deposition was distributed throughout the corn canopy at the 1 September sampling period (Fig. 1). Similar patterns were observed for the 1-m height increments at the other sampling periods. The inventories of Pu at different height increments generally reflected the relative biomass at that increment with some increased inventories in the upper increments due to increases in the concentrations of ^{238}Pu from atmospheric deposition (Table 6). The increased concentrations in the upper height increments may have resulted from greater surface area to mass ratios. The mean (± S. E.) surface area to mass ratios for corn vegetation borne at heights > 1 m is 16 ± 1 mm^3 g^{-1}, whereas the mean ratio for vegetation borne at heights < 1 m is 10 ± 1 mm^3 g^{-1} (data from Pinder et al. 1988a).

The concentrations of ^{238}Pu for plants grown in the polyethylene enclosures and harvested on 1 September were similar to those for ^{238}Pu attributed to atmospheric deposition (Table 6). The mean ^{238}Pu concentrations for whole plants (± S. E. for n = 5 samples) were 1.2 ± 0.3, 0.76 ± 0.29 and 0.38 ± 0.03 for the enclosures located at 0.15, 0.35 and 0.5 km from the stack, respectively. The similar concentrations support the accuracy of the partitioning computations. A similar correspondence of concentrations from enclosures and those from partitioning computations was observed for soybeans (Adriano et al. 1982).

The Plutonium Contents of Corn Due to Resuspension

Although the Pu inventories of the vegetation due to resuspension (Table 5) are not required for estimating model parameters, the inventories and the distribution of the resuspension inventories in the corn canopy are important for interpreting R_{sg} and understanding differences in estimates of R_{sg} among crops. The inventories due to resuspension ranged from 3 % to 21 % of those for deposition with little difference among fields in the relative importance of resuspension and deposition. Inventories from resuspension averaged 10 % of those from deposition. The inventories attributed to resuspension for Fields 1 and 2 were similar to those observed for wheat and soybean crops.

The inventories of ^{238}Pu due to resuspension showed relatively little change among sampling times. The major exception occurred for the 1 August sampling period on Field 2 where three plots had inventories > 50 mBq m^{-2}. Such unusually large and possible outlier values can occur for resuspension due to spatial and temporal variation in ISR for deposition and resuspendible materials as well as analytical errors in determining Pu concentrations (Pinder and McLeod 1989).

The ^{238}Pu due to resuspension for the 1 September sample was not uniformly distributed in the corn canopy but was concentrated in the < 1 m height increments (Fig.

1). Similar patterns were observed in the 1-m increments at other sampling periods. Greater resuspension near ground levels is consistent with the results of Dreicer et al. (1984) for tomato plants and the results of Pinder and McLeod (1988) for sunflowers grown on Field 2.

Plutonium Contents of Mechanically-Harvested Grain

The concentrations of ^{238}Pu and $^{239,240}Pu$ were easily measurable in samples of grain from the mechanical harvesting (Table 6) because of the relatively large sample masses available for analysis. The ^{238}Pu concentrations were usually > 1 μBq g^{-1} and were greater than those expected due to root uptake. For an assumed concentration ratio of 10^{-6} for root uptake, the expected concentrations for Fields 1, 2 and 3 are 0.008, 0.002 and 0.001 μBq ^{238}Pu g^{-1}. These concentrations are less than those observed for wheat and soybeans on Fields 1 and 2. Total ^{238}Pu concentrations for wheat grain from Fields 1 and 2 were 14 $\pm$ 2 and 7 $\pm$ 1 μBq g^{-1}, respectively (McLeod et al., 1980). Concentrations for soybeans were 3 $\pm$ 1 and 4 $\pm$ 1 μBq g^{-1}, respectively (Adriano et al., 1982).

The ^{238}Pu concentrations for combined grains appeared to be greater than those observed for the hand-separated grains, but the differences are not distinct because of the number of hand-separated grain samples with Pu concentrations near or below the detection limits. This complicates the interpretation of possible Pu transfers from vegetation to grain occurring in the combine and the estimation of T_{vg}. A measurable increase in Pu concentrations between hand-separated and mechanically-harvested grain is needed to indicate transfer within the combine. Because 1) mechanically-harvested grains had greater concentrations than those expected due to root uptake and 2) the Pu present in hand-separated grain could have resulted from cross contamination from outer leaves (see discussion above), it was assumed that all the Pu content of mechanically-harvested grain resulted from transfers within the combine. The resulting estimates of T_{vg}, the fraction transferred, were 1.8 X 10^{-5}, 1.0 X 10^{-3} and 1.0 X 10^{-3} for Fields 1, 2 and 3,

respectively. The relatively low value for Field 1 resulted from the poor yield of grain.

There was little evidence to indicate resuspension of Pu to grain surfaces. The ^{238}Pu concentrations for grains were always greater than the $^{239,240}Pu$ concentrations, and partitioning of the concentrations into components due to atmospheric deposition and resuspension indicated negligible contributions from resuspension (Table 6). None of the mechanically-harvested grain samples from Fields 1 and 3 and only 2 of the 5 samples from Field 2 had measurable quantities of Pu attributed to resuspension. Estimates of R_{sg} for Fields 1, 2 and 3 were 0, 1.3×10^{-8} and 0, respectively.

DISCUSSION

The drought conditions during the later part of the growing season have the potential to affect the parameter estimates. Greater vegetation biomass and grain yields may have resulted in greater estimates for I_v, U_{sg} and T_{vg}. The corn on Field 1 was most affected by drought, and the estimates of I_v, U_{sg} and T_{vg} from Field 1 are less than those from the Fields 2 and 3. To reduce the potential impacts of drought on parameter estimates, the estimates from Field 1 will be omitted from comparisons of parameters and Pu behaviors among crops.

Comparison of Model Parameters

Comparison of parameter estimates of corn with those for wheat and soybeans are presented in Table 8. Parameter estimates of I_v, U_{sg} and T_{vg} for wheat and soybeans are drawn from Pinder et al. (1988b). Estimates of R_{sg} for wheat and soybeans have been computed from the data of McLeod et al. (1980) and Adriano et al. (1982). Estimates of I_v are similar among crops, especially given the relatively large standard errors for soybeans and corn, whereas estimates of U_{sg}, T_{vg} and R_{sg} appear to differ among crops.

The different estimates of U_{sg} result from principally from differences in concentration ratios. The concentration ratios for wheat and soybeans are taken from the data of Romney et al. (1981) for the acidic Lyman and Malbis soils. The ratio for wheat grain was 1.6×10^{-7}. The ratio for beans was 1.1×10^{-6}. There was less variation among yields. Yields of wheat grain, beans and corn grain were 120, 100 and 140 g m^{-2}.

The estimate of T_{vg} for corn is less than those for wheat and soybeans. The lower estimate for corn probably reflects differences in the structure and operation of the combines. For wheat and soybeans, the combines clip plants near the ground surface and almost all the vegetation and surface Pu inventories are taken into the combine. In contrast, a corn combine usually removes only the upper portion of the plants where the

ears are located. As a result, only a portion of the corn biomass and surface ^{238}Pu inventory enters the combine, and not all of the surface Pu contamination is available to be transferred to grain. If T_{vg} is recomputed for only that portion of the Pu inventory for the > 1-m tall sections of corn plants, the mean (± S. E.) for Fields 2 and 3 is 0.0025 ± 0.0007 which is similar to that for soybeans.

To more directly compare the movement of atmospheric deposition to grains among crops, the fraction of the yearly deposition present on grain at harvest was computed. This fraction may be expressed as I_g where $I_g = I_v \times T_{vg}$. The mean I_g (± S. E. for n = 2 fields for each crop) were 1.3 (± 0.1) X 10^{-4} for wheat grain, 4.1 (± 2.6) X 10^{-5} for soybeans, and 3.9 (± 1.1) X 10^{-5} for corn. These values indicate greater transfer of atmospheric deposition to grains for wheat than for soybeans and corn. The fractions transferred for soybeans and corn are similar. The greater transfer for wheat occurs due to greater transfer from vegetation surfaces to grain surfaces in the combine (i. e., > T_{vg}).

The mechanics of combining which result in relatively low T_{vg} may also account for the relatively small R_{sg} for corn grain. Less transfer of resuspended Pu from vegetation surfaces to grain surfaces may have occurred for corn because the 1-m high setting of the combine head excluded the < 1-m high portions of the corn plants which contained the majority of the resuspended Pu (Figure 2). Moreover, the high setting may have reduced the intake of soil particles resuspended into the air during the combining activity.

Although some differences in parameter estimates occurred, these differences were smaller than those that might be expected given the differences in plant morphologies among crops. Greater similarity occurred between corn and soybeans, a monocot and a dicot, than between the two dicots, wheat and corn. The differences among crops appear to be more the result of plant height and combine design than differences in plant morphology.

The Relative Importances of Root Uptake, Atmospheric Deposition and Resuspension

The contributions of atmospheric deposition, resuspension and root uptake to the ^{239}Pu concentrations for grains for a corn agroecosystem are presented in Figure 3 for a hypothetical scenario of a nuclear facility operating over a 30-year period and depositing 100 and 1 mBq ^{239}Pu m^{-2} d^{-1} onto sites that already contained 75 Bq ^{239}Pu m^{-2} due to global fallout. These deposition rates were used because they encompass the range of rates observed on the SRS from areas close to the point of the release to areas where deposition rates are similar to global levels (Corey et al. 1982).

During the first year of operation at each deposition rate, the principal mode of contamination is the interception and retention of atmospheric deposition. Resuspension is less important than deposition, and the contamination resulting from root uptake is largely negligible compared to surface contamination from deposition and resuspension. As deposition continues, the Pu inventories in the soil at the higher deposition rate increase with concomitant increases in the relative importances of resuspension and uptake. The maximum contribution of resuspension to grain occurs in the 30th year of operation where resuspension contributes approximately 1 % as much Pu as deposition. Plutonium inventories in the soil do not increase appreciably at the lower deposition rate, and resuspension contributes approximately 3 % as much Pu to grain as deposition throughout the operation of the facility.

Greater resuspension (i. e., > R_{sg}) was observed for wheat and soybeans than for corn, but resuspension for these crops is also less important than deposition at 100 mBq m^{-2} d^{-1} (Figure 4). Resuspension for wheat and soybeans contributes < 10 % as much as deposition in the 30th year of operation. At deposition rates of 1 mBq m^{-2} d^{-1}, resuspension of previously deposited Pu contributes ≤ 50 % as much Pu to grain as deposition for wheat and soybeans (Fig. 4).

Resuspension in years 1 to 10 at the higher deposition rate may be somewhat greater than that depicted in Figure 3. Greater resuspension may occur because a large portion of

the soil inventory would be recent deposition that may be more readily suspended into the atmosphere due to its presence on the soil surface (Anspaugh et al. 1975). Once a plowing or discing event occurs, however, the recent deposition would be mixed into the soil (Adriano et al. 1982).

Although parameter estimates varied among crops, the crops demonstrated similar relative importances of deposition, resuspension and root uptake. This similarity occurs because estimated I_v were > 0.01, estimated T_{vg} were > 0.001, and estimated R_{sg} were > estimated U_{sg}. These values and patterns result in similar relative importances of deposition, resuspension and root uptake among agroecosystems.

The relatively small contributions of resuspension after 30 years at the greater deposition rate in Figure 4 contrasts with observations near H-Area where contributions of resuspension to wheat grain and beans may account for 30 to 50 % of those for deposition after only 20 years of operation (McLeod et al. 1980; Adriano et al. 1982). The greater importance of resuspension at H-Area results from previous periods of greater atmospheric release and deposition (Pinder et al. 1979). Much of the ^{238}Pu inventory of H-Area soils apparently resulted from an accidental release in 1969 (McLendon 1975). The ^{238}Pu inventories in the soil at H-Area represent approximately 100 years of deposition at current rates. Thus, periods of unusually large releases to the atmosphere may change the relative importances of pathways in subsequent years.

After shutdown of the facility, which is assumed to occur after 30 years, contamination processes would be limited to resuspension and root uptake. The Pu contents due to these processes after shutdown would be similar to those depicted in Figure 4 for the 30th year of operation. Resuspension would be of greater importance than root uptake. The Pu contents of grain at the higher deposition rate would be increased due to accumulation of Pu from deposition in the soil. Soil inventories would increase from 75 to 1170 Bq m^{-2} at the higher deposition rate. The fraction of the ^{239}Pu exported from the system per year with harvested corn grain after shutdown would be 6.9 X 10^{-9} (= R_{sg} + U_{sg})

which is 4000 X less than the loss rate per year of ^{239}Pu due to radioactive decay.

Although surface contamination predominates the Pu content of grains, it must be remembered that much of the surface Pu may be removed in food processing operations. The respective fates of surface and internal Pu contamination in food processing may vary among crops and processed foods, and little appears to be known about the fate of surface contamination in processing operations. Aarkrog's (1978) studies of Pu fallout in the 1960's suggests that about 25 % of the surface Pu content of grain can be incorporated into whole-grain food products. If 25 % of the surface Pu content of grain due to deposition and resuspension is incorporated into processed foods, surface Pu would still contribute the majority of the Pu content of processed foods for both deposition rates in Figures 3 and 4.

Assimilation of Pu from the gastrointestinal tract is affected by chemical and physical form (ICRP 1979) and may be greater for Pu incorporated into plant tissues via root uptake than for Pu in inorganic form (Sullivan et al. 1980). Estimates of the fraction of the ingested Pu that is assimilated range from 10^{-3} for readily available Pu to 10^{-5} for relatively insoluble Pu (ICRP 1979). Although the range of assimilation fractions is well defined, the nature of Pu-bearing particles on plant surfaces, and consequently, the appropriate assimilation fraction is dependent upon variable factors including the form of release and possible aging of Pu-bearing particles in the environment. If 1) 25 % of the surface Pu is retained in processed foods, 2) the assimilation fraction for surface Pu is 10^{-5}, and 3) the assimilation fraction for Pu from root uptake is 10^{-3}, then surface Pu accounts for most of the assimilated Pu at the higher deposition rate. In the 30th year of operation, 4 X as much Pu is ingested from surface contamination as from root uptake at 100 mBq m^{-2} d^{-1}. At the lower deposition rate, surface Pu accounts for one-half as much assimilated Pu as root uptake.

There is a further complication in assessing the relative importances of external and internal contamination in the amounts of assimilated Pu. The processes which cause

surface contamination (i. e., deposition and resuspension) can also result in internal contamination of grains through foliar absorption and translocation to grain. Cataldo et al. (1980) observed that approximately 5 X 10^{-5} of the Pu deposited on bush bean leaves was absorbed and translocated to roots and seeds. For the scenarios in Figure 3, foliar absorption and translocation to grain of 1 X 10^{-5} of the Pu deposited on leaves and stems to seeds would result in ^{239}Pu concentrations in corn grain of 0.096 and 0.000096 μBq g^{-1} for deposition rates of 100 and 1 mBq m^{-2} d^{-1}, respectively. These concentrations would not measurably increase the total Pu content of grain, but they would significantly alter the content of Pu incorporated into internal tissues. At the higher deposition rate, Pu concentrations from foliar uptake would be 15 X those for root uptake. At the lower deposition rate, foliar uptake would account for 2 X the concentration from root uptake.

Whereas physical processes and surface contamination dominate Pu transport processes in agroecosystems, uncertainties about 1) the fate of surface Pu in food processing, 2) the relative assimilation of surface and internal Pu, and 3) the potential importance of foliar uptake mechanisms limit our understanding and appreciation of the transport of Pu from grain-producing agroecosystems to the human diet.

CONCLUSIONS

These results imply some general conclusions concerning Pu mobility in agroecosystems. Where Pu releases to the atmosphere are on-going, atmospheric deposition will likely be the principal contamination mechanism. Where Pu releases and deposition have occurred in the past, resuspension is likely to be the predominant mechanism in moving Pu to grain crops. The greater relative importance of resuspension versus root uptake has been noted for a number of other systems (Watters et al. 1980; White et al. 1981; Arthur and Alldredge, 1982; Romney et al. 1987; Watters 1987) and is likely to be the case for most sites of Pu contamination except possibly for tree species where foliage is borne at considerable heights above the ground surface (Dahlman and McLeod 1977; Pinder et al. 1987).

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TABLE 1. Concentrations (mBq g^{-1}) of Pu isotopes in the 0 - 50 mm and 50 - 150 mm sections of the soil in Fields 1, 2 and 3. Data are means ± standard errors for n = 10 samples.

FIELD	SOIL SECTION (mm)	CONCENTRATIONS ^{238}Pu	$^{239,240}Pu$
1	0 - 50	11 ± 1	35 ± 7
	50 - 150	5.9 ± 0.8	16 ± 3
2	0 - 50	2.9 ± 0.3	8.6 ± 1.2
	50 - 150	1.7 ± 0.2	4.8 ± 0.7
3	0 - 50	1.3 ± 0.1	5.8 ± 1.1
	50 - 150	1.1 ± 0.2	3.7 ± 0.7

TABLE 2. Corn biomass (g m^{-2}) in 1-m height increments for Fields 1, 2 and 3. Data are means ± standard errors for 10 samples.

DATE AND HEIGHT INCREMENT	BIOMASS		
	Field 1	Field 2	Field 3
		--- Leaf and Stem Biomass ---	
15 June			
0.0-1.0	398 ± 74	236 ± 48	
1.0-2.0	53 ± 12	87 ± 25	
>2.0	1 ± 1	5 ± 5	
Total	451 ± 83	328 ± 76	
1 August			
0.0-1.0	359 ± 44	414 ± 39	
1.0-2.0	367 ± 55	921 ± 52	
>2.0	67 ± 12	223 ± 21	
Total	793 ± 91	1557 ± 81	
1 September			
0.0-0.5	178 ± 28	102 ± 10	108 ± 8
0.5-1.0	131 ± 18	99 ± 8	99 ± 8
1.0-1.5	104 ± 12	199 ± 49	107 ± 8
1.5-2.0	66 ± 11	100 ± 29	57 ± 4
>2.0	31 ± 6	59 ± 11	34 ± 5
Total	510 ± 66	480 ± 34	404 ± 25

Table 2. Continued.

DATE AND HEIGHT INCREMENT	BIOMASS		
	Field 1	Field 2	Field 3
		--- Leaf and Stem Biomass ---	
27 October			
0.0-1.0	127 ± 26	148 ± 28	147 ± 31
1.0-2.0	81 ± 21	106 ± 33	53 ± 15
>2.0	7 ± 5	24 ± 9	4 ± 4
Total	215 ± 47	278 ± 66	204 ± 42
		--- Ear Biomass ---	
1 September			
0.0-0.5			
0.5-1.0		3 ± 3	
1.0-1.5	14 ± 9	110 ± 30	15 ± 10
1.5-2.0	4 ± 3	313 ± 67	106 ± 30
>2.0		15 ± 8	10 ± 8
Total	18 ± 9	442 ± 67	131 ± 36
27 October			
0.0-1.0			
1.0-2.0	2 ± 2	283 ± 90	92 ± 36
>2.0			
Total	2 ± 2	283 ± 90	92 ± 36

TABLE 3. The concentrations (mBq g^{-1}) of ^{238}Pu and $^{239,240}Pu$ in corn grown on Fields 1, 2 and 3. Data are means ± standard errors for leaf and stem biomass and ear biomass in height increments (m) above the ground surface.

DATE AND HEIGHT INCREMENT	FIELD 1			FIELD 2			FIELD 3		
	n	^{238}Pu	$^{239,240}Pu$	n	^{238}Pu	$^{239,240}Pu$	n	^{238}Pu	$^{239,240}Pu$
				--- Leaf and Stem Samples ---					
15 June									
0.0-1.0	10	0.88±0.16	0.69±0.11	10	0.09±0.02	0.09±0.02			
1.0-2.0	9	1.3 ±0.3	0.92±0.26	9	0.14±0.03	0.13±0.04			
>2.0									
1 August									
0.0-1.0	10	0.78±0.23	0.46±0.10	10	0.37±0.05	0.23±0.03			
1.0-2.0	10	0.80±0.12	0.49±0.09	10	0.27±0.04	0.19±0.03			
>2.0	8	1.4 ±0.5	0.63±0.16	10	0.56±0.05	0.34±0.03			
1 September									
0.0-0.5	5	0.54±0.09	0.38±0.10	10	0.38±0.06	0.28±0.05	10	0.29±0.05	0.32±0.06
0.5-1.0	5	1.3 ±0.3	0.83±0.33	10	0.39±0.04	0.27±0.03	9	0.34±0.08	0.18±0.03
1.0-1.5	5	0.91±0.19	0.49±0.16	10	0.95±0.13	0.43±0.06	10	0.17±0.03	0.07±0.01
1.5-2.0	4	2.0 ±0.8	1.1 ±0.6	10	0.65±0.13	0.30±0.05	9	0.35±0.04	0.17±0.01
>2.0	3	2.8 ±1.1	1.2 ±0.7	10	1.0 ±0.2	0.46±0.06	9	1.4 ±0.4	0.40±0.12
27 October									
0.0-1.0	6	0.64±0.07	0.44±0.14	8	0.60±0.08	0.41±0.04	7	0.53±0.05	0.41±0.07
>2.0	7	1.3 ±0.2	0.72±0.22	7	0.96±0.14	0.52±0.09	4	0.54±0.13	0.28±0.08

Table 3. Continued.

DATE AND HEIGHT INCREMENT	FIELD 1			FIELD 2			FIELD 3		
	n	^{238}Pu	$^{239,240}Pu$	n	^{238}Pu	$^{239,240}Pu$	n	^{238}Pu	$^{239,240}Pu$
					--- Ear Samples ---				
1 September									
0.0-0.5									
0.5-1.0				1	0.01	0.01	2	0.05±0.05	0.04±0.03
1.0-1.5	2	0.04±0.03	0.02±0.01	3	0.04±0.02	0.02±0.02	7	0.03±0.03	0.06±0.04
1.5-2.0				10	0.02±0.01	0.01±0.01			
>2.0				1	0.01	0.02			
27 October									
0.0-1.0									
>1.0	1	0.02	0.02	7	0.04±0.01	0.02±0.01	5	0.06±0.02	0.05±0.02

TABLE 4. The biomass (g dry mass per ear) and the the Pu concentrations (μBq g^{-1}) of the tissues of corn ears collected from Field 2 on 27 October 27. Data are means ± standard errors for n = 4.

TISSUE	BIOMASS	CONCENTRATIONS	
		^{238}Pu	$^{239,240}Pu$
Whole Ears	128 ± 18	47 ± 17	13 ± 5
Outer Leaves	7 ± 1	680 ± 52	330 ± 24
Inner Leaves	8 ± 1	26 ± 6	13 ± 4
Cob	22 ± 3	2 ± 2	< 2
Grain	91 ± 13	1	< 1

TABLE 5. The mean inventories (mBq m^{-2}) of ^{238}Pu attributed to atmospheric deposition and resuspension on Fields 1, 2 and 3. Data are means and standard errors are computed from n = 10 samples except for Field 1 on 27 October when n = 5.

FIELD	TISSUE	SAMPLING DATE			
		15 June	1 August	1 September	27 October
		--- Atmospheric Deposition ---			
1	Leaves and stems	443 ± 177	630 ± 136	519 ± 150	171 ± 39
	Ears			1.0 ± 0.8	< 0.1
2	Leaves and Stems	33 ± 11	500 ± 60	302 ± 30	219 ± 68
	Ears			14 ± 8	14 ± 6
3	Leaves and Stems			134 ± 17	10 ± 25
	Ears			1.0 ± 0.6	3.7 ± 1.5
		--- Resuspension ---			
1	Leaves and Stems	72 ± 32	68 ± 31	56 ± 37	21 ± 12
	Ears			< 0.1	< 0.1
2	Leaves and Stems	6.3 ± 1.8	29 ± 8	8.4 ± 1.7	6.1 ± 1.9
	Ears			0.2 ± 0.1	0.5 ± 0.4
3	Leaves and Stems			11 ± 7	9.4 ± 2.8
	Ears			0.4 ± 0.2	3.8 ± 3.4

TABLE 6. Concentrations (mBq g^{-1}) of ^{238}Pu attributed to deposition for leaf and stem biomass from Fields 1, 2 and 3 for the 1 September sampling period. Data are presented for height increments of 0.5 m and the whole plant which represents a mean of the concentrations at different height increments weighted for the biomass in that increment. Data are means and standard errors for n = 5 samples for Field 1 and n = 10 samples for Fields 2 and 3.

HEIGHT INCREMENT (m)	FIELDS 1	2	3
0.0 - 0.5	0.47 ± 0.09	0.34 ± 0.06	0.21 ± 0.06
0.5 - 1.0	1.1 ± 0.3	0.36 ± 0. 04	0.30 ± 0.08
1.0 - 1.5	0.70 ± 0.13	0.94 ± 0. 16	0.17 ± 0.03
1.5 - 2.0	1.5 ± 0.7	0.65 ± 0. 13	0.30 ± 0.05
> 2.0	1.7 ± 0.9	1.0 ± 0. 2	1.3 ± 0.4
Whole Plant	0.82 ± 0.16	0.64 ± 0.06	0.31 ± 0.05

TABLE 7. The ^{238}Pu and $^{239,240}Pu$ concentrations ($\mu Bq\ g^{-1}$) in mechanically-harvested grain and the partitioning of the ^{238}Pu concentrations into components due to atmospheric deposition and resuspension for Fields 1, 2 and 3. The ^{238}Pu concentrations were partitioned using the isotopic ratios of ^{238}Pu to $^{239,240}Pu$. Data are means ± 1 standard error for n = 5.

FIELD	CONCENTRATIONS		PARTITIONING OF ^{238}Pu CONCENTRATIONS	
	^{238}Pu	$^{239,240}Pu$	Deposition	Resuspension
1	2.4 ± 0.2	0.76 ± 0.22	2.4 ± 0.2	0.0[a]
2	1.1 ± 0.1	0.58 ± 0.10	1.1 ± 0.1	0.1
3	1.7 ± 0.3	0.61 ± 0.11	1.7 ± 0.3	0.0[a]

[a] No detectable contribution from resuspension

Table 8. Estimates of parameter values for the Pu transfer model presented in Figure 1. The model parameters are: 1) I_v, the fraction of the yearly deposition that is retained on the vegetation at harvest; 2) T_{vg}, the fraction of the Pu on vegetation attributed to atmospheric deposition that is transferred to grain during combining; 3) U_{sg}, the fraction of the soil inventory absorbed by roots and translocated to grain; and 4) R_{sg}, the fraction of the soil inventory of Pu resuspended to grain surfaces. The parameter values for wheat and soybeans are the means ± standard errors for values computed for data from Fields 1 and 2 (Pinder et al., 1989. The values for corn represent means and standard errors of values from Fields 1, 2 and 3.

PARAMETER	CROP		
	Wheat	Soybeans	Corn
I_v	0.030 ± 0.001	0.020 ± 0.013	0.037 ± 0.011
T_{vg}	0.0045 ± 0.0002	0.0021 ± 0.0001	0.0010 ± 0.0001
U_{sg}	$9.9\ (\pm\ 0.3) \times 10^{-11}$	$5.7\ (\pm\ 0.2) \times 10^{-10}$	$7.3\ (\pm\ 4.1) \times 10^{-10}$
R_{sg}	$1.6\ (\pm\ 0.1) \times 10^{-7}$	$7.4\ (\pm\ 2.3) \times 10^{-8}$	$6.2\ (\pm\ 6.2) \times 10^{-9}$

Figure 1. A model of Pu transport in agroecosystems involving processes occurring while the crop is growing in the field and processes occurring when the crop is mechanically harvested using a combine. Transport processes in the field include interception and retention of atmospheric deposition and root uptake and translocation of Pu to grain. Processes occurring during the combining include transfer of Pu-bearing particles from deposition from the surface of the vegetation to the surface of the grain. Resuspension, which is indicated as a single vector, probably involves two separate processes. these are described in the text. Atmospheric deposition is expressed as Bq m^{-2} y^{-1}, and the soil inventory and quantity of Pu harvest are expressed as Bq m^{-2}. The effects of radioactive decay for ^{239}Pu are negligible and are omitted from the model. The model is defined by four parameters including: 1) I_v, the fraction of the yearly deposition retained on vegetation surfaces at harvest; 2) T_{vg}, the fraction of Pu on vegetation surfaces transferred to grain during combining; 3) U_{sg}, the fraction of the soil inventory absorbed by roots and translocated to grain; and 4) R_{sg}, the fraction of the soil inventory of Pu resuspended to grain.

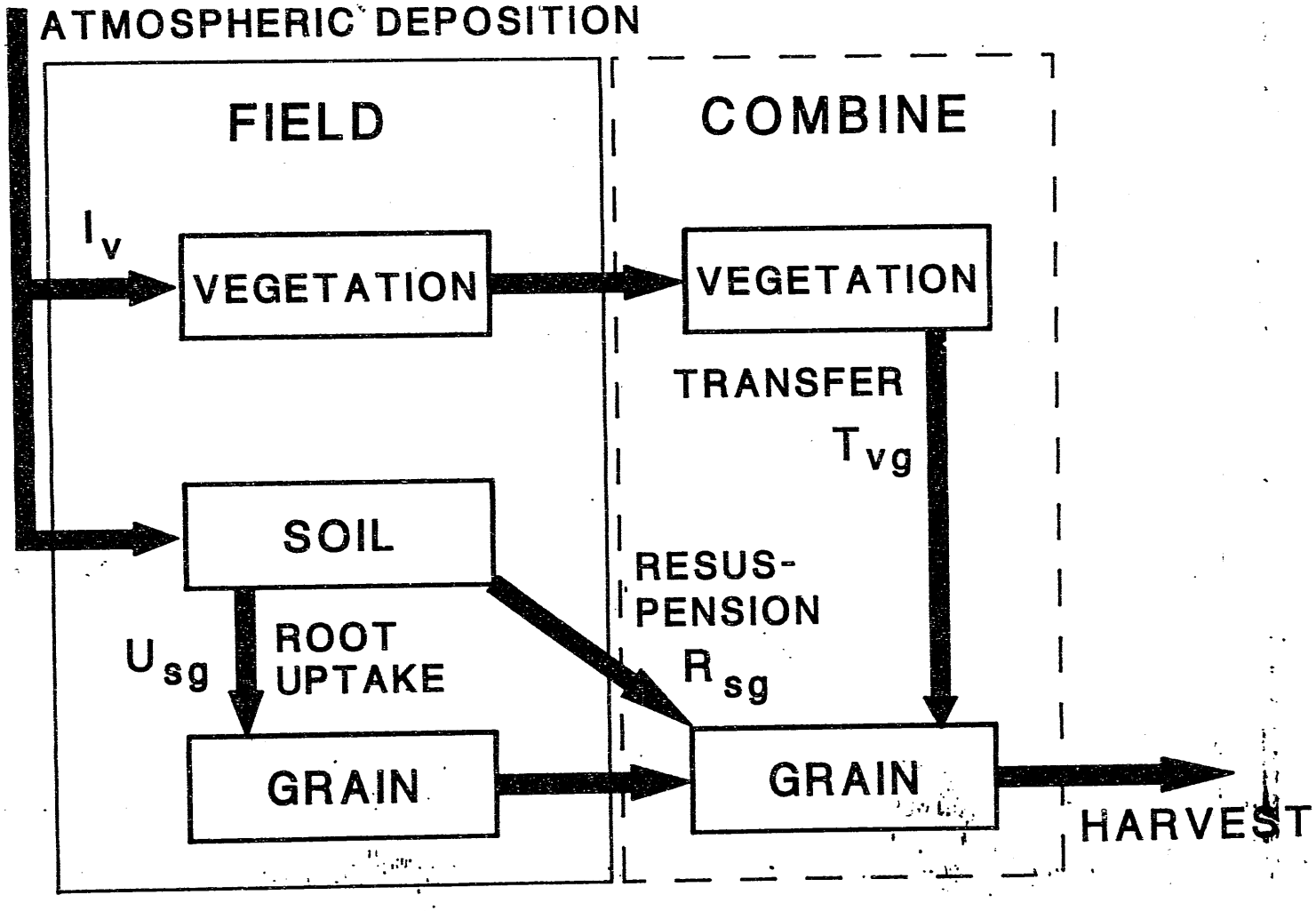

ATMOSPHERIC DEPOSITION
FIELD
COMBINE
I_v
VEGETATION
VEGETATION
TRANSFER
T_{vg}
SOIL
RESUS-
PENSION
R_{sg}
U_{sg}
ROOT
UPTAKE
GRAIN
GRAIN
HARVEST

Figure 2. The distribution of the inventories (mBq m^{-2}) of ^{238}Pu attributed to atmospheric deposition and resuspension in the canopy of corn sampled on 1 September from Fields 1, 2 and 3. Note the changes in scales among fields and between deposition and resuspension. Data are means (vertical bars) ± standard error (horizontal bars) for n = 5 samples from Field 1 and n = 10 samples from Fields 2 and 3.

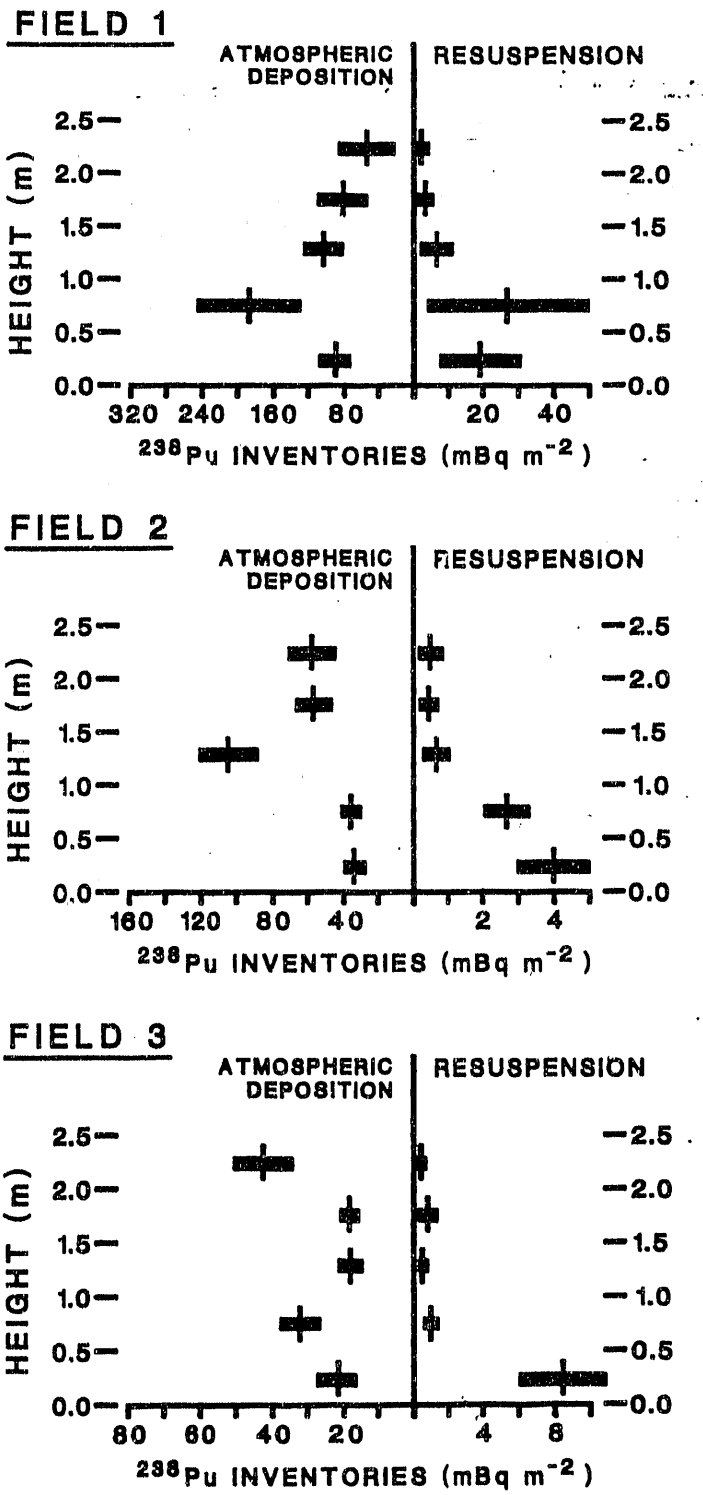

FIELD 1
ATMOSPHERIC DEPOSITION
RESUSPENSION
HEIGHT (m)
2.5
2.0
1.5
1.0
0.5
0.0
320 240 160 80
20 40
238Pu INVENTORIES (mBq m-2)
FIELD 2
ATMOSPHERIC DEPOSITION
RESUSPENSION
HEIGHT (m)
160 120 80 40
2 4
238Pu INVENTORIES (mBq m-2)
FIELD 3
ATMOSPHERIC DEPOSITION
RESUSPENSION
HEIGHT (m)
80 60 40 20
4 8
238Pu INVENTORIES (mBq m-2)

Figure 3. The ^{239}Pu concentrations for corn grains due to atmospheric deposition, resuspension and root uptake for a hypothetical scenario where a nuclear facility operates for 30 years and deposits either 100 mBq ^{239}Pu m^{-2} d^{-1} (Fig. A) or 1 mBq^{239}Pu m^{-2} d^{-1} (Fig. B) onto fields already containing 75 Bq $^{239}Pu \cdot m^{-2}$ due to global fallout. Predictions are computed using the model depicted in Figure 1 and parameter estimates from Table 8.

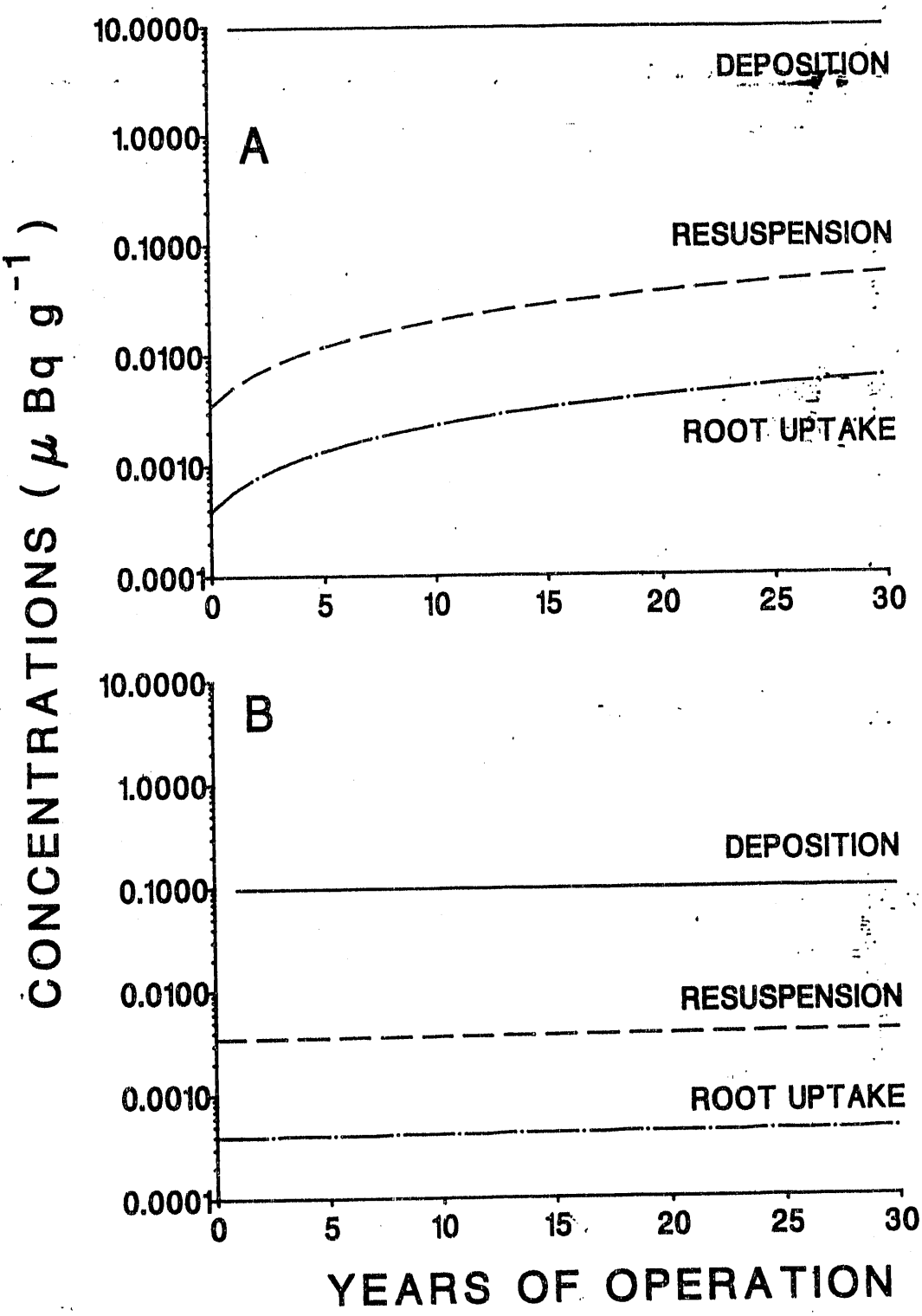
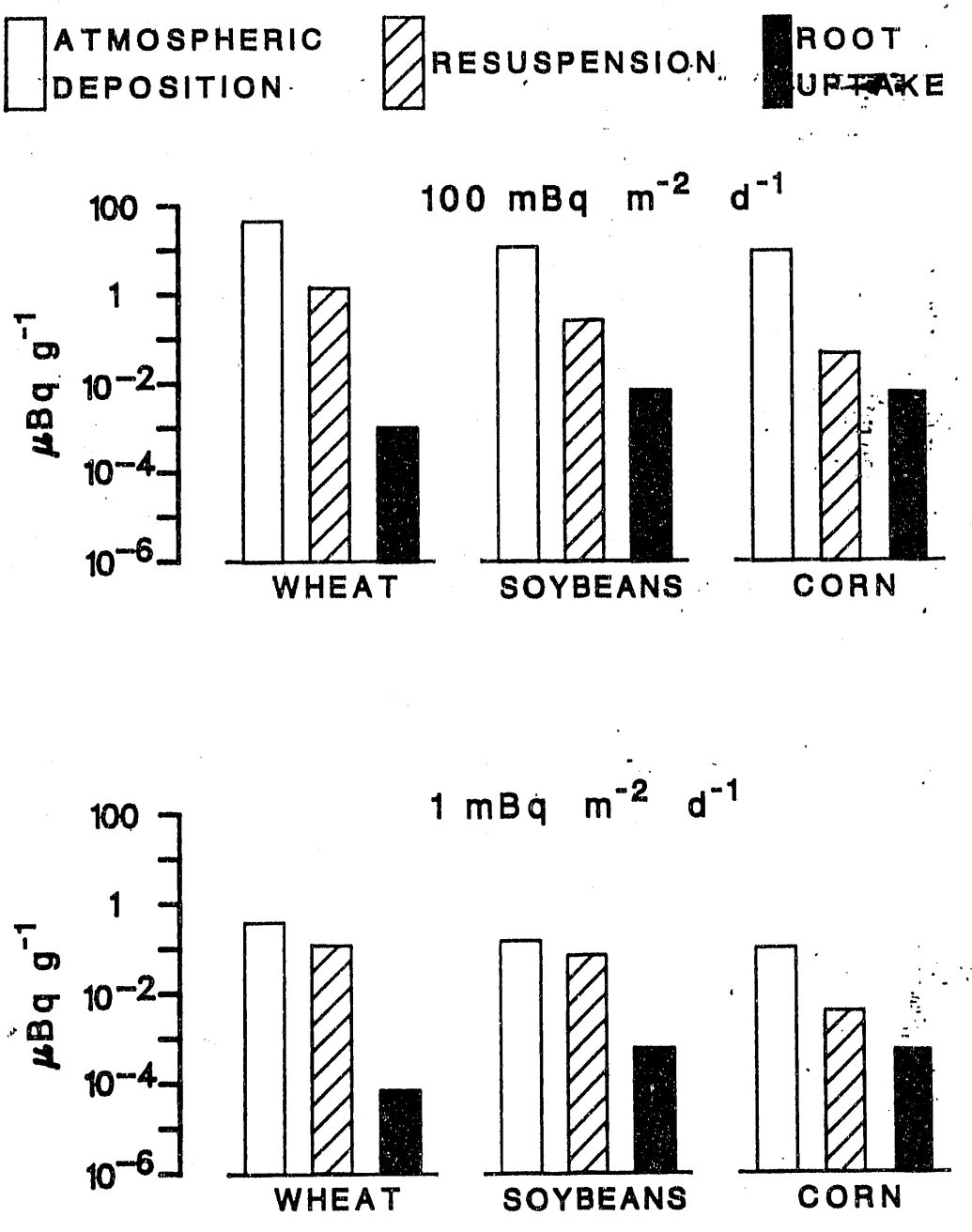

10.0000
1.0000
0.1000
0.0100
0.0010
0.0001
A
DEPOSITION
RESUSPENSION
ROOT UPTAKE
0
5
10
15
20
25
30
B
DEPOSITION
RESUSPENSION
ROOT UPTAKE
CONCENTRATIONS (μ Bq g⁻¹)
YEARS OF OPERATION

Figure 4. The ^{239}Pu concentrations for grains from wheat, soybean and corn agroecosystems as a result of atmospheric deposition, resuspension and root uptake for a hypothetical scenario where a nuclear facility operates for 30 years and deposits either 100 or 1 mBq^{239}Pu m^{-2} d^{-1} onto fields already containing 75 Bq ^{239}Pu m^{-2} due to global fallout. Predictions are computed using the model depicted in Figure. 1 and parameter estimates from Table 8. Predictions are presented for the 30th year of operation.


ATMOSPHERIC DEPOSITION
RESUSPENSION
ROOT UPTAKE
100 mBq m-2 d-1
μBq g-1
100
1
10-2
10-4
10-6
WHEAT
SOYBEANS
CORN
1 mBq m-2 d-1
μBq g-1
100
1
10-2
10-4
10-6
WHEAT
SOYBEANS
CORN

END

DATE
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