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PLUTONIUM CONTENTS OF FIELD CROPS IN THE SOUTHEASTERN UNITED STATES *

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INTRODUCTION

Research on the transuranic nuclides in the environment has gained momentum in recent years because of the proposed increase in production and utilization of Pu in the nuclear fuel cycle. Because of high toxicity and long half-life of Pu, its health hazard to man should be evaluated. Although inhalation intake has been considered the major pathway by which Pu reaches man (Bennett, 1976), ingestion of Pu-contaminated foodstuff through the soil-to-plant pathway should be critically evaluated because of Pu's long persistence and general immobility in the environment (Francis, 1973). Numerous studies have been conducted to determine plant uptake of Pu, both under field and controlled conditions (Francis, 1973; Price, 1972; Hanson, 1975; Bennett, 1976). However, most of these experiments were done in pot culture and when experiments were done in the field only non-agricultural vegetation was involved. This general lack of information on field grown crops is apparent from the proceedings of the 1976 International Symposium on the Transuranium Nuclides in the Environment held in San Francisco (IAEA STI/PUB/410).

The main purpose of this paper is to compare Pu contents of crops grown in two different natural environments in the southeastern United States contaminated by low levels of Pu. The fields at the Savannah River Plant near Aiken, South Carolina have been receiving Pu at low chronic levels from an emission stack of a reprocessing facility since 1955, while the field plots at the Oak Ridge National Laboratory are located at the White Oak Creek bed which received its Pu from the Manhattan Project Operations in 1944.

SITE DESCRIPTION:

The Savannah River Plant

The Savannah River Plant (SRP) site is located on the upper Coastal Plain in Aiken, Allendale and Barnwell counties, South Carolina. The site is bordered on the southwest by the Savannah River, and is about evenly divided between two distinct physiographic subregions: the Pleistocene Coastal Terraces (below 82 m in elevation) and the Aiken Plateau (above 82 m and rising to 121 m on the southwest boundary). The Coastal Terraces include the "low terrace" broad floodplain of the Savannah River covered with a dense swamp forest and the "high terrace" having a level-to-gently rolling topography. The latter was extensively cultivated before the establishment of SRP in those areas where soils and drainage were favorable. The Aiken Plateau is quite hilly and deeply dissected by small streams. Soils in the Aiken Plateau are mostly sandy and/or gravelly and low in fertility. The Coastal Terraces subregion soil has more clay with greater productivity potential.

SRP is on a reservation of 77,830 hectares, with public access controlled since its acquisition in 1951. The reservation consists of freshwater streams, old fields, and forest with most of the old fields in the upper Coastal Terraces. Wildlife, including a deer herd of about 6,000, abounds.

For over 20 years, this integrated nuclear complex has included nuclear reactors (three of the original five are operating at present), two nuclear fuel reprocessing plants, a fuel fabrication facility, a heavy water production unit, and nuclear and environmental research laboratories (Fig. 1).

It also includes an ecological research laboratory to assess the effects of nuclear technology on the environment and its biota. The reprocessing plants (F and H), along with global fallout, are the sources of the transuranic elements that enter the SRP environs. Each source releases Pu of unique isotopic composition: 25 and 95 α % ^{238}Pu[1/] from the F and H plants, respectively, and 10 α % ^{238}Pu from global fallout. These isotopic differences provide a convenient basis for studying the origin and transport of Pu in various SRP ecosystems.

An agricultural study area was established adjacent to the H reprocessing plant in 1975. The relatively flat field was divided into two sections, the South Field (145 m x 30 m) and the North Field (105 m x 30 m), with a 40 - m buffer zone between them. The fields lie northeast of a 62 - m high emission stack, where small quantities of Pu (< 3.6 Ci) have been released to the atmosphere since its operation began in 1955. The nearest point of the South Field is about 170 m away from the stack. The reddish soils are well drained with textures ranging from sandy clay loam to sandy loam and are slightly acidic, with pH generally between 5 and 6.

The climate of the SRP area consists of mild winters and long, warm, humid summers. Temperatures average about 9°C in the winter and 30°C in the summer. The average annual temperature is 18°C. The average annual relative humidity and rainfall are 70% and 120 cm, respectively. The maximum annual precipitation recorded was 187 cm in 1929, the minimum 71 cm in 1933 (Langley and Marter, 1973).

1/ $^{238}Pu = \frac{^{238}Pu \text{ alpha activity}}{\text{Total Pu alpha activity}} \times 100$

White Oak Creek Floodplain, Oak Ridge:

Manhattan Project Operations in 1944 produced treated wastes containing Pu, Am and Cm. Following soda ash treatment to precipitate various ions, the liquid effluents were released to White Oak Creek (WOC). Trace quantities of ^{238}Pu, ^{239}Pu, ^{241}Am, and ^{244}Cm were deposited along the water course, in an intermediate retention pond, and finally in White Oak Lake. Deposition of these elements in sediments of a retention pond occurred over a 6-month period in 1944. From the time the pond was drained in late 1944, no additional radioactivity is believed to have been deposited at this location of the WOC floodplain.

The alluvial soil of the floodplain is representative of bottomlands of the Tennessee Ridge and Valley Province. Azonal characteristics predominate because of concurrent erosion and deposition of materials during periodic floods. The soil profile remains relatively undeveloped although accumulation of humus is evident from a dark brown coloration in the 0- to 3- cm zone.

As a temporary holding basin 30 years ago, sedimentation during the 6 months of impoundment contributed new sedimentary materials. A Tennessee Valley Authority (personal communication, T. C. Bounds) survey in 1951 reported that approximately 2,100 m^3 of sediment had been deposited at this locality. Distributed uniformly over the research site, this volume represented an increment of approximately 9 cm of new sediment. However, equal deposition was unlikely, and the exact history of sedimentation associated with the 1944 impoundment is unknown.

Textural analysis indicate that soils are silty loam (72% silt, 24% clay) containing almost no sand or gravel (Tamura, 1976). While this

texture is representative of the floodplain, isolated gravel lenses occur irregularly across the floodplain and within the soil profile.

The soil reaction is neutral to mildly alkaline as pH ranges from 7.1 to 7.6. The mild alkalinity is probably caused by the soda ash method of waste treatment employed before effluents were released from laboratory operations. Data have not been obtained on base exchange or soil fertility, although on the basis of plant growth and crop performance, the site possesses high fertility potential.

RESULTS AND DISCUSSION

Field Crops

The Savannah River Plant:

Wheat: The South Field was divided into 18 equal plots and the North Field into 12 equal plots in the fall of 1975. The fields were limed, cultivated, fertilized, and planted to wheat following standard agricultural methods. Resuspendible* and soil samples were collected prior to and following land preparation. Foliage samples were collected in March and April. At harvest in June, plants were separated into grain and straw. Wheat grain samples were obtained by two techniques. The first approach was to carefully collect wheat heads by hand from each of the 30 plots, and separate the grain with a laboratory thrashing machine. This machine was carefully cleaned between samples from each plot to minimize the amount of dust that would adhere to the grain. The second

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* Resuspendible matter (0-0.1 cm depth) was collected in filter bags using a portable vacuum cleaner with suction velocity at 6 m/sec. The entire sample was used for analysis.

approach was to harvest the grain with a tractor-pulled combine. Grain collected by this method was exposed to the usual soil and dust stirred up by harvest activities. Total biomass for the crop (grain plus hull plus straw) was estimated as 3,615 and 3,545 kg/hectare for the South and North Fields, respectively. Wheat was also grown 37 km from the reprocessing plants, as a control.

Data from the Pu analyses are summarized in Table 1. The results are separately presented for the South and North Fields because of significant differences in Pu concentrations in the resuspendible matter and top soil and potentially different deposition patterns from emission stack on the two fields. There appears to be no significant changes in the Pu concentrations of wheat foliage with time on either the South or the North Field. The Pu content averaged 4×10^{-2} pCi/g dry weight of foliage or straw.

The isotopic composition of the foliage, as indicated by the % ^{238}Pu, changed significantly with time (Table 1). In March the wheat plants were short and the foliage not yet dense. At this stage, it appeared that contamination of the foliage primarily originated from the soil, as in rain splash or resuspendible matter. As the plants grew taller and the foliage became denser they were able to intercept fallout particles more efficiently. Also the dense foliage minimized rainfall energy upon impact on the ground. In April and June the foliage had much higher ^{238}Pu as compared with March samples, suggesting that contamination primarily originated from ~~fallout particles from the~~ current stack emission~~s stack~~. The Pu concentration of straw was approximately 300 times greater than that of the laboratory-threshed grain and 40 times greater than that of the field-combined grain.

The lower grain levels compared to the straw values indicate the possibility that the grain is shielded from atmospheric deposition. The Pu concentrations in foliage, straw, and grain samples were slightly higher in the South Field.

The straw from off-plant control site had only $(8 \pm 2) \times 10^{-4}$ pCi/g (data not shown), about two orders of magnitude lower than SRP field samples. The thrashed grain from the off-site, however, has the same Pu contents as the SRP thrashed samples. However, the high α percentage of 62 ± 15 for off-site thrashed grain should be noted, which is disparate from the straw value. This high value apparently was caused by cross-contamination during thrashing of the SRP samples, since all thrashed samples had similar values.

The C. R.'s for the June straw were 5×10^{-2} and 1×10^{-1} for the South and North Fields, respectively. Similar values (1×10^{-1}) were obtained for off-plant samples. The C. R. for the combined grain averaged 2×10^{-3} from the two fields, whereas, the thrashed grain averaged 2×10^{-4}.

In Table 2, Pu contents of field- and glasshouse-grown wheat plants were compared. The soils used in the glasshouse study came from the surface layer (0-20 cm) of the South Field and had 1.96 pCi total Pu/g dry weight and 21 α % ^{238}Pu. Thus, only the South Field data are presented for comparison with glasshouse data. The glasshouse-grown plants had total Pu contents one order of magnitude less than the field-grown plants.

¶ The combined field-grown grain was sifted using a soil screen, to evaluate the effectiveness of removing soil particles on reducing the Pu contents. Data from Tables 1 and 2 for the unsifted combined grain are remarkably

similar. The sifted combined grain had a factor of two less total Pu content than the unsifted grain.

Soybeans: The soybean data from the field and glasshouse are presented in Table 2. Total Pu concentrations in glasshouse-grown plants were an order of magnitude less than field-grown plants. The C. R. for plants grown in the glasshouse was a factor of 10 lower than plants grown in the field. The glasshouse-plants (whole parts) were sampled when the bean pods were still green and before the plants started defoliating. The field-grown plants were sampled approximately 1 month before the field was combined, the period when the plants had the maximum biomass. The combined grain had two orders of magnitude lower Pu content than the vegetative parts.

The bean plants had similar Pu concentrations (10^{-2} pCi/g dry wt.) and α percentages as the wheat straw (whole plants without the grain). Consequently, the two crops had similar concentration ratios. However, the soybean grain had slightly lower Pu contents than the wheat grain.

Corn: Plutonium contents of field- and glasshouse-grown corn are presented in Table 2. Corn leaves were sampled from the field when the plants had their maximum biomass. The leaves were sampled from two heights: 0-1 m from the ground and 1-2 m (or 1 m to the top). Plants in the glasshouse were sampled when the ears had matured. Leaves of glasshouse-grown corn had total Pu contents one order of magnitude lower than those of field-grown corn. Leaves from the 1-2 m section of the corn plants had Pu

contents almost a factor of 2 higher than leaves from the lower section (0-1 m). This suggests that the upper foliage partially filters and retains Pu-bearing particles from atmospheric deposition.

Shelled corn from the glasshouse study had slightly lower Pu concentrations than the field-combined grain (Table 2). However, the grain from the glasshouse had a C. R. an order of magnitude lower than the field-combined grain. The unsifted combined grain had a factor of 2 higher total Pu contents than did the sifted. Apparently, the sifting separated approximately 50% of Pu-bearing particles and/or foreign matter from the grain.

Relative Contribution of Root Uptake to Plutonium Contents of Field Crops: The comparison of the data on crops grown in the field and in the glasshouse (Table 3) provides an estimate of the plutonium uptake via the root pathway. The results indicate that Pu contamination of the field crops is primarily external. For wheat and corn samples, approximately 97% of the total contamination was external. For soybeans, contribution from root uptake appeared greater than 10%. No plausible explanation can be offered at this time for this discrepancy.

White Oak Creek Floodplain, Oak Ridge:

Forage and Vegetable Crops: Forage and vegetable crops harvested from a field plot located on the contaminated floodplain contained measurable quantities of Pu. Concentrations of ^{239,240}Pu in foliage of bushbean, soybean, tomato, lettuce, radish and millet ranged from 0.01 to 0.33

pCi/g dry weight (Table 4). Concentrations of Pu in fruit of these species were lower by at least an order of magnitude than the foliage concentrations. Limited data on root crops indicated that the Pu concentrations of the edible portion were similar to that of foliage.

The Pu concentrations of foliage and fruit appeared to represent internally-bound Pu because surface contamination was removed prior to radiochemical analysis. Samples of bushbean, soybean and tomato were washed and rinsed in a sonic bath. Lettuce was handwashed using representative food preparation techniques. This procedure effectively removed surface-bound Pu because, by microscopic examination, no particles were observed on any sample except soybean pods where hirsute structures effectively retained surface contaminants. The Pu concentration of soybean fruit, which included the pod, was an order of magnitude greater than beans alone. However, concentrations between whole snapbeans and the shelled beans were similar. Because surface contamination was deliberately removed from foliage and fruit, the Pu content of these vegetables is attributed to assimilation by the root pathway. Other results in support of root assimilation are discussed elsewhere (Dahlman, et. al. 1976; Dahlman and McLeod, 1977).

Radish and carrot samples were hand washed prior to radiochemical analysis. The washing techniques were similar to routine cleaning procedures that would be employed prior to table preparation or cooking of garden vegetables. Concentration of Pu in these root-vegetables ranged from 0.31 to 0.41 pCi/g. The concentration in the root of vegetables was similar to that of the foliage. Although surfaces of radish and

carrot samples were not examined microscopically for the presence of particles, the tissue contained Pu concentrations equivalent to those of foliage which had been vigorously cleaned. Hence, the root vegetables were considered to be relatively free of soil particles.

Peeling the skins of potatoes and beets removed most of the internally-bound plutonium. Potato skin averaged 0.53 pCi/g as compared to 0.006 pCi/g for peeled potatoes. This indicates that about 99% of the Pu was removed by peeling. Beets skins had even greater concentrations of Pu, 1.3 pCi/g for skin versus 0.005 pCi/g for peeled beets. Thus, peeling beet skin removed about 99.5% of total Pu in the beet.

The uptake pattern for the soybean plants at the WOC Floodplain is presented in Table 5. The Pu concentration in the foliage after 8 weeks of growth was only 0.02 pCi/g dry weight. By the 12th week, the Pu concentration had increased five times (0.10 pCi/g) and increased ten times (0.19 pCi/g) by the 16th week. Thus, the increase in Pu concentration is related to the extent of the root system and nutrient requirement. The foliage biomass was at maximum at around the 16th week. Correspondingly, the C. R. of the foliage increased by an order of magnitude from the 8th to the 16th week.

Dose-to-Man from Ingestion of Plutonium-Contaminated Foodstuff: The dose to a person from ingesting wheat or other foods (Tables 2 and 4) can be estimated using the nomograph in Fig. 2. For example, if a person ate 2×10^5g of wheat grain containing 2 fCi Pu/g dry grain, the 70-year bone dose commitment would be ~0.5 mrem. Likewise, if a person ate 2×10^5g of peeled Irish potatoes (approximately equivalent to 2,000 kg

wet potatoes) containing 6 fCi Pu/g dry weight, the 70-year dose-to-bone would be ~ 1.0 mrem. These doses are low compared with the 130 mrem dose from background radiation (Klement, et. al., 1972). The basic data used for these calculations come from ICRP publications (ICRP 2, 19 & 20).

Soils

The Savannah River Plant:

As with plant materials, the resuspendible matter and soils from the South Field (closer to the stack) had higher Pu concentrations than the North Field (Table 6). The differences in concentrations are more pronounced in the resuspendible matter where the South Field had Pu levels one order of magnitude higher than the North Field. Soils (0-5 cm) from both fields had Pu levels about three orders of magnitude higher than off-plant soils. Cultivation of the fields result in more uniform Pu concentrations in the soil profile, increasing the Pu at the deeper depths (5-15 cm). Cultivation also altered the isotopic composition of the resuspendible portion to approach the soil values. After the initial cultivation, the α percentage for the resuspendible matter decreased from an average of 50 to an average of 27 for the two fields.

White Oak Creek Floodplain, Oak Ridge:

Concentrations of Pu in the floodplain soil generally ranged from 10 to 150 pCi/g dry weight over a 3-ha area (Fig. 3). The highest concentrations occur behind the former dike, along WOC and at the upper end of the floodplain. Concentrations of Pu are less than 25 pCi/g near the margins of the floodplain. The ^{239}Pu concentration of soil at a garden plot site (W35, S295) was 63 $\pm$ 0.4 (S.E.) pCi/g.

A typical profile distribution of Pu shows that the highest concentration of Pu is observed in the uppermost zone of the profile (Duguid, 1975). Occasionally, however, where higher concentration of Pu occurs in the subsoil, this atypical distribution is attributed to variable deposition of sediments over an initial Pu deposit rather than Pu cycling by leaching processes.

Plutonium is not distributed uniformly between the silt and clay fractions of the soil. Although 72% of the soil is silt, this fraction contains 60% of the Pu; whereas the clay (24% of the soil) contains 40% of the Pu. In this case, the high affinity of Pu for colloids, the clay fraction, may be responsible for Pu enrichment in clay.

The results of various attempts to extract Pu from the soil indicate that more than 95% of the Pu can be removed by hot 8 M HNO_3; milder HNO_3 treatments removed smaller amounts of Pu (Auerbach, 1975). These treatments indicate that Pu recovery from the floodplain soil does not require rigorous treatment such as HNO_3-HF or carbonate fusion.

Even milder treatment with 0.1 M citric acid will remove 16 to 24% of the soil Pu. Contact with a CHELEX resin removes 11%. Humic acid solubilized by the presence of Na+ from a Na-saturated CHELEX resin contained 5% of the total Pu after 4 weeks. The carbonate treatment removed 54% of the soil Pu. Further analysis of the carbonate extract showed that at least 90% of the Pu behaved as Pu(IV) (Bondietti and Sweeton, 1977; Bondietti, et. al., 1976).

GENERAL DISCUSSION

The various CR values for the agricultural vegetation at SRP are generally higher than those obtained with indoor-grown plants. Schulz, et. al. (1976a, b) obtained values for barley in the order of 10^{-5} for vegetative material and grain was a factor of 20-100 lower. In wheat grain, they obtained CR values ranging from 1.1×10^{-7} to 3.8×10^{-6}. Lipton and Goldin (1976) obtained CR values for pea plants in the order of 10^{-4}. Natural plant species not subject to contamination from atmospheric releases or resuspension were observed to have CR in the order of 10^{-6} to 10^{-4} (Francis, 1973; Hanson, 1975), much lower than values obtained at SRP. However, in arid, windy environments, dust and soil particles can become airborne and may be deposited and retained in leaves causing Pu CR values to approach 10^{0} (Romney, et. al., 1975). Earlier studies at SRP indicated that deposition on the surfaces of the leaves and stems was the principal mechanism of Pu contamination of natural vegetation (Adriano and Pinder, 1977; McLendon, et. al., 1976). The Pu concentrations of all ecosystem components decreased as the distance from reprocessing plants increased (McLendon, 1975; McLendon, et. al., 1976). Thus, considerable external contamination of plants through atmospheric release and resuspension is a complicating factor in field data interpretation.

The CR values from the glasshouse studies at SRP are in the order 10^{-4} to 10^{-3} for the vegetative materials and, in general, are similar or slightly higher than the values obtained by Schulz et. al. (1976) and Lipton and Goldin (1976). Our glasshouse results suggest that ^{238}Pu appeared to be more available than ^{239}Pu, as indicated by relatively higher

^{238}Pu percentage values in the vegetative samples than in the soil. The percent ^{238}Pu ranged from 33 $\pm$ 6 for soybeans to 55 $\pm$ 11 for corn. The soils used for growing these crops had only 21% ^{238}Pu. This differential availability between the two radionuclides has been observed also at the Trinity Site ecosystem, where changing ratios of $^{238}Pu/^{239}Pu$ in soils, vegetation, and animal components were obtained (Hanson, 1975). Although the ^{238}Pu data in the various ecosystem components were not conclusive, earlier studies by McLendon, et. al. (1976) at SRP support evidence presented in other studies that there is an apparent increase in the biological availability of ^{238}Pu relative to that of ^{239}Pu in the environment. Hanson has already discussed the possible mechanisms causing this differential availability.

In general, Pu concentrations in vegetative parts of agricultural plants at SRP, where the primary factor of contamination is external, are similar to those of the forage and vegetable foliage cultured at the Oak Ridge floodplain ecosystem, where contamination was caused primarily by root uptake. However, the CR values of vegetative parts of crops at SRP were about one order of magnitude higher than those from Oak Ridge.

Plutonium data on the edible portions of root crops are almost nonexistent. Thus, it is difficult to compare the data from the Oak Ridge floodplain with anyone. Peeled potatoes grown on soils receiving only global fallout Pu had a CR of 3×10^{-4} (Bennett, 1976). This is similar to the Oak Ridge data. The Oak Ridge data suggest that peeling subterranean crops removes most of the Pu, as high as 99.5%, in the case of beets. Whether Pu in these vegetative tissues occurs as a free ion is still unknown.

SUMMARY

Agricultural crop experiments at the Savannah River Plant conducted adjacent to a reprocessing facility which releases approximately 2.4 mCi of total Pu per year through an emission stack and at the White Oak Creek floodplain at Oak Ridge where Pu was deposited in 1944 from the Manhattan Project weapon development revealed the following:

1. Pu concentrations and isotopic compositions in winter wheat grown in 1975 at SRP were affected by distance from the stack, plant height, and method of processing the grain. In general, vegetative materials closer to the stack (South Field) have Pu concentrations a factor of 2 higher than samples farther away (North Field). The straw from the wheat grown on-plant had approximately two orders of magnitude higher Pu contents than the off-plant straw. Grain harvested by a combine has an order of magnitude higher Pu content than laboratory-thrashed grain. When the plant height was short, as in March, the sources of contamination were the stack emission and the resuspendible and soil materials, as indicated by an average 38 α % ^{238}Pu. Later in the season as the plants matured and grew taller, emission became the principal source of contamination as indicated by a higher alpha percentage.

2. Pu concentrations of the foliage of soybeans and corn grown in the South Field at SRP were similar to the Pu concentrations of the June wheat straw (in the order of 10^{-2} pCi/g dry wt.) collected from the same field. However, the corn foliage had higher α % ^{238}Pu than did the wheat and soybean foliage, reflecting the minimal contribution of resuspension to the corn canopy. Pu contents of the combined grain of the three crops were

basically similar. Sifting the combined grain separated about 50% of the Pu-bearing particles and/or foreign matter from the grain, resulting in Pu concentrations in the sifted grain a factor of 2 lower than the unsifted grain.

3. Preliminary results from the SRP glasshouse studies indicate that approximately 3% of the total contamination of field-grown crops adjacent to a reprocessing facility was contributed by root uptake. Also it appeared from glasshouse results that ^{238}Pu was more available than ^{239}Pu.

4. Cultivation of the fields at SRP prior to planting wheat caused the Pu concentrations in the soil profile to become more uniform, increasing the Pu concentrations at the deeper depths (5-15 cm). It also caused the α percentage for the resuspendible portion to approach the soil values. Subsequent cultivations caused greater uniformity in Pu concentration in the top soil.

5. Concentrations of $^{239,240}Pu$ in the foliage of forage and vegetable crops grown at Oak Ridge White Oak Creek floodplain ranged from 0.01 to 0.33 pCi/g. This represents the amounts of Pu taken up by plants exclusively via the root pathway from soils with Pu concentrations ranging from 10 to 150 pCi/g and averaging 63 pCi/g. [redacted] Pu concentrations in fruit of these species were lower by at least an order of magnitude than the foliage concentrations.

6. Peeling the skins of potatoes and beets grown at WOC floodplain removed approximately 99% of the internally-bound Pu.

7. Pu contents of soybean foliage was related to the maturity stages, maximum when the foliage biomass was at maximum.

8. At the WOC floodplain, silt constitutes 72% of the soil mass, and clay only 24%. However, 40% of Pu is bound to the clay fraction, suggesting the high affinity of Pu for colloids. At least 95% of this Pu can be removed by hot 8 M HNO_3.

9. In general, the CR values of vegetative parts of crops at SRP were about one order of magnitude higher than those at Oak Ridge, suggesting the influence of aerial deposition of Pu at the SRP site.

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Table 1. Plutonium concentration and isotopic composition of wheat grown near a reprocessing facility (H-Area) at SRP.

	South Field				North Field			
	Sample n	Total Pu pCi/g	% Pu-238	CR	Sample n	Total Pu pCi/g	% Pu-238	CR
Wheat:								
Foliage, March	6	$(5.2 \pm 1.0) \times 10^{-2}$	38 ± 4	3×10^{-2}	4	$(2.0 \pm 0.60) \times 10^{-2}$	38 ± 2	6×10^{-2}
Foliage, April	18	$(2.1 \pm 0.42) \times 10^{-2}$	61 ± 3	1×10^{-2}	12	$(9.8 \pm 1.2) \times 10^{-3}$	66 ± 3	3×10^{-2}
Straw, June	18	$(8.8 \pm 1.5) \times 10^{-2}$	58 ± 2	5×10^{-2}	12	$(3.9 \pm 1.8) \times 10^{-2}$	55 ± 3	1×10^{-1}
Grain (combined)	8	$(2.0 \pm 0.64) \times 10^{-3}$	56 ± 4	1×10^{-3}	7	$(0.9 \pm 0.19) \times 10^{-3}$	55 ± 5	3×10^{-3}
Grain (thrashed)	18	$(3.0 \pm 0.52) \times 10^{-4}$	60 ± 3	2×10^{-4}	12	$(1.4 \pm 0.43) \times 10^{-4}$	64 ± 6	3×10^{-4}

Values are means $\pm$ 1 standard error

Table 2. Plutonium concentration and isotopic composition of vegetative parts and grain of crops grown near a reprocessing facility (H-Area) and glasshouse at SRP. The soils used in the glasshouse were from the top layer of the south field.

	Sample n	South Field[1] Total Pu, pCi/g	% Pu-238	CR	Sample n	Glasshouse[2] Total Pu, pCi/g	% Pu-238	CR
Wheat:								
Straw, June	18	$(8.8\pm1.5)\times10^{-2}$	58 ± 2	6×10^{-2}	10	$(3.0\pm0.47)\times10^{-3}$	41 ± 2	2×10^{-3}
Grain, combine-unsifted	7	$(3.0\pm0.98)\times10^{-3}$	50 ± 6	2×10^{-3}	--	--	--	--
Grain, combine-sifted	6	$(1.6\pm0.86)\times10^{-3}$	47 ± 4	1×10^{-3}	--	--	--	--
Soybeans:								
Vegetative, whole plants	14	$(5.2\pm0.45)\times10^{-2}$	53 ± 2	3×10^{-2}	10	$(5.6\pm0.51)\times10^{-3}$	33 ± 2	3×10^{-3}
Grain, combine	9	$(5.5\pm1.3)\times10^{-4}$	41 ± 3	3×10^{-4}	--	--	--	--
Corn:								
Leaves, 0-1 m height	10	$(2.5\pm0.35)\times10^{-2}$	63 ± 3	2×10^{-2}	6	$(1.1\pm0.16)\times10^{-3}$	55 ± 4	6×10^{-4}
Leaves, 1-2 m height	10	$(4.2\pm0.57)\times10^{-2}$	70 ± 1	3×10^{-2}	--	--	--	--
Grain, combine-unsifted	5	$(1.7\pm0.31)\times10^{-4}$	63 ± 2	1×10^{-4}	6	$(1.2\pm0.33)\times10^{-4}$	76 ± 2	6×10^{-5}
Grain, combine-sifted	5	$(8.5\pm1.1)\times10^{-5}$	78 ± 4	5×10^{-5}	--	--	--	--

[1] The total Pu content of 1.61 pCi/g dry wt. of the 0-15 cm soil depth for the South Field for the wheat crop was used. This same value was used for the soybean and corn calculations.

[2] The glasshouse soils had the following average contents: 1.96 pCi total Pu/g dry wt. (n=9) and 21% Pu-238 (n=9). From the glasshouse, the following samples were used: wheat - whole plant without grain, soybeans - whole plant without grain; and corn - leaves from the whole plant and hand-shelled grain.

[3] The same combined grains were sifted through a soil screen.

Table 3. Comparison of total plutonium contents (fCi/g dry wt.) of vegetative materials from crops grown on a field (South Field) near a reprocessing facility (H-Area) and in a glasshouse at SRP. The soils used in the glasshouse were from the top layer of the South Field.

Vegetative Material	South Field	Glasshouse	Difference	% External Content[1]
Wheat straw	88.0	3.0	85.0	97
Soybeans, whole plants	52.2	5.6	46.6	89
Corn leaves[2]	33.5	1.1	32.4	97

[1]Calculated from the equation:

$$\frac{\text{Field content} - \text{Glasshouse content}}{\text{Field content}} \times 100$$

[2]Fiberglass mats were placed on top of the soil in the corn pots in the glasshouse to prevent plant contamination by resuspension.

Table 4. Plutonium concentration and concentration ratio (CR) of field crops from the Oak Ridge WOC Floodplain.

Crop	Sample n	Total Pu[a] pCi/g	CR[b]
Foliage:			
Bushbean	8	$(1.1 \pm 0.3) \times 10^{-1}$	2×10^{-3}
Soybean	24	$(1.0 \pm 0.2) \times 10^{-1}$	2×10^{-3}
Tomato	3	$(3.3 \pm 0.7) \times 10^{-1}$	5×10^{-3}
Lettuce	3	$(8.0 \pm 2.0) \times 10^{-2}$	1×10^{-3}
Radish	7	$(1.0 \pm 0.5) \times 10^{-1}$	2×10^{-3}
Millet	14	$(1.0 \pm 0.3) \times 10^{-2}$	2×10^{-4}
Fruit			
Bushbean,			
Whole bean	7	$(7.0 \pm 2.0) \times 10^{-3}$	1×10^{-4}
Shelled bean	5	$(1.0 \pm 0.4) \times 10^{-2}$	2×10^{-4}
Tomato	5	$(4.0 \pm 2.0) \times 10^{-3}$	6×10^{-5}
Soybean,			
Whole bean	7	$(1.5 \pm 0.3) \times 10^{-2}$	2×10^{-4}
Shelled bean	3	$(1.7 \pm 1.2) \times 10^{-2}$	3×10^{-4}
Subterranean:			
Radish	5	$(4.1 \pm 0.6) \times 10^{-1}$	6×10^{-3}
Carrot	1	3.1×10^{-1}	5×10^{-3}
Irish Potato,			
whole	3	$(8.0 \pm 1.0) \times 10^{-2}$	1×10^{-3}
peeled	2	$(6.0 \pm 1.0) \times 10^{-3}$	1×10^{-4}
skin only	4	$(5.3 \pm 1.2) \times 10^{-1}$	8×10^{-3}
Beet,			
peeled	3	$(5.0 \pm 1.0) \times 10^{-3}$	8×10^{-5}
skin only	2	$(1.3 \pm 0.5) \times 10^{0}$	2×10^{-2}

[a]Values are means ± 1 standard error which includes analytical error of approximately 10%.

[b]CR values are based on soil concentration of 63 ± 0.4 (SE) pCi/g.

Table 5. Plutonium concentration and concentration ratio (CR) for soybean foliage harvested at different stages of growth (ORNL WOC Floodplain).

Growth Stage	Sample n	Total Pu pCi/g	CR[a]
8 weeks	8	$(2.0 \pm 0.4) \times 10^{-2}$	3×10^{-4}
12 weeks	8	$(1.0 \pm 0.3) \times 10^{-1}$	2×10^{-3}
16 weeks	8	$(1.9 \pm 0.4) \times 10^{-1}$	3×10^{-3}

[a]CR values are based on soil concentration of 63 ± 0.4 (SE) pCi/g.

Table 6. Plutonium concentration and isotopic composition of soils in an agricultural study area adjacent to a reprocessing facility at SRP as affected by cultivation.

	SOUTH FIELD			NORTH FIELD		
	Sample n	Total Pu, pCi/g	% Pu-238	Sample n	Total Pu, pCi/g	% Pu-238
Prior to cultivation						
Suspendible	18	$(1.3 \pm 0.38) \times 10^{1}$	51 ± 2	12	$(1.6 \pm 0.26) \times 10^{0}$	49 ± 3
0-5 cm	21	$(3.2 \pm 0.79) \times 10^{0}$	29 ± 3	14	$(1.0 \pm 0.10) \times 10^{0}$	19 ± 1
6-15 cm	18	$(1.2 \pm 0.28) \times 10^{-1}$	30 ± 2	12	$(9.4 \pm 1.8) \times 10^{-2}$	20 ± 2
After 1st cultivation						
Suspendible	18	$(4.8 \pm 0.57) \times 10^{0}$	28 ± 1	12	$(8.5 \pm 1.1) \times 10^{-1}$	27 ± 2
0-5 cm	21	$(3.0 \pm 0.59) \times 10^{0}$	30 ± 3	14	$(6.3 \pm 0.48) \times 10^{-1}$	29 ± 2
6-15 cm	18	$(9.2 \pm 3.4) \times 10^{-1}$	22 ± 2	12	$(1.7 \pm 0.40) \times 10^{-1}$	25 ± 3
After 2nd cultivation						
Suspendible	18	$(4.6 \pm 0.69) \times 10^{0}$	34 ± 2	12	$(9.3 \pm 1.4) \times 10^{-1}$	30 ± 1
0-5 cm	18	$(4.4 \pm 1.1) \times 10^{0}$	31 ± 3	12	$(7.0 \pm 1.0) \times 10^{-1}$	28 ± 3
6-15 cm	18	$(4.7 \pm 1.5) \times 10^{-1}$	30 ± 3	11	$(3.8 \pm 1.0) \times 10^{-1}$	21 ± 3

Values are means $\pm$ 1 standard error.

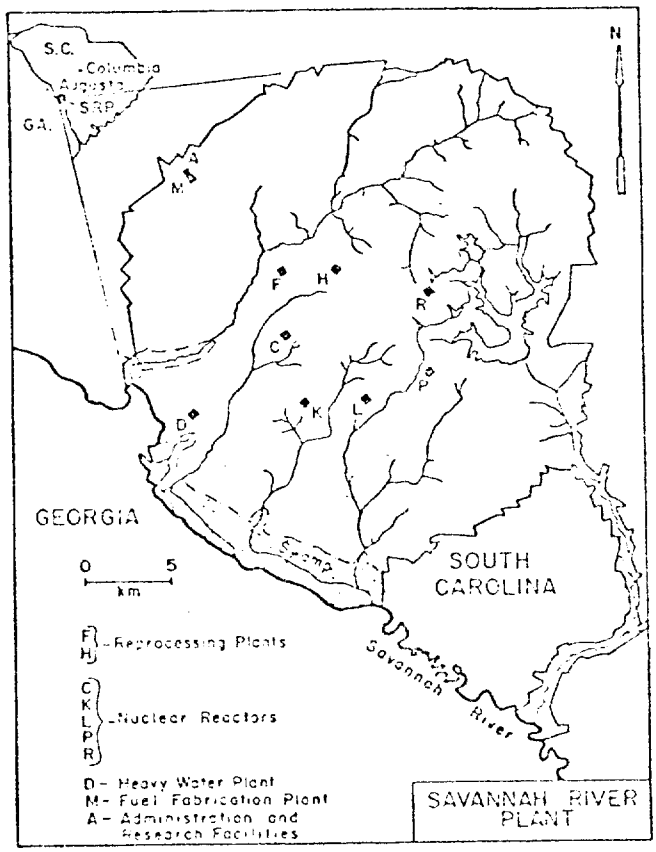

Fig. 1. Locations of the two reprocessing plants (H and F) at the Savannah River Plant. The agricultural study area is near the H-Area.

Fig. 2. Nomograph to calculate dose-to-bone from consumption of Pu-contaminated foodstuff. The dose is that which will be received by the bone during a 70-year lifespan. The following assumptions from the ICRP publications were used: effective absorbed energy per disintegration = 270 Mev (ICRP 2); fraction from gastrointestinal tract to blood = 3×10^{-5}, fraction from blood to bone = 0.45, half-life in bone $\simeq$ 100 yr. (ICRP 19); mass of bone = 5×10^{3} g (ICRP 20).

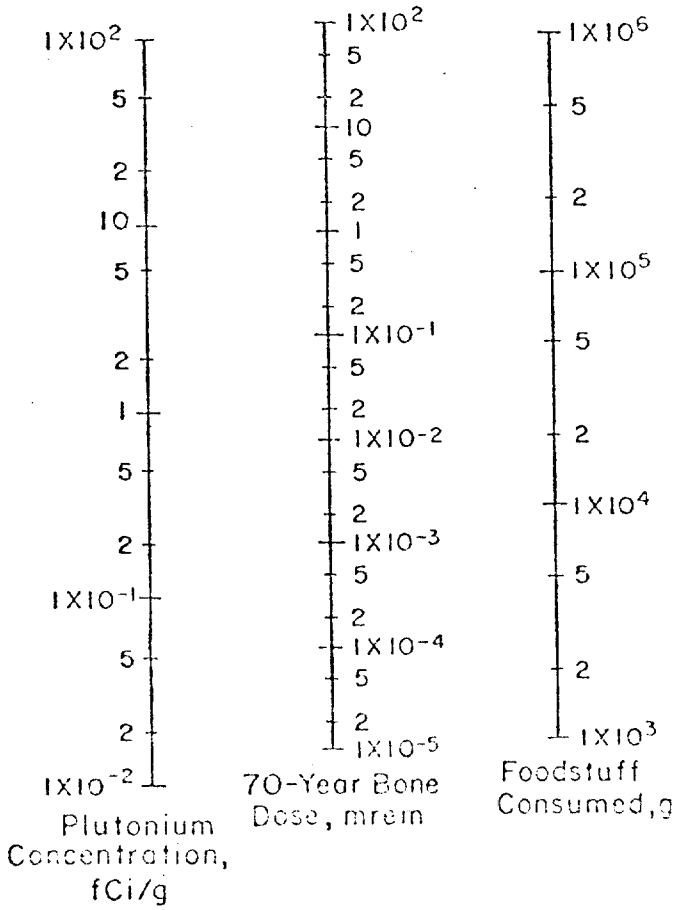
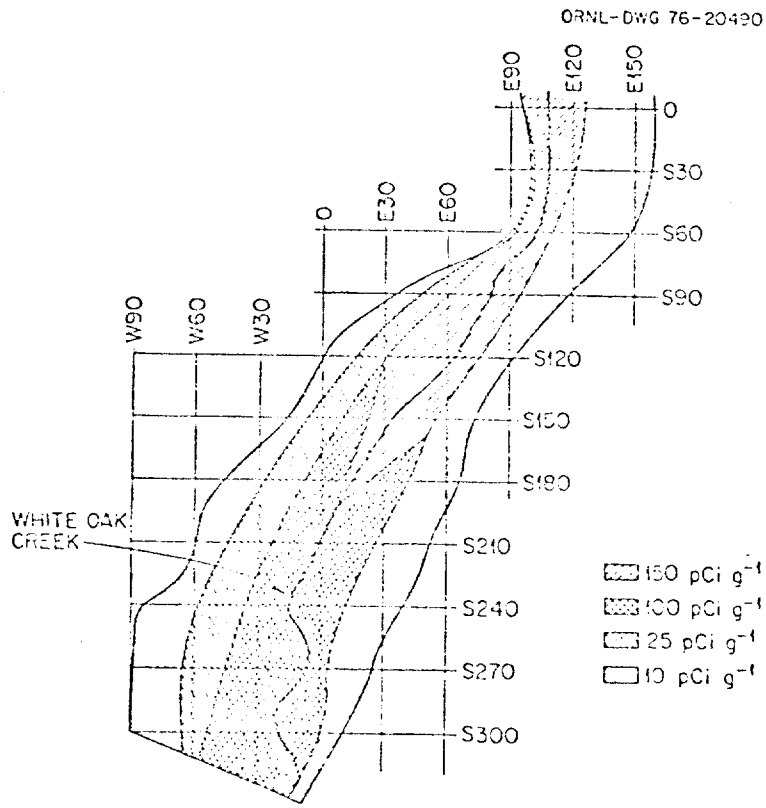
Fig. 3. Areal distribution of Pu at the contaminated Oak Ridge Floodplain. Grid size is 30 m x 30 m (900 m^2).