RADIONUCLIDE CONTENT OF VEGETATION AND SOIL ON AN INTEGRATED NUCLEAR COMPLEX

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A Summer Study Conducted During 1974

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INTRODUCTION

Detailed environmental plutonium studies have been conducted to evaluate the availability to growing plants of plutonium used in conjunction with safety shots and other weapons test programs at the U. S. Energy Research and Development Administration's Nevada Test Site. Such detailed studies have not been conducted in the southeastern U.S.

As an undergraduate student trainee at the Savannah River Laboratory, I was asked to conduct preliminary environmental plutonium studies to obtain information necessary to conduct the detailed studies that would be implemented in the future. The objective of my studies was to determine the relationships between the content in the soil and vegetation of plutonium, strontium, and cesium. The sampling locations were chosen to represent background areas and an area adjacent to the exhaust stack for a chemical reprocessing plant that had been operated continuously for 20 years. My responsibilities included experimental design, sample collection, sample analysis, and data interpretation. As a consequence of my preliminary studies, the plutonium, cesium, and strontium concentrations in soil and vegetation samples are known, so that appropriate sample sizes are obtained for accurate analysis. This information was used to scale plot sizes and evaluate sample size for a detailed 1.75 acre experiment in the vicinity of the exhaust stack. The purpose of the latter experiment is to obtain a detailed understanding of the effect of nuclear fuel reprocessing on the surrounding environment and man as well as to obtain fundamental information necessary for the environmental impact statements for the planned breeder programs and nuclear power plants.

PROJECT DESCRIPTION

One of the main objectives of this study was to determine relationships between the content of plutonium, cesium, and strontium in plant matter and the

*Student Trainee during the summer of 1974

content of these radionuclides in the soil. Since so much variation in plant uptake is due to differing soil characteristics, special attention was also given to soil characterization. This soil analysis included soil pH; particle size analysis; percent sand, silt, and clay; cation exchange capacity; and total exchangeable bases. Plant matter was analyzed for stable ion content. And finally, both plant material and soils were radiochemically analyzed for plutonium, cesium, and strontium.

SAMPLING

Soil and vegetation were collected from two locations on the Savannah River Plant in July 1974. The control, or background location, was at the plant boundary, the sample from the fuel reprocessing area was within 150 meters of the exhaust stack. Only one type of vegetation was sampled because this was only a preliminary study. The plant chosen was lespedesia, a common South Carolina weed. It was chosen because it was abundant and available at each of the locations.

Each sampling location was carefully examined, and an area with a maximum amount of lespedesia was chosen to sample. The lespedesia is a weed averaging approximately 50 cm in height and flowers in July. It was clipped within 1/2 inch from the soil. Care was taken to see that plant samples did not touch the soil. Samples were transported to the laboratory in sealed polyethylene bags.

In the same area, and as close to the sampled lespedesia as possible, soil samples were taken. The total depth sampled was 15 cm, which is approximately the root zone and thus would represent what is available to the growing plants. The soil sample was divided into two sampling depths: 0-5 cm and 5-15 cm. The ground was cleared of litter before the samples were taken. A trowel technique was used for soil sampling. A large hole, 5 cm deep, was dug. The soil from this hole constituted the 0-5 cm sample. Inside this hole, a smaller 10 cm one was dug. This soil was the 5-15 cm sample. A flat mason's trowel was used as a tool, and a ruler was employed to ensure uniformity and accuracy. These samples were also transported in sealed polyethylene bags.

SAMPLE PREPARATION

Nearly all of the vegetation collected was used in some phase of sample analysis. Upon return to the laboratory, the lespedesia was placed in a 3-liter beaker and rinsed three times with distilled water. The sample was agitated during the washing to remove particles on, but not incorporated in the vegetation. This consideration was necessary because this was an uptake study. After washing, the vegetation was dried at 75°C for 24 hours and finally ignited overnight in a muffle furnace at 550°C. This treatment gave a feasible sample size for direct gamma spectroscopy, put the vegetation in the proper form for other radiochemical techniques, and destroyed organics which would have complicated the

stable ion analysis.

More soil was collected than needed for analysis. Consequently, a second sampling technique was used. The soils were first mixed in their polyethylene bags. Then they were put through a 10-mesh sieve. Clay particles were broken down by hand, but all others too large to pass through the sieve were discarded. The sieved soils were sampled using the quartering method. The soils were spread out and divided into quarters, and an approximately equal amount was taken from each quarter. The soils were dried either in flat open pans on a hot plate or in an oven at 105°C overnight. Finally, the samples were ground, using a mortar and pestle, into fine dust.

METHODOLOGY

Radiochemical Analyses:

The samples of both soil and vegetation were analyzed for cesium-137 using gamma pulse height analysis. A system which utilized the Ge-Li crystal for detection was used to give high resolution. Long counting times were necessary for most vegetation samples due to small sample size and low activity. These analyses were made with a low-background gamma spectroscopy facility available at the Savannah River Laboratory.

The samples of both soil and vegetation were analyzed for strontium-90 using a stripping technique followed by counting of the beta activity which had grown in during a specific time period. Briefly, this technique involved leaching of the samples. Then strontium was precipitated as the oxalate. The precipitate was separated, ignited in a muffle furnace, and redissolved. The yttrium-90 was stripped using HCl and HDEHP. Finally, it was allowed to build-up for 15 days, and the sample was again stripped. The yttrium-90 from the buildup was mounted on a planchet and counted. From this, the strontium-90 concentration was calculated. Percent recovery was determined by spiking one of the replicate samples with a known amount of strontium-90.

The samples of soil and vegetation were analyzed for plutonium, using different procedures. The method of choice for soil analysis involved a leach procedure followed by separation using an ion exchange column. The elute was finally adjusted to the proper pH and electroplated to platinum. Counting was done on a solid state alpha spectrometer to resolve ^{236}Pu , ^{238}Pu , and $^{239-240}\text{Pu}$. An internal spike of ^{236}Pu is used to calculate percent recovery.

The method of plutonium analysis for vegetation involved dissolution of the ashed plant material and then double separation. The first separation was with TIOA-xylene, and the second used HCl. The organic residue was then destroyed using nitric acid and hydrogen peroxide. Finally, the solution was mounted on a planchet and counted in an alpha spectrometer.

Samples of both soil and vegetation were analyzed for stable ion content using a neutron activation method. The samples were exposed to a californium-252 neutron source. After some decay time, they were analyzed using gamma pluse height analysis with a Ge-Li detection system.

Soil samples were further analyzed for soil characteristics such as soil pH; particle size; percent sand, silt, and clay; and cation exchange capacity. Standard methods were used in these determinations such as can be found in *Methods of Soil Analysis* Vols. I and II, published by the American Society of Agronomy.

Results:

Table I presents the radionuclide contents of the vegetation and soil from two sampling locations. There is an elevated content of the measured radionuclides in the vicinity of the reprocessing facility. In addition, the table presents the relative concentration factor (pCi/g dry vegetation divided by pCi/g dry soil) between the vegetation and the soil. Studies conducted under greenhouse conditions where foliar deposition of soil particles is unlikely, generally report values of 10^{-3} - 10^{-4} for this value for plutonium, much lower than the 10^{-0} to 10^{-2} values observed here. The reason for this statistically significant difference is unclear although it suggests foliar contamination from soil particles containing small quantities of plutonium. The strontium and cesium values range within reported values.

Table 1. Radionuclide content of soil and vegetation

	Plutonium		Cesium		Strontium	
	pCi/g	RCF*	pCi/g	RCF*	pCi/g	RCF*
<u>Control</u>						
Vegetation	0.002		0.19		0.007	
0-5 cm	<0.004	>0.5	1.48	0.1	0.05	0.1
5-15 cm	<0.004	>0.5	0.25	0.8	0.03	0.2
<u>Adjacent to reprocessing facility</u>						
Vegetation	0.15		0.43		0.22	
0-5 cm	13.2	0.01	4.46	0.1	0.5	0.4
5-15 cm	1.1	0.1	2.11	0.2	0.24	0.9

*RCF - relative concentration factor (pCi/g dry weight vegetation divided by pCi/g dry soil)

In order to determine the suitability of washing for removing surface particulates, a study was conducted to evaluate the efficiency of the wash technique. The data gathered were inconclusive, but they did show that activity does decrease with washing. In future studies of plant uptake, either washing techniques should be improved and shown efficient or experimental design should include precautions concerning depositions of particulates on foliage.

Besides the information on soil and vegetation contents of plutonium, cesium, and strontium, other valuable knowledge has been gained from this study. Areas at the Savannah River Plant suitable for field research concerning plutonium in the environment have been identified. Greater understanding of soil and vegetation sampling problems has been gained. It is now possible to scale further studies so the statistically best results can be obtained (i.e., instruments can be operated at their most accurate level due to adequate sample planning).

Further research has been suggested by this project. Some of the questions raised are: Does radionuclide content depend upon the age of the plant? How is the radionuclide content distributed in the plant, and how is this distribution related to type of plant (e.g., legume, leafy, etc.)? In what form is plutonium available to plants? Does plutonium have an analog in nature which will help regulate its uptake (such as the Sr-Ca analogous pair)?

It is my hope that this pilot project has answered a few questions and raised enough of them so that it will be of benefit at the Savannah River Plant.