Americium-241 in Surface Soil Associated with the Hanford Site and Vicinity

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May 1981

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SUMMARY

Various kinds of surface soil samples were collected and analyzed for Americium-241 (^{241}Am) to examine the feasibility of improving soil sample data for the Hanford Surface Environmental Surveillance Program. Results do not indicate that a major improvement would occur if procedures were changed from the current practices.

Conclusions from this study are somewhat tempered by the very low levels of ^{241}Am (<0.10 pCi/g dry weight) detected in surface soil samples and by the fact that statistical significance depended on the type of statistical tests used. In general, the average concentration of ^{241}Am in soil crust (0 to 1.0 cm deep) was greater than the corresponding subsurface layer (1.0 to 2.5 cm deep), and the average concentration of ^{241}Am in some onsite samples collected near the PUREX facility was greater than comparable samples collected 60 km upwind at an offsite location.

CONTENTS

SUMMARY	iii
INTRODUCTION	1
METHODS	3
SAMPLING SITES	3
SAMPLING STRATEGY	3
SAMPLE PREPARATION AND ANALYSIS	4
RESULTS AND DISCUSSION	5
SAMPLE RESULTS	5
Quality Control	9
CONCLUSIONS	13
REFERENCES	15

TABLES

1. Samples Collected at Each Sampling Site	4
2. Sample Descriptions and Individual Sample Values	6
3. Comparisons Between Different Types of Soil Samples	7
4. Comparing Variability Between Soil Crust (0 to 2.5 in.) and Bare Soil (0 to 2.5 cm)	10
5. Quality Control Data Evaluation	11

AMERICIUM-241 IN SURFACE SOIL ASSOCIATED WITH THE HANFORD SITE AND VICINITY

INTRODUCTION

Soil samples collected for the Hanford Environmental Surveillance Program are assumed to represent an average condition over the area sampled. Five shallow cylinders of surface soil (4.0 cm diameter, 2.5 cm deep) are collected at a pre-selected sampling location and then mixed together to form a composite sample for analysis. Samples are usually collected from bare soil, which may contain portions of root systems, pieces of moss colonies, and other minute plants growing on the soil surface. The moss colonies dry out during warm weather and appear as a crust over the soil surface. For the purpose of this report, the moss colonies and other minute plants found on the soil surface will be referred to as "soil crust."

Soil crust is long-lasting when there is little or no disturbance by man. Individual colonies could be expected to exist for many years. Moreover, the rough and irregular surface of soil crust may be a better material for trapping and retaining airborne particles than bare soil. If this is so, then sampling and analyzing soil crust may provide soil samples that are more sensitive in terms of detecting very low levels of radionuclides in the environment. Increased sensitivity would be particularly useful in detecting and following trends in the buildup of trace quantities of radionuclides in soil.

The primary purpose of this study was to examine the feasibility of improving soil sample data for the Hanford Surface Environmental Surveillance Program by sampling and analyzing soil crust instead of bare surface soil as is the current sampling practice. A secondary purpose was to compare onsite surface soil concentrations of ^{241}Am with those determined for a distant offsite location. Americium-241 was selected for study because of its long half-life (458 y) and potential longevity in the environment. There is only limited information about ^{241}Am in the Hanford environs.

Criteria for deciding if soil crust was a preferred sampling medium included demonstrating that:

- statistical variation associated with replicates of soil crust is equal to or less than the variation among replicates of bare soil
- soil crust is significantly more efficient than bare soil at trapping and retaining airborne particles (i.e., higher in ^{241}Am concentration)
- it is as easy or easier to collect, handle, and analyze samples of soil crust as it is samples of bare soil.

METHODS

SAMPLING SITES

Surface soils near the Hanford chemical separations and waste management facilities (200 Areas) are known to contain slightly higher concentrations of $^{239-240}\text{Pu}$ than other locations, both onsite and off (Corley et al. 1972). Also, very small quantities of ^{241}Am could occur in local surface soils from the release of trace amounts of ^{241}Am or ^{241}Pu from the PUREX facility during the years of fuel reprocessing (1956 to 1972). Thus, an onsite location was selected for soil sampling, about 500 m east of the 90-m tall PUREX gaseous effluent discharge stack. An offsite location was chosen near Moxee (Yakima Co.), Washington, at the Washington State Plant Introduction Station, approximately 60 km west and generally upwind from Hanford. The vegetation (sagebrush-cheatgrass) was similar at each site and neither site showed heavy use or grazing pressure. The soil crust consisted of dense clumps of moss, with a few lichens and other minute plants. Clumps frequently occurred in protected areas such as beneath shrubs and toward the north sides of shrubs.

SAMPLING STRATEGY

A single study plot at each site consisted of a rectangular area about 80 x 160 m. Each plot was divided crosswise into 20 strips about 8 m wide and 80 m long. Each strip was then divided into 20 equal-sized subplots, about 4 m wide and 8 m long, five of which were chosen at random to serve as sampling locations. Samples collected from the five subplots were composited by sample type (Table 1). Thus, each study plot yielded 20 composited samples of each type of sample. One additional composited sample of soil crust was collected from between shrubs at each site from five randomly selected subplots and later spiked with a known amount of ^{241}Am to serve as a quality control sample. These 10 spiked samples were submitted blind for radiochemical analysis. Two aliquots from each spiked sample were analyzed for ^{241}Am .

TABLE 1. Samples Collected at Each Sampling Site

<u>Sample Type and Depth</u>	<u>Beneath Shrubs</u>	<u>Between Shrubs</u>
Soil crust 0 to 1.0 cm	Yes	Yes
Next soil layer 1.0 to 2.5 cm	Yes	Yes
Bare soil 0 to 2.5 cm	No	Yes

SAMPLE PREPARATION AND ANALYSIS

All samples were dried at 105 C for at least 48 hrs and delivered to U.S. Testing Company, Inc. (UST) for radiochemical analysis. UST developed a new analytical technique to improve detection to about 0.05 pCi/g dry weight (U.S. Testing 1980). A 50 g aliquot from each sample was ashed, acid-leached and the ^{241}Am extracted using cation and anion exchange resins and liquid-liquid chromatography. Final solutions were electrodeposited and the plates counted by alpha spectrometry for Americium activity. Americium-243 was used as an internal standard to determine yield.

RESULTS AND DISCUSSION

SAMPLE RESULTS

Data are arranged in Table 2 to show the minima, maxima, standard error, mean, and median ^{241}Am concentrations for each type of sample collected. An obvious feature of the data is the very low concentration of ^{241}Am in all surface soil samples (<0.10 pCi/g dry) for both onsite and offsite locations.

A statistical treatment of the data was used to study these questions:

1. Do samples of soil crust (0 to 2.5 cm) contain greater concentrations of ^{241}Am than samples of bare soil (0 to 2.5 cm)?
2. Do samples of soil crust (0 to 1.0 cm) contain greater concentrations of ^{241}Am than corresponding deeper soil layers (1.0 to 2.5 cm)?
3. Do samples collected beneath shrubs contain greater concentrations of ^{241}Am than samples collected between shrubs?
4. Do samples collected onsite contain greater concentrations of ^{241}Am than samples collected offsite?
5. Do replicate samples of the total soil crust layer (0 to 2.5 cm) have less variability than replicate samples of bare soil samples (0 to 2.5 cm)?

Questions 1 through 4 were addressed by doing a series of t-tests on appropriate data sets. The comparisons made and the t-test results are summarized in Table 3.

Paired sample t-tests (Snedecor and Cochran, 1980, p. 84) were used to compare the total soil crust layer versus bare soil (Table 3). The 0 to 2.5 cm bare soil concentration for each strip was subtracted from a weighted average of the 0 to 1.0 cm soil crust plus the 1.0 to 2.5 cm soil layer concentrations for that strip, i.e.:

$$\text{Difference for the } i\text{th strip} = \left[(2X_{1i} + 3X_{2i}) / 5 \right] - X_{3i} \quad ,$$

where

$$\begin{aligned} X_{1i} &= 0 \text{ to } 1.0 \text{ cm soil crust concentration} \\ X_{2i} &= 1.0 \text{ to } 2.5 \text{ cm soil layer concentration} \\ X_{3i} &= 0 \text{ to } 2.5 \text{ cm bare soil concentration.} \end{aligned}$$

TABLE 2. Sample Descriptions and Individual Sample Values

Location	Position	Sample Type	Code	Range of Individual Sample Values ^(a)	Standard Error ^(b)	Mean	Median
pCi ²⁴¹ Am/g dry soil							
				-0.02 0.0 0.02 0.04 0.06 0.08 0.10			
Onsite	Beneath Shrubs	Soil Crust (0-1.0 cm)	A		0.0047	0.037	0.035
		Soil Layer (1.0-2.5 cm)	B		0.0040	0.021	0.020
	Between Shrubs	Soil Crust (0-1.0 cm)	C		0.0048	0.035	0.034
		Soil Layer (1.0-2.5 cm)	D		0.0047	0.024	0.021
		Bare Soil (0-2.5 cm)	E		0.0060	0.027	0.017
Offsite	Beneath Shrubs	Soil Crust (0-1.0 cm)	F		0.0047	0.021	0.019
		Soil Layer (1.0-2.5 cm)	G		0.0043	0.014	0.007
	Between Shrubs	Soil Crust (0-1.0 cm)	H		0.0056	0.031	0.024
		Soil Layer (1.0-2.5 cm)	I		0.0058	0.021	0.017
		Bare Soil (0-2.5 cm)	J		0.0058	0.018	0.014

(a) Each dot marks the ²⁴¹Am concentration measured in a single sample. Negative values are due to background exceeding the sample count.

(b) Standard error = $\left[\frac{1}{n(n-1)} \sum_{i=1}^n (x_i - \bar{x})^2 \right]^{1/2}$

TABLE 3. Comparisons Between Different Types of Soil Samples

Comparisons	Test	t-test ^(a)	Degrees of Freedom	α
SOIL CRUST PLUS SOIL LAYER <u>VS</u> BARE SOIL ^(b)				
Onsite beneath shrubs	$(2A + 3B)/5$ <u>vs</u> E	0.05	19	0.96
Onsite between shrubs	$(2C + 3D)/5$ <u>vs</u> E	0.16	19	0.87
Offsite beneath shrubs	$(2F + 3G)/5$ <u>vs</u> J	-0.58	19	0.57
Offsite between shrubs	$(2H + 3I)/5$ <u>vs</u> J	1.1	19	0.29
SOIL CRUST <u>VS</u> SOIL LAYER				
Onsite beneath shrubs	A <u>vs</u> B ^(c)	2.6*	38	0.01
Onsite between shrubs	C <u>vs</u> D ^(c)	1.6	38	0.12
Offsite beneath shrubs	F <u>vs</u> G ^(c)	1.1	38	0.28
Offsite between shrubs	H <u>vs</u> I ^(c)	1.3	38	0.20
BENEATH SHRUBS <u>VS</u> BETWEEN SHRUBS				
Onsite soil crust	A <u>vs</u> C ^(c)	0.35	38	0.73
Onsite soil crust plus soil layer	$(A + B)$ <u>vs</u> $(C + D)$ ^(d)	-0.04	78	0.97
Offsite soil crust	F <u>vs</u> H	-1.5	38	0.14
Offsite soil crust plus soil layer	$(F + G)$ <u>vs</u> $(H + I)$	-1.8	78	0.08
ONSITE <u>VS</u> OFFSITE				
Beneath shrubs soil crust	A <u>vs</u> F	2.5*	38	0.02
Beneath shrubs soil layer	B <u>vs</u> G	1.3	38	0.20
Beneath shrubs soil crust plus soil layer	$(A + B)$ <u>vs</u> $(F + G)$	2.6*	78	0.01
Between shrubs soil crust	C <u>vs</u> H	0.47	38	0.64
Between shrubs soil layer	D <u>vs</u> I	0.40	38	0.69
Between shrubs soil crust plus soil layer	$(C + D)$ <u>vs</u> $(H + I)$	0.60	78	0.55
Bare Soil	E <u>vs</u> J	1.2	38	0.24

(a) Significance at the two-tailed alpha level shown in last column is noted with*.

(b) See text for explanation of notation and the paired t-tests performed.

(c) t-tests for paired samples were also computed for these comparisons. Conclusions were the same as for the independent sample t-test.

(d) The notation $(C + D)$ means that data sets C and D (Table 1) were pooled together to form a single data set of size 40.

The paired sample t-test is then

$$t_{19} = \frac{\bar{X}}{S/\sqrt{20}}$$

where $\bar{X}$ = average of the 20 differences
S = standard deviation of the 20 differences
19 = degrees of freedom.

The remaining values given in Table 3 are for "independent sample" t-test (Snedecor and Cochran, 1980, p. 89) and were computed as

$$t_{n_1 + n_2 - 2} = \frac{\bar{x}_1 - \bar{x}_2}{S_{\bar{x}_1 - \bar{x}_2}}$$

where $S_{\bar{x}_1 - \bar{x}_2}$ = standard deviation of $\bar{x}_1 - \bar{x}_2$,

$\bar{x}_1$ and $\bar{x}_2$ are the two sample means being compared, and

n_1 and n_2 are the number of data values in the two groups.

These test results suggest the following:

- soil crust material is significantly higher in average ^{241}Am concentration than the corresponding subsurface layer for samples collected onsite from beneath shrubs (Test A vs B)
- soil crust material collected onsite from beneath shrubs is significantly higher in average ^{241}Am concentration than comparable samples collected offsite (Test A vs F)
- the total 0 to 2.5 cm layer of surface soil collected onsite from beneath shrubs is significantly higher in ^{241}Am concentration than comparable samples collected offsite [Test (A+B) vs (F+G)].

The same conclusions were obtained when the Wilcoxon Rank Test (Hollander and Wolfe, 1973, p. 68) was used in place of the independent sample or paired t-tests to account for data distributions that may not be normal. We also note that if Bonferroni t-tests (Miller, 1966, p. 67) are used to take into account the fact that an occasional significant result is expected even when no true differences exists (in any of the comparisons), then none of the t-tests in Table 3 are statistically significant. Hence, statistical tests do not in themselves provide a completely convincing case for significant differences. More highly significant and consistent differences might have been obtained had more than 20 samples per data set been collected.

Question 5 above was addressed statistically by comparing the sample variances of the total soil crust layer (weighted average of 0 to 1.0 cm and 1.0 to 2.5 cm) concentrations with the bare soil (0 to 2.5 cm) concentrations. One-tailed F-tests were performed by computing (Snedecor and Cochran, 1980, p. 252):

$$F_{n_1-1, n_2-1} = s_2^2 / s_1^2 ,$$

where s_2^2 = sample variance of the $n_2 = 20$ bare soil concentrations, and

s_1^2 = sample variance of the $n_1 = 20$ soil crust concentrations.

The results of these tests are shown in Table 4. Three of the four F-tests were statistically significant. This suggests that the true variability of bare soil (0 to 2.5 cm) concentrations may be greater than that for samples of the total soil crust layer (0 to 1.0 cm plus 1.0 to 2.5 cm). If these results were confirmed by additional studies, it might be preferable to sample soil crust rather than bare soil since more precise estimates of average concentrations would be obtained.

Quality Control

The quality control data for the 10 spiked samples submitted for ^{241}Am laboratory analyses are given in Table 5. The bias for the 10 samples ranged

TABLE 4. Comparing Variability Between The Total Soil Crust Layer (0 to 2.5 cm) and Bare Soil (0 to 2.5 cm)

Comparisons ^(a)	$F = s_2^2/s_1^2$ ^(b)
Onsite Beneath Shrubs (2A + 3B)/5 vs E	0.00073/.00021 = 3.5**
Onsite Between Shrubs (2C + 3D)/5 vs E	0.00073/.00025 = 2.9*
Offsite Beneath Shrubs (2F + 3G)/5 vs J	0.00068/.00019 = 3.6**
Offsite Between Shrubs (2H + 3I)/5 vs J	0.00068/.00050 = 1.4 NS

(a) We are testing $H_0: \sigma_1^2 = \sigma_2^2$ versus $H_A: \sigma_1^2 < \sigma_2^2$

where σ_1^2 = true variance of crust material samples (0 to 2.5 cm), and

σ_2^2 = true variance of bare soil samples (0 to 2.5 cm).

(b) F = "F" statistic with 19 and 19 degrees of freedom.

* = Statistically significant at the = .05 one-tailed level.

** = Statistically significant at the = .01 one-tailed level.

NS = Non-significant.

from -32% (-0.27 pCi/g) to +4% (+0.03 pCi/g) with an average of -7.7 (-0.065 pCi/g). Seven of the ten biases were negative (average lab result less than the spiked value). Two of these seven were about 30% low. The other eight biases were within +10% of the true value. The 20 individual deviations (lab results minus spiked value) ranged from -34% (-0.29 pCi/g) to +13% (+0.10 pCi/g). These are acceptable results considering the low concentration and high degree of difficulty of the radiochemical procedure.

TABLE 5. Quality Control Data Evaluation

Sample	Spiked Value pCi/g	Lab Results	Lab Results Minus Spiked Value	
		pCi/g	pCi/g	%
1	0.66	0.56	- 0.10	- 15
		0.69	+ 0.03	+ 4.5
		Ave = 0.63	Bias = - 0.03	- 4.5
2	0.75	0.71	- 0.04	- 5.3
		0.85	+ 0.10	+ 13
		Ave = 0.78	Bias = + 0.03	+ 4.0
3	0.78	0.69	- 0.09	- 12
		0.81	+ 0.03	+ 3.8
		Ave = 0.75	Bias = - 0.03	- 3.8
4	0.81	0.71	- 0.10	- 12.3
		0.79	- 0.02	- 2.5
		Ave = 0.75	Bias = - 0.06	- 7.4
5	0.81	0.76	- 0.05	- 6.2
		0.89	+ 0.08	+ 10
		Ave = 0.82	Bias = + 0.01	+ 1.8
6	0.84	0.59	- 0.25	- 30
		0.61	- 0.23	- 27
		Ave = 0.60	Bias = - 0.24	- 29
7	0.85	0.56	- 0.29	- 34
		0.60	- 0.25	- 29
		Ave = 0.58	Bias = - 0.27	- 32
8	0.85	0.80	- 0.05	- 5.9
		0.85	0.0	0.0
		Ave = 0.83	Bias = - 0.02	- 2.4
9	1.04	0.85	- 0.19	- 18
		1.09	+ 0.05	+ 4.8
		Ave = 0.97	Bias = - 0.07	- 6.7
10	1.12	1.06	- 0.06	- 5.4
		1.23	+ 0.11	+ 9.8
		Ave = 1.15	Bias = + 0.03	+ 2.7

Average of the 10 biases =
- 0.065 pCi/g (- 7.7%).

CONCLUSIONS

Results from this study have shown that:

- Very low levels of ^{241}Am (<0.10 pCi/g dry) were detected in various types of surface soil samples collected on the Hanford Site as well as offsite.
- Although not statistically significant in every case, the average ^{241}Am concentrations for all soil crust (0 to 1.0 cm) samples were greater than the corresponding subsurface (1.0 to 2.5 cm) samples.
- Although not statistically significant, the onsite average ^{241}Am concentrations for all five categories of samples were greater than corresponding offsite averages.
- Soil crust (0 to 1.0 cm) material consisting of mosses and other minute plants may contain higher ^{241}Am concentrations than the corresponding subsurface layer (1.0 to 2.5 cm) for samples collected onsite from beneath shrubs.
- Soil crust (0 to 1.0) samples collected onsite from beneath shrubs may be higher in ^{241}Am concentration than comparable samples collected offsite.
- The total 0 to 2.5 cm soil crust layer collected onsite from beneath shrubs may be higher in ^{241}Am concentration than comparable samples collected offsite (there was no statistical difference for samples of soil crust or bare soil collected between shrubs).
- In general, the difference in dispersion (variance) between most data sets was not large. However, significant differences in variance between bare soil and soil crust samples were obtained.

The primary purpose of this study was to examine the feasibility of improving soil sample data for the Hanford Surface Environmental Surveillance Program by sampling soil crust material. Results do not indicate that a major improvement would occur if procedures were changed. However, collecting soil

crust material from beneath shrubs may be useful when sampling for trace elements to detect trends and the possibility of a gradual environmental buildup. Also, more precise estimates of average ^{241}Am concentrations may be possible using soil crust rather than bare soil sample results. The secondary purpose of this study was to compare onsite surface soil concentrations of ^{241}Am with those determined for a distant offsite location. Statistically significant results indicate that some onsite soil samples are higher in ^{241}Am than comparable offsite samples.

No difference was noted in handling the various sample types when compared to current environmental surveillance practices. Analytical results for quality control samples indicated acceptable accuracy for ^{241}Am determinations.

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