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DP-MS-79-20

**MIGRATION OF TRANSURANIC NUCLIDES IN EARTHEN BURIAL
TRENCHES AT THE SAVANNAH RIVER PLANT**

by

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An outline of a paper invited for presentation at a Symposium on "Migration of Transuranic Nuclides in the Environment" to be held at the National ANS Meeting, San Francisco, California, November 13-15, 1979.

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The talk to be given at the National ANS Meeting is presented in outline form. The outline details items to be emphasized as each slide is presented. The slides support and also expand on all information given in the extensive summary (attached) which was approved by Savannah River Laboratory (SRL) management and the Department of Energy, Savannah River Operations Office (DOE-SR) prior to submission and acceptance by the ANS.

Slide 1: Presents objectives of waste migration studies at SRL and points out incentives and/or benefits to be derived from information gathered in these studies; particular emphasis is placed on need for migration data to develop accurate dose-to-man modeling parameters.

Slide 2: Summarizes known or estimated information about unencapsulated transmission (TRU) waste burials at SRP. TRU nuclides are of concern in any modeling studies because of their long half-life. Plutonium is the primary TRU nuclide of interest in present studies, although americium and curium also are buried in the Savannah River Plant Burial Ground.

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* The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy.

Slide 3: Summarizes SRP site characteristics that can have potential effect on extent and rate of radionuclide migration. These characteristics, in combination, are unique to the SRP site. Thus, studies at the SRP site are required to quantify the extent and rate of migration. Studies at other sites can guide or direct SRP studies, but cannot give complete characterization for the SRP site.

Slide 4: Map locates Burial Ground within SRP separations area. A natural watershed divide provides ground water flow towards Four Mile Creek from the Burial Ground. Water table under the Burial Ground is at ~ 40-feet. Wastes are buried in trenches ~ 20-feet deep and are covered with ~ 4 feet of clean soil. The coring study is intended to determine how quickly TRU nuclides are leaching from wastes and moving through the SRP soil matrix to the water table and/or creeks.

Slide 5: Summarizes trench sampling strategy: Survey phase is complete; resampling phase is underway; prospecting phase to locate hotspots (> 1 Ci) has not been initiated. Hotspots in the Burial Ground can be located from SRP records based on known burial coordinates. Soil cores obtained from survey and resampling phases are the bases for data presentations in this talk.

- Slide 6. Self-explanatory description of coring samples; although Shelby tubes were more difficult to open (they had to be physically cut open), they were used quite extensively because of generally higher soil recoveries, which will be explained more fully in subsequent slides. One single-coring generally consisted of from 8 to 10 core segments (depending on type of sampler used) in order to sample the complete 20-foot depth horizon of the waste trenches.
- Slide 7. Shows schematic of coring layout to test recoveries of various core sampler types. Intent was to pass each core sampler through similar trench locations to minimize any effect of variable soil/waste composition on observed recovery parameters.
- Slide 8: Tabulates actual recovery of soil in split-core and 3-in. Shelby tube segments as a function of depth. The observed soil/waste composition in all of the cores collected for this evaluation were fairly similar, as had been hoped by the specific coring layout described in the previous slide. In general, Shelby tubes gave highest recoveries at any depth. Two-in. Shelby tubes were also evaluated, but recoveries were comparable to split-barrel cores. These data led to extensive use of 3-in. Shelby tubes in much of the coring program.

Slide 9: The photograph points out a specific problem which reduced soil recoveries during coring, even when utilizing Shelby tube cores. In this case, plastic waste has plugged the end of the core. When this happens, it is not difficult to visualize that the waste simply pushes through or compresses the waste/soil matrix until it is driven to its full range (2 to 2.5 feet) and then retrieved from the hole. If no further waste is present, the subsequent core segment can generally be expected to have a higher soil recovery; but in many cases, waste occupies a significant depth horizon in the trench (i.e., several feet) so that several cores may have very low recovery due to waste pluggage at the end of the cores.

Slide 10: Photograph showing an interesting waste strike-screwdriver surrounded by some soil. Soil recovery in this case is fairly low. This slide is presented primarily because of uniqueness of particular waste recovered.

Slide 11: Summarizes coring data obtained in survey coring phase and in resampling phase for one particular trench. The resampled trench had yielded significant waste and activity strikes in the survey phase. The trench was resampled to determine the suitability of that trench for a planned vegetation uptake study.

More extensive coring in that trench resulted in significantly higher waste strike frequencies than had been observed in the survey phase. Average soil recovery from all core segments relative to total core length is presented for the cores from the re-sampling phase to indicate approximate waste/soil volume occurring in the trench.

Slide 12: Graphically summarizes observed recoveries in each of nineteen cores from the resampling phase and demonstrates that average soil recoveries for all segments decrease as observed waste strikes within one complete 20-foot core length increase. This observation is not surprising because locations having high packaged waste volumes would be expected to have low soil volumes,

Slide 13: Demonstrates that soil recovery in each core segment varies relative to sampling depth. This slide complements the previous slide in that lowest recoveries coincide with regions where the highest percentage of waste strikes is experienced (particularly in the 6-to-18 foot horizon). The histogram shows distribution of waste strikes as a function of depth for the resampled trench; waste distribution is fairly uniform throughout the 6-to-18 foot horizons.

Slide 14: Tabular data on waste strike distribution and activity strike distribution as a function of depth. Data from survey phase and resampling phase have very similar distributions. It should be remembered that the number of waste strikes was significantly higher than the number of probable activity strikes, as pointed out in a previous slide (Slide 10).

Slide 15: Table depicting distribution of all waste strikes in terms of actual waste composition type. Waste types observed are those that would typically be expected from a nuclear production and/or research site like SRP. The distributions in the survey coring and the trench resampling phase were found to be very similar, as shown in the slide.

Slide 16: This slide points out a somewhat surprising finding - the fact that wood and paper wastes were well-preserved despite long burial in the solids. The preserved wood and paper wastes were found at various depths throughout the trench horizon (as shown in the previous slide) and not just at great depths (16-to-20 feet, for example), where anaerobic conditions might be expected to reduce waste decomposition rates.

Slide 17: Shows paper form obtained from core. Form is known to have been used only prior to 1960. Note surprisingly

well-preserved composition. In this case, paper came from great depth, but similar observations were made at depths closer to the surface.

Slide 18: Slide showing well-preserved wood waste - same comments as in Slide 16.

Slide 19: Graph shows radioactivity found in soil for one particular core. The schematic below the graph summarizes the soil and waste recoveries observed in this particular core. Activity could not be assigned to either soil or waste using gross counting equipment. However, careful analysis showed low-levels of activity in the soil. The radioactivity was an average value for all of the soil found in the segment because the total soil recovery was homogenized prior to analysis. In this case, one core segment, designated "A" in the core schematic, had zero soil recovery. Soil collected just beneath that waste strike had the highest concentration of activity observed in this core. The activity dropped off rapidly and was ~ 0.1 pCi/g in the undisturbed soil horizon below 20 feet. Similar activity distributions were observed in many of the collected cores.

Slide 20: These data are similar to those on the previous slide for a different core. In this case, several segments (A, B, C) had zero or very low soil recoveries. Most

of the radioactivity measured in the soil was below levels where waste was found. Increased activity in soil from 18- to 20-foot horizon is probably due to leaching from the waste found in 16- to 18-foot horizon. However, neither waste nor soil probed any activity with gross counting equipment. Soil below 20 feet was not collected in this case.

Slide 21: This photograph is included to demonstrate that soil in close proximity to fairly highly contaminated waste does not contain extremely high radioactivity levels. The waste shown in this core segment probed $\sim 5 \times 10^5$ counts/minute. Although the efficiency of the counting equipment used to probe the waste is unknown, this activity level probably corresponds to ~ 300 to 700 nCi/g waste. Analysis of a sample of soil from below the waste (taken after blending ~ 6 inches of soil to right of waste in the slide) measured 811 pCi/g (0.8 nCi/g), which is a relatively small fraction of the total activity found on the waste. Most of the radioactivity in the soil probably was located in the first few inches of soil contacting the waste. However, the sampling procedure was not controlled to prove this point. More careful sampling procedures will be used in the future to determine conclusively the range of migration of radionuclides from high-activity waste strikes.

Slide 22: This table summarizes average radioactive contents of soils recovered from the resampling phase. Some of the soils having highest activities (as determined by probing) were not measured accurately; thus those activities were not included in obtaining the averages for these data. The important point is that a higher concentration of plutonium was found in the lower horizons of the trench (below ~ 10 feet), which may be an indication of leaching of radioactivity from waste. In most cases, average soil concentrations are well below 10 nCi/g.

Slide 23: Corings within one foot of a central core location containing waste that had a high amount of activity (~ 10 counts/minute) generally did not have high activities in the peripheral cores, as shown in the table. Thus, sites with high-activity waste are generally found in only highly localized volumes, rather than distributed widely throughout the trenches.

Slide 24: The distribution of plutonium among soil particle sizes was determined as follows: Each core segment was homogenized, and a 25-g aliquot of each was treated with hydrogen peroxide to destroy organic matter. The treated soil was dispersed by shaking overnight with 0.02M sodium pyrophosphate. Sand (particle size > 50 μ m) was separated by wet-sieving through a 325-mesh

screen. Silt (particle size between 2 μm and 50 μm) was separated from the clay suspension by centrifuging. Clay (particle size $<2 \mu\text{m}$) was collected by flocculating with 1M magnesium acetate and centrifuging. Each soil fraction was analyzed for plutonium content. Additionally, all of the liquid that had contacted the soil (sodium pyrophosphate dispersing solution plus distilled water rinses (the supernate) was collected and analyzed for plutonium. Most of the plutonium (75.8%) was found in the clay fraction. Sand and silt fractions averaged $\sim 11\%$. The supernate averaged 2.4% of the plutonium. Results are shown in the table presented in the slide. Most of the plutonium was in the clay fraction because of the large surface area of the small particles, indicating that soil particle coatings are sorbing the plutonium.

Slide 25: The binding sites of plutonium on the soil were determined by sequentially extracting duplicate 10-g aliquots of soil samples. Ion-exchanged, reductant-soluble, and organic acid fractions were removed from the soil. This procedure has been used by several investigators. The ion-exchangeable phase was removed by stirring the soil for 24 hours at room temperature with 500 mL of 0.1N magnesium chloride. The metallic oxide (reductant-soluble) phase was removed by stirring

the soil for 15 minutes at 90°C in a solution composed of 10 g sodium dithionite dissolved in 400 mL of 0.3M sodium citrate. The organic phase (humic-fulvic acids) was removed by stirring the residue overnight at room temperature with 500 mL of 0.1N sodium hydroxide. The undissolved residue remaining after these treatments was also analyzed for plutonium. Results showed that the major activity was associated with the metallic oxide phases of the soil, rather than being ion-exchanged or in the organic phases. Tests were made reversing the order of the dithionite-citrate and sodium hydroxide extraction procedures to determine if higher fractions of activity might be associated with organic phases that might be removed by the dithionite-citrate treatment. No difference in results was observed.

Slide 26: To assess the plutonium leachability, 10-g samples of each of the six homogenized soil core segments were continuously shaken for two hours with 100-mL volumes of either distilled water, 0.1M HNO₃, 0.1M HCl, 1M ammonium acetate, or 1M ammonium carbonate + 0.01M ethylenediaminetetraacetic acid (EDTA). Distilled water removed a low fraction of the plutonium from the soil. In almost all cases, the distribution coefficient, K_d , from these batch desorption experiments

was greater than 1000 (average K_d for all SRP soil studied is 1600, with a range of 300 to 4000). The dilute mineral acids removed higher fractions of plutonium, indicating that the solubilized plutonium may have been present in the soil as a non-polymeric hydroxide, or it may have been sorbed on a soil constituent solubilized by the acids.

Extraction of soils with ammonium acetate has been used to predict the availability of micronutrients to plants. One molar ammonium acetate solution removed less than 1% of the plutonium from most samples. This low extractability implies that plutonium is not ionically bound to Burial Ground soil, and suggests a low availability of plutonium to plants.

The 1M ammonium carbonate solution containing 0.01M EDTA extracted the most plutonium (28.5% to 74.6%) from the soils, probably because of formation of EDTA or carbonate complexes with plutonium. High extractability as EDTA or carbonate complexes implies that plutonium is not sorbed on the soil as a polymeric hydrous oxide.

These results are in general agreement with results from the determination of plutonium binding sites presented in the previous slide. The plutonium is

not ion-exchanged (and thus not easily leached with water). The plutonium found in the reductant-soluble (predominantly iron and manganese oxide coatings on the soil particles) and crystalline phases of the soil matrix is not expected to be soluble in the soil-water system. Thus, the bulk of the plutonium now on these soils will be immobile except for possible movement of soil particles containing plutonium. Transportation of soil particles has not been investigated extensively at the SRP site.

Slide 27: Points made are self-explanatory and are supported by all the data presented in talk.

Slide 28: This slide will be presented to mention other programs underway at SRL that compliment the coring data program and provide data input to the dose-to-man modeling program. These programs include controlled field studies (lysimeters, tree studies, and the well monitoring program) and laboratory studies (soil column tests). Waste migration studies comprise a significant effort at the SRP site.

SLIDE 1

Waste Migration Program
Sampling of Earthen Burial Trenches

Study Objectives

- Determine Current Extent of TRU Migration
- Determine Chemical Species and Mineral Association in Soils of Plutonium
- Determine Waste/Soil Ratios
- Improve Estimate of TRU Inventory

Incentive/Benefits

- Quantify Migration Risk; Predict Dose-to-Man
- Guide Planning for Surveillance/Decommissioning of Burial Ground

SLIDE 2

Unencapsulated Alpha-Waste Burials

- All Waste Prior to 1965
- Total Estimated Inventory; 4,000 Ci
  - 2600 Ci ^{238}Pu
  - 400 Ci ^{239}Pu

SLIDE 3

SRP Site Characteristics

- Humid Environment
- Sandy-Clay Soils
- Acidic Water/Soils
- Geology/Hydrology

SLIDE 4

AVERAGE ELEVATION OF THE WATER TABLE AT SRP DURING 1968

Note: Water Table Depth at the Burial Ground is 35 to 40 ft.

SLIDE 5

Trench Sampling Strategy

- Survey Phase (Complete)
  - Random Sampling (3 cores/31 trenches)
  - Waste/Radionuclide Inventory
- Resampling Phase (Partially Complete)
  - High TRU Locations Based on Survey Phase
- Prospecting Phase (not initiated)
  - Locate "Hotspots" (>1 Ci)
  - Known Burial Coordinates (SRP Records)

SLIDE 6

Soil Samplers

Split Barrel

Sample Size:

- 4.75-cm ID (2-inches) x 0.61 meters (24-inches)
- Standard Soil Sampling Tool
- Easy Soil Removal

Shelby Tube

Larger Dimensions:

- 7.6-cm ID (3-inches) x 0.76 meters (30-inches)
- Thin-Walled
- Higher Soil Recovery
- More Difficult Soil Removal

SLIDE 7

SCHEMATIC OF TEST CORE LAYOUT

SLIDE 8

Average Soil Recovery of Different Coring Tools

<u>2-in. Split-Barrel</u>		<u>3-in. Shelby Tube</u>	
<u>Depth</u>	<u>% Recovery</u>	<u>Depth</u>	<u>% Recovery</u>
0-2'	75.0	0-2.5'	95.0
2-4'	58.3	2.5-5.0'	66.6
4-6'	42.4	5.0-7.5'	49.2
6-8'	31.9	7.5-10.0'	56.7
8-10'	29.2	10.0-12.5'	69.2
10-12'	34.7	12.5-15.0'	78.3
12-14'	58.3	15.0-17.5'	80.8
14-16'	20.2	17.5-20.0'	100.0
16-18'	39.6	20.0-22.5'	100.0
18-20'	73.6	22.5-25.0'	100.0
20-22'	91.7		
22-24'	89.6		
Average Recovery 53.7±24.3%		Average Recovery 79.6±18.9%	
Average Recovery 39.3±13.5%		Average Recovery 75.1±18.5%	
between 2-18'		between 2-18'	

CORE 19-81

11-13 FT.

CORE 22-254

18-20 FT

SLIDE 11

CORING DATA SUMMARY

	<u>Survey Phase</u> <u>(31 Trenches)</u>	<u>Resample Phase</u> <u>(One Trench)</u>
Number of Cores Collected	107	19
Number Segments Collected	930	159
Linear Feet of Trench Sampled	15,000	100
% Segments Having Waste Strikes*	28	46
% Segments Having Activity (>200 c/m)	11	13
% Soil Recovery**	-	48

* Segments of 4 to 20 ft depth horizontal only

** Recovery Relative to Total Volume Cored - 0 to 20 ft Depth

SLIDE 12

**SOIL RECOVERY FROM RESAMPLING PHASE
vs OBSERVED WASTE STRIKES IN SINGLE CORE**

SLIDE 13

SOIL/WASTE RECOVERIES vs TRENCH DEPTH

SLIDE 14

Distribution of Waste/Activity in
Alpha Waste Trenches

<u>Depth, ft</u>	<u>% Distribution of Waste Strikes</u>		<u>% Distribution of Activity Strikes</u>	
	<u>Survey</u>	<u>Resampling</u>	<u>Survey</u>	<u>Resampling</u>
0 to 2	-	-	-	-
2 to 4	<1	1	1	-
4 to 6	8	9	5	6
6 to 8	11	15	7	13
8 to 10	11	13	11	13
10 to 12	13	18	18	6
12 to 14	13	13	16	19
14 to 16	12	15	14	25
16 to 18	14	12	12	13
18 to 20	13	4	11	6
20 to 22	5	-	5	-

SLIDE 15

COMPOSITION OF WASTE RECOVERED

<u>Material Type</u>	<u>Waste Fragments, Per Cent of Total Number</u>	
	<u>Survey Phase</u>	<u>Resampling Phase</u>
Rubber	36	35
Plastic	27	29
Paper	17	22
Metal	9	1
Glass	5	9
Wood	5	3
Cloth	1	0

SLIDE 16

Distribution of Paper and Wood Waste
Strikes as A Function of Trench Depth*

<u>Depth, ft</u>	<u>Per Cent of Observed Waste Strikes</u>	
	<u>Paper</u>	<u>Wood</u>
4 to 6	2	8
6 to 8	10	8
8 to 10	10	15
10 to 12	15	22
12 to 14	25	31
14 to 16	12	-
18 to 20	12	8
20 to 22	2	-
Total Number of Strikes	41	13

*Total Number of Segments
Evaluated: 1089

SLIDE 17

Photograph of Recovered Paper Fragments
From Core 8-215 (20 to 22 Ft. Depth)
Negative No. DPSTF-1-13426-5

SLIDE 18

Photograph of Recovered Wood
Fragments from Core 21-96 (18-20 Ft Depth)
Negative No. DPSTF-1-13660-3

SLIDE 19

SOIL ACTIVITY vs DEPTH IN CORE SAMPLE

SLIDE 20

SOIL ACTIVITY vs DEPTH IN CORE SAMPLE

21L-27

SLIDE 22

Average Measured Soil Activity
Vs. Trench Depth - Resampling Phase

<u>Depth, ft</u>	<u>Measured Activity, p Ci/g</u>	<u>Core Segments Measured For Average</u>
0 to 2	-	-
2 to 4	0.16	6
4 to 6	0.37	11
6 to 8	0.50	6
8 to 10	0.83	7
10 to 12	0.81	8
12 to 14	10.9	8
14 to 16	8.2	6
16 to 18	12.9	6
18 to 20	8.5	3
20 to 22	0.23	1

SLIDE 23

Horizontal Distribution of High Activity
Waste Strikes in Trenches

Location:	<u>Center</u>	<u>South*</u>	<u>West*</u>	<u>North*</u>	<u>East*</u>
<u>Trench 1</u>					
Highest Probable Activity, (cts/min)	200,000	None	300	None	1000
Highest Soil Activity, p Ci/g	405	-	2	-	38
% Cores with Waste	35	50	50	75	75
<u>Trench 2</u>					
Highest Probable Activity (cts/min)	100,000	None	None	None	None
Highest Soil Activity, p Ci/g	-	-	-	-	-
% Cores with Waste	50	25	10	50	25

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*Peripheral Cores Taken at One-Foot Radius Around Center Core

SLIDE 24

PLUTONIUM DISTRIBUTION ON ALPHA TRENCH SOILS

<u>Fraction</u>	<u>% of Soil</u>	<u>% of Pu</u>
Sand	55.7 ± 5.2	11.4 ± 6.6
Silt	5.2 ± 1.2	10.4 ± 3.6
Clay	39.1 ± 5.4	75.8 ± 5.7
Supernate	-	2.4 ± 1.6

SLIDE 25

Plutonium Speciation in Burial Ground Soil

Percent of Pu in Fraction

<u>Sample</u>	<u>MgCl₂</u>	<u>Dithionite</u>	<u>NaOH</u>	<u>Residue</u>
C-17	0.3	80	0.3	5.4
Core, 4,8-10'	0.6	79	0.5	10.7
" 4,10-12'	1.8	82	1.9	4.0
" 5,0-2'	3.4	76	2.3	4.2
" 7,0-2'	6.3	73	6.9	20.0

SLIDE 26

Alpha-Trench Soil Leaching Tests

<u>Leach Solution</u>	<u>% of Pu Removed in Leach</u>
Dist. H ₂ O	0.7 ± 1.1
0.1N HNO ₃	13.4 ± 12.0
0.1N HCl	14.1 ± 10.0
1N NH ₄ OAc	1.1 ± 1.6
1M (NH ₄) ₂ CO ₃ + 0.01M EDTA	53.5 ± 18.4

SLIDE 27

Summary

- Leaching and Migration of TRU Nuclides in Soil From Wastes Buried 15 to 25 Years are minimal.
- Plutonium Concentrations in Soil are Mostly $<10\text{nCi/g}$.
- Recovered Waste Types are Representative of Expected Plant and Laboratory Wastes.
- High-Activity Sites are Highly Localized.
- Wastes are Surprisingly Well-Preserved.
- Plutonium is sorbed on Metallic Oxide Coatings of Soil Clays.

SLIDE 28

Experimental Programs at SRL/SRP Supporting
Dose-to-Man Program

Field Studies (Leaching/migration in Natural Environment)

Lysimeters

Vegetation Uptake

Well Monitoring

Trench Coring

Lab Studies (Mechanisms Controlling Leaching/Migration)

Soil/Waste Chemistry

Mini-Lysimeters/Soil Columns