ANALYSIS OF ENVIRONMENTAL SAMPLES FROM THE MOUND LAB. AREA
- Progress Report -

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ABSTRACT

All of the available analytical results from the 8 sediment cores and 16 water samples obtained from water bodies in the vicinity of Mound Laboratory during July, 1974, are reported here. These include Pu^{238} in both sediment and water, Am^{241} in sediment and H^3 in water. The quality of these analyses was found to be acceptable with the exception of an approximately 30% bias in the accuracy of the Pu^{238} data.

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MASTERINTRODUCTION

In July of 1974 a field sampling program was carried out in Miamisburg, Ohio, near Mound Laboratory. The objective of this exercise was to obtain undisturbed, large area sediment cores and water samples from local contaminated water bodies.¹ All of the samples are to be analyzed for Pu^{238} , the major contaminating radionuclide, and some for Pu^{239} , Pu^{241} , Am^{241} and H^3 . Other physical-chemical measurements, such as density, particle size, solubility etc. are being made on selected sediment samples. Hopefully, with these data, useful information concerning the fresh water geochemistry of these transuranics (and tritium) will be obtained.

In all, 8 sediment cores were obtained and divided into 120 depth increments of between 1 and 4 cm thickness. A total of 16 samples

USERDA, HASL ENVIRONMENTAL QUARTERLY
HASL-291 (1975)

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of pond, canal, river and drinking water were taken. The sampling locations are shown diagrammatically in Figure 1. Details of the coring procedure, the equipment and the samples obtained were reported earlier.² Each sediment depth increment was further divided either into two approximate halves (labeled "A" and "B") or into a half ("A") and two quarters ("B" and "C"). Samples of sediment and water have been distributed to a number of laboratories including Woods Hole Oceanographic Institution (WHOI), Oak Ridge National Laboratory (ORNL) and Mound Laboratory in addition to the Health and Safety Laboratory (HASL).

This report summarizes the progress of the analytical programs, to date.

RADIOCHEMICAL ANALYSIS

A. Sediment

To minimize the probability of laboratory contamination during drying, homogenizing and aliquoting of the sediment, we decided to leach the entire "A" half of each incremental sample. These vary from about 50 to 200 grams of dry material. The material is rinsed from the plastic container with distilled water and 7.5N HNO₃, into a 3 liter beaker. The sample is then leached by the standard HASL procedure,³ through step 5. At this point, the volume is in excess

of 2 liters and complete oxidation of the organic matter is difficult and unnecessary. The clear filtered solution is quantitatively aliquoted into appropriate size samples (generally between 2 and 10 ml.) depending upon the anticipated activity concentration. These are analyzed for Pu^{238} and $\text{Pu}^{239+240}$ by essentially the HASL method described in reference (3) starting with step 6. Most of the samples are being analyzed under contract with LFE Environmental Analysis Laboratories, Richmond, California. Ten to twenty percent of the total are being analyzed at HASL. Tables 1A through 1G summarize all completed results.

A procedure for analyzing Pu^{241} in the sediment leach aliquots by liquid scintillation spectrometry is being investigated by HASL's Radiochemistry Division. No final data are available at this time.

Americium²⁴¹ is being analyzed on a limited number of samples by non-destructive counting of its .059 Mev x-ray on a Ge(Li) diode system at HASL. All of the data completed at this time are listed in Table 2.

B. Water

Tritium analysis of the water samples was carried out at HASL by direct liquid scintillation counting of 2 ml aliquots of distillate. The results so far available are listed in Table 3.

Analyses of the water for Pu^{238} , $\text{Pu}^{239+240}$ and Am^{241} are in progress at WHOI. One barrel of each water sample was acidified on arrival at WHOI and small (500 - 1000 ml) aliquots were analyzed for plutonium. These results are listed in Table 4; Am^{241} data from these will be reported later. Similar size aliquots were removed from the duplicate barrel and filtered without prior acidification. These will be analyzed for the same nuclides.

QUALITY CONTROL

The quality of the reported sediment analyses, in this program, is monitored by use of quality control samples, submitted with each batch of unknowns, to the analysts. They are analyzed "blind", that is, the analysts cannot readily differentiate the controls from the unknowns. Four types of control samples are being used:

(a) Solution Blanks are simply aliquots of acid solution, at about the same pH as the real samples; they alert us to contamination problems which occur post leach.

(b) Sediment Blanks are aliquots of a leach solution from a relatively low activity sediment prepared identically to the Miamisburg sediments. These serve to indicate contamination during as well as post leaching.

(c) Standards are solution blanks to which known amounts of Pu^{239} have been added, as a measure of the accuracy of the analysis.

(d) Replicates are two or more aliquots from the same leach solution. In the normal sense, when submitted to one laboratory, the results of replicate analyses indicate the precision. We also submit them to different laboratories as an indication of possible intercalibration differences.

Table 5 lists all of the quality control data so far completed. No serious problems in the quality of the data have appeared in this early stage of the program; in general the results are reassuring although a systematic error of about 30% may be manifested in the accuracy of Pu^{239} sediment results. We cannot be certain as yet whether this reflects a high bias in the analysis or a low one in the standards submitted.

REFERENCES

- (1) Many public announcements were made concerning the offsite contamination.
e.g., Cincinnati Post, October 3, 1974
- (2) Volchok, H. L.
Sediment Sampling Near Mound Laboratory - July 1974
USAEC Report HASL-286, October 1, 1974 (p. I-71)
- (3) Radiochemical Determination of Plutonium in Soil by Leaching
HASL-Procedures Manual (HASL-300)
Edited by John H. Harley, pg. E-Pu-06-01

Figure 1. Water and sediment sampling locations near Mound Laboratory.

Table 1A

Pu²³⁹ in South Pond Sediment - Station 1

<u>Depth Increment</u>	<u>gms leached</u>	<u>pCi/gm</u>	<u>± σ</u>
0 - 2	155	24.8	1.7
2 - 3	58	48.5	2.5
3 - 4			
4 - 5	154	14.2	0.8
5 - 6			
6 - 7	89	23.0	1.6
7 - 8			
8 - 9	151	40.8	1.7
9 - 10			
10 - 11	91	63.8	3.2
11 - 12			
12 - 13	99	92.3	6.5
13 - 14			
14 - 15	103	166	8
15 - 16			
16 - 17	81	112	6
17 - 18			
18 - 19	67	88.5	2.7
19 - 20			
20 - 21	128	70.7	3.5
21 - 22			

Table 1B

Pu²³⁸ in North Pond Sediment - Station 2

<u>Depth Increment (cm)</u>	<u>gms leached</u>	<u>pCi/gm</u>	<u>± σ</u>
0 - 2	155	16.5	1.0
2 - 4			
4 - 6	131	10.2	0.5
6 - 8	145	7.7	0.1
8 - 10			
10 - 12			
12 - 14			
14 - 16			
16 - 18			
18 - 20			
20 - 22			
22 - 24			
24 - 26	216	0.87	0.39

TABEL 1C

Pu²³⁸ in North Pond Sediment - Station 3

<u>Depth Increment</u> (cm)	<u>gms leached</u>	<u>pCi/gm</u>	<u>± σ</u>
0 - 2	148	21.0	1.3
2 - 4			
4 - 6	184	15.6	0.8
6 - 8	137	9.1	0.4
8 - 10			
10 - 12			
12 - 14			
14 - 16			
16 - 18			
18 - 20			
20 - 22			
22 - 24			
24 - 26			

TABLE 1D

Pu²³⁸ in the Great Miami River Sediment - Station 4

<u>Depth Increment (cm)</u>	<u>gms leached</u>	<u>pCi/gm</u>	<u>± σ</u>
0 - 2	218	0.63	0.04
2 - 4			
4 - 6			
6 - 8	164	0.36	0.01
8 - 10	156	0.45	0.03
10 - 12			

TABLE 1E

Pu²³⁹ in the North Canal Sediment - Station 5

<u>Depth Increment (cm)</u>	<u>gms Leached</u>	<u>pCi/cm</u>	<u>± σ</u>
0 - 3	153	230	10
3 - 5			
5 - 7	132	259	18
7 - 9			
9 - 11			
11 - 13			
13 - 15			
15 - 17			
17 - 19			
19 - 21			
21 - 23			
23 - 25			
25 - 27			
27 - 29	125	473	34
29 - 31			

TABLE 1F

Pu²³⁸ in the North Canal Sediment - Station 6

<u>Depth Increment . (cm)</u>	<u>gms leached</u>	<u>pCi/gm</u>	<u>± σ</u>
0 - 2			
2 - 4	102	378	16
4 - 6			
6 - 8			
8 - 10			
10 - 12			
12 - 14			
14 - 16			
16 - 18			
18 - 22			
22 - 26			
26 - 30			
30 - 34			
34 - 38			
38 - 42			
42 - 46			
46 - 50			
50 - 54			

TABLE 1G

Pu²³⁹ in the North Canal Sediment - Station 7

<u>Depth Increment (cm)</u>	<u>gms Leached</u>	<u>pCi/gm</u>	<u>± σ</u>
0 - 3	131	335	13
3 - 5	102	438	39
5 - 7			
7 - 9	139	589	18
9 - 11			
11 - 13			
13 - 15	135	504	25
15 - 17			
17 - 19	211	678	27
19 - 23			
23 - 27			
27 - 31			
31 - 35			

TABLE 2

Am²⁴¹ in North Canal Sediment

<u>Station No.</u>	<u>Depth Increment (cm)</u>	<u>pCi/cm</u>	<u>± σ</u>
5	0 - 3	.216	.013
5	5 - 7	.230	.012
5	27 - 29	.250	.018
6	2 - 4	.237	.021
6	30 - 34	2.47	.07
7	0 - 3	.099	.030
7	3 - 5	.248	.022
8	39 - 43	3.18	.25

TABLE 3

Tritium Concentrations in Mound Area Water Samples

<u>Sample</u>	<u>Station</u>	<u>nCi/l¹</u>	<u>± c</u>
<u>Great Miami River</u>	4A	<0.7	
" " "	4B	<0.3	
<u>South Pond</u>	1A	49	5
" "	1B	49	5
" "	1C	48	5
<u>North Pond</u>	2A	49	5
" "	2B	49	5
" "	3	49	5
<u>North Canal</u>	5	50	5
" "	6A	48	5
" "	6B	48	5
" "	7	45	5
" "	8A	55	5
" "	8B	50	5
<u>Drinking Water</u>	-	11	2

¹All values are average of 2 determinations.

TABLE 4

Pu Concentrations in Mound Area Water Samples

Sample	Station	$\frac{^{239}\text{Pu}}{^{235}\text{Pu}} \pm \sigma$	
		^{239}Pu	^{235}Pu
Great Miami River	4A	$5 \times 10^{-3} \pm .5 \times 10^{-3}$	$9 \times 10^{-5} \pm 5 \times 10^{-5}$
North Pond	3	$.14 \pm .01$	$< .5 \times 10^{-3}$
" "	2A	$.31 \pm .03$	$< .5 \times 10^{-3}$
South Pond	1A	$.54 \pm .03$	$< .3 \times 10^{-3}$
North Pond	5	$2.9 \pm .2$	$70 \times 10^{-3} \pm 50 \times 10^{-3}$
" "	6A	$7.3 \pm .3$	$< .5 \times 10^{-3}$
" "	7	$2.3 \pm .1$	$5 \times 10^{-3} \pm 3 \times 10^{-3}$
" "	8A	$7.2 \pm .2$	$14 \times 10^{-3} \pm 5 \times 10^{-3}$

¹ Except for the river, stations are listed in north to south order.

TABLE 5

The Quality of the AnalysesA. Blank Solutions

<u>HASL No.</u>	<u>Lab</u>	<u>dpm/Sample</u>	
		<u>Pu²³⁸</u>	<u>Pu²³⁹</u>
SE-9	LFE	.18±.05	< .04
SE-13	LFE	.06±.03	.04±.03
SE-29	HASL	.15±.02	.02±.02
SE-37	LFE	.20±.10	< .04

B. "Blank" Sediment

SE-8	LFE	.13±.04	1.4±.1
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C. Standards

<u>HASL No.</u>	<u>Lab</u>	<u>dpm Pu²³⁸</u>	
		<u>Added</u>	<u>Found</u>
SE-15	LFE	10.3	13.9±0.6
SE-27	HASL	19.3	22.8±1.1
SE-40	LFE	22.3	31.1±3.1

D. Replicates

<u>HASL No.</u>	<u>Station/depth</u>	<u>Lab.</u>	<u>dpm/Pu²³⁸</u>
SE-3	5/0-3	LFE	119±5
SE-6	"	"	117±5
SE-24	"	HASL	116±3
SE-19	4/0-2	LFE	30.0±1.8
SE-20	"	"	30.2±1.8
SE-32	1/12-13	LFE	52.3±1.4
SE-36	"	"	49.3±3.5
SE-14	1/8-9	LFE	34.2±1.4
SE-30	"	"	33.5±2.3