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**Preliminary Inventory of $^{239,240}\text{Pu}$, ^{90}Sr ,
and ^{137}Cs in Waste Pond No. 2 (3513)**

T. Tamura
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J. O. Duguid

ENVIRONMENTAL SCIENCES DIVISION
Publication No. 1039

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PRELIMINARY INVENTORY OF $^{239,240}\text{Pu}$, ^{90}Sr , and ^{137}Cs
IN WASTE POND NO. 2 (3513)

T. Tamura, O. M. Sealand, and J. O. Duguid

ENVIRONMENTAL SCIENCES DIVISION
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ABSTRACT

TAMURA, T., O. M. SEALAND, and J. O. DUGUID. 1977. Preliminary inventory of $^{239,240}\text{Pu}$, ^{90}Sr , and ^{137}Cs in Waste Pond No. 2 (3513). ORNL/TM-5802. Oak Ridge National Laboratory, Oak Ridge, Tennessee. 30 pp.

Preliminary inventory of the $^{239,240}\text{Pu}$, ^{90}Sr , and ^{137}Cs was made in Waste Pond No. 2 (Facility No. 3513) in preparation for decommissioning of the facility. Based on the activity in the recovered cores and the areal distribution of bottom sediments, the inventory of $^{239,240}\text{Pu}$ was approximately 80 g equivalent to 5 Ci. The ^{90}Sr inventory was 0.24 g equivalent to 33.6 Ci. The ^{137}Cs inventory was 2.3 g or 200 Ci.

The least squares fit of the power curve $y = a x^b$ shows that significant relationship exists among the three nuclides in the pond. The coefficient of determination, r^2 , was 0.79 for Cs-Pu, 0.84 for Sr-Pu, and 0.81 for Cs-Sr. The regression lines were determined using all the data sets ($n = 58$); if three suspected points of the Cs-Pu plot are rejected, the r^2 increases to 0.91. Assumptions used to derive the inventory and the radionuclide relationships are discussed.

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Introduction

Plans have been made to discontinue use of Waste Pond No. 2 (3513). This pond has served as a waste-receiving pond since 1944; however, in recent years it has received only the liquid effluent from the low-level treatment plant (Building 3518) and has served as a settling basin for any solids leaving the plant. With a new ion-exchange plant to treat low-level wastes, Waste Pond No. 2 is no longer needed.

The purpose of this survey was to ascertain the amounts of $^{239,240}\text{Pu}$, ^{90}Sr , and ^{137}Cs which are present in the bottom sediment of the pond. The specific objective of the sampling was to establish the amount of $^{239,240}\text{Pu}$ present in order to decide how the pond should be decommissioned. This report contains a preliminary inventory of the plutonium as well as an inventory of the radiostrontium and radiocesium.

Sampling and Analysis

Waste Pond No. 2 is situated between the low-level treatment plant (3518) to the west and two smaller ponds (3539, 3540) to the east. To the north, Waste Pond No. 1 (3524) serves as an equilization pond for low-level waste prior to treatment in the plant (3518). To the south,

White Oak Creek flows westward and receives the overflow from Waste Pond No. 2. The dimensions of the pond estimated from the chain link fence on the top of the bank that forms its perimeter are approximately 70 meters by 76 meters. The dimensions of the pond based on the current pool level, are approximately 67 meters by 67 meters. The pond was constructed with inward sloping sides; the dimensions as measured at the bottom of the slope (at the pond bottom) are approximately 200 feet by 200 feet.

A grid system was designed for sampling the pond. In the north-south direction, a line was drawn through the center of the pond, and two additional lines were drawn 15 meters distance from the center line. The east-west grid was installed with the center line drawn midway across the pond. Two additional lines were drawn 12 meters from the center line; and two more lines drawn 12 meters away from the previous line. This arrangement provided 15 intersections where the core samples were to be taken. The grid system and the core labeling are shown in Fig. 1. Note that the sampling system does not extend to the outer limits of the pond. Furthermore the dimensions shown in Fig. 1 include the flat bottom area of the pond and exclude any slope areas overlain by the normal level of the water. In this initial survey it was felt that more extensive sampling would not be justified but that any further studies should include the sloped sides of the pond.

Aluminum tubing of 3.5 cm (1 3/8 inches) inside diameter was used to core the 15 reconnaissance samples. The tubing was driven into the sediment until the hard bottom of the original floor was reached. When the tubing was removed, the bottom of the core was sealed with a cork and the core was frozen in a bath containing a mixture of diesel fuel

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Fig. 1. Transect lines and identifying code for core samples taken in Waste Pond No. 2.

and dry ice. A tubing larger in diameter than the aluminum coring tube was permanently placed in the freezing mixture; the coring tube was inserted into the larger tubing for freezing. In this way, the cores were not directly in contact with the freezing solution. Upon freezing, the tubing containing the sediment core was double-bagged in plastic and placed in a deep freeze for storage.

To prepare the sediment for analysis, the frozen core was placed on the apron of a sink connected to the radioactive waste disposal system. The double bag that covered the core was slit at the plugged end, and the core was allowed to thaw at room temperature for approximately 30 minutes. This "loosened" the sediment from the wall of the tubing and permitted the sediment to be pushed out with minimum of effort.

The exposed sediment was sliced into segments, depending on visual appearance, which suggested differences in sediment character. In general, the segments consisted of a very loose lightweight surface (zone 1), followed by a dark organic-appearing zone (zone 2), below which was a dark organic-appearing zone intermixed with whitish-gray strata (zone 3); the latter increased in prominence with depth (zone 4) and was followed by a dense compact zone (zone 5). In some cores several zones were missing or were too limited for separate sampling. Each slice was placed in a plastic bottle, weighed, and centrifuged at 3000 rpm for 20 minutes, and the supernate was decanted. The volume of supernate was recorded, and the wet solid was reweighed and then placed in a 105°C oven to dry. The dried weight of the solid was also determined and recorded.

The dried sediment was sent to the Analytical Chemistry Division for plutonium analysis as well as for ^{137}Cs and ^{90}Sr determinations. The $^{239,240}\text{Pu}$ was extracted with hot 8 M HNO_3 and precipitated on lanthanum fluoride. The lanthanum fluoride was transferred to a steel plate and was counted in a gas flow proportional counter. The plate was also counted in an alpha spectrometer, but only a semiquantitative estimate of the $^{239,240}\text{Pu}$ concentration could be obtained. In all samples the results showed that greater than 80% of the alpha activity was associated with an energy of 5.15 MeV, presumably $^{239,240}\text{Pu}$. In order to evaluate the reliability of 80% as a valid proportion of $^{239,240}\text{Pu}$ in the total alpha activity in the sediment, several samples were analyzed for plutonium by the HF-HNO_3 digestion technique with ^{236}Pu tracer as recommended by the USAEC regulatory guide.¹ A comparison of the results of the analyses is shown in Table 1. The values listed in Table 1 include the 80% total alpha activity reported by Analytical Chemistry (AC) as being 5.15 MeV. Measured $^{239,240}\text{Pu}$ activity using a solid state detector in the Environmental Sciences (ES) Division is also shown in the table. The ratios show that the values by AC are both positive and negative relative to the ES values. The mean value of 97.6 has a standard error of about 6%. In calculating the inventory, the data used were determined by Analytical Chemistry using 80% for the plutonium concentration.

The ^{137}Cs analysis was made by counting the dried sediment in a sodium iodide well crystal. Strontium-90 was determined by hot 8 M nitric acid extraction, fuming nitric acid precipitation, purification, and final strontium oxalate precipitate that was counted in an end-

Table 1. Comparison of plutonium analysis by Analytical Chemistry (AC) and by Environmental Sciences (ES) Divisions

Sample Code	AC (dpm/g)	ES (dpm/g)	$\frac{AC}{ES} \times 100$
1-1-1	32,640	32,575	100
-2	10,000	12,330	82
-3	2,110	3,300	64
1-4-2	68,160	52,020	131
-3	8,400	9,015	93
-4	10,815	9,525	114
2-1-1	14,910	20,845	72
-2	5,790	7,185	81
3-3-1	36,800	33,525	110
-2	36,160	33,405	108
-3	22,400	22,625	99
-4	1,085	925	117
			<u>97.6 ± 12.6*</u>

* ± refers to the half-width of the 90% confidence interval.

window proportional counter. Of the three radionuclides, ^{137}Cs analysis was the most reliable since it involved direct gamma counting; ^{90}Sr analysis was more reliable than $^{239,240}\text{Pu}$ since the latter involved alpha detection and the standardization of the analytical procedure was not established. The reliability of the analyses was used in deciding the independent variable when the relationships of the three nuclides were investigated.

Inventory

To obtain the inventory of $^{239,240}\text{Pu}$, and ^{90}Sr (henceforth referred to as plutonium, cesium, and strontium), the activity of each increment of core was determined. These values expressed as disintegrations/minute/gram are shown in Table 2. In addition, the length of each section of core, the gross weight, the volume of water removed after centrifugation, and the oven-dried weight of the increment are given in the table. The relatively low moisture loss of the bottom increment of the cores indicates that the samples represent substrate material. The exceptions are cores 1-3, 2-3, and 2-5. Core 1-3 appears to have been mislabeled, and from the radionuclide and moisture values 1-3-3 represents the bottom increment. Core 2-3 is the shortest core removed (25 inches) and appears not to have penetrated the substrate. Core 2-5 is the longest core recovered (48.3 inches); the lower activity of segment 2-5-5 in plutonium, cesium, and strontium and the higher percentage of dried sediment suggest being near or at the substrate. Segments 2-5-1 and 2-5-2 were inadvertently combined, and the analysis represents these two segments. In calculating the inventory, no corrections were made for the incomplete recovery of cores 2-3 and 2-5.

Table 2. Plutonium, cesium, and strontium concentrations in bottom sediment of waste pond no. 2 (3513)

Sample Code	Section Length (in.)	Gross Weight (g)	Centrifuged Volume of H ₂ O (ml)	Oven Dried Weight (g)	Plutonium (dpm/g)	Cesium (dpm/g)	Strontium (dpm/g)
1-1-1	11	212.65	117	44.10	32,640	1.52×10^6	1.75×10^5
-1	10	324.55	181	81.70	10,100	3.80×10^4	2.09×10^5
-3	7	197.60	39	109.25	2,110	5.28×10^2	1.15×10^3
-4	5	228.70	5.3	167.90	128	9.50×10^2	3.02×10^3
				402.95			
1-2-1	8	313.40	197	46.25	27,040	2.03×10^6	1.07×10^5
-2	8	215.40	112	56.25	13,860	4.69×10^5	6.58×10^4
-3	11.5	349.70	30	251.70	520	1.25×10^4	$\leq 4.80 \times 10^2$
				354.20			
1-3-1	12	280.80	202	33.60	24,960	9.48×10^5	1.48×10^5
-2	13	---	192	87.20	13,330	7.02×10^3	1.43×10^2
-3	8	333.25	48	199.15	≤ 77	3.41×10^3	$\leq 5.69 \times 10^2$
-4	6	199.80	99	40.45	19,840	1.27×10^6	1.28×10^5
				326.80			
1-4-1	8.5	159.35	114	22.02	139,520	3.58×10^5	1.45×10^5
-2	8	166.20	104	22.90	68,160	1.72×10^6	3.01×10^5
-3	6	141.43	84.5	27.85	8,400	7.26×10^5	7.39×10^4
-4	6	97.40	65	14.60	10,815	2.33×10^5	7.20×10^4
-5	6	221.85	56	132.60	1,360	6.11×10^4	1.49×10^4
				219.97			

∞

Table 2 (contd)

Sample Code	Section Length (in.)	Gross Weight (g)	Centrifuged Volume of H ₂ O (ml)	Oven Dried Weight (g)	Plutonium (dpm/g)	Cesium (dpm/g)	Strontium (dpm/g)
1-5-1	17	297.20	219	33.65	34,560	2.33×10^6	2.14×10^5
-2	6	177.78	111	43.78	9,170	3.26×10^5	5.29×10^4
-3	6	196.15	52	120.65	2,830	1.52×10^5	2.74×10^4
				<u>198.08</u>			
2-1-1	9	341.2	202	75.20	14,910	5.63×10^5	9.80×10^4
-2	6	127.5	61	30.55	5,790	1.01×10^6	6.80×10^4
-3	7.5	130.5	36	44.90	5,070	9.96×10^4	8.53×10^3
-4	7.5	337.0	6.2	194.40	337	1.16×10^3	$\leq 3.99 \times 10^2$
				<u>345.05</u>			
2-2-1	12	232.2	152	30.00	34,560	2.62×10^6	3.04×10^5
-2	10	253.8	171	45.55	15,730	3.64×10^6	9.79×10^4
-3	6	225.2	89	93.10	2,930	2.14×10^5	2.86×10^4
-4	6	257.0	2.8	213.55	≤ 179	1.05×10^3	$\leq 3.18 \times 10^2$
				<u>382.20</u>			
2-3-1	6	251.6	174	35.70	35,200	1.15×10^6	2.01×10^5
-2	8	185.0	128	18.55	6,704	5.90×10^5	5.64×10^4
-3	11	127.1	74	24.30	41,120	2.94×10^6	4.67×10^5
				<u>78.55</u>			
2-4-1	5	131.3	98	16.42	8,590	2.68×10^5	1.23×10^5
-2	12.5	226.1	144	40.53	68,320	1.34×10^6	2.02×10^5
-3	6	151.4	74	26.90	18,720	2.14×10^6	2.28×10^5
-4	5	214.9	112	68.78	3,570	8.94×10^4	5.29×10^3
-5	5	245.3	3.2	206.11	≤ 224	5.30×10^3	5.03×10^2
				<u>358.74</u>			

Table 2 (contd)

Sample Code	Section Length (in.)	Gross Weight (g)	Centrifuged Volume of H ₂ O (mL)	Oven Dried Weight (g)	Plutonium (dpm/g)	Cesium (dpm/g)	Strontium (dpm/g)
2-5-1	28.5	---	541	10.67	37,440	1.54×10^6	1.85×10^5
-2							
-3	11.5	213.7	142	33.95	39,520	1.91×10^6	2.06×10^5
-4	3.5	134.0	95	13.15	13,375	3.16×10^6	3.01×10^5
-5	4.8	114.4	61	27.21	7,375	6.22×10^5	7.13×10^4
				<u>84.98</u>			
3-1-1	10	130.8	77	24.20	58,240	1.53×10^6	1.24×10^5
-2	12	291.8	(190)*	24.20	11,230	1.37×10^6	8.31×10^4
-3	8	263.6	62	150.10	≤ 155	5.40×10^3	2.34×10^2
				<u>198.50</u>			
3-2-1	11.8	285.2	229	27.35	8,850	3.81×10^5	1.16×10^5
-2	12	298.4	190	51.40	59,840	2.10×10^6	1.44×10^5
-3	11	231.2	114	44.30	7,665	4.33×10^5	3.12×10^4
-4	8	282.3	58	168.85	655	1.28×10^4	$\leq 2.98 \times 10^2$
				<u>291.90</u>			
3-3-1	10.5	183.6	131	20.35	36,800	1.20×10^6	1.71×10^5
-2	11.5	291.5	185	44.45	36,160	1.20×10^6	2.01×10^5
-3	6	275.6	175	49.40	22,400	9.18×10^4	1.37×10^3
-4	6	316.0	91	181.90	1,084	4.55×10^4	4.84×10^3
				<u>296.10</u>			

Table 2 (contd)

Sample Code	Section Length (in.)	Gross Weight (g)	Centrifuged Volume of H ₂ O (ml)	Oven Dried Weight (g)	Plutonium (dpm/g)	Cesium (dpm/g)	Strontium (dpm/g)
3-4-1	4.5	123.5	80	21.12	8,370	3.72×10^5	6.59×10^5
-2	15.8	225.1	146	48.26	32,640	4.38×10^4	1.49×10^5
-3	3.5	96.6	53	14.84	156,800	2.36×10^5	2.77×10^4
-4	3.5	93.5	56	26.68	1,400	3.17×10^4	2.19×10^3
-5	3.5	84.7	4.5	66.24	≤ 102	5.34×10^3	5.42×10^2
				177.14			
3-5-1	13.8	268.6	159	44.50	32,320	2.50×10^6	2.20×10^5
-2	6	210.2	106	25.80	12,540	6.34×10^5	1.45×10^5
-3	8	328.8	4	237.65	175	7.53×10^3	$\leq 3.09 \times 10^2$
				307.95			

*Number in parentheses obtained from loss in weight of centrifuged soils.

The activity per segment and the total activity per core expressed in microcuries are given in Table 3. The core with the highest plutonium activity is core 1-4; in this core, 2.34 μCi of plutonium was present. The highest cesium activity is 118.18 μCi in core 2-2. The highest strontium activity is 16.91 μCi in core 1-1.

The activity of the core was plotted on the pond transect and contour lines drawn to delineate three levels of plutonium activity, three levels of cesium activity, and four levels of strontium activity. The activity range and the mean activity of each range are shown in Table 4; the contour lines delineating the zones are shown in Figs. 2, 3, and 4 for plutonium, cesium, and strontium, respectively. It should be noted that the sampling did not extend to the outer edges of the pond. The contour lines drawn to the outer perimeter are based on the assumption that the concentration at the perimeter is equal to that at the nearest sampling point on the grid. For a more accurate representation of the nuclide distribution, additional samples should be taken not only at the 200 feet limit but also at the sloping bank beneath the water level.

To determine the inventory of the radionuclides, it is necessary to obtain the areal distribution of each zone. For this distribution, the contour zones were drawn on high-grade tracing paper and the respective zones cut and weighed. The percentage of each zone was calculated from the percentage weight of each zone (Table 4). The total area of the pond was 5.8×10^6 inches²; the area of each core was 1.5 inches².

The calculated inventory of plutonium is 0.9 Ci, 1.5 Ci, and 2.6 Ci, respectively, for the three zones in order of increasing concentrations. The estimate is 5.0 Ci in the pond. The specific activity of ^{239}Pu is

Table 3. Plutonium, Cesium, and Strontium contents in the bottom sediment

Sample Code	Plutonium ($\mu\text{Ci}/\text{section}$)	Cesium ($\mu\text{Ci}/\text{section}$)	Strontium ($\mu\text{Ci}/\text{section}$)
1-1-1	0.63	29.91	3.42
-2	0.37	13.84	7.63
-3	0.10	2.58	5.63
-4	0.01	0.07	0.23
	<u>1.11</u>	<u>46.40</u>	<u>16.91</u>
1-2-1	0.54	41.92	2.21
-2	0.35	11.79	1.65
-3	0.06	1.41	0.05
	<u>0.95</u>	<u>55.12</u>	<u>3.91</u>
1-3-1	0.38	14.20	2.22
-2	0.54	27.32	5.58
-3	0.01	0.30	0.05
-4	0.36	22.95	2.31
	<u>1.29</u>	<u>64.77</u>	<u>10.16</u>
1-4-1	1.38	6.07	1.42
-2	0.71	17.59	3.08
-3	0.10	9.02	0.90
-4	0.07	1.52	0.15
-5	0.08	3.62	0.88
	<u>2.34</u>	<u>37.82</u>	<u>6.43</u>
1-5-1	0.54	35.00	3.21
-2	0.18	6.38	1.04
-3	0.15	8.17	1.47
	<u>0.87</u>	<u>49.55</u>	<u>5.72</u>
2-1-1	0.49	18.88	3.29
-2	0.08	13.75	0.93
-3	0.10	2.00	0.17
-4	0.03	0.10	0.03
	<u>0.71</u>	<u>34.73</u>	<u>4.42</u>
2-2-1	0.45	35.09	4.07
-2	0.32	74.11	1.99
-3	0.12	8.88	1.19
-4	0.02	0.10	0.03
	<u>0.91</u>	<u>118.18</u>	<u>7.28</u>

Table 3 (contd)

Sample Code	Plutonium ($\mu\text{Ci}/\text{section}$)	Cesium ($\mu\text{Ci}/\text{section}$)	Strontium ($\mu\text{Ci}/\text{section}$)
2-3-1	0.54	18.30	3.21
-2	0.05	4.87	0.47
-3	0.45	31.88	5.04
	<u>1.04</u>	<u>55.05</u>	<u>8.72</u>
2-4-1	0.06	1.96	0.90
-2	1.25	24.24	3.66
-3	0.22	25.71	2.74
-4	0.11	2.75	0.16
-5	0.02	0.49	0.05
	<u>1.66</u>	<u>55.15</u>	<u>7.51</u>
2-5-1	0.18	7.32	0.88
-2			
-3	0.58	28.93	3.12
-4	0.08	18.57	1.77
-5	0.09	7.54	0.87
	<u>0.93</u>	<u>62.36</u>	<u>6.64</u>
3-1-1	0.64	16.52	1.34
-2	0.12	14.82	0.90
-3	0.01	0.36	0.02
	<u>0.77</u>	<u>31.70</u>	<u>2.26</u>
3-2-1	0.11	4.64	1.42
-2	1.41	48.21	3.30
-3	0.16	8.57	0.62
-4	0.05	0.96	0.02
	<u>1.73</u>	<u>62.38</u>	<u>5.36</u>
3-3-1	0.34	10.89	1.55
-2	0.73	23.79	3.99
-3	0.50	2.02	3.02
-4	0.09	3.70	0.39
	<u>1.66</u>	<u>40.40</u>	<u>8.95</u>
3-4-1	0.08	3.51	6.21
-2	0.73	0.94	3.21
-3	1.05	1.56	0.18
-4	0.01	0.38	0.03
-5	0.00	0.16	0.02
	<u>1.87</u>	<u>6.55</u>	<u>9.65</u>

Table 3 (contd)

Sample Code	Plutonium ($\mu\text{Ci}/\text{section}$)	Cesium ($\mu\text{Ci}/\text{section}$)	Strontium ($\mu\text{Ci}/\text{section}$)
3-5-1	0.64	49.55	4.37
-2	0.15	7.32	1.67
-3	0.02	0.80	0.03
	<u>0.81</u>	<u>57.67</u>	<u>6.07</u>

Table 4. Selected activity ranges and areal extent of activity zones in Waste Pond No. 2.

		Plutonium	Cesium	Strontium
Zone no.1	Activity range (dpm/core)	$< 2 \times 10^6$	$< 1 \times 10^8$	$< 1 \times 10^7$
	Mean activity (μCi)	0.81	27.7	3.53
	Areal coverage (%)	29.3	22.0	11.1
2	Activity range (dpm/core)	$2 - 3 \times 10^6$	$1 - 2 \times 10^8$	$1 - 2 \times 10^7$
	Mean activity (μCi)	1.06	56.7	6.71
	Areal coverage (%)	35.9	76.1	70.8
3	Activity range (dpm/core)	$> 3 \times 10^6$	$> 2 \times 10^8$	$2 - 3 \times 10^7$
	Mean activity (μCi)	1.85	118.3	9.60
	Areal coverage (%)	34.8	1.9	12.6
4	Activity range (dpm/core)	---	---	$> 3 \times 10^7$
	Mean activity (μCi)			16.9
	Areal coverage (%)			5.5

Fig. 2. Activity distribution of plutonium in sediment of Waste Pond No. 2.

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Fig. 3. Activity distribution of cesium in sediment of Waste Pond No. 2.

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Fig. 4. Activity distribution of strontium in sediment of Waste Pond No. 2.

0.0613 Ci/g or 16.3 g/Ci; the estimated mass of plutonium then is $5.0 \times 16.3 = 81.5$ g.

The calculated inventory of cesium is 23.8 Ci, 168.2 Ci, and 8.8 Ci, respectively, for the three zones. The estimated total is 200.8 Ci. The specific activity of ^{137}Cs is 87.0 Ci/g or 0.0115 g/Ci; the estimated total mass of cesium then is $200.8 \times 0.0115 = 2.3$ g.

The calculated inventory of strontium is 1.5 Ci, 23.8 Ci, 4.7 Ci, and 3.6 Ci, respectively, for the four zones. The estimated total is 33.6 Ci in the pond. The specific activity of ^{90}Sr is 141 Ci/g or 0.0071 g/Ci; the estimated total mass of strontium then is $33.6 \times 0.0071 = 0.24$ g.

According to the Radioactive Waste Management Guide on fissionable material, waste containing concentrations of ^{239}Pu above 10 $\mu\text{Ci/kg}$ must be stored to permit retrieval.² To estimate the concentration on the basis of weight, the activity was calculated on the basis of recovered core weight. For this calculation the contribution of the bottom segment was omitted if the plutonium activity per gram was less than 10% of the next upper zone. In these cases the bottom section represents the original bed of the pond or the limestone liner. An exception is core 1-3; it is believed the last two sections had been mislabeled, and thus the third rather than the fourth section was omitted in the calculations. The results of the calculations are shown in Table 5. On the basis of the 10 $\mu\text{Ci/kg}$ guideline, the contour representing this concentration was drawn for the pond and is shown in Fig. 5. The distribution suggests that about two-thirds of the pond area exceeds 10 $\mu\text{Ci/kg}$.

Table 5. Plutonium concentration based on weight of sediment in core

Sample Code	Oven Dried Weight of Core (g)	Sediment Weight (g)	Plutonium in Sediment	
			(μCi)	($\mu\text{Ci/kg}$)
1-1	402.95	235.05	1.10	4.7
1-2	354.20	102.50	0.89	8.7
1-3	326.80	127.65	1.28	10.0
1-4	219.97	87.37	2.26	25.9
1-5	198.08	198.08	0.87	4.4
2-1	345.05	150.65	0.68	4.5
2-2	382.20	168.65	0.89	5.3
2-3	78.55	78.55	1.04	13.2
2-4	358.74	152.63	1.64	10.7
2-5	84.98	84.98	0.93	10.9
3-1	198.50	48.40	0.76	15.7
3-2	291.90	123.05	1.68	13.6
3-3	296.10	114.20	1.57	13.8
3-4	177.14	110.90	1.87	16.9
3-5	307.95	70.30	0.79	11.2

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Fig. 5. Distribution of plutonium in Waste Pond No. 2 based on the weight of sediment.

Radionuclide Relationships

Although the major purpose of the reconnaissance survey of the pond was to obtain information on the fissionable plutonium, the data obtained for ^{137}Cs and ^{90}Sr suggest the possible relationships of these nuclides with plutonium. Such relationships, if existing, may be useful in reducing the cost of analysis for plutonium in a given system when additional or future samples are to be analyzed. In addition, if the pond serves as a source term for the plutonium downstream in White Oak Creek, establishing relationships of the nuclides may provide clues or differential leaching or possibly new source terms for radioactivity in the stream.

An arithmetic plot of the data in Table 2 did not show a significant relationship, a logarithmic plot of the data indicated a linear relationship. To establish the best-fit relationship and the coefficient of determination, r^2 , the equation $y = ax^b$ was solved. Cesium concentration was assigned as the independent variable when plotted against strontium and plutonium, and strontium was the independent variable when plotted against plutonium. This assignment was made since the most reliable analysis was cesium and the least reliable was plutonium.*

Figure 6 is a plot of the Cs-Pu relationship. The power curve was converted to the linear form by the expression

$$\ln y = \ln a + b(\ln x) \quad (1)$$

*R. R. Rickard, Analytical Chemistry Division, ORNL, personal communication.

Fig. 6. Relationship of cesium and plutonium in the sediments of ORNL Pond No. 2.

where y is the plutonium concentration, x is the cesium concentration, and a and b are the regression coefficients. The least square regression line for the relationship is

$$Pu = 0.50 Cs^{0.77} \quad (2)$$

and the coefficient of determination, r^2 , is 0.79. The correlation coefficient is $\sqrt{r^2}$, or 0.89. The standard error of estimate of y on x, or the unexplained error, $S_{y.x}$, is 0.90.

Similarly the regression lines for the Sr-Pu and Cs-Sr were determined. For Sr-Pu the line is

$$Pu = 2.32 Sr^{0.75} \quad (3)$$

$$r^2 = 0.84$$

$$S_{y.x} = 0.76$$

and for Cs-Sr the line is

$$Sr = 0.46 Cs^{0.91} \quad (4)$$

$$r^2 = 0.81$$

$$S_{y.x} = 1.0$$

and the scatter diagrams with the regression line included are shown in Figs. 7 and 8. Inspection of Figs. 6, 7, and 8 shows several points considerably off the regression line. By comparing the data of the suspect sample plotted in the three figures, the indicated direction of error is established. Thus in the Cs-Pu relationship (Fig. 6) plotted points lie considerably above the regression line. Inspection of the Sr-Pu and Cs-Sr plots of the same samples suggests that of the three

Fig. 7. Relationship of strontium and plutonium in the sediments of ORNL Pond No. 2.

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Fig. 8. Relationship of cesium and strontium in the sediments of ORNL Pond No. 2.

points, two are due to overestimating the plutonium and the third to underestimating the cesium. The criterion for rejecting the data was arbitrarily set at a deviation greater than three standard errors from the regression line. On this basis the three points in Fig. 2 were considered invalid, and the regression line was computed without these suspected values. The regression line is ($n = 55$):

$$Pu = 0.35 Cs^{0.79} \quad (5)$$

$$r^2 = 0.91$$

$$S_{y.x} = 0.59$$

These relationships suggest that cesium and strontium might be used to estimate the plutonium concentration in the pond. In future surveys such as the Floodplain of White Oak Creek, White Oak Lake and Clinch River, investigators should determine the Cs-Pu and the Sr-Pu relationships. Such relationships could reduce the cost of analysis of later studies, and the regression line equations may show preferential dissolution occurring in different areas or under different weathering conditions.

With two of the plutonium values (samples 1-4-1 and 3-4-3) suspected of being overestimated, the inventory of plutonium in the pond was recalculated. The plutonium concentrations for these two samples were assigned average values based on the regression lines of the Cs-Pu and the Sr-Pu relationships. The recalculated mean concentrations of Zones 1, 2, and 3 (as shown in Table 4) were 0.82, 1.08, and 1.68 μCi , and the respective areas were 34%, 49%, and 17% of the

total area. The inventory of plutonium is 4.2 Ci, or 68 grams of $^{239,240}\text{Pu}$. This value may be compared with 5.0 Ci, or 81 grams, using all measured values of plutonium.

Conclusions

The initial reconnaissance survey of Waste Pond No. 2 revealed the presence of $^{239,240}\text{Pu}$ in the bottom sediment. A considerable portion of the pond sediment exceeds the 10 $\mu\text{Ci/kg}$ level. A portion of the bottom sediment was not evaluated in this survey, and for a more accurate assessment of the plutonium, samples should be taken from the outer perimeter of the pond. Approximately 80 grams of $^{239,240}\text{Pu}$ (5.0 Ci) are estimated to be in the bottom sediment. In addition 2.3 g of ^{137}Cs (200 Ci) and 0.24 g of ^{90}Sr (33 Ci) are estimates to be present in the sediment.

The least squares fit of the power curve $y = a x^b$ shows that a significant relationship exists among the three nuclides in the pond sediment. The coefficient of determination, r^2 , was 0.79 for Cs-Pu, 0.84 for Sr-Pu, and 0.81 for Cs-Sr. The regression lines were determined using all the data sets ($n = 58$); if three suspected points of Cs-Pu relationship are rejected, the r^2 increases to 0.91.

It is recommended that further studies be made of Waste Pond 2. In addition to taking further samples for analyses, the leachability of the radionuclides should be investigated to determine if the elimination of the low-level treatment plant (3518) effluent (causing a reduction in pH of the Pond) will increase the leaching of plutonium and strontium from the sediment.

Reference

1. United States Atomic Energy Commission Regulatory Guide,
"Measurement of Radionuclides in the Environment: Sampling
and Analysis of Plutonium in Soil," Regulatory Guide 4.5,
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2. United States Atomic Energy Commission Manual, "Radioactive
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