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Title: Characterization of Uranium and Plutonium in Surface-Waters and Sediments Collected at the Rocky Flats Facility

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Characterization of Uranium and Plutonium in Surface-Waters and Sediments Collected at the Rocky Flats Facility

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Abstract

This study was initiated to characterize actinides in environmental samples collected at the Rocky Flats Plant (RFP) Thermal Ionization Mass Spectrometry (TIMS) measurement techniques were used to measure the plutonium and uranium content of water and sediment samples collected from the ponds used to control surface-waters on-site at RFP. TIMS was also used to separate the uranium into anthropogenic and naturally occurring components The results of these studies are presented

INTRODUCTION

The Rocky Flats Plant (RFP) is a Department of Energy (DOE) facility where plutonium and uranium components were manufactured for nuclear weapons. During plant operations radioactivity was inadvertently released into the environment. Plutonium has been detected in air. soil and water samples at RFP. The largest single contributor to plutonium in the environment is resuspension of contaminants originating at the OU2/903 Pad.[1] Between January and June 1968 oil drums containing cutting oil contaminated with plutonium were removed from the OU2/903 Pad. Some of these drums had leaked and high winds spread the plutonium. Seed. *et al.* estimate that 6.1 Ci of plutonium were lost to the soil.[2] Uranium releases have also occurred. Actinides may be introduced into surface-waters at RFP by wind deposition of contaminated soil and erosion of topsoil by rain and snow melt A portion of the surface-water collected at RFP is discharged off site. This provides a potential pathway to transport plutonium and uranium into reservoirs that serve as public water supplies. This study was initiated to characterize the plutonium and uranium present in surface-waters at RFP.

SURFACE-WATER HYDROLOGY

Figure 1 illustrates the creeks, ponds and reservoirs in the RFP vicinity. Their functions are described below. Three drainage basins and natural ephemeral streams transverse RFP. The Woman Creek drainage basin traverses and drains the southern portion of the site. The Rock Creek drainage basin drains the northwestern portion of the plant complex. The Walnut Creek drainage basin traverses the western, northern, and northeastern portions of the RFP site. Three ephemeral streams are tributary to Walnut Creek. The streams are Dry Creek, North Walnut Creek and South Walnut Creek.

Figure 2 gives a schematic representation of the surface-water systems at RFP. Dams, detention ponds, diversion structures, and ditches have been constructed at RFP to control the release of plant discharges and surface (storm water) runoff. The ponds located downstream of the plant complex on North Walnut Creek are designated A-1 through A-4. Ponds on South Walnut Creek are designated B-1 through B-5. The ponds in the Woman Creek drainage basin are designated C-1 and C-2.

Ponds A-1 and A-2 are currently used to collect seep and culvert flows and precipitation runoff from the northern area of the plant site. Ponds A-1 and A-2 are now being operated as non-discharge ponds. Volumes of water are controlled by overpond spray evaporation. Pond A-3 collects surface-water diverted around Ponds A-1 and A-2 and a substantial portion of the North Walnut Creek and northern plant site runoff. Pond A-3 is operated in a detain, sample, analyze and release mode. Water released from Pond A-3 is transported to Pond A-4. Ponds B-1 and B-2 are currently used to collect suspect flows or upsets from the sewage treatment plant (STP) operated at RFP. These ponds are currently operated as non-discharge retention ponds. Water from Ponds B-1 and B-2 is transferred to A-2 after characterization. Pond B-3 receives treated effluent from the STP. Water from Pond B-3 is released into Ponds B-4 and B-5. Pond B-4 is operated as a flow-through pond. Pond B-5 is a terminal pond. Pond C-1 is located on Woman Creek and receives natural flows. An interceptor ditch (South Interceptor Ditch) is located between and parallel to Woman Creek and the southern boundary of the plant complex. The South Interceptor Ditch drains into Pond C-2. Any offsite discharge from RFP is from the terminal ponds (Ponds A-4, B-5 or C-2).

Figure 1
Rocky Flats Plant and Surrounding Area Drainage

Rocky Flats Plant and Surrounding Area Drainage

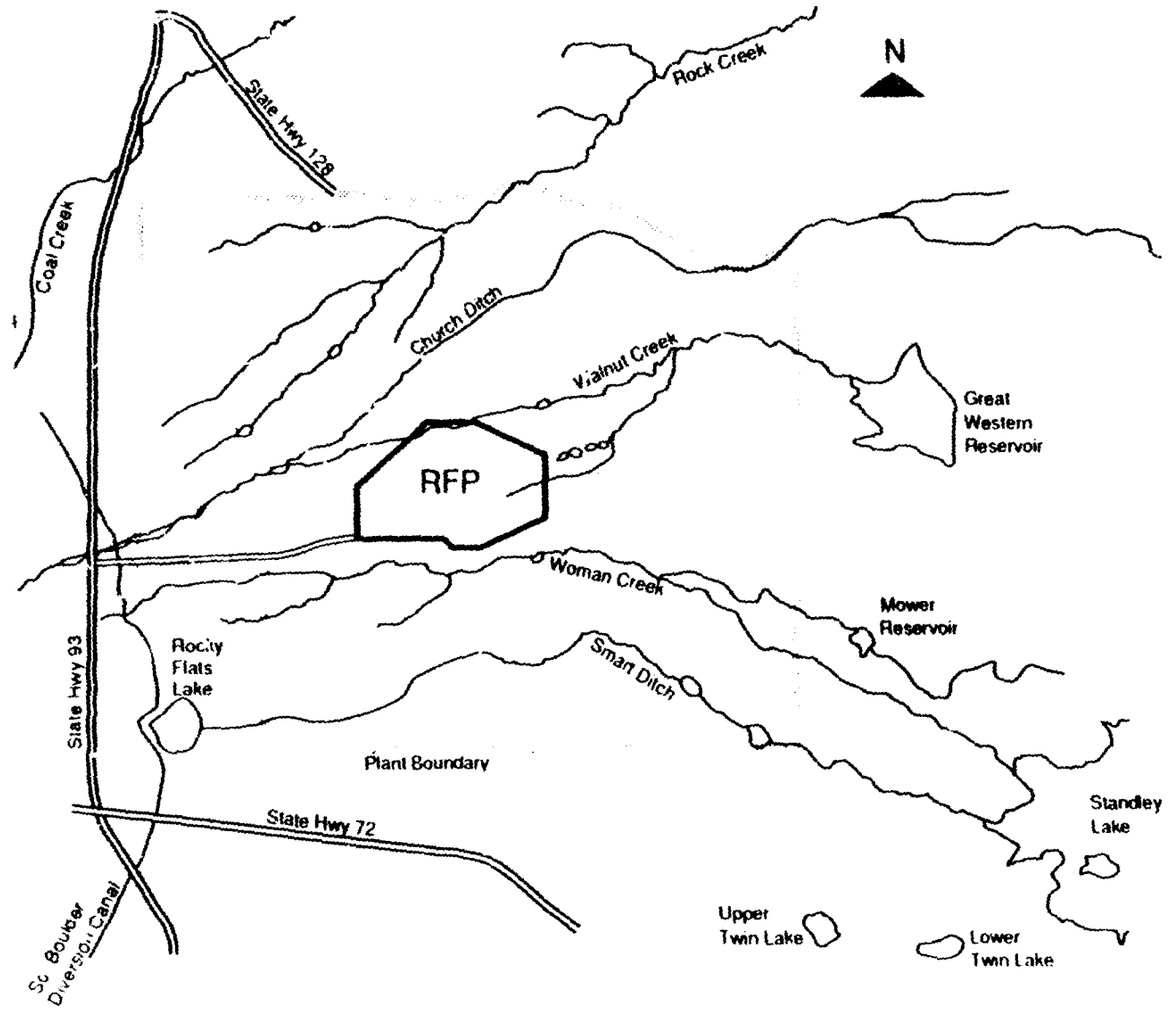
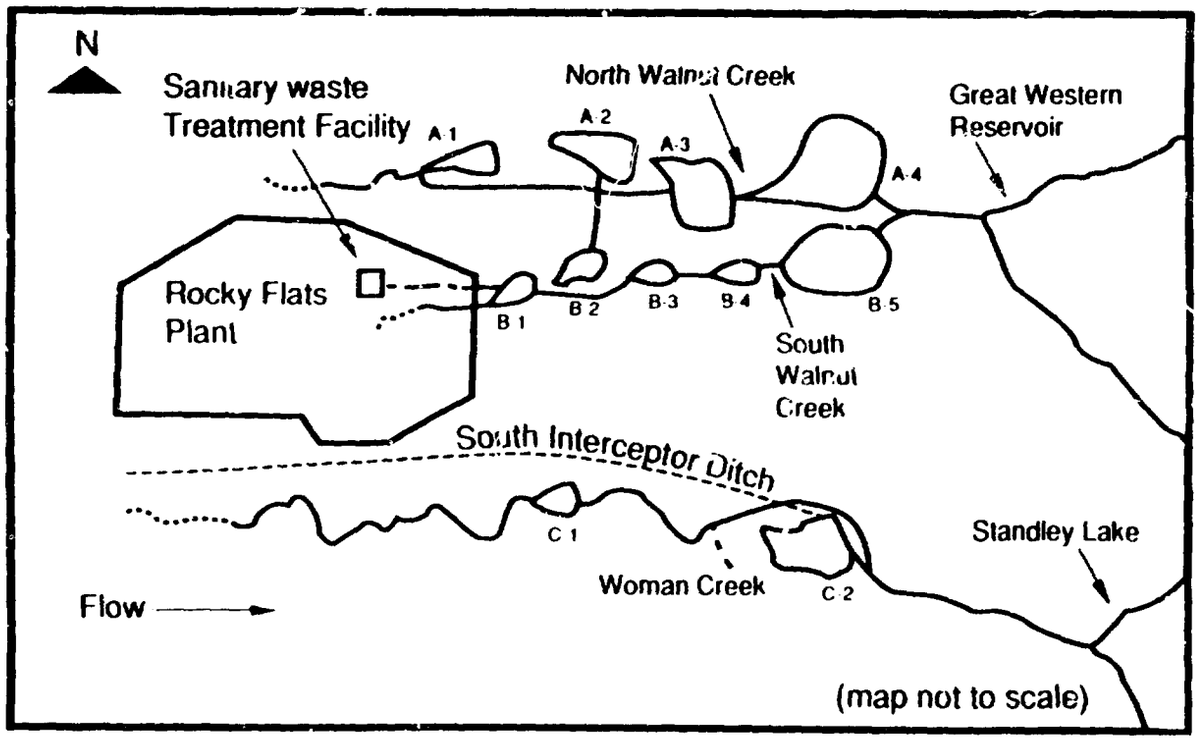


Figure 2
Detailed Schematic of Rocky Flats Plant Pond System

Detailed Schematic of Rocky Flats Plant Pond System



EXPERIMENTAL

Water samples were stabilized in the field with nitric acid. Water samples were collected monthly from Ponds A-4, B-5 and C-2. Ponds A-1, A-2, A-3, B-1, B-2, B-3, B-4 and C-1 were sampled quarterly. Sediment samples were collected from each of the ponds. Uranium and plutonium were quantified by TIMS measurement techniques. Mass spectrometers measure the number of atoms in a sample. The numbers of atoms are converted to activities by means of the relationship:

$$A = N \lambda$$

A is the activity expressed in disintegrations per unit time. N is the number of atoms of the particular radionuclide and λ is the decay constant that is equal to $0.693/t_{1/2}$ ($t_{1/2}$ is the half-life of the radionuclide). Where there is more than one isotope for the element contributing to the total activity, the total activity of the contributing isotopes is determined by the summation of the individual components. The half-lives for the radionuclides used for the calculations are given in Table I.

Table I. Half-lives of Radionuclides Used for Calculating Activities

Radionuclide	Half-Life (years)
Uranium-234	2.450×10^5
Uranium-235	7.040×10^8
Uranium-236	2.342×10^7
Uranium-238	4.468×10^9
Plutonium-239	2.411×10^4
Plutonium-240	6.560×10^3

RESULTS AND DISCUSSION

Table II summarizes the uranium and plutonium alpha activities measured in surface-waters collected from ponds at RFP.

Table II. Alpha Activities Measured In Surface-Waters at RFP

Date Sampled	Pond	Uranium pCi/L	Plutonium pCi/L
02/24/93	A1	0.73	0.0571
05/12/93	A1	2.1[illegible]	0.0133
08/24/93	A1	15.79	0.0261
11/25/92	A2	7.25	0.6572
02/24/93	A2	3.50	0.0257
05/12/93	A2	5.59	0.0032
08/24/93	A2	6.41	0.0059
11/25/92	A3	5.14	0.0251
02/24/93	A3	4.63	0.0030
05/12/93	A3	4.48	0.0037
08/24/93	A3	4.54	0.0195
10/25/92	A4	1.55	0.0016
11/24/92	A4	0.10	0.0061
12/14/92	A4	1.25	0.0091
01/14/93	A4	1.50	0.0022
02/23/93	A4	3.33	0.0010
03/17/93	A4	2.21	0.0025
04/21/93	A4	2.42	0.0036
05/12/93	A4	2.53	0.0014
06/15/93	A4	3.53	0.0026
07/15/93	A4	1.63	0.0014
08/24/93	A4	0.90	0.0179
02/24/92	B1	2.87	0.3130
11/25/92	B1	0.70	2.9334
05/17/93	B1	3.40	0.1175
08/24/93	B1	3.80	0.6855
11/25/92	B2	6.95	3.0902
02/24/93	B2	4.79	0.0190
05/17/93	B2	10.97	0.2180
08/24/93	B2	5.36	0.3526
11/25/92	B3	0.10	0.1582
02/24/93	B3	0.47	0.1020
05/13/93	B3	0.32	0.0144
08/24/93	B3	0.20	0.2285
02/24/93	B4	1.14	0.0777
05/13/93	B4	1.46	0.0352
08/24/93	B4	0.20	0.0465
10/23/92	B5	0.65	0.0055
11/24/92	B5	0.67	0.0111
12/14/92	B5	0.66	0.0062
01/14/93	B5	0.78	0.0078
02/23/93	B5	0.93	0.0058
03/17/93	B5	1.05	0.0043
04/21/93	B5	1.75	0.0101
05/13/93	B5	1.08	0.0022
06/15/93	B5	0.41	0.0018
07/15/93	B5	0.65	0.0033
08/23/93	B5	0.59	0.0057
12/13/92	C1	1.59	0.0049
02/23/93	C1	1.85	0.0084
05/13/93	C1	1.48	0.0073
08/23/93	C1	1.96	0.0566
10/21/92	C2	1.90	0.0547
11/25/92	C2	1.98	0.0270
12/14/92	C2	2.12	0.0241
01/14/93	C2	2.37	0.0142
02/23/93	C2	1.43	0.0043
03/17/93	C2	1.23	0.0204
04/15/93	C2	2.68	0.0181
05/13/93	C2	3.20	0.0305
06/15/93	C2	1.96	0.0403
07/15/93	C2	2.91	0.0289
08/23/93	C2	2.50	0.0968

Uranium

The uranium activities measured in the RFP surface-water samples are higher than the uranium activities measured in some of the reservoirs in the vicinity. The uranium activity levels in the surface-waters collected at RFP are lower than the uranium activities in some wells in the Colorado. Table III reports the highest uranium concentration measured in the RFP ponds during this study. The uranium concentrations in the local reservoirs are the mean uranium concentrations measured by RFP in 1988.[6] The values reported for the selected wells and springs in Colorado were single measurements.[7,8]

The presence of ^{236}U in the surface-water samples collected at RFP and the variable $^{238}U/^{235}U$ atom ratios detected in water samples collected from the holding ponds prove that anthropogenic uranium is present. Table IV summarizes the atom percent of ^{234}U, ^{235}U, ^{236}U and ^{238}U present in each surface-water sample collected at RFP.

Table III. Uranium Concentrations in Waters

Location	Description	Uranium content pCi/L
Pond A-1	RFP Holding Pond	15.79
Pond A-2	RFP Holding Pond	7.25
Pond A-3	RFP Holding Pond	5.14
Pond A-4	RFP Holding Pond	3.53
Pond B-1	RFP Holding Pond	3.80
Pond B-2	RFP Holding Pond	10.97
Pond B-3	RFP Holding Pond	0.47
Pond B-4	RFP Holding Pond	1.46
Pond B-5	RFP Holding Pond	1.75
Pond C-1	RFP Holding Pond	1.96
Pond C-2	RFP Holding Pond	3.20
Dillon	Reservoir	0.5
Great Western	Reservoir	2.05
Ralston	Reservoir	0.9
Standley	Reservoir	1.8
*La Junta	Well	497
*Pueblo	Well	168

* The wells listed in Table III do not serve as public water supplies.

T ble IV. Atom Percent of Uranium Isotopes Present in Surface-Waters

Date Sampled	Pond	% U-234	% U-235	% U 236	% U-238
Natural Uranium		0.0057	0.7204	0.0000	99.2739
02/29/93	A1	0 0040	0.7138	0.0020	99.2802
05/12/93	A1	0.0045	0.5549	0.0018	99.4388
08/24/93	A1	0.0052	0.4339	0.0020	99.5589
11/25/92	A2	0.0037	0.4869	0.0019	99.5075
02/24/93	A2	0.0040	0.5568	0.0018	99.4373
05/12/93	A2	0.0045	0.5652	0.0018	99.4284
08/24/93	A2	0.0043	0.4259	0.0018	99.4777
11/25/92	A3	0.0037	0.4477	0.0017	99.5520
02/29/93	A3	0.0042	0.5385	0.0016	99.4557
05/12/93	A3	0.0041	0.4897	0.0017	99.5045
08/24/93	A3	0.0039	0.4801	0.0018	99.5142
10/23/92	A4	0.0046	0.5377	0.0011	99.4566
11/24/92	A4	0.0020	0.6604	0.0003	99.3373
12/14/92	A4	0.0046	0.5798	0.0011	99.4144
01/14/93	A4	0.0055	0.5827	0.0012	99.4107
02/23/93	A4	0.0047	0.5490	0.0014	99.4448
03/17/93	A4	0.0046	0.5425	0.0014	99.4516
04/21/93	A4	0.0049	0.5588	0.0012	99.4351
05/12/93	A4	0.0052	0.5693	0.0012	99.4243
06/15/93	A4	0.0049	0.5445	0.0017	99.4488
07/15/93	A4	0.0045	0.5417	0.0011	99.4527
08/23/93	A4	0.0053	0.6001	0.0012	99.3935
11/25/92	B1	0.0064	0.6649	0.0006	99.3282
02/24/93	B1	0.0068	0.6573	0.0010	99.3348
05/13/93	B1	0.0064	0.6473	0.0011	99.3451
08/24/93	B1	0.0059	0.6340	0.0014	99.3588
11/25/92	B2	0.0068	0.7225	0.0014	99.2693
02/23/92	B2	0.0050	0.5553	0.0015	99.4382
05/13/93	B2	0.0048	0.5400	0.0024	99.4528
11/25/92	B3	0.0030	0.6737	0.0017	99.3216
02/24/93	B3	0.0045	0.6349	0.0011	99.3595
05/13/93	B3	0.0055	0.6018	0.0010	99.3916
08/24/93	B3	0.0091	0.6143	0.0009	99.3757
11/25/92	B4	0.0060	0. 6462	0.0011	99.3468
02/24/93	B4	0.0049	0.6290	0.0011	99.3650
05/13/93	B4	0.0045	0.6229	0.0011	99.3715
08/24/93	B4	0.0043	0.6480	0.0018	99.3458

Table IV. Atom Percent of Uranium Isotopes Present in Surface-Waters

Date Sampled	Pond	% U-234	% U-235	% U-236	% U-238
Natural Uranium		0.0057	0.7204	0.0000	99.2739
10/23/93	B5	0.0049	0.6716	0.0006	99.3229
11/24/92	B5	0.0056	0.6788	0.0005	99.3153
12/14/92	B5	0.0065	0.6721	0.0001	99.3213
01/14/93	B5	0.0058	0.6413	0.0009	99.3521
02/23/93	B5	0.0057	0.6508	0.0005	99.3341
03/17/93	B5	0.0059	0.6615	0.0007	99.3319
04/21/93	B5	0.0059	0.6568	0.0007	99.3366
05/13/93	B5	0.0058	0.6593	0.0007	99.3342
06/15/93	B5	0.0042	0.5679	0.0008	99.4272
07/15/93	B5	0.0052	0.6569	0.0011	99.3368
08/23/93	B5	0.0050	0.6728	0.0006	99.3216
12/14/92	C1	0.0078	0.7175	0.0000	99.2747
02/24/93	C1	0.0074	0.7170	0.0002	99.2754
05/13/93	C1	0.0073	0.7242	-----	99.2684
08/23/93	C1	0.0076	0.7176	0.0000	99.2748
10/23/92	C2	0.0046	0.5242	0.0018	99.4694
11/25/92	C2	0.0044	0.5308	0.0020	99.4628
12/14/92	C2	0.0045	0.5552	0.0017	99.4386
01/14/93	C2	0.0048	0.5370	0.0018	99.4564
02/23/93	C2	0.0049	0.5534	0.0017	99.4400
03/17/93	C2	----	0.5502	-----	99.4498
04/15/93	C2	0.0049	0.5471	0.0016	99.4464
05/13/93	C2	0.0048	0.5465	0.0016	99.4471
06/15/93	C2	0.0045	0.5668	0.0011	99.4276
07/15/93	C2	0.0048	0.5469	0.0016	99.4466
08/24/93	C2	0.0048	0.5426	0.0020	99.4471

Naturally occurring uranium contains 0.7204 atom percent ^{235}U. Most of the surface-water samples measured at RFP were depleted in ^{235}U. The amount of ^{235}U present in depleted uranium varies. The majority of depleted uranium produced in this country contains 0.2% ^{235}U to 0.5% ^{235}U.[9] Table V illustrates the maximum amount of depleted uranium that is present in each water sample collected at RFP. The values derived assume a simple two component system. The data reported in Table V assume that the depleted uranium released by RFP contains 0.5% ^{235}U. This conservative estimate determines the maximum amount of depleted uranium present in the surface-waters.

Estimates are made using the following relationship:

$$(N238/N235)obs = \{(N238/N235)dep\}\{F\} + \{(N238/N235)nat\}\{1-F\}$$

(N238/N235)obs is the $^{238}U/^{235}U$ atom ratio measured in the sample, (N238/N235)dep is the $^{238}U/^{235}U$ atom ratio in the depleted uranium and (N238/N235)nat is the $^{238}U/^{235}U$ atom ratio in naturally occurring uranium. F is the fraction of depleted uranium in the sample and {1-F} is the fraction of the uranium that is naturally occurring.

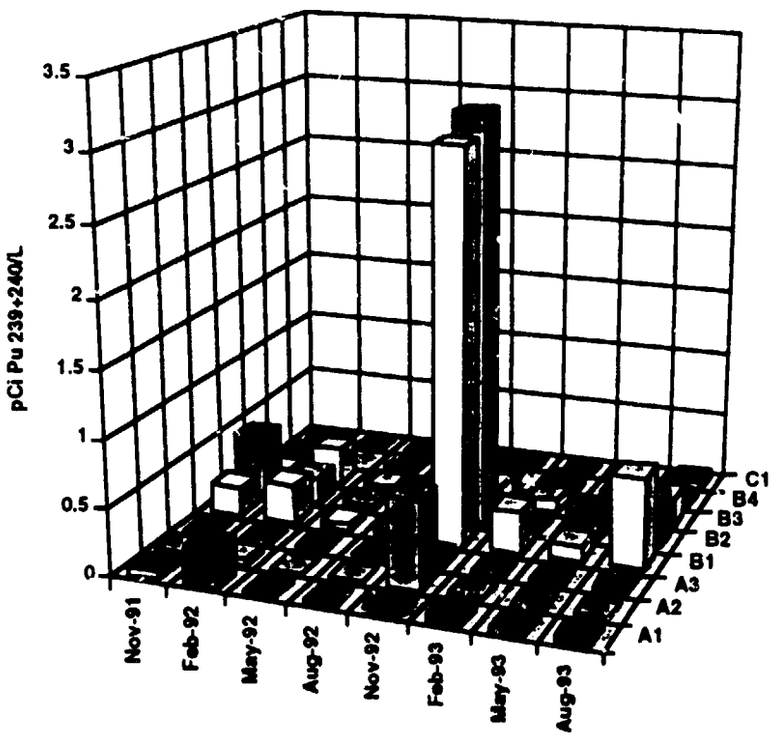
Table V. Maximum Percentage of Uranium in Water Attributable to Release of Depleted Uranium at RFP

Date Sampled	Pond	Maximum Percentage of Depleted Uranium	Date Sampled	Pond	Maximum Percentage of Depleted Uranium
02/29/93	A1	2	05/13/93	B3	35
05/12/93	A1	50	08/24/93	B3	36
08/24/93	A1	100	11/25/92	B4	19
11/25/92	A2	81	05/13/93	B4	26
05/12/93	A2	46	02/29/93	B4	25
02/29/93	A2	50	08/24/93	B4	19
08/24/93	A2	66	10/23/92	B5	13
11/25/92	A3	100	11/24/92	B5	10
02/29/93	A3	57	12/14/92	B5	12
05/12/93	A3	80	01/14/93	B5	21
08/24/93	A3	100	02/23/93	B5	18
10/23/92	A4	81	03/17/93	B5	18
11/24/92	A4	17	04/21/93	B5	17
12/14/92	A4	41	05/13/93	B5	16
01/14/93	A4	40	06/15/93	B5	46
02/23/93	A4	53	07/15/93	B5	16
03/17/93	A4	58	08/23/93	B5	12
04/21/93	A4	49	12/03/92	C1	1
05/12/93	A4	45	05/13/93	C1	0
06/15/93	A4	54	02/23/93	C1	1
07/15/93	A4	56	08/23/93	C1	0
08/23/93	A4	31	10/23/92	C2	63
11/25/92	B1	14	11/25/92	C2	60
05/13/93	B1	23	12/14/92	C2	51
02/24/93	B1	17	01/14/93	C2	58
08/24/93	B1	23	02/23/93	C2	51
11/25/92	B2	0	03/17/93	C2	52
05/13/93	B2	57	04/15/93	C2	53
02/24/93	B2	50	05/13/93	C2	53
08/24/93	B2	56	06/15/93	C2	46
11/25/92	B3	12	07/15/93	C2	51
02/24/93	B3	23	08/24/93	C2	54

Plutonium

The plutonium content of Ponds A-1, A-2, A-3, B-1, B-2, B-3, B-4 and C-1 was measured quarterly. The results are presented in Figure 3.

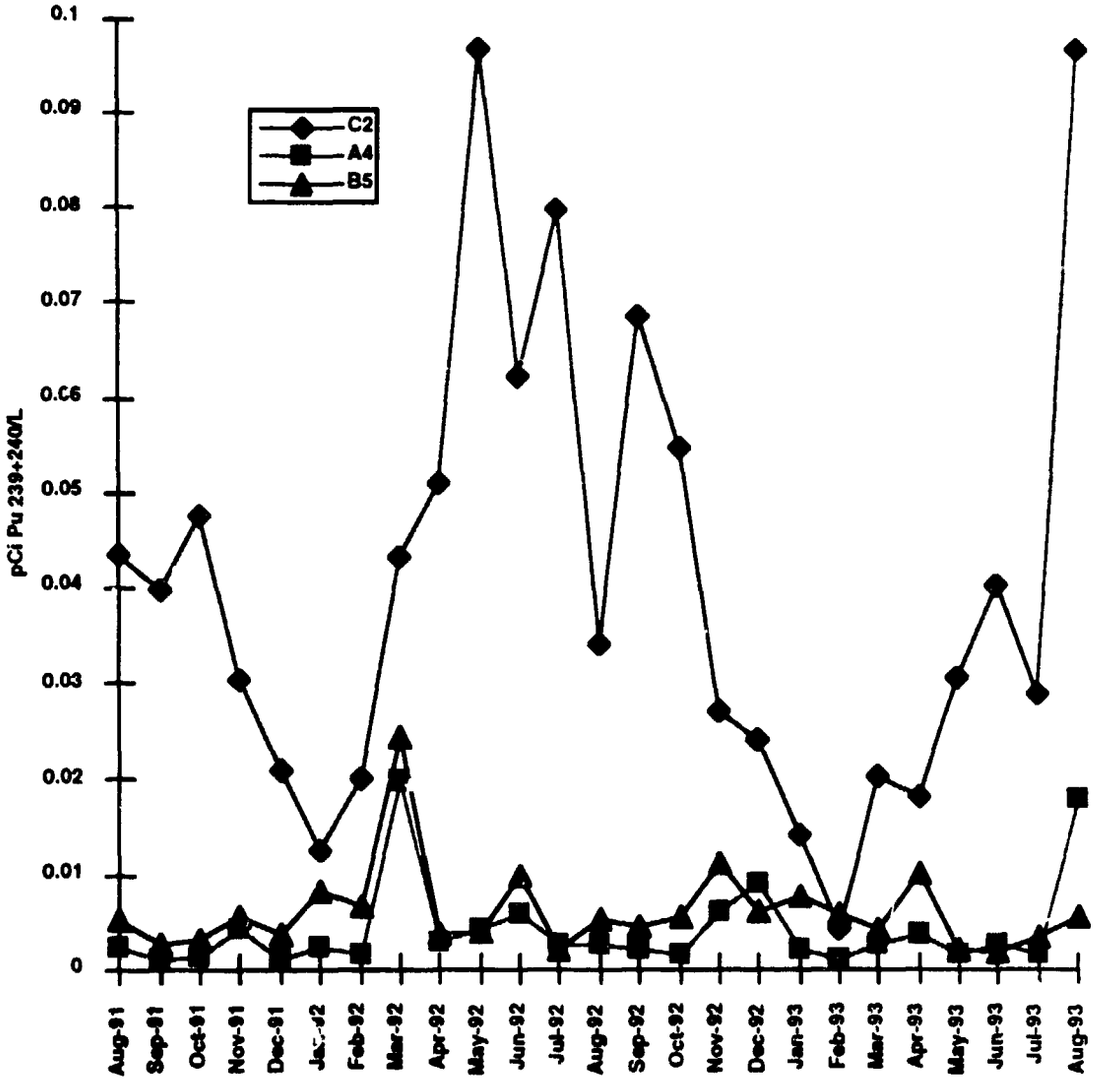
Figure 3.
Plutonium Concentrations in Holding Ponds at RFP that are Analyzed Quarterly



The plutonium concentrations observed in Ponds A-2, B-1 and B-2 during November, 1992 are significantly higher than any other measurements of RFP pond waters. These samples contained significant (multiple grams) quantities of sediments. The sediments in the ponds contain more plutonium on a per gram basis than the waters. Sediments may be re-suspended by wind or they may be incorporated during sampling. Inclusion of sediments in water samples can bias analytical results.

Ponds A-4, B-5, C-2 were sampled monthly. The plutonium concentrations in these ponds are presented in Figure 4.

Figure 4
Plutonium Concentrations in Locations Analyzed Monthly



There appears to be a seasonal variation in the plutonium concentrations in Pond C-2. Plutonium concentrations are higher in the spring and summer than winter.

Sediment samples were collected from each of the ponds at RFP. The samples were analyzed for uranium and plutonium content. The results are summarized in Table VI.

Table VI. Actinide Elements Detected in Sediments and Soils

Description	Uranium pCi/g	Plutonium pCi/g
A-1 Pond Sediment	4.37	2.375
A-2 Pond Sediment	6.01	2.829
A-3 Pond Sediment	1.67	0.674
A-4 Pond Sediment	1.17	0.105
B-1 Pond Sediment	8.74	152.256
B-2 Pond Sediment	4.16	12.973
B-3 Pond Sediment	6.50	11.335
B-4 Pond Sediment	1.26	1.062
B-5 Pond Sediment	0.97	0.348
C-1 Pond Sediment	0.57	>0.002
C-2 Pond Sediment	2.75	2.728

Comparison of the plutonium activities reported in Table II for water samples to the plutonium activities reported in Table VI for sediment samples indicates that one gram of sediment from a holding pond contains approximately 50 times more plutonium than 1 liter of water from the pond. The highest plutonium concentrations were detected in sediments collected from Ponds B-1, B-2 and B-3. The sediment collected from Pond C-2 contained 2.73 pCi $^{239+240}$Pu/g.

Depleted uranium was detected in the sediments collects from the A and B series ponds. Table VII reports the maximum percentage of uranium present in each sediment sample attributable to depleted uranium. The same algorithm was used for water and sediment samples.

Table VII. Maximum Percentage of Uranium in Sediments Attributable to Depleted Uranium

Description	Maximum Percentage of Uranium Attributable to Release of Depleted Uranium From RFP
A-1 Pond Sediment	100
A-2 Pond Sediment	100
A-3 Pond Sediment	21
A-4 Pond Sediment	28
B-1 Pond Sediment	80
B-2 Pond Sediment	40
B-3 Pond Sediment	65
B-4 Pond Sediment	8
B-5 Pond Sediment	8
C-1 Pond Sediment	0
C-2 Pond Sediment	91

CONCLUSIONS

One gram of sediment from a holding pond contains approximately 50 times more plutonium than 1 liter of water from the pond. The upper Ponds A-1, A-2, A-3, B-1, B-2, B-3, and B-4 contain measurable quantities of plutonium. The plutonium concentrations in these ponds ranged from 0.004 to 3.09 pCi $^{239+240}$Pu/L. The plutonium concentrations in waters collected from Pond C2 appear to vary seasonally.

The upper Ponds A-1, A-2, A-3, B-1, B-2, B-3, and B-4 contain measurable quantities of depleted uranium. The uranium concentrations ranged from 0.2 to 15.8 pCi/L. Essentially 100% of the uranium in Pond A-1 and Pond A-2 originated as depleted uranium. All other ponds, except Pond C-1, contain mixtures of naturally occurring and depleted uranium. Approximately half of the uranium present in Ponds A-4 and C-2 originated as depleted uranium. Approximately 20% of the uranium in the waters collected from Pond B-5 originated as depleted uranium. The uranium in the sediments collected from Pond C-1 is naturally occurring uranium. Approximately 50% of the uranium detected in the waters and 90% of the uranium detected in the sediment sample collected from Pond C-2 were anthropogenic.

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