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Estimation of Uranium and Cobalt-60 Distribution Coefficients and Uranium-235 Enrichment at the Combustion Engineering Company Site in Windsor, Connecticut

Environmental Assessment Division
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The following is a list of the acronyms, abbreviations, and units of measure used in this document.

ACRONYMS

AEC	U.S. Atomic Energy Commission
DOE	U.S. Department of Energy
FUSRAP	Formerly Utilized Sites Remedial Action Program
Kd	distribution coefficient
NRC	U.S. Nuclear Regulatory Commission
RESRAD	*res*idual *rad*ioactive material guideline computer code

RADIONUCLIDES

Co-60	cobalt-60
U-232	uranium-232
U-234	uranium-234
U-235	uranium-235
U-238	uranium-238

UNITS OF MEASURE

cpm	count(s) per minute
g	gram(s)
h	hour(s)
ha	hectare(s)
km	kilometer(s)
L	liter(s)
µg	microgram(s)
µm	micrometer(s)
µR	microroentgen(s)
m	meter(s)
M	molar
mg	milligram(s)
mi	mile(s)
mL	milliliter(s)
pCi	picocurie(s)

ESTIMATION OF URANIUM AND COBALT-60 DISTRIBUTION COEFFICIENTS AND URANIUM-235 ENRICHMENT AT THE COMBUSTION ENGINEERING COMPANY SITE IN WINDSOR, CONNECTICUT

by

Y. Wang, K.A. Orlandini, and C. Yu

ABSTRACT

Site-specific distribution coefficients for uranium isotopes and cobalt-60 (Co-60) and the fraction of uranium-235 (U-235) enrichment by mass were estimated for environmental samples collected from the Combustion Engineering Company site in Windsor, Connecticut. This site has been identified for remedial action under the U.S. Department of Energy's (DOE's) Formerly Utilized Sites Remedial Action Program (FUSRAP). The authority of DOE at the Combustion Engineering site is limited to (1) Building 3; (2) other activities or areas associated exclusively with Building 3 (such as sewer lines); or (3) contamination that is exclusively highly enriched uranium (i.e., enriched to more than 20% in the U-235 isotope). In this study, 16 samples were collected from the Combustion Engineering site, including 8 soil samples, 4 sediment samples, 3 water samples, and 1 water plus sludge sample. These samples were analyzed for isotopic uranium by alpha spectrometry and for Co-60 by gamma spectrometry. The site-specific distribution coefficient for each isotope was estimated as the ratio of extractable radionuclide activity in the solid phase to the activity in the contact solution following a 19-day equilibration. The uranium activity measurements indicate that uranium-234 (U-234) and uranium-238 (U-238) were in secular equilibrium in two soil samples and that soil and sediment samples collected from other sampling locations had higher U-234 activity than U-238 activity in both the solid and solution phases. The site-specific distribution coefficient (Kd) ranged from 82 to 44,600 mL/g for U-238 and from 102 to 65,900 mL/g for U-234. Calculation of U-235 enrichment by mass indicated that four soil samples had values greater than 0.20; these values were 0.37, 0.38, 0.46, and 0.68. Cobalt-60 activity was detected in only three sediment samples. The measured Co-60 activity in the solid phase ranged from 0.15 to 0.45 pCi/g and that in the water phase of all three samples combined was 4 pCi/L. The Kd value for Co-60 in the site brook sediment was calculated to be 70 mL/g.

1 INTRODUCTION AND BRIEF HISTORY

The Combustion Engineering Company has operated a facility in Windsor, Connecticut, partly as a contractor on nuclear reactor and fuel projects for the U.S. Atomic Energy Commission (AEC), predecessor agency of the U.S. Department of Energy (DOE). Beginning in 1955 and continuing for over a decade, Combustion Engineering Company served as a direct contractor to the AEC and as a subcontractor to other firms for a number of projects involving the use of highly enriched uranium provided by the AEC. The company has also conducted nuclear research, development, and production activities at the site, under licenses issued by the U.S. Nuclear Regulatory Commission (NRC) and its predecessor. These activities have not involved the use of significant quantities of highly enriched uranium.

1.1 SITE DESCRIPTION AND SETTING

The Combustion Engineering Company site is located at 2000 Day Hill Road (previously known as 1000 Prospect Hill Road) in Windsor. The location is 13 km (8.1 mi) north of Hartford, Connecticut, and within 5 km (3.1 mi) of Bradley International Airport (Figure 1). Interstate 91 runs to the east of the site, and a brook runs east to west across the northern end of the site, joining the Farmington River northeast of the site. The site covers approximately 445 ha (1,100 acres) of land and contains more than a dozen buildings and several smaller support facilities. The site includes various wooded areas and three ponds (Figure 2). The potentially contaminated area covers nearly 243 ha (600 acres) (Moulton 1995). The remaining area of 202 ha (499 acres) is across the highway from the main complex and consists of undeveloped property that was never used for AEC activities.

Portions of the Combustion Engineering Company site that were previously used for AEC activities include Building 3 and may include Buildings 5 and 6 and their associated drainpipes and sewer lines, the waste storage pad area, the drum burial site, and a site brook. The waste storage pad area, located at the interior of the site, consists of a 110 m × 220 m (360 ft × 720 ft) clearing in a wooded area. It has a slightly sloping terrain. The drum burial pit, which covers an area about five times smaller than the waste storage pad area, is located on level terrain between two steep embankments in a wooded area. Building 3 was used for uranium fuel fabrication. The other areas listed above may have been involved in AEC production activities.

1.2 SITE HISTORY

During the late 1940s and early 1950s, Combustion Engineering Company supplied certain nonnuclear component parts to AEC reactor projects (Combustion Engineering 1991). The Combustion Engineering site was used during the 1950s and 1960s for nuclear research, fabrication of nuclear fuel from highly enriched uranium, and construction of naval reactor prototypes. A 1955 agreement between Combustion Engineering Company and the AEC initiated the company's

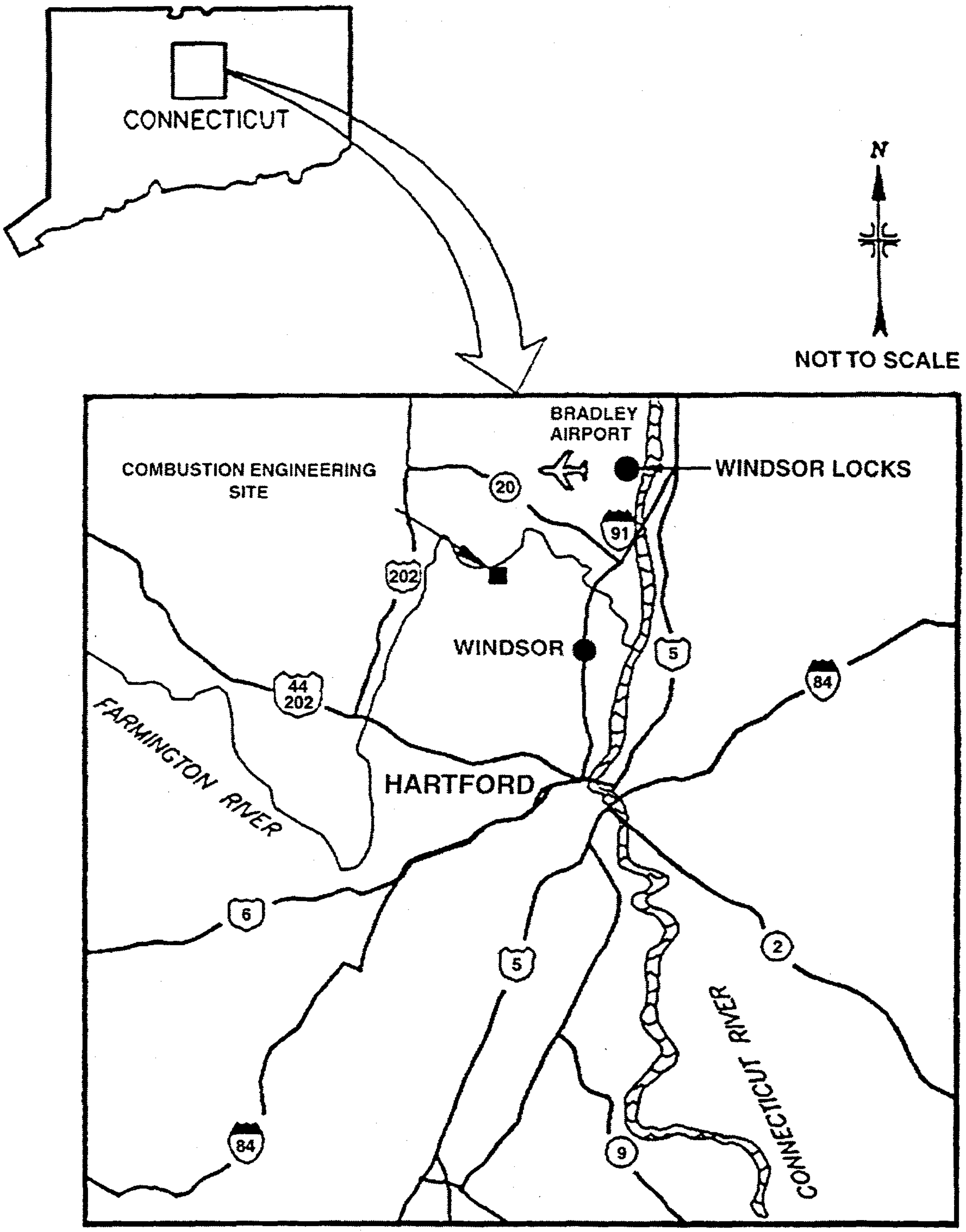

FIGURE 1 Location of the Combustion Engineering Company Site in Windsor, Connecticut (Source: Oak Ridge Institute for Science and Education 1994)

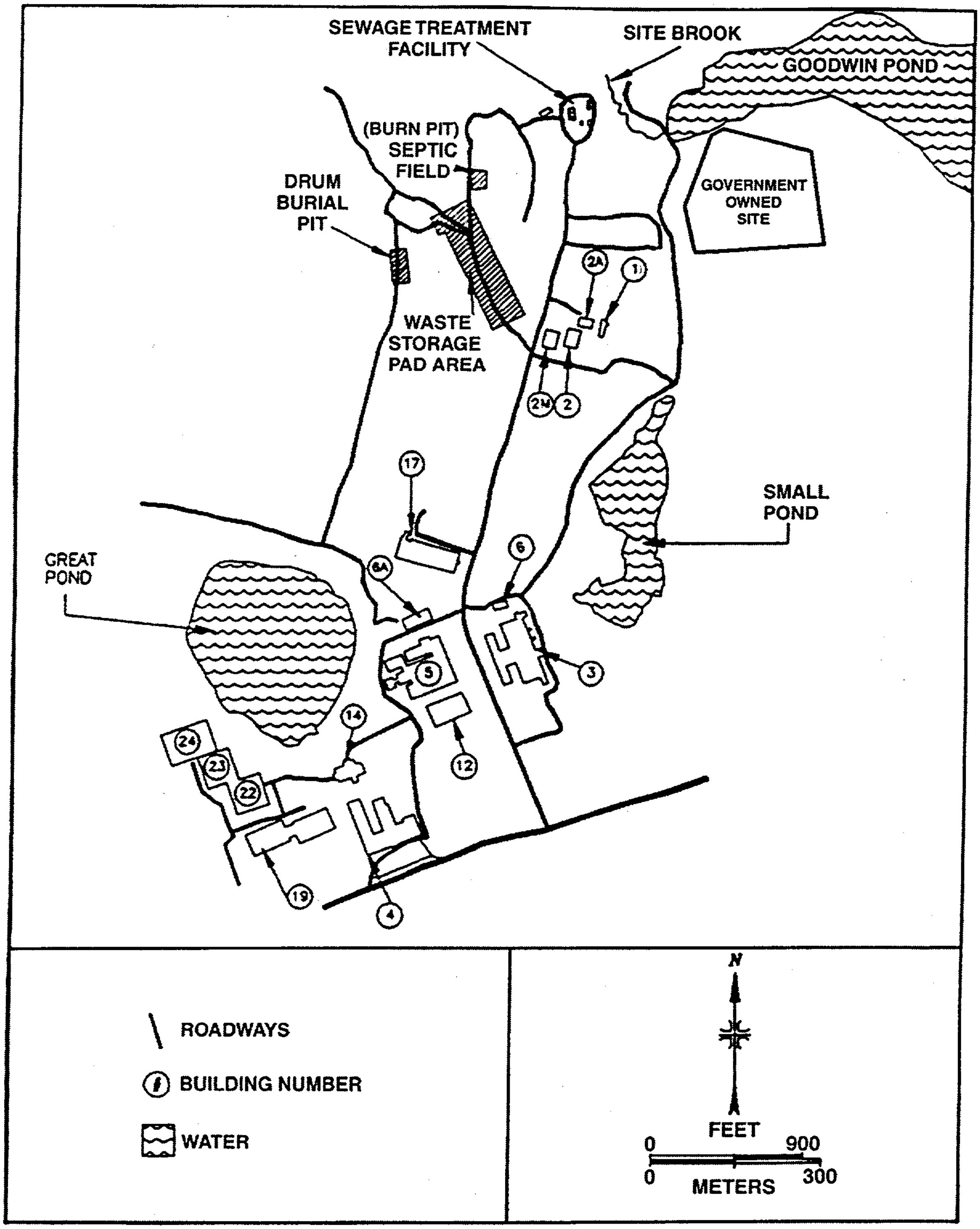

FIGURE 2 Map of the Combustion Engineering Site Showing Principal Site Features and Areas of Suspected Contamination (Source: Oak Ridge Institute for Science and Education 1994)

involvement in AEC's naval reactor program. Beginning in 1955 and continuing for over a decade, Combustion Engineering Company served as a direct contractor to the AEC and as a subcontractor to other firms for a number of projects involving the use of highly enriched uranium provided by the AEC. The uranium furnished for use at the Combustion Engineering site varied from approximately 5% to more than 90% enrichment of uranium-235 (U-235) (Williams 1994).

The Combustion Engineering site was later subdivided, with the 4-ha (9.8-acre) naval reactor prototype site becoming a U.S. government property. The Combustion Engineering Company retained ownership of the remainder of the property.

Radiological surveys conducted at the Combustion Engineering site in the early 1980s identified an area contaminated with thorium and uranium (enrichments ranging from 1 to 80%) in the burn and drum storage area (referred to as a septic field in Figure 2). This area was remediated by Combustion Engineering in 1986, and a confirmatory survey was conducted by Oak Ridge Associated Universities in 1989. The survey concluded that the area was within then-current NRC guidelines for thorium and uranium in soil (Landis 1989).

Combustion Engineering Company currently operates a nuclear fuel manufacturing facility licensed by the NRC (license number SNM-1067) on the Windsor site. At the request of DOE's Office of Environmental Restoration and Waste Management, the Oak Ridge Institute for Science and Education conducted a designation survey at the site during 1993. This survey identified several interior and exterior locations as containing highly enriched uranium with greater than 20% U-235 enrichment (Oak Ridge Institute for Science and Education 1994). The exterior areas included the waste storage pad area, drum burial pit, grounds north of Building 3, sanitary sewer and industrial drain lines, and the site brook and its banks. The Combustion Engineering site has been designated for remedial action by DOE under its Formerly Utilized Sites Remedial Action Program (FUSRAP). However, DOE's authority at the site is limited to the following: (1) Building 3; (2) other facilities or areas associated exclusively with Building 3 (i.e., sewer lines); or (3) contamination that is exclusively highly enriched uranium (i.e., enriched to more than 20% in the isotope U-235). As a result, although certain facilities and areas of the Combustion Engineering site have been designated for remedial action by DOE under FUSRAP, Combustion Engineering Company will conduct any needed remedial action at areas affected by its commercial activities.

1.3 REMEDIAL ACTIVITIES

Remedial action activities at the Combustion Engineering site will follow the guidelines established in DOE Order 5400.5 (DOE 1990). The DOE *res*idual *rad*ioactive material guideline (RESRAD) computer code (Yu et al. 1993) is used to derive residual radionuclide guidelines on a site-specific basis. This report presents the cobalt-60 (Co-60) and uranium distribution coefficients and the U-235 enrichment in environmental samples collected from selected locations at the Combustion Engineering site.

2 ENVIRONMENTAL SAMPLE ANALYSIS

Sixteen samples were collected from the Combustion Engineering site in this study (Figures 3 and 4). Table 1 lists sample identification numbers, sampling locations, and exposure rates of these environmental samples. The samples were analyzed for isotopic uranium by alpha spectrometry and for Co-60 by gamma spectrometry. A site-specific distribution coefficient (Kd) was calculated for Co-60, uranium-238, uranium-235, and uranium-234. The fraction of U-235 enrichment by mass was also calculated for these samples.

2.1 MEASUREMENT OF URANIUM ACTIVITY IN SOLID PHASE

To measure uranium activity in the solid phase, aliquots of all soil and sediment samples were digested three times with hot, strong (>8 M) nitric acid. After the repeated acid digestion, the extracted radionuclides in solution were separated from the residue by centrifugation. The soil and sediment residues were further digested by concentrated hydrofluoric-nitric acids to solubilize the more chemically resistant and less available uranium in these samples.

A known amount of isotopically pure uranium-232 (U-232) was added to the leachate to monitor the recovery of the uranium isotopes present. A two-step ion-exchange process was applied to isolate the uranium and remove iron interference in the leachate. The eluted uranium isotopes were electrodeposited onto a stainless steel plate for final activity measurement by alpha spectrometry. If the total iron in a sample aliquot was less than 0.4 mg, only a single processing step was needed to separate the uranium prior to electrodeposition. Details of the procedure for preparing the uranium sample plate for activity measurement by alpha spectrometry can be found in Fried et al. (1985), Nelson and Orlandini (1986), and Kaplan et al. (1994).

2.2 MEASUREMENT OF URANIUM ACTIVITY IN SOLUTION PHASE

To measure uranium activity in the solution phase, uranium was leached from the soil or sediment samples into the solution phase by contacting 20 g of soil or sediment with 15 to 20 mL of water sample collected from the site brook (at sampling location 11) for a 19-day equilibration. The solid-solution mixture was separated by centrifugation and filtration with a 0.45-μm filter. The filtrate was then acidified, and a known amount of high-purity U-232 was added to the solution as the yield monitor. A one-step ion-exchange process was applied to separate the uranium isotopes from the aqueous matrix. The separated uranium was then electrodeposited onto a stainless steel plate for final activity measurement by alpha spectrometry.

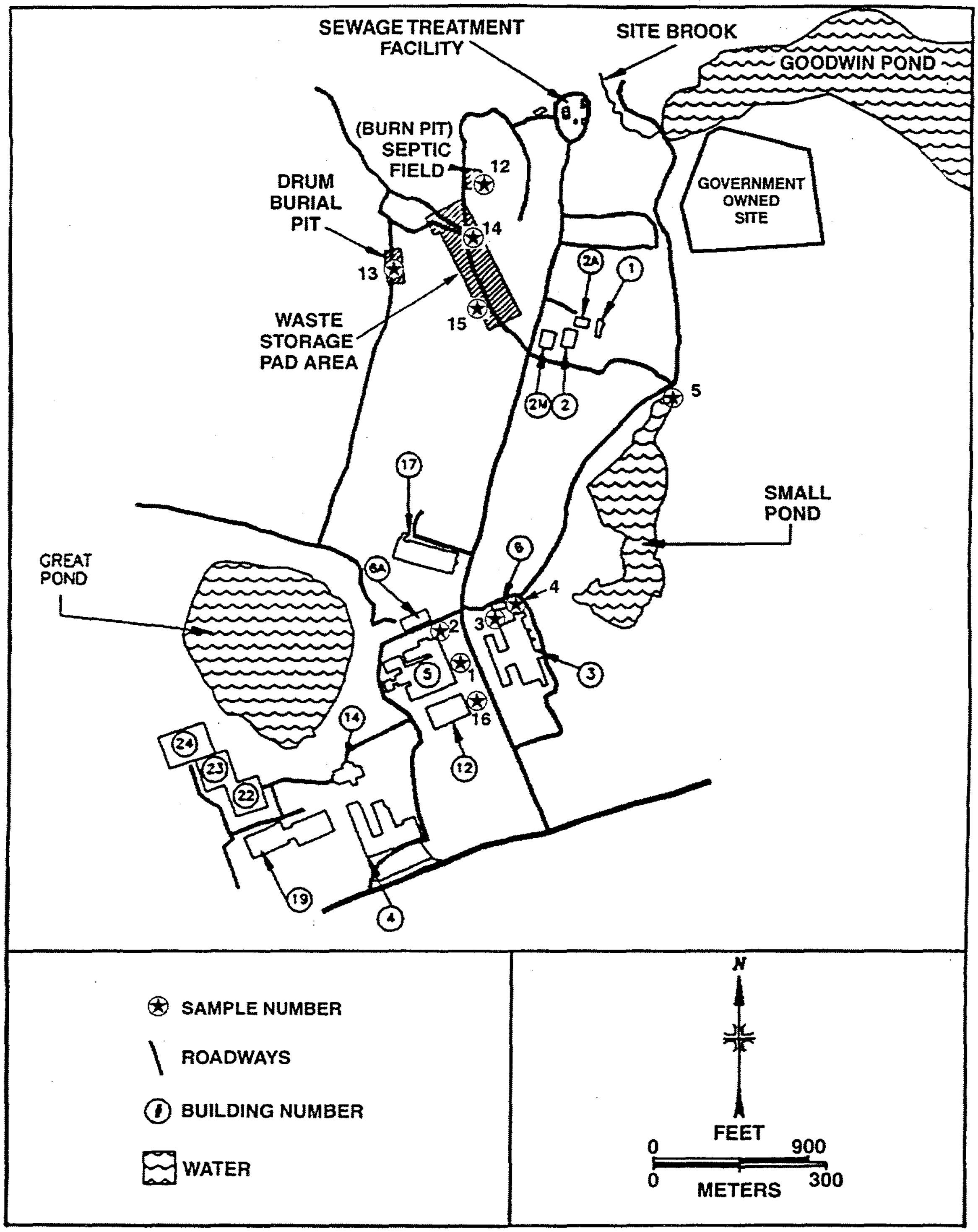

FIGURE 3 Sampling Locations for the Environmental Samples Collected at the Combustion Engineering Company Site

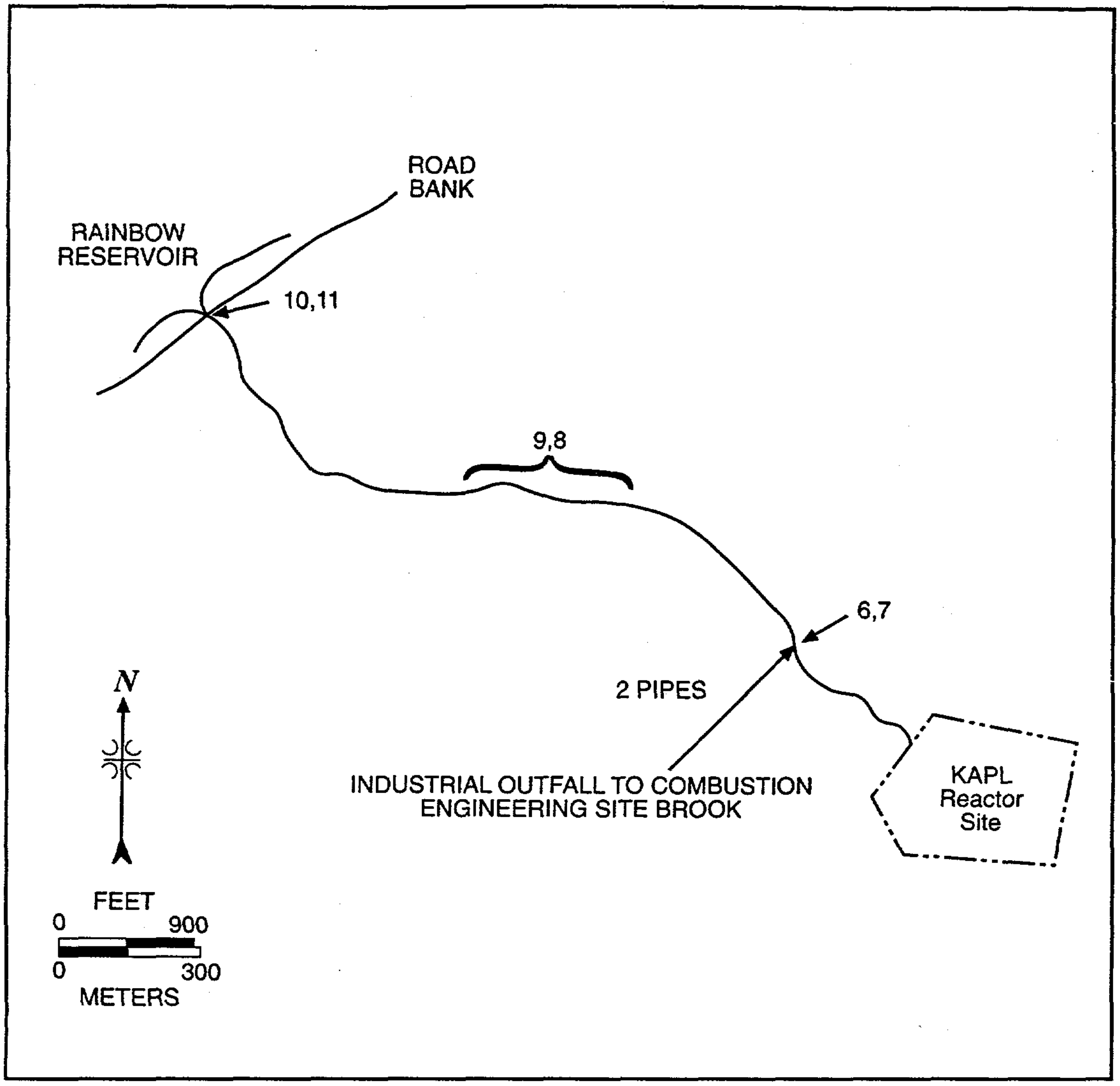

FIGURE 4 Sampling Locations at the Site Brook on the Combustion Engineering Company Site

TABLE 1 Description of Samples Collected at the Combustion Engineering Site, 1995

Sample Number	Matrix	Sampling Location	Exposure Rate[a] (μR/h)	Gross Gamma[b] (cpm)	Sample Description
1	Soil	East side of Building 5	12	300	
2	Soil	Northeast corner of Building 5	17	300	
3	Soil	Between Buildings 3 and 6	19	350	
4	Soil	East side of Building 6	18	850	
5	Water	Northeast end of small pond	10	200	A representative water sample and suspended solids in long-term contact.
6	Sediment	Industrial outfall to site brook	11	340	Sample taken from bottom of site brook.
7	Water	Industrial outfall to site brook	11	340	Site brook is a flowing stream containing some inputs from past operations. The radionuclide content in solution might not be in equilibrium with the underlying solids at any point.
8	Sediment	Site brook	11	250	Sample taken from bottom of site brook.
9	Sediment	Site brook	10	180	Sample taken from bottom of site brook.
10	Sediment	Site brook at Rainbow reservoir	10	200	
11	Water	Site brook at Rainbow reservoir	10	200	Site brook is a flowing stream containing some inputs from past operations. The radionuclide content in solution might not be in equilibrium with the underlying solids at any point.
12	Soil	East edge of burn pit	20	600	
13	Soil	Middle of drum burial pit	12	200	
14	Organic soil	East end of storage pad	17	500	
15	Soil	West end of storage pad	14	250	
16	Water and sludge	East of Building 12	11	200	

[a] Measured with Eberline ASP-1 W/GM at 1 m above the ground.

[b] Measured with Ludlum Mod 18 by holding the probe on the surface of the sample bag.

2.3 MEASUREMENT OF COBALT-60 ACTIVITY

Cobalt-60 activity was measured in a similar manner to uranium activity. First, the aqueous phase in the sediment samples was separated from the solid phase by filtration with a 0.45-µm filter. The Co-60 activity was then measured by gamma spectrometry using an intrinsic germanium detector with 120% relative efficiency. A counting time of less than 1 day was used to obtain the measurement.

3 DATA INTERPRETATION AND DISCUSSION

The activities of uranium isotopes in solid and solution phases, the calculated distribution coefficients (Kd values), and the U-235 enrichment in each sample are summarized in Table 2. The Kd for each isotope was calculated as the ratio of extractable activity in the solid phase to the activity in the contact solution following a 19-day equilibration. The uranium activities reported here were measured from leaching the soil and sediment with natural water and strong acid. The mass fraction of U-235 enrichment was calculated as the fraction of U-235 mass in the uranium-238 and uranium-235 (U-238+U-235) mass. The U-234 mass was not significant and was not included in this calculation.

Results of the activity measurements (Table 2) indicate that U-234 and U-238 were in secular equilibrium (within counting errors) in soil samples collected from sampling locations 1 and 2. Soil and sediment samples collected from other locations had higher U-234 activity than U-238 activity in both the solid and solution phases. This excess U-234 and U-235 activity may possibly be related to past site operations.

The Kd values ranged from 82 to 44,600 mL/g for U-238 and from 102 to 65,900 mL/g for U-234 (Table 2). The highest Kd for U-238 was measured in a water sample collected from the northeast end of a small pond (sampling location 5) containing naturally suspended solids in long-term contact. The suspended solids in this water sample were separated by filtration (0.45-µm filter), and the uranium isotopes in the solution and solid phases were measured. The Kd values of uranium isotopes calculated from this water sample were considered to be equilibrium values.

Results of the calculations indicate that the Kd values for soil samples collected from sampling locations 3, 13, and 15 were higher for U-238 than U-234, whereas the Kd values for sediment samples collected from sampling locations 5, 6, 8, 9,10, and 16 were higher for U-234 than U-238. A likely explanation is that U-238 and U-234 are associated with different chemical species in soil and sediment. The natural sources and operational sources of uranium are in different physical and chemical conditions, which are not equilibrated with each other.

Results of the mass fraction calculations of U-235 enrichment (Table 2) indicate that four soil samples had U-235 enrichment greater than 0.20. These four soil samples were collected from sampling location 3 between Buildings 3 and 6, sampling location 4 at the east side of Building 6, sampling location 14 at the east end of the storage pad, and sampling location 15 at the west end of the storage pad. The values for U-235 enrichment by mass were calculated to be 0.37, 0.46, 0.68, and 0.38, respectively.

Measurements of the more chemically resistant, less available or chemically refractory forms of uranium in 13 soil and sediment residues are summarized in Table 3. In two residues (sample numbers 1 and 2), U-234 and U-238 were in secular equilibrium (within counting errors). The U-234 activities were higher than the U-238 activities in the other 11 soil and sediment

TABLE 2 Measurements of Extractable Uranium Isotope Activities in Solid and Solution Phases of Samples Collected at the Combustion Engineering Site

Sample Number	Phase[a]	Activity			U-235 Enrichment by Mass[b]
		U-238	U-235	U-234	
1	Solid (pCi/g)	0.375 ± 0.024	Normal[c]	0.375 ± 0.024	"Natural" or "normal" uranium[c]
	Solution (pCi/L)	0.066 ± 0.023	Normal	0.086 ± 0.027	
	Kd (mL/g)	5,680 ± 2,040	Normal	4,360 ± 1,370	
2	Solid (pCi/g)	0.506 ± 0.018	Normal	0.506 ± 0.018	"Natural" or "normal" uranium
	Solution (pCi/L)	0.186 ± 0.048	Normal	0.228 ± 0.049	
	Kd (mL/g)	2,720 ± 710	Normal	2,220 ± 480	
3	Solid (pCi/g)	0.435 ± 0.044	1.670 ± 0.170	59.2 ± 6.0	0.37 ± 0.07
	Solution (pCi/L)	0.024 ± 0.007	0.193 ± 0.035	7.14 ± 0.18	
	Kd (mL/g)	18,100 ± 5,600	8,650 ± 1,800	8,290 ± 870	
4	Solid (pCi/g)	3.67 ± 0.39	20.0 ± 1.0	774 ± 0	0.46 ± 0.06
	Solution (pCi/L)	0.163 ± 0.037	0.876 ± 0.088	37.7 ± 0.6	
	Kd (mL/g)	22,500 ± 5,600	22,800 ± 2,500	20,500 ± 300	
5	Solid (pCi/g)	1.56 ± 0.06	0.017 ± 0.003	2.70 ± 0.10	0.002 ± 0.001 in suspended solid
	Solution (pCi/L)	0.035 ± 0.005	0.0014 ± 0.0002	0.041 ± 0.006	
	Kd (mL/g)	44,600 ± 6,600	12,100 ± 2,700	65,900 ± 9,900	
6	Solid (pCi/g)	0.657 ± 0.069	0.159 ± 0.021	5.85 ± 0.61	0.036 ± 0.009
	Solution (pCi/L)	0.022 ± 0.006	0.0043 ± 0.0020	0.100 ± 0.017	
	Kd (mL/g)	29,900 ± 8,800	37,000 ± 18,000	58,500 ± 11,700	
7	Solution (pCi/L)	0.110 ± 0.009	0.005 ± 0.001	0.160 ± 0.008	0.007 ± 0.001 in water

TABLE 2 (Cont.)

Sample Number	Phase[a]	Activity U-238	Activity U-235	Activity U-234	U-235 Enrichment by Mass[b]
8	Solid (pCi/g)	2.01 ± 0.20	0.774 ± 0.077	30.1 ± 3.0	0.06 ± 0.01
	Solution (pCi/L)	0.139 ± 0.035	0.040 ± 0.020	0.705 ± 0.083	
	Kd (mL/g)	14,500 ± 3,900	19,400 ± 9,900	42,700 ± 6,600	
9	Solid (pCi/g)	3.97 ± 0.15	1.96 ± 0.11	63.3 ± 0.6	0.071 ± 0.006
	Solution (pCi/L)	0.736 ± 0.164	0.200 ± 0.050	8.05 ± 0.56	
	Kd (mL/g)	5,390 ± 1,200	9,800 ± 2,510	7,860 ± 550	
10	Solid (pCi/g)	0.395 ± 0.032	0.154 ± 0.020	6.03 ± 0.13	0.057 ± 0.01
	Solution (pCi/L)	4.83 ± 0.27	1.43 ± 0.15	58.9 ± 2.8	
	Kd (mL/g)	82 ± 8	108 ± 18	102 ± 5	
11	Solution (pCi/L)	0.130 ± 0.011	0.022 ± 0.004	0.752 ± 0.022	0.025 ± 0.008 in water
12	Solid (pCi/g)	0.325 ± 0.030	0.054 ± 0.012	2.24 ± 0.08	0.024 ± 0.007
	Solution (pCi/L)	0.028 ± 0.007	0.005 ± 0.002	0.153 ± 0.024	
	Kd (mL/g)	11,600 ± 3,100	10,800 ± 5,000	14,600 ± 2,400	
13	Solid (pCi/g)	0.395 ± 0.040	0.17 ± 0.03	6.10 ± 0.34	0.06 ± 0.02
	Solution (pCi/L)	0.034 ± 0.011	0.031 ± 0.010	1.18 ± 0.08	
	Kd (mL/g)	11,600 ± 3,900	5,480 ± 2,020	5,170 ± 450	
14	Solid (pCi/g)	11.0 ± 0.6	147 ± 2	5,490 ± 10	0.68 ± 0.04
	Solution (pCi/L)	4.25 ± 0.78	61.8 ± 6.1	2,660 ± 20	
	Kd (mL/g)	2,590 ± 490	2,380 ± 240	2,060 ± 20	

TABLE 2 (Cont.)

Sample Number	Phase[a]	Activity			U-235 Enrichment by Mass[b]
		U-238	U-235	U-234	
15	Solid (pCi/g)	0.543 ± 0.048	2.13 ± 0.10	73.5 ± 0.6	0.38 ± 0.04
	Solution (pCi/L)	0.168 ± 0.032	1.11 ± 0.09	41.7 ± 1.3	
	Kd (mL/g)	3,230 ± 680	1,920 ± 360	1,760 ± 60	
16	Solid (pCi/g)	0.886 ± 0.069	0.249 ± 0.036	9.19 ± 0.40	0.04 ± 0.01 in solid
	Solution (pCi/L)	0.056 ± 0.004	0.0073 ± 0.0011	0.233 ± 0.009	0.02 ± 0.01 in solution
	Kd (mL/g)	15,800 ± 1,700	34,100 ± 7,100	39,400 ± 2,300	

[a] pCi = picocurie = 10^{-12} curie. The extractable uranium activities in the solid phase were determined from 8 M nitric acid extraction. The extractable uranium activities in the solution phase were determined by extraction with a representative site brook water sample.

[b] The relative fraction of U-235 by mass is calculated as the fraction of U-235 mass in the (U-238+U-235) mass. The U-234 mass was not significant and was not included in this calculation.

[c] "Natural" or "normal" uranium has 0.007 U-235 by mass of total uranium. Sample numbers 1 and 2 have slightly increased U-235 fractions (but less than 0.01).

Note: Specific activities for U-235 = 2.162 pCi/μg and for U-238 = 0.333 pCi/μg.

residues. This measurement of excess U-234 activity is consistent with the uranium isotope activity measurement in the nitric acid extractable phase (Table 2).

Measurements of Co-60 activity in sediment samples are listed in Table 4. Cobalt-60 activity was detected in only three sediment samples, which were collected from sampling locations 6, 8, and 9 at the site brook. The measured Co-60 activity in the solid phase ranged from 0.15 to 0.45 pCi/g. Cobalt-60 activity in the water phase was measured from the combined water fractions of the three samples listed. The Kd value for Co-60 in the site brook sediment was calculated to be 70 ± 20 mL/g. These measurements were based on a one-day gamma count. If necessary, extended counting could be used to detect lesser amounts.

TABLE 3 Measurements of Uranium Isotope Activities in Soil and Sediment Residues of Samples Collected at the Combustion Engineering Site

Sample Number	Activity (pCi/g)		
	U-238	U-235	U-234
1	0.180 ± 0.020	0.008 ± 0.003	0.203 ± 0.017
2	0.159 ± 0.014	0.006 ± 0.001	0.153 ± 0.013
3	0.261 ± 0.018	0.186 ± 0.015	6.23 ± 0.34
4	0.264 ± 0.018	0.798 ± 0.044	28.1 ± 1.6
6	0.130 ± 0.011	0.227 ± 0.014	7.45 ± 0.44
8	0.216 ± 0.015	0.380 ± 0.022	13.8 ± 0.8
9	0.293 ± 0.019	1.11 ± 0.05	42.1 ± 2.0
10	0.116 ± 0.010	0.140 ± 0.011	5.81 ± 0.26
12	0.245 ± 0.014	0.023 ± 0.004	0.691 ± 0.031
13	0.365 ± 0.016	0.370 ± 0.016	13.3 ± 0.6
14	7.11 ± 0.51	88.3 ± 1.8	3,170 ± 10
15	0.184 ± 0.012	0.112 ± 0.009	3.92 ± 0.17
16	0.514 ± 0.035	0.252 ± 0.019	8.41 ± 0.58

TABLE 4 Measurements of Cobalt-60 Activity in Sediment Samples Collected at the Combustion Engineering Site

Sample Number	Solid Phase (pCi/g)	Water Phase (pCi/L)	Kd (mL/g)
6	0.15 ± 0.01	-	-
8	0.22 ± 0.01	-	-
9	0.45 ± 0.03	-	-
Average	0.27 ± 0.03	4 ± 1[a]	70 ± 20

[a] Combined water fractions of the three samples listed.

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