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GEOCHEMICAL INFORMATION FOR SITES CONTAMINATED WITH LOW-LEVEL RADIOACTIVE WASTES: III - WELDON SPRING STORAGE SITE

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PREFACE

This document is the third in a series of topical reports describing geochemical information developed at Oak Ridge National Laboratory for Surplus Facility Management Program (SFMP) and Formerly Utilized Site Remedial Action Program (FUSRAP) sites. The preceding documents were:

SMFP or FUSRAP site	Report number
Niagara Falls Storage Site	ORNL-6083
St. Louis Airport Storage Site	ORNL-6097

GEOCHEMICAL INFORMATION FOR SITES CONTAMINATED WITH LOW-LEVEL RADIOACTIVE WASTES: III - WELDON SPRING STORAGE SITE

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ABSTRACT

The Weldon Spring Storage Site (WSSS), which includes both the chemical site and the quarry, became radioactively contaminated as the result of wastes that were being stored from operations to recover uranium from pitchblende ores in the 1940s and 1950s. The U.S. Department of Energy (DOE) is considering various remedial action options for the WSSS. This report describes the results of geochemical investigations carried out at Oak Ridge National Laboratory (ORNL) to support these activities and to help quantify various remedial action options. Soil and groundwater samples were characterized, and uranium and radium sorption ratios were measured in site soil/groundwater systems by batch contact methodology.

Soil samples from various locations around the raffinate pits were found to contain major amounts of silica, along with illite as the primary clay constituent. Particle sizes of the five soil samples were variable (50% distribution point ranging from 12 to 81 μm); the surface areas varied from 13 to 62 m^2/g. Elemental analysis of the samples showed them to be typical of sandy clay and silty clay soils. Groundwater samples included solution from Pit 3 and well water from Well D. Anion analyses showed significant concentrations of sulfate and nitrate (>350 and >7000 mg/L, respectively) in the solution from Pit 3. These anions were also present in the well water, but in lower concentrations.

Uranium sorption ratios for four of the soil samples contacted with the solution from Pit 3 were moderate to high (~300 to ~1000 mL/g). The fifth sample had a ratio of only 12 mL/g. Radium sorption ratios for the five samples were moderate to high (~500 to ~1000 mL/g). These values indicate that soil at the WSSS may show favorable retardation of uranium and radium in the groundwater.

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1. INTRODUCTION

The Weldon Spring Storage Site (WSSS), which includes the chemical plant and quarry, is located in St. Charles County, Missouri,

approximately 13 miles southwest of the city of St. Charles and 25 miles due west o. St. Louis. Its location with respect to the Missouri River is shown in Fig. 1.[1] Details of the site, including raffinate pits and sampling wells, are presented in Fig. 2.[2] The chemical plant is situated in the eastern portion of the former Weldon Spring Ordnance Works, which produced TNT and DNT from 1941 to 1944. The four raffinate pits on the site, which have a total capacity of ~130 million gal, contain ~6 million ft^3 of waste, including an estimated 150 tons of uranium and 75 tons of thorium. The site has been managed by the DOE as a part of the Surplus Facility Management Program (SFMP).

Several investigators have reported on the geology, hydrology, and potential radiation hazard to the nearby environment for this site.[3-10] An extensive assessment of the WSSS (both the quarry and chemical plant sites) was made by Lawrence Berkeley Laboratory staff members in 1980,[11] and their general conclusion was that no immediate threat to the environment existed. However, the assessment stated that the raffinate pits, located directly over a regional water table, were separated from it by a minimum of only about 20 ft of silty clay. It was their opinion that, on a long-term basis, the raffinate pits did pose a potential hazard to the environment.

The philosophy, or approach, used in the experimental work reported here has been developed by taking into consideration the technical and theoretical aspects of geochemical sorption processes and apparent solubility limit conditions which might be encountered in near-surface disposal of low-level radioactive wastes.[12] Necessary programmatic information and funding limitations were major controlling factors in the collection of the data described in this report.

Since the use of sorption distribution coefficient (Kd) values infers thermodynamic equilibrium, we have chosen to report radionuclide sorption data in terms of sorption ratio (Rs) values, which are calculated as empirical ratios. An Rs value is simply the ratio of the concentration of the species on the test solid adsorbent divided by the concentration of the solute in the test solution after contact. Since volume is being divided by mass in the calculation of the sorption ratio, the units are mL/g.[12]

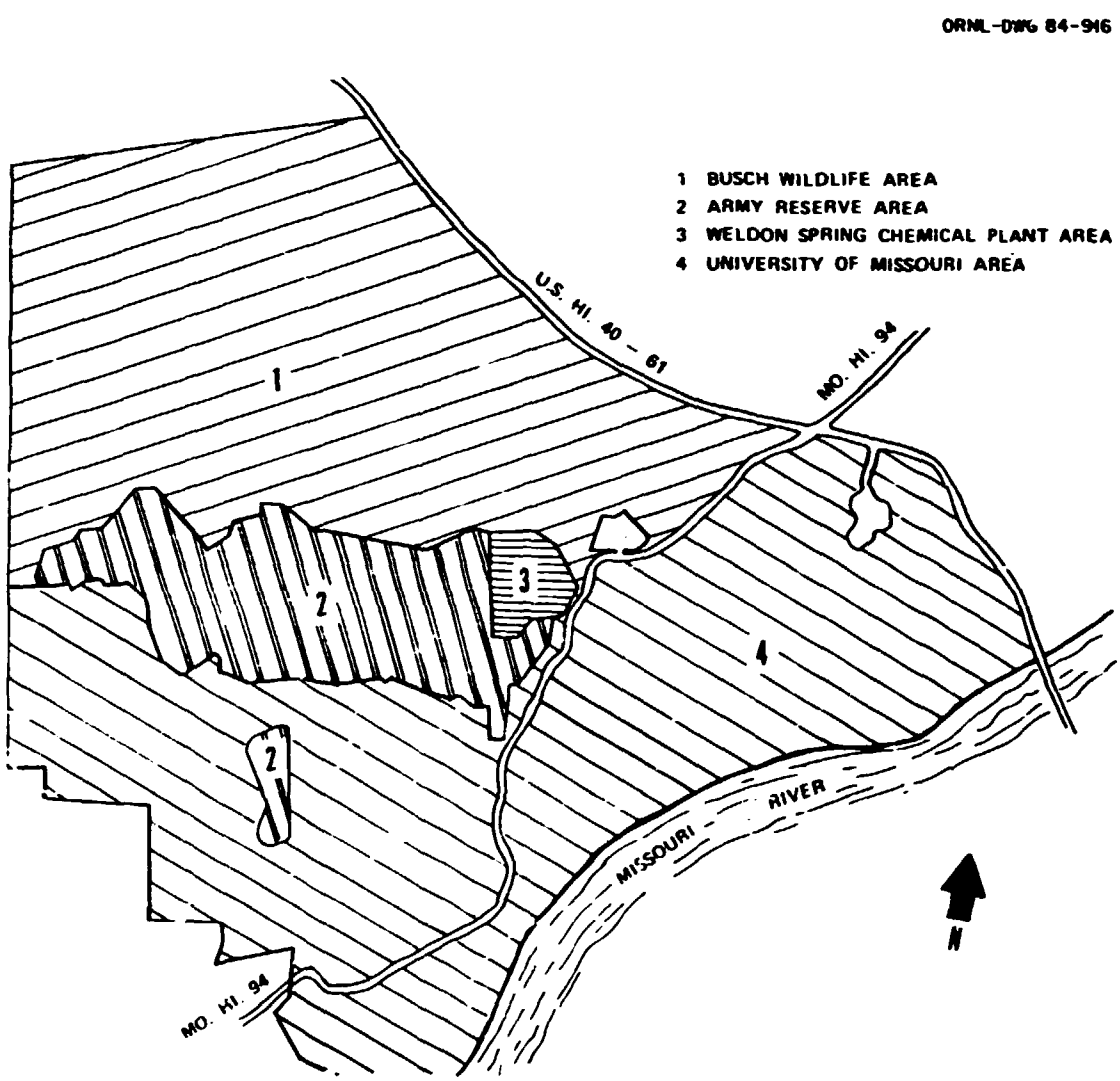

Fig. 1. Property adjacent to the Weldon Spring Chemical Plant site. Source: Department of the Army, Office of Department of Army Project Manager for Chemical Demilitarization and Installation Restoration, Assessment of Weldon Spring Chemical Plant in St. Charles County, Missouri, March 1976.

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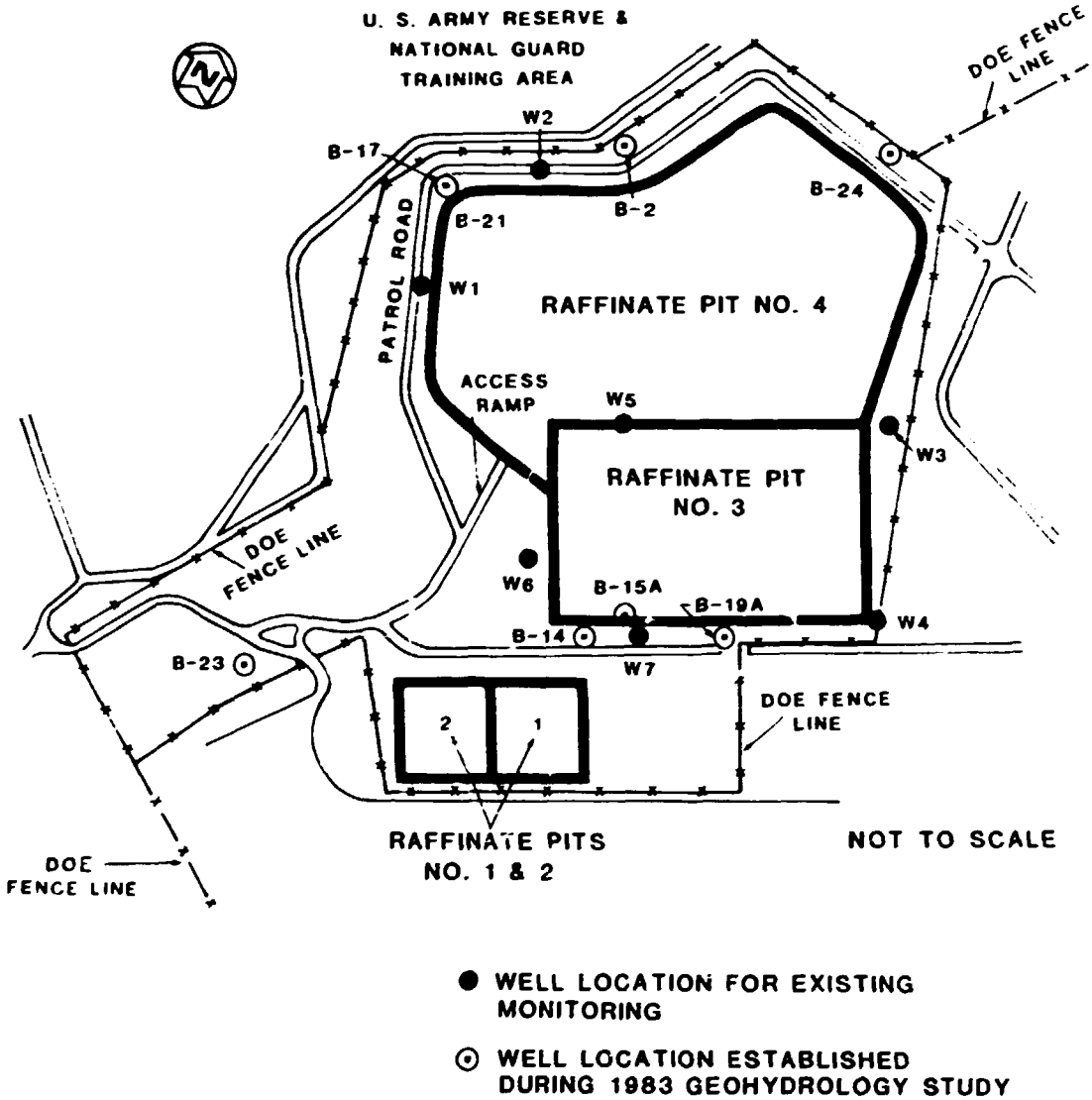

Fig. 2. Raffinate pit and groundwater sampling locations in the vicinity of the Weldon Spring Chemical Plant site. Source: Bechtel National, Engineering Evaluation of Alternatives for the Disposition of the Weldon Spring Raffinate Pits Site, DOE/OR/20722-5 (DE84010849), April 1984.

The information provided here is designed to characterize the soil in the vicinity of the raffinate pits at the Weldon Spring Chemical Plant with respect to its capability for retaining any radionuclides (contained in the pit residues) which might become solubilized.

2. SITE CHARACTERIZATION

2.1 DESCRIPTION OF SITE MATERIALS RECEIVED

Five soil samples, one water sample from a raffinate pit (Pit 3), and three groundwater samples from a monitoring well (Well D) from the WSSS chemical plant area were provided by Bechtel National, Oak Ridge, Tennessee. Brief descriptions of these samples are given in Table 1. The liquid sample bottles were filled to overflowing and then capped to exclude any air.

2.2 SOIL CHARACTERIZATION

The five soil samples (sampling locations shown in Fig. 3) were weighed, dried by lyophilization, reweighed, and then lightly ground in a Braun mill to pass a 50-mesh screen (100% <297 μm). The moisture contents of these samples, shown in Table 2, ranged between 12 and 19%.

Particle-size distribution analyses for the five soil samples were performed by the Technical Services Department at the Oak Ridge Gaseous Diffusion Plant. Micromerograph techniques were employed to obtain the data that are summarized in Table 3. Details are presented graphically in Figs. A-1 through A-5, in Appendix A. As shown in Table 3, the 50% distribution point ranged from 18.4 to 80.9 μm. Employing a standard definition of soil types,[13] all the WSSS soil samples can be categorized as clayey sand or sandy silty clay. The presence of appreciable amounts of sand is consistent with the high quartz content observed in the x-ray diffraction (XRD) data. The density of each of the five soil samples was found to be 2.7 g/cm^3.

X-ray diffraction techniques were also employed to identify the crystalline phases in the five soil samples. The results showed that quartz was the major crystalline component (~85%) in all of the soil samples and that illite, a typical soil clay mineral, was present at low levels (~15%).

Table 1. Source and description of WSSS samples received from Bechtel National[a]

	Source	Location	Description	Elevation (ft)
Soil samples				
WSSS-1	Fill station at 1:1 slope	14 + 20	Silty clay (brown/gray): background, 8782 cpm; in situ, 13,674 cpm	650
WSSS-2	Fill station	12 + 30	Silty clay (brown): background, 8782 cpm; in situ, 8364 cpm	644
WSSS-3	Foundation station	11 + 70	Silty clay (black): background, 8782 cpm; in situ, 7726 cpm	-
WSSS-4	Dike fill station (new - from dike repair)	12 + 80	Silty clay: background, 7660 cpm; in situ, 12,042 cpm	646
WSSS-5	Station 24 ft from D/S toe foundation	13 + 70	Silty clay (black/gray): background, 8782 cpm; in situ, 14,093 cpm	-
Groundwater samples				
Pit 3-1	Pit 3		pH of 8.6	
Pit 3-2	Pit 3		pH of 8.7	
Pit 3-3	Pit 3		pH of 8.6	
Well water	Well D		pH of 7.07	

[a]Information supplied by Bechtel National.

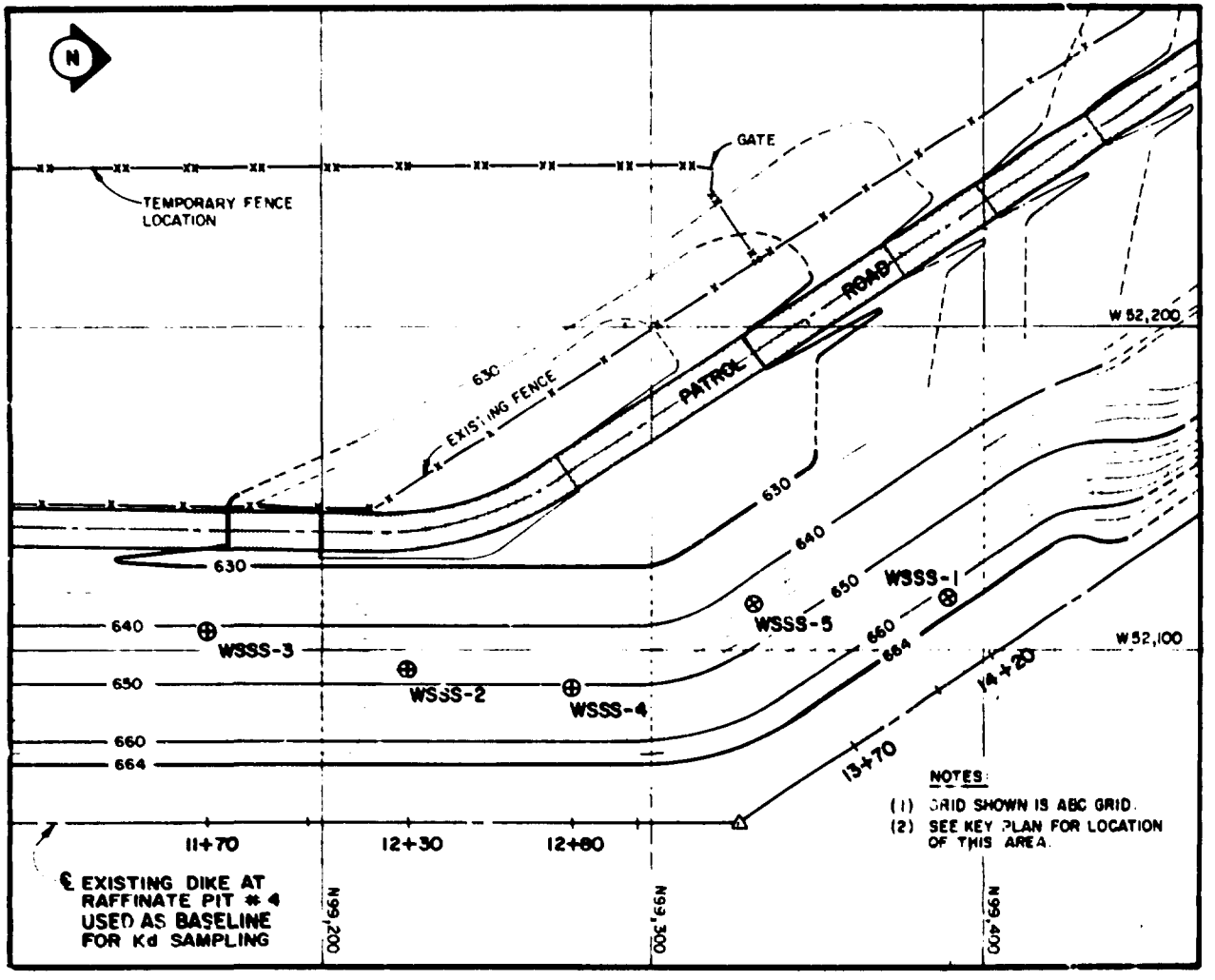

Fig. 3. Soil sampling locations at the WSSS. The section of dike shown is adjacent to, and west of, Raffinate Pit 4. The grid, as designated by the Atomic Energy Commission, is 28° 19' 33" east of true north. This figure was prepared by Bechtel National, Inc. (BNI), based on the following reference drawings (BNI): AA-DD02-C-04 and AA-DD05-C-05.

Table 2. Weight loss on drying soil samples from the Weldon Spring Storage Site

Sample designation	Source	Location	Total dry weight[a] (g)	Weight loss on drying (%)
WSSS-1	Fill station at 1:1 slope	14 + 20	1751	14
WSSS-2	Fill station 34 ft east of D/S toe	12 + 30	1738	12
WSSS-3	Foundation station	11 + 70	1455	16
WSSS-4	Dike fill station 60 ft from toe	12 + 80	1690	19
WSSS-5	Station 24 ft from D/S toe	13 + 70	2098	18

[a]Samples dried by lyohilization.

Table 3. Particle-size distribution[a] analysis of soil samples from the Weldon Spring Storage Site

Sample designation	Density (g/cm^3)	Particle size (μm)	
		50%[b]	10%[b]
WSSS-1	2.7	80.9	16.8
WSSS-2	2.7	24.8	3.3
WSSS-3	2.7	12.0	2.5
WSSS-4	2.7	46.7	5.0
WSSS-5	2.7	18.4	2.5

[a]Equal to, or less than, the stated value.

[b]Distribution point.

Surface area analysis of the five soil samples by the nitrogen adsorption Brunauer-Emmett-Teller (BET) technique also showed the soil samples to be significantly varied. The values obtained (Table 4) ranged from 13.4 to 62.3 m^2/g.

The five soil samples were analyzed for elemental content by inductively coupled plasma (ICP) spectrometry. The resulting data, presented in Table 5, indicate a composition typical of that for a highly leached sandy clay soil. The concentrations of alkaline-earth elements (calcium and magnesium) are somewhat lower than normal in these soil samples.

2.3 GROUNDWATER CHARACTERIZATION

The aqueous samples from Pit 3 and the well water from Well D were analyzed for elemental content by ICP spectrometry. The data are shown for three samples from Pit 3 and the sample from Well D both as-received (Table 6) and after filtration through 0.8-μm filter media (Table 7). No major differences between the as-received and filtered samples are apparent; that is, no significant colloidal material was present. The major elements detected in Pit 3 samples were calcium, magnesium, potassium, and sodium. These elements were also prominent in Well D water, but were found at slightly lower concentrations.

Table 4. Surface area analysis of soil samples from the Weldon Spring Storage Site

Sample designation	Sampling site	Location	Surface area[a] (m^2/g)
WSSS-1	Fill station	14 + 20	46.5
WSSS-2	Fill station	12 + 30	34.8
WSSS-3	Foundation station	11 + 70	13.4
WSSS-4	Dike fill station	12 + 80	62.3
WSSS-5	Station	13 + 70	27.9

[a]Surface area was measured by BET nitrogen adsorption after samples had been dried by lyophilization.

Table 5. Elemental analysis (μg/g) of WSSS soil samples by using inductively coupled plasma spectrometry

Element	Sample designation				
	WSSS-1	WSSS-2	WSSS-3	WSSS-4	WSSS-5
Ag	<5.	<5.	<5.	<5.	<5.
Al	69,000.	58,000.	48,000.	75,000.	53,000.
B	<19.	<19.	<19.	<19.	<19.
Ba	470.	520.	570.	420.	540.
Be	2.	1.	1.	1.	1.
Ca	4,700.	4,000.	3,700.	4,100.	4,000.
Cd	<2.	<2.	<2.	<2.	<2.
Co	8.	12.	9.	5.	10.
Cr	72.	53.	43.	65.	50.
Cu	17.	16.	12.	12.	12.
Fe	34,000.	27,000.	20,000.	34,000.	26,000.
Ga	20.	21.	<18.	27.	19.
Hf	20.	22.	19.	23.	21.
K	5,700.	7,400.	9,600.	5,200.	7,900.
Mg	4,800.	4,200.	3,100.	5,300.	3,800.
Mn	110.	560.	780.	180.	760.
Mo	100.	97.	80.	73.	110.
Na	4,500.	7,300.	9,000.	5,200.	7,000.
Ni	34.	31.	<28.	<28.	<28.
Pb	<65.	<65.	<65.	<65.	<65.
Sb	<35.	<35.	<35.	<35.	<35.
Se	<110.	<110.	<110.	<110.	<110.
Sr	74.	87.	99.	64.	91.
Ti	3,600.	3,700.	3,600.	3,700.	3,500.
V	89.	74.	58.	88.	69.
Zn	73.	55.	50.	62.	52.
Zr	130.	140.	160.	130.	140.

Table 6. Elemental analysis (mg/L) of as-received WSSS water samples by using inductively coupled plasma spectrometry

Element	Sample designation				
	Pit 3-1	Pit 3-2	Pit 3-3	Pit 3 (av)	Well D
Ag	0.0038	0.0034	0.0031	0.0034	<0.0030
Al	<0.0150	0.0386	<0.0150	<0.0023	0.0212
As	<0.0600	<0.0600	<0.0600	<0.0600	<0.0600
B	<0.1283	0.1306	0.1307	0.1299	0.0262
Ba	0.0989	0.0960	0.0963	0.0971	0.1368
Be	0.0005	0.0012	0.0011	0.0009	<0.0002
Ca	888.2	896.6	894.1	893.0	47.71
Cd	<0.0041	<0.0041	<0.0041	<0.0041	<0.0041
Cr	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020
Cu	0.0046	0.0050	0.0051	0.0049	0.0594
Fe	<0.0050	<0.0050	<0.0050	<0.0050	0.0246
K	93.2	95.8	95.0	94.7	7.01
Li	2.84	2.92	2.89	2.88	0.0083
Mg	213.4	215.0	212.4	213.6	13.3
Mn	<0.0050	<0.0050	0.0057	<0.0052	0.2313
Mo	2.20	2.23	2.21	2.22	<0.0080
Na	1150.0	1150.0	1150.0	1150.0	20.4
Ni	<0.0040	<0.0040	<0.0040	<0.0040	0.0212
P	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020
Pb	<0.0020	<0.0020	<0.0020	<0.0020	0.1974
Si	<0.010	<0.010	<0.010	<0.0010	0.051
Sr	2.17	2.21	2.21	2.20	0.1703
Ta	<0.050	<0.050	0.050	<0.050	<0.050
Ti	<0.0010	<0.0010	<0.0010	0.0010	<0.0010
V	0.4875	0.4992	0.4935	0.4934	<0.0030
Zn	<0.0020	<0.0020	<0.0020	<0.0020	0.1421
Zr	<0.0010	0.0012	<0.0010	<0.0011	<0.0010

Table 7. Elemental analysis (µg/mL) of filtered[a] WSSS water samples by using inductively coupled plasma spectrometry

Element	Sample designation	
	Pit 3	Well D
Ag	0.0025	0.0030
Al	0.0319	0.0154
As	<0.0600	<0.0600
B	0.1296	0.0286
Ba	0.0958	0.1398
Be	0.0012	<0.0002
Ca	892.3	48.9
Cd	<0.0041	<0.0041
Cr	<0.0020	<0.0020
Cu	0.0048	0.0719
Fe	<0.0050	<0.0050
K	95.2	6.87
Li	2.91	0.0079
Mg	212.4	13.56
Mn	<0.0050	0.1813
Mo	2.21	0.0104
Na	1150.0	20.6
Ni	0.0064	0.0184
P	<0.0020	0.0312
Pb	<0.0020	0.1162
Si	0.3136	6.27
Sr	2.22	0.1710
Th	<0.050	<0.050
Ti	<0.0010	<0.0010
V	0.4780	<0.0030
Zn	<0.0020	0.0753
Zr	<0.0010	<0.0010

[a]Filtered through 0.8-µm filter media.

The results of anion analysis (by ion chromatography), along with pH values for the aqueous samples from Pit 3 and the well water from Well D, are summarized in Table 8. All of the samples from Pit 3 show relatively high concentrations of nitrate and sulfate (>7000 and >350 mg/L, respectively). There is a significant concentration of bicarbonate, especially in the water from Well D (104 mg/L). The pH of each sample was measured onsite after withdrawal, and the values obtained were confirmed within 0.1 unit at the time the samples were received at ORNL. However, the pH tended to be significantly lower (1.1 $\pm$ 0.5 pH units) when measured 3 weeks later, probably due to exposure of the sample to air and subsequent dissolution of CO_2.

3. RADIONUCLIDE SORPTION MEASUREMENTS - SOIL/GROUNDWATER SYSTEMS

3.1 URANIUM(VI) SORPTION/SOLUBILITY VALUES

Uranium(VI) sorption isotherms for the five WSSS soil samples were determined by using the procedure outlined in Appendix B. The uranium sorption behavior for these soil samples is summarized in Table 9. Detailed data for these sorption isotherm tests are shown in Tables C-1 through C-5 and Figs. C-1 and C-2, in Appendix C. In tests with low initial uranium concentration (5 µg/mL), the Rs values varied from 280 to 1300 mL/g for four of the five samples; the fifth sample exhibited a low value of 12 mL/g, which was probably due to its low-clay and high-quartz contents. At the highest initial uranium concentration (10,000 µg/mL), the Rs values varied from 0.72 to 1.6 mL/g. The test pH values were relatively low because the pH was not adjusted but allowed to equilibrate naturally; this is probably the reason that no solubility limit was noted even at a uranium concentration of 10,000 µg/mL. At least four of the five soil samples could probably be characterized as having sufficient sorption capacity to significantly retard the migration of uranium.

3.2 RADIUM SORPTION/SOLUBILITY VALUES

Radium Rs values were also determined for the five WSSS soil samples. Groundwater from Raffinate Pit 3 (see Tables 6-8 for composition), spiked with the desired concentration of ^{226}Ra, was used in each test. The

Table 8. Major anion analysis of aqueous samples from the Weldon Spring Storage Site

Sample designation	pH		Concentration of major anions (mg/L)									
	Initial[a]	Delayed[b]	HCO_3^-	CO_3^{2-}	OH^-	Cl^-	SO_4^{2-}	PO_4^{3-}	F^-	NO_2^-	NO_3^-	Br^-
					As received							
Pit 3-1	8.6	7.2	40	0	0	18	361	<40	5	14	7583	<50
Pit 3-2	8.7	7.3	40	0	0	18	362	<40	5	11	7203	<50
Pit 3-3	8.6	6.9	39	0	0	19	379	<40	13	10	7175	<50
Well D	7.1	6.9	104	0	0	32	75	<4	1	<2	13	<5
					After filtration[c]							
Pit (composite)	-	7.4	40	0	0	18	362	<40	8	14	7207	<50
Well D	-	7.8	106	0	0	32	80	<4	1	<2	20	<5

[a]Measured onsite at the time of sample withdrawal; confirmed within 0.1 pH unit at the time samples were received at ORNL.

[b]Measured after ~3 weeks.

[c]Through 0.8-μm filter media.

Table 9. Summary of uranium(VI) sorption behavior for soil samples from the Weldon Spring Storage Site

Sample designation	Location of sampling station	Initial U conc.[a] (µg/mL)	After contact	
			pH	Sorption ratio (mL/g)
WSQDS-1	14 + 20	Low	7.6	1.2E1
		High	3.1	8.0E-1
WSQDS-2	12 + 30	Low	6.7	2.8E2
		High	3.2	1.2E0
WSQDS-3	11 + 70	Low	6.3	1.3E3
		High	3.4	1.6E0
WSQDS-4	12 + 80	Low	6.4	1.2E3
		High	3.1	7.2E-1
WSQDS-5	13 + 70	Low	6.0	1.3E3
		High	3.3	7.8E-1

[a]Low = 5; high = 10,000.

procedure for obtaining these Rs values is similar to that used in the determination of sorption isotherms (Appendix B). Only single sorption values are given for each of the soil samples; a summary of these values is presented in Table 10.

The radium Rs values shown in Table 10 represent an average material balance of approximately 82%, which is acceptable for radium analysis. These values, which range from 660 to 18,000 mL/g, indicate very effective adsorption of the radium by all of the WSSS soil samples. In summary, the sorption ratio for each of the five soil samples indicates favorable retardation of radium; that is, it may be high enough to retard this radionuclide from migrating into the environment to any significant degree.

Table 10. Summary of radium sorption behavior for soil samples from the Weldon Spring Storage Site

Sample designation	Location of sampling station	Initial Ra conc. (pCi/mL)	After contact	
			pH	Rs (mL/g)
WSSS-1	14 + 20	100,000	7.0	1.1E3
WSSS-2	12 + 30	100,000	7.0	1.5E3
WSSS-3	11 + 70	100,000	7.0	6.6E2
WSSS-4	12 + 80	100,000	7.0	1.2E3
WSSS-5	13 + 70	100,000	7.0	1.8E3

4. ACKNOWLEDGMENTS

The authors wish to acknowledge the excellent technical assistance of G. C. Young, who conducted most of the laboratory tests reported in this document; the Materials and Chemistry Department, ORGDP, for x-ray diffraction, particle size distribution, and surface area analyses of the soil; and the Analytical Chemistry Division, ORNL, for ICP and ion chromatographic analyses. Appreciation is also expressed to H. J. Alexander, Bechtel National, for excellent liaison and to Martha Dawson and Martha Stewart for secretarial and editorial assistance, respectively, in the preparation of this report.

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6. APPENDIXES

Appendix A.

PARTICLE-SIZE DISTRIBUTION ANALYSIS CURVES

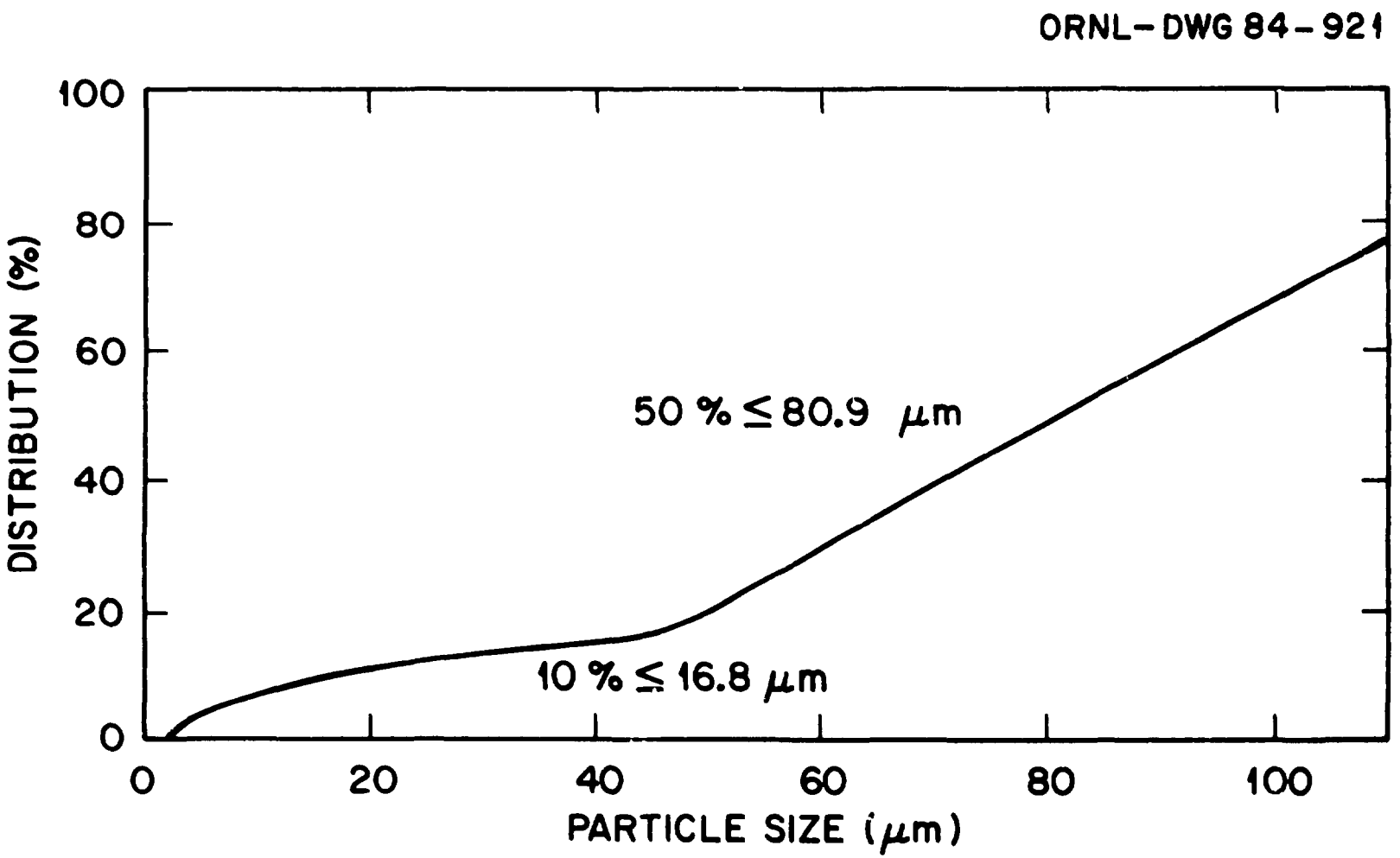

Fig. A-1. Particle size distribution of soil sample WSSS-1 from fill station (14 + 20) at the Weldon Spring Storage Site.

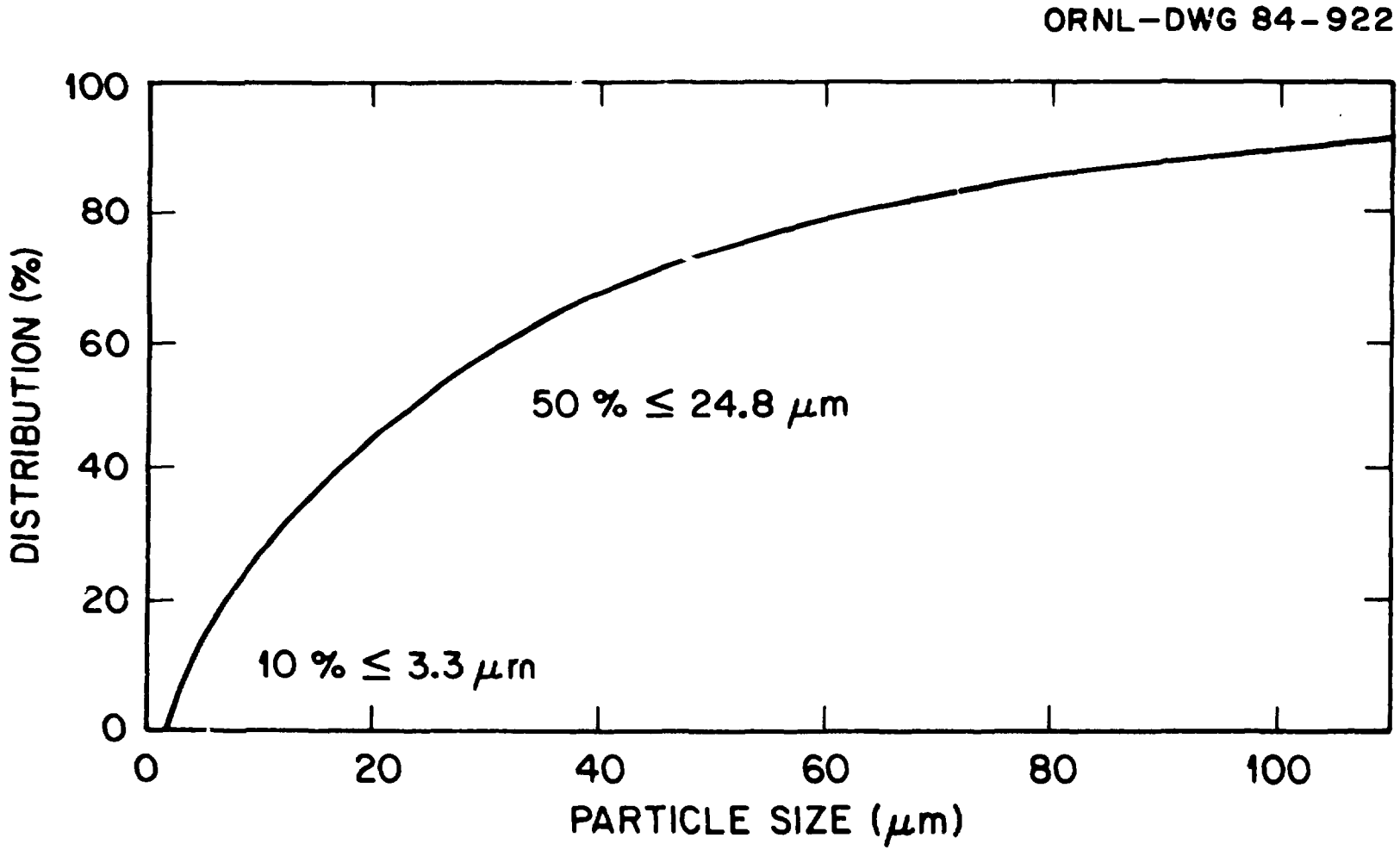

Fig. A-2. Particle size distribution of soil sample WSSS-2 from fill station (12 + 30) at the Weldon Spring Storage Site.

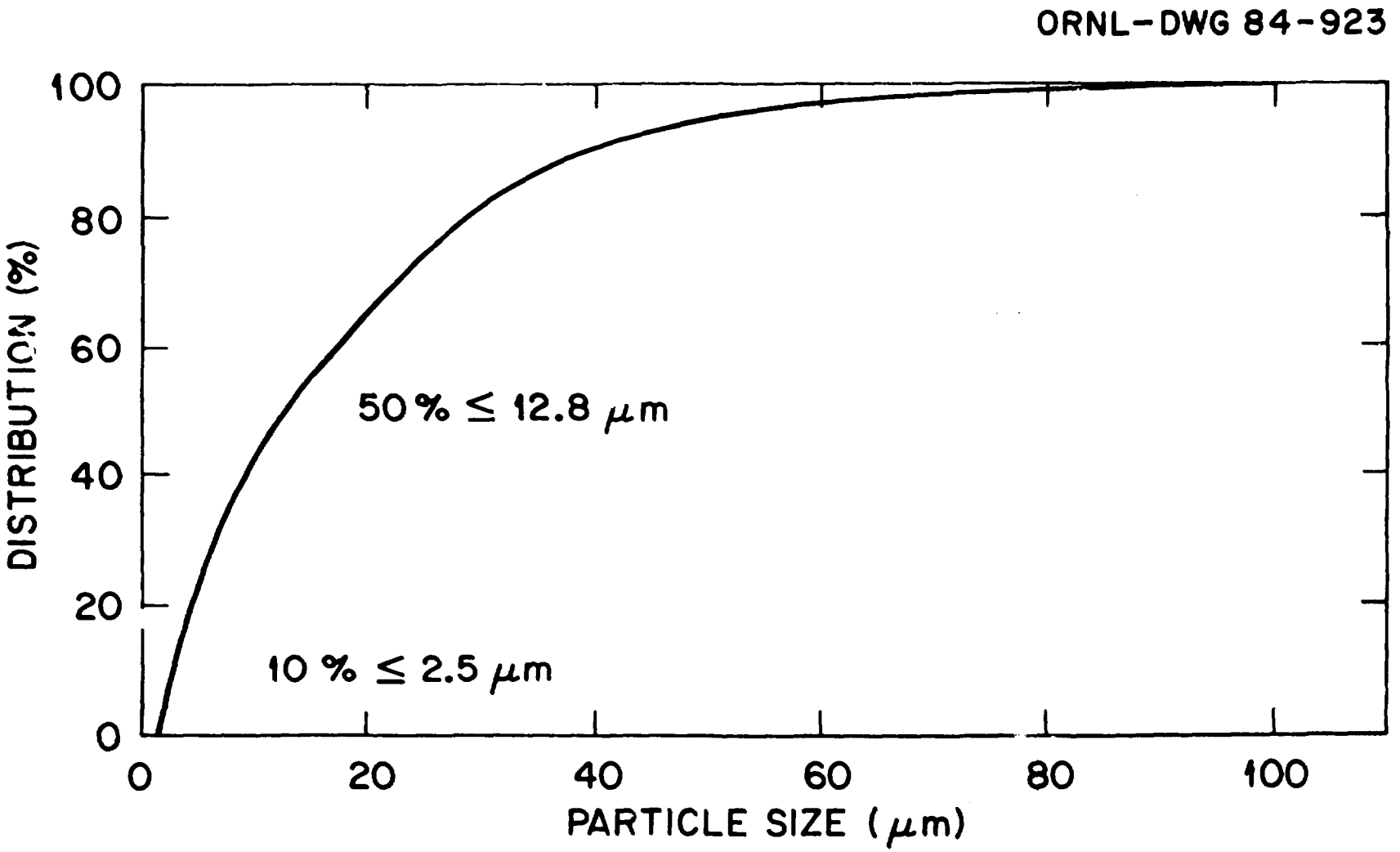

Fig. A-3. Particle size distribution of soil sample WSSS-3 from foundation station (11 + 70) at the Weldon Spring Storage Site.

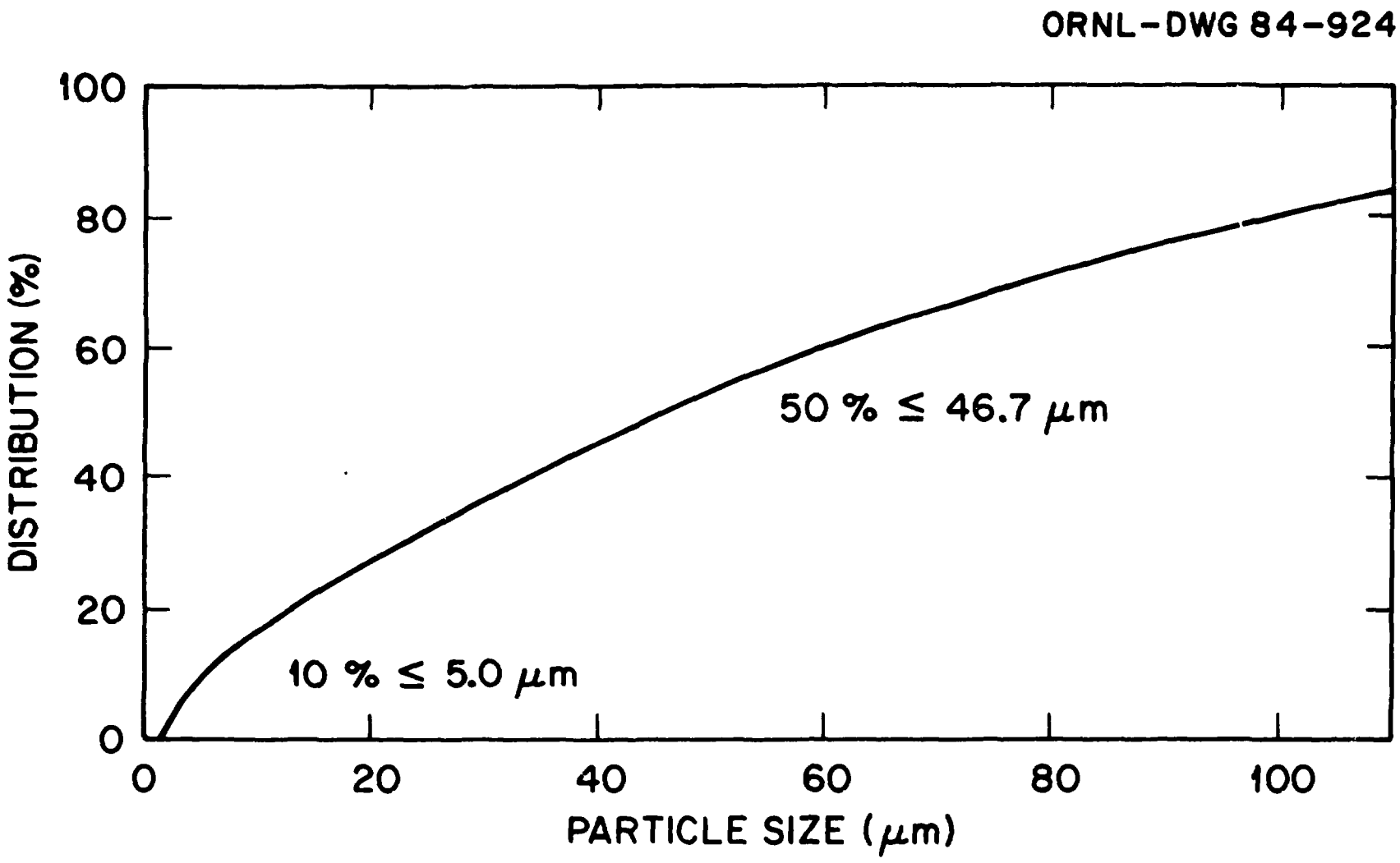

Fig. A-4. Particle size distribution of soil sample WSSS-4 from dike fill station (12 + 80) at the Weldon Spring Storage Site.

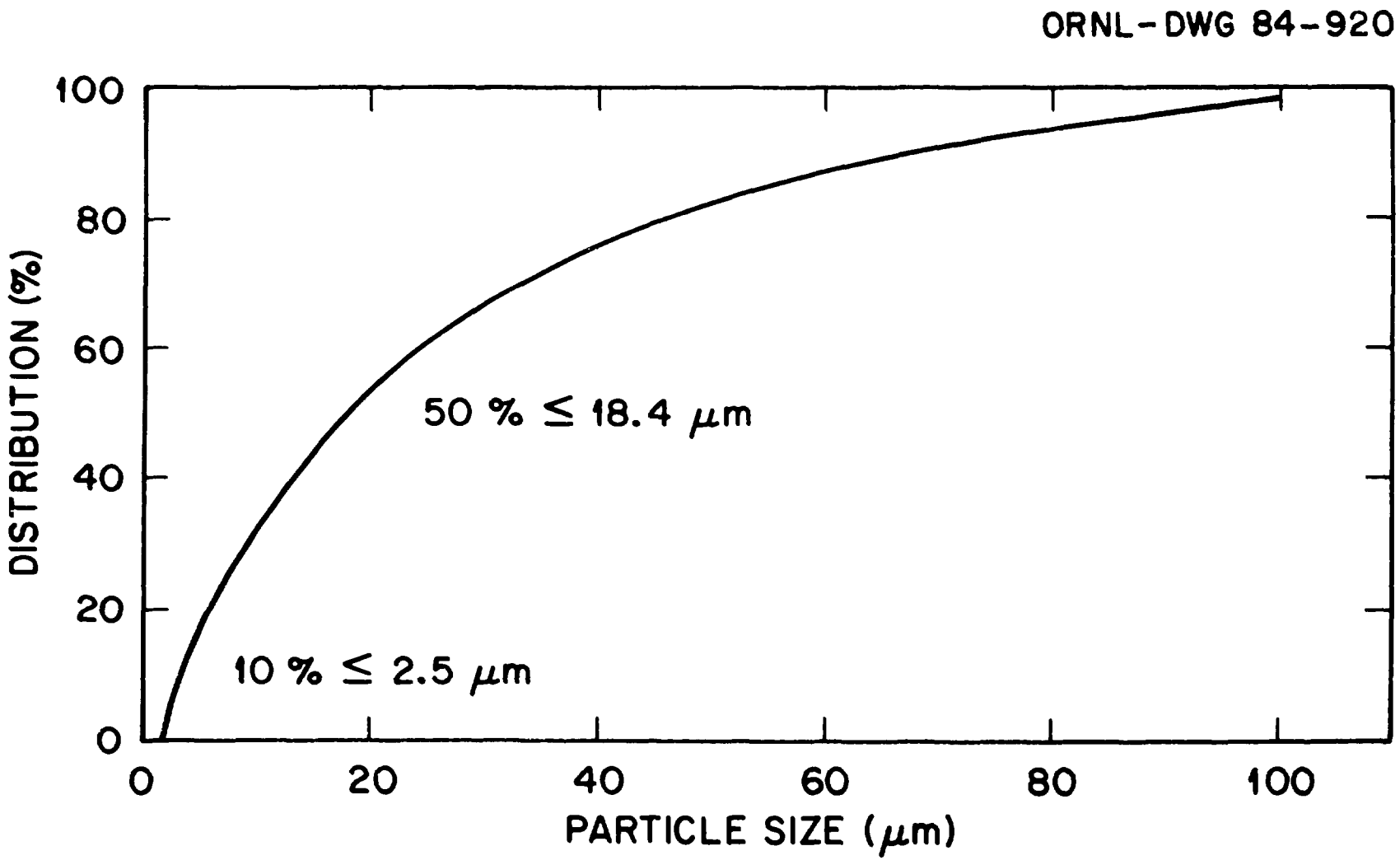

Fig. A-5. Particle size distribution of soil sample WSSS-5 from station (13 + 70) at the Weldon Spring Storage Site.

Appendix B.

EXPERIMENTAL METHODS

B.1. EXPERIMENTAL PROCEDURE FOR THE DETERMINATION OF SORPTION ISOTHERMS

To provide data for sorption isotherms, all tests are conducted by contacting 2.0 g of soil with 10 mL of the appropriate well water in polystyrene centrifuge tubes for 3 h in an attempt to establish equilibrium between the soil and the well water. An argon atmosphere is maintained above the slurry to prevent any additional source of carbonate from being introduced into the sample or air oxidation of reducing components. The appropriate amount of uranium (or other metal ion) is then added to the slurry to obtain the desired initial concentration. The concentration of the uranium(VI), or other metal-ion, stock solutions is chosen so that the addition of 0.1 mL to the 10 mL of well water will give the desired initial concentration. The radioactive tracer (^{233}U in the case of uranium) is subsequently added to the sample. The activity of the radionuclide stock solution is also adjusted to a level such that the addition of 0.1 mL of the solution to the 10 mL of the well water will give an initial counting level of ~100,000 cpm/mL. The samples are then contacted on a shaker for ~18 h. Various contact times are used in sorption rate studies. After the sample has been removed from the shaker, the solids are allowed to settle and the pH of the supernate is measured. If there is any noted departure from the predetermined experimental pH level (the natural pH established between the soil and well water) being investigated, an adjustment is made to bring the pH to the desired level. If pH adjustment has been necessary on any of the samples, a period of recontacting is employed. After each sample has reached a satisfactory pH level, it is placed in a Sharples centrifuge and centrifuged at 5000 rpm for ~15 min. For the determination of uranium concentration, 1.0 mL of the clear supernate is removed and mixed with 10 mL of Instagel scintillation counting mixture in a counting vial. The alpha activity is then determined by counting in a Packard Tri-Carb scintillation spectrometer. Appropriate standards and blanks are also counted.

B.2. COMPUTER PROGRAM FOR CALCULATING SORPTION OR DESORPTION RATIOS

```
00010   PRINT "PROGRAM FOR CALCULATING ISOTHERM"
00020   PRINT "THIS PROGRAM REQUIRES THE FOLLOWING VARIABLES:"
00030   PRINT "(1) C, ACTIVITY OF INITIAL TRACER IN CTS/M/ML"
00040   PRINT "(2) VS, VOLUME OF SAMPLE AND (3) VRS, VOLUME OF RESIDUAL
        SOLUTION
"
00050   PRINT "(4) A, ACTIVITY OF FINAL SOLUTION IN CTS/MIN/ML"
00060   PRINT "(5) W, WEIGHT OF SAMPLES"
00070   PRINT "(6) M, CONCENTRATION OF INITIAL NUCLIDE IN MOLES/L"
00080   PRINT "(7) N, ANY ACTIVITY ADDED BY NUCLIDE"
00090   PRINT "TO END ENTRIES, GIVE 0 FOR ACTIVITY OF SAMPLE"
00100   Y$="N"
00110   DIM A(60),W(60),E(60),B(60),V(60),A1(60)
00115 DIM N(60)
00120   DIM G(60),H(60),J(60),K(60),R(60),V1(60)
00130   PRINT "STANDARD TRACER CTS/MIN/ML ";
00140   INPUT C
00150   I=1
00160   FOR I=1 TO 60
00170   PRINT "ACTIVITY OF SAMPLE IS (CPM/ML)";
00180   INPUT A(I)
00190   IF A(I)=0 THEN 300
00200   PRINT "ACTIVITY ADDED BY NUCLIDE IS ";
00210   INPUT N(I)
00220   PRINT "SAMPLE VOLUME, RESIDUAL VOLUME ARE (MLS)";
00230   INPUT V(I),E(I)
00240   PRINT "WEIGHT OF SAMPLE IS (GM)";
00250   INPUT W(I)
00260   PRINT "CONCENTRATION NUCLIDE IN MOLES/L IS ";
00270   INPUT B(I)
00280   NO=I
00290   NEXT I
00300   FOR I=1 TO NO
00310   V1=V(I)+E(I)
00320   C=N(I)+C
00330   H1=B(I)/C
00340   S=V(I),V1
00350   S1=C*S
00360   R(I)=V1*(S1-A(I))/(A(I)*W(I))
00370   H(I)=A(I)*H1
00380   G(I)=R(I)*H(I)
00390   J(I)=.4343*LOG(G(I,)
00400   K(I)=.4343*LOG(H(I))
00410 NEXT I
00420   PRINT
00430   PRINT "SAMPLE       CONC.      MASS          VOL.          RES.
"
00440   PRINT "CTS/MIN/ML   MOLES/L    GRAMS         MLS.          VOL.
"
00450   PRINT
```

```
00460   FOR I=1 TO NO
00465   PRINT A(I),B(I),W(I),V(I),E(I)
00480   NEXT I
00490   PRINT
00500   PRINT
00510   PRINT"AMOUNT        AMOUNT                                    D
"
00520   PRINT"ADSORBED      IN SOLUTION    LOG          LOG         OR R
S"
00530   PRINT"MOLS/KG       MOL/L          ADSORBED     SOL
OR RD"
00540   PRINT
00550   FOR I=1 TO NO
00555   PRINT G(I),H(I),J(I),K(I),R(I)
00570   NEXT I
00580   PRINT
00590   PRINT "DESORPTION (Y/N) ";
00600   INPUT Y$
00620   IF Y$="N" THEN 820
00630   FOR I=1 TO NO
00640   PRINT "NEW VOLUME IS ";
00650   INPUT V1(I)
00660   PRINT "NEW RESIDUAL VOLUME IS ";
00670   INPUT E(I)
00680   PRINT "NEW ACTIVITY OF SAMPLE IS ";
00690   INPUT A1(I)
00695   H1=B(I)/C
00700   A=R(I)*W(I)*A(I)
00710   A1=E(I)*A(I)
00720   S=A1(I)*(V(I)+E(I))
00730   R1=(A+A1-S)/W(I)
00740   R(I)=R1/A1(I)
00750   H(I)=A1(I)*H1
00760   G(I)=R(I)*H(I)
00770   J(I)=.4343*LOG(G(I))
00780   K(I)=.4343*LOG(H(I))
00790   A(I)=A1(I)
00800   NEXT I
00801   PRINT
00802   PRINT"SAMPLE        CONC.        MASS        VOL.       RES."
00803   PRINT"CTS/MIN/ML    MOLES/L      GRAMS       MLS.       VOL."
00804   PRINT
00805   FOR I=1 TO NO
00806   PRINT A1(I),B(I),W(I),V1(I),E(I)
00807   NEXT I
00810   GO TO 490
00820   END
```

B.3. PROGRAM FOR THE CALCULATION OF COUNT RATE AND STANDARD DEVIATION

```
PROGRAM NAME: CSIG'.BAS
00005 PRINT "THIS PROGRAM CALCULATES STANDARD DEVIATION FOR COUNTING"
00007 PRINT "FOR SAMPLES COUNTED 1 TO 10 TIMES"
00010 PRINT "TO TERMINATE PROGRAM TYPE 0 FOR TIMES COUNTED"
00020 PRINT "HOW MANY TIMES WAS SAMPLE COUNTED";
00030 INPUT N
00035 IF N = 0 THEN 410
00040 PRINT "ENTER TOTAL COUNT (INCL. BKG.) AND COUNTING TIME"
00041 PRINT "IN PAIRS, ONE PAIR PER LINE"
00042 S1 = 0
00043 S2 = 0
00044 S3 = 0
00050 FOR I=1 TO N
00060 INPUT C1(I),T1(I)
00070 R(I) = C1(I)/(T1(I)**2)
00100 S1 = S1 + R(I)
00110 S2 = S2 + C1(I)
00115 S3 = S3 + T1(I)
00116 R1 = S2/S3
00125 NEXT I
00126 G1 = (SQRT(S1))/N
00130 PRINT "HOW MANY TIMES WAS BKG COUNTED";
00140 INPUT M
00150 PRINT "ENTER BKG COUNT AND COUNTING TIME IN PAIRS"
00155 PRINT "ONE PAIR PER LINE"
00156 S4 = 0
00157 S5 = 0
00158 S6 = 0
00160 FOR J = 1 TO M
00170 INPUT B(J),T2(J)
00180 D(J) = B(J)/(T2(J)**2)
00190 S4 = S4 + T2(J)
00200 S5 = S5 + D(J)
00210 S6 = S6 + B(J)
00215 NEXT J
00220 G2 = (SQRT(S5))/M
00230 R2 = S6/S4
00240 D2 = R1-R2
00241 X = S2/(S3**2)
00242 Y = S6/(S4**2)
00243 G = SQRT(X+Y)
00244 P = G*100/D2
00250 IF D2 <= 0 THEN 370
00251 F1 = G1**2
00252 F2 = G2**2
00260 G3 = SQRT(F1+F2)
00262 A = S2/N
00264 P2 = G3*100/A
```

```
00270 PRINT "TOTAL TIME SAMPLE COUNTED";S3
00280 PRINT "TOTAL TIME BKG COUNTED   ";S4
00290 PRINT "TOTAL SAMPLE COUNTS      ";S2
00300 PRINT "TOTAL BACKGROUND COUNTS ";S6
00310 PRINT "GROSS SUMMATION SAMPLE COUNT RATE";R1
00315 PRINT "GROSS SUMMATION BKG COUNT RATE IS";R2
00320 PRINT "STANDARD DEVIATION BASED ON SUMMED COUNTS IS";G
00330 PRINT "STANDARD DEVIATION BASED ON SEPARATE COUNTS IS";G3
00340 PRINT "PERCENT SD BASED ON SUMMED COUNTS IS";P
00350 PRINT "PERCENT SD BASED ON SEPARATE COUNTS IS";P2
00360 GO TO 380
00370 PRINT "TOTAL COUNT RATE <=BACKGROUND = ";D2
00380 PRINT
00390 PRINT "************************************"
00400 GO TO 20
00410 END
```

Appendix C.

URANIUM(VI) SORPTION ISOTHERM DATA FOR SOIL SAMPLES FROM THE WELDON SPRING STORAGE SITE

Table C-1. Uranium(VI) sorption isotherm data obtained by contacting soil sample WSSS-1 with solution from Pit 3

Sampling location: fill station (14 + 20)

Initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (mL/g)
5	7.6	6.1E-9	7.6E-8	-8.21	-7.12	1.2E1
10	7.1	1.3E-8	1.5E-7	-7.87	-6.84	1.1E1
20	7.1	3.1E-8	2.7E-7	-7.51	-6.57	9.0E0
30	7.3	4.9E-8	3.9E-7	-7.30	-6.41	8.0E0
40	7.3	7.0E-8	5.0E-7	-7.15	-6.30	7.1E0
50	7.1	9.1E-8	6.1E-7	-7.04	-6.22	6.7E0
100	7.3	2.1E-7	1.1E-6	-6.69	-5.96	5.4E0
200	7.4	4.2E-7	2.1E-6	-6.38	-5.67	5.1E0
300	7.0	1.5E-7	5.7E-6	-6.82	-5.25	3.8E1
400	6.6	3.1E-7	7.0E-6	-6.51	-5.15	2.3E1
500	6.3	1.2E-7	1.0E-5	-6.92	-5.00	8.4E1
1,000	5.0	7.6E-7	1.8E-5	-6.12	-4.76	2.3E1
5,000	3.7	1.6E-7	1.4E-5	-4.74	-4.84	8.0E-1
10,000	3.1	3.7E-7	3.0E-5	-4.44	-4.52	8.0E-1

Table C-2. Uranium(VI) sorption isotherm data obtained by contacting soil sample WSSS-2 with solution from Pit 3

Sampling location: fill station (12 + 30)

Initial U conc. (µg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (mL/g)
5	6.7	3.8E-10	1.1E-7	-9.42	-6.98	2.8E2
10	6.7	8.1E-10	2.1E-7	-9.09	-6.68	2.6E2
20	6.7	2.0E-9	4.2E-7	-8.70	-6.38	2.1E2
30	6.9	3.3E-9	6.3E-7	-8.48	-6.20	1.9E2
40	6.6	5.5E-9	8.3E-7	-8.26	-6.08	1.5E2
50	6.7	6.1E-9	1.0E-6	-8.21	-5.98	1.7E2
100	6.6	2.1E-8	2.0E-6	-7.67	-5.69	9.5E1
200	6.4	1.4E-8	4.2E-6	-7.85	-5.38	3.0E2
300	6.3	1.6E-8	6.3E-6	-7.80	-5.20	4.0E2
400	6.1	2.5E-8	8.4E-6	-7.60	-5.07	3.4E2
500	5.7	5.0E-8	1.1E-5	-7.30	-4.98	2.1E2
1,000	4.8	6.5E-7	1.8E-5	-6.19	-4.74	2.8E1
5,000	3.7	1.6E-5	2.9E-5	-4.81	-4.54	1.9E0
10,000	3.2	3.4E-5	4.3E-5	-4.47	-4.37	1.3E0

Table C-3. Uranium(VI) sorption isotherm data obtained by contacting soil sample WSSS-3 with solution from Pit 3

Sampling location: foundation station (11 + 70)

Initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (mL/g)
5	6.2	8.0E-11	1.1E-7	10.10	6.97	1.3E3
10	6.1	1.8E-10	2.1E-7	9.74	6.67	1.2E3
20	6.2	4.7E-10	2.3E-7	9.33	6.37	9.0E2
30	6.2	5.3E-10	6.4E-7	9.27	6.19	1.2E3
40	6.0	7.2E-10	8.5E-7	9.14	6.07	1.2E3
50	6.2	1.1E-9	1.1E-6	8.97	5.97	9.9E2
100	6.1	2.4E-9	2.1E-6	8.61	5.67	8.8E2
200	6.0	5.2E-9	4.3E-6	8.28	5.37	8.2E2
300	6.0	1.0E-8	6.4E-6	7.99	5.20	6.3E2
400	5.8	1.3E-8	8.5E-6	7.88	5.07	6.4E2
500	5.4	2.6E-8	1.1E-5	7.59	4.98	4.1E2
1,000	4.7	3.1E-7	2.0E-5	6.51	4.70	6.4E1
5,000	3.7	1.4E-5	3.8E-5	4.86	4.42	2.7E0
10,000	3.4	3.3E-5	5.2E-5	4.49	4.29	1.6E0

Table C-4. Uranium(VI) sorption isotherm data obtained by contacting soil sample WSSS-4 with solution from Pit 3

Sampling location: dike fill station (12 + 80)

Initial U conc. (µg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (mL/g)
5	6.4	9.2E-11	1.1E-7	-10.03	-6.97	1.2E3
10	6.2	3.7E-10	2.1E-7	-9.43	-6.67	5.8E2
20	6.3	8.6E-10	4.2E-7	-9.06	-6.37	4.9E2
30	6.3	1.2E-9	6.4E-7	-8.91	-6.20	5.2E2
40	6.3	1.8E-9	8.5E-7	-8.74	-6.07	4.6E2
50	6.3	2.2E-9	1.1E-6	-8.65	-5.97	4.7E2
100	6.1	4.6E-9	2.1E-6	-8.34	-5.67	4.6E2
200	5.9	1.1E-8	4.2E-6	-7.94	-5.37	3.7E2
300	5.7	2.6E-8	6.3E-6	-7.59	-5.20	2.4E2
400	5.4	5.6E-8	8.3E-6	-7.25	-5.08	1.5E2
500	4.8	3.0E-7	9.2E-6	-6.52	-5.04	3.0E1
1,000	4.5	1.8E-6	1.2E-5	-5.75	-4.91	6.9E0
5,000	3.6	2.0E-6	7.4E-6	-4.71	-5.13	3.8E-1
10,000	3.1	3.7E-5	2.7E-5	-4.43	-4.57	7.2E-1

Table C-5. Uranium(VI) sorption isotherm data obtained by contacting soil sample WSSS-5 with solution from Pit 3

Sampling location: station 24 ft from D/S toe foundation (13 + 70)

Initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (mL/g)
5	6.0	8.1E-11	1.1E-7	-10.09	-6.97	1.3E3
10	6.0	2.2E-10	2.1E-7	-9.65	-6.67	9.6E2
20	6.0	4.6E-10	4.3E-7	-9.33	-6.37	9.2E2
30	6.0	7.2E-10	6.4E-7	-9.14	-6.19	8.9E2
40	6.0	3.0E-9	8.4E-7	-8.52	-6.07	2.8E2
50	6.3	1.2E-9	1.1E-6	-8.92	-5.97	8.8E2
100	5.9	3.4E-9	2.1E-6	-8.47	-5.67	6.2E2
200	6.1	1.7E-8	4.2E-6	-7.77	-5.38	2.5E2
300	5.7	1.1E-8	6.4E-6	-7.98	-5.20	6.0E2
400	5.7	1.7E-8	8.5E-6	-7.76	-5.07	5.0E2
500	5.3	2.7E-8	1.1E-5	-7.57	-4.98	3.9E2
1,000	4.7	3.2E-7	2.0E-5	-6.50	-4.70	6.2E1
5,000	3.7	1.6E-5	2.4E-5	-4.78	-4.61	1.5E0
10,000	3.3	3.7E-5	2.9E-5	-4.43	-4.54	7.8E-1

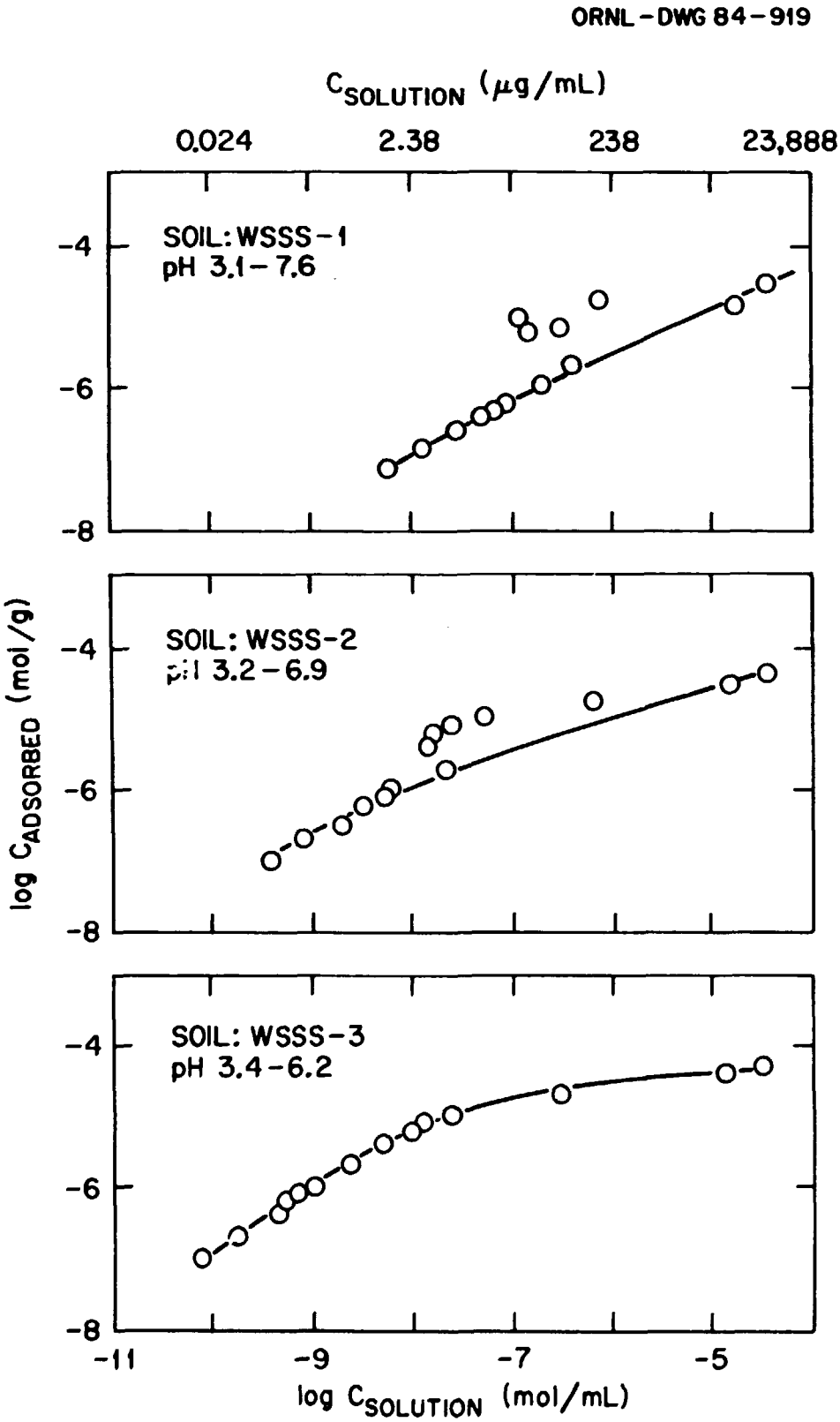

Fig. C-1. Uranium(VI) sorption isotherms for soil samples WSSS-1, WSSS-2, and WSSS-3 from the Weldon Spring Storage Site.

ORNL-DWG 84-918

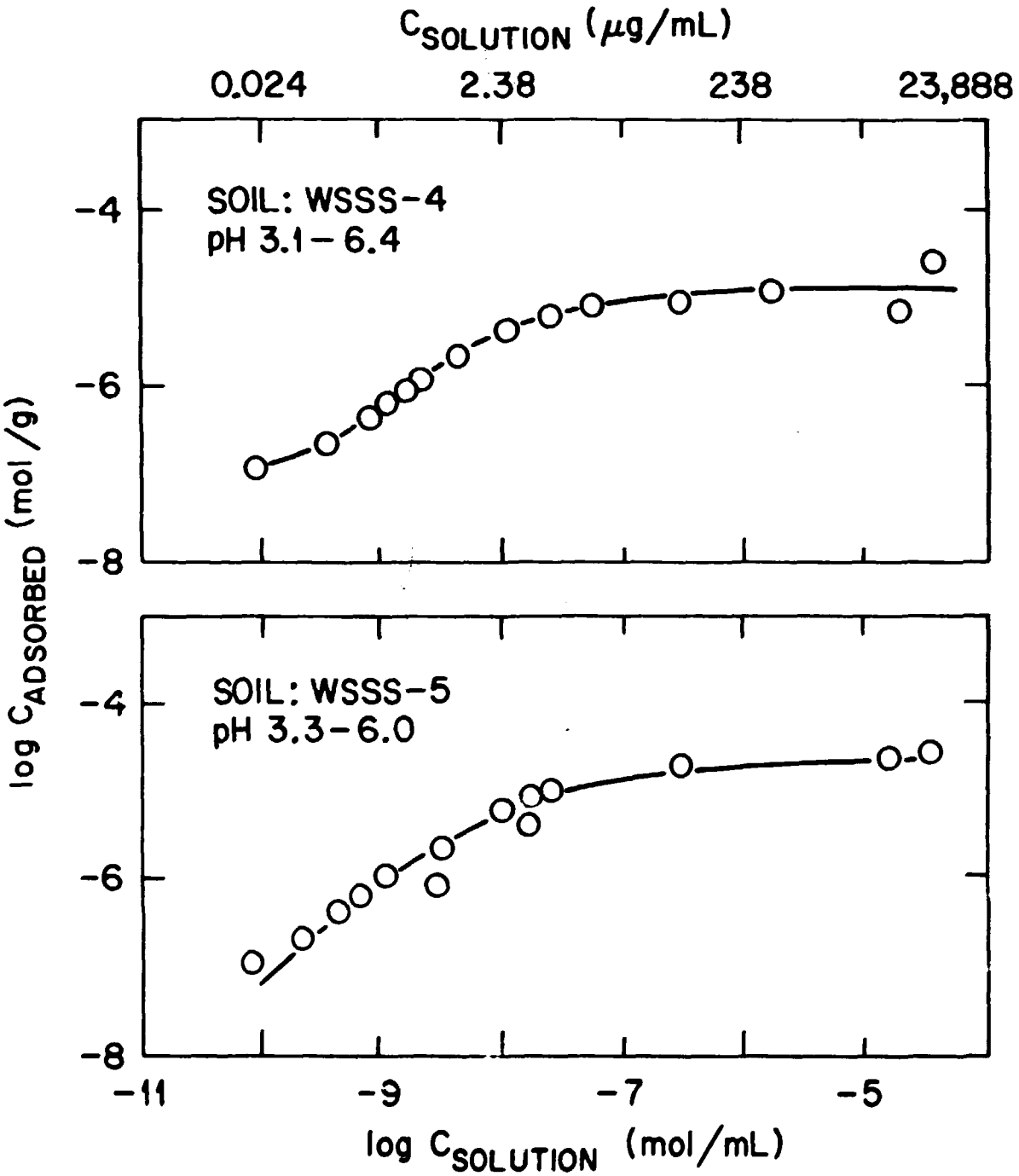

Fig. C-2. Uranium(VI) sorption isotherms for soil samples WSSS-4 and WSSS-5 from the Weldon Spring Storage Site.

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