ORNL-6083
Dist. Category UC-70A

Chemical Technology Division

GEOCHEMICAL INFORMATION FOR SITES CONTAMINATED WITH LOW-LEVEL RADIOACTIVE WASTES: I - NIAGARA FALLS STORAGE SITE

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ORNL--6083

DE85 003718

Date of Issue: November 1984

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831
operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under Contract No. DE-AC05-84OR21400

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
NTIS price codes—Printed Copy: A07 Microfiche A01

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PREFACE

This is the first in a series of three reports that document geochemical information obtained for three sites contaminated with low-level radioactive wastes. Other reports in this series will be published at a later date.

GEOCHEMICAL INFORMATION FOR SITES CONTAMINATED WITH LOW-LEVEL RADIOACTIVE WASTES: I - NIAGARA FALLS STORAGE SITE

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ABSTRACT

The Niagara Falls Storage Site (NFSS) became radioactively contaminated as a result of wastes that were being stored from operations carried out to recover uranium from pitchblende ore in the 1940s and 1950s. The U.S. Department of Energy (DOE) is considering various remedial action options for the NFSS. This report describes the results of geochemical investigations performed to help provide a quantitative evaluation of the effects of various options. NFSS soil and groundwater samples were characterized; and uranium and radium sorption ratios, as well as apparent concentration limit values, were measured in site soil/groundwater systems by employing batch contact methodology.

Near-surface soils were typically lacustrine clay or silty-clay types. Appreciable amounts of dolomite appeared to be present. A core profile changed from sandy loam at the surface to dark clay with chert at the 13.7-m (45-ft) depth just above the underlying red shale bedrock. Radiochemical analysis of the soil core revealed contamination as high as 2600 μg/g for uranium and 8300 pCi/g for ^{226}Ra at depths near the original site surface. The core profile showed no evidence of downward migration of uranium or radium during the 30-year history of waste storage in the R-10 pile. Lateral migration was not explored.

Analysis of groundwater from the soil indicated that it was a dilute sodium/calcium/magnesium sulfate solution, while groundwater from the bedrock was much more concentrated in sodium chloride/sulfate. These analytical differences suggest that there is little vertical communication between bedrock and soil groundwaters.

Batch contact methodology was used to construct sorption isotherms for uranium with three types of near-surface soils and a soil core profile from the surface to bedrock. Radium sorption ratios were measured at one radium concentration. All tests employed well water from the NFSS.

Poor uranium sorption behavior was exhibited by all soil/groundwater systems; maximum sorption ratios ranged from 3.9 to 9.0 L/kg at the lowest uranium solution concentrations tested and decreased to 1 L/kg at higher concentrations. One sample of soil at the 13.7-m (45-ft) depth (just above bedrock) showed high uranium sorption. Uranium was very soluble in soil/groundwater systems; the apparent concentration limit was greater than 6 g/L. The high solubility was shown to be due to the formation of the soluble uranyltricarbonate anionic complex. Very high radium sorption ratios (up to 11,200 L/kg) were obtained.

The results suggest that any uranium which is in solution in the groundwater at the NFSS may be poorly retarded due to the low uranium sorption ratio values and high solubility measured. Further, appreciable concentrations of uranium in groundwater could be attained from soluble wastes. Release of uranium via groundwater migration could be a significant release pathway. Solubilized radium would be expected to be effectively retarded by soil at the NFSS as a result of the very high radium sorption ratios observed.

The addition of iron oxyhydroxide to NFSS soils resulted in much higher uranium sorption ratios. Additional field testing of this potential remedial action additive could be desirable.

1. EXECUTIVE SUMMARY

The NFSS is one of the radioactively contaminated sites being addressed by DOE in its Surplus Facility Management Program (SFMP). These sites were contaminated (primarily with natural uranium and uranium decay-chain products such as radium) as a result of the processing of pitchblende ore for the recovery of uranium in the 1940s and 1950s or the subsequent storage of the wastes generated by these operations. The NFSS is the current storage site for most of the pitchblende processing wastes. These wastes were collected during the 1950s and 1960s from other locations throughout the United States and transferred to the NFSS; no pitchblende processing was performed at the NFSS. Within the SFMP activities, the DOE is evaluating various remedial action alternatives for these radioactively contaminated sites. Alternatives for the NFSS may range from maintenance at essentially the current conditions to various degrees of decontamination and possible return of the site to other uses.

Determination of geochemical information for the NFSS is one of the activities involving the Surplus Facility Management and the Formerly Utilized Sites Remedial Action Programs. Such information is necessary for the evaluation of various remedial action alternatives. This report describes the results of the investigation of two geochemical aspects: (1) characterization of NFSS soils and groundwaters, and a uranium and radium soil core profile; and (2) laboratory measurement of uranium and radium sorption behavior and apparent concentration limit values in NFSS soil/groundwater systems. These data, particularly the radioactive element sorption ratio and solubility values, are needed for the Environmental Impact Statement (EIS) in order to quantify calculated radioactive element mobility predictions for various remedial action alternatives.

1.1 SOIL CHARACTERIZATION

Seven near-surface soil samples and one auger-drilled soil core profile from the surface through the main waste disposal surface pile (the R-10 pile) to the bedrock were analyzed. The near-surface soil samples showed similar properties, which were characteristic of lacustrine clay or silty-clay soils. The moisture content ranged from 10 to 27% and the surface area from 18.8 to 29.3 m^2/g. Analysis of the samples showed the presence of major amounts of aluminum, calcium, iron, potassium, magnesium, silicon, and sodium — elements that are typical of clay soils. The near-surface soils had high carbonate contents (7.8 to 15.1%), which are consistent with the calcium and magnesium contents and suggest the presence of appreciable amounts of dolomite. The presence of dolomite is reasonable since the NFSS location is several miles northeast and below the dolomite escarpment east of the city of Niagara Falls. Quartz, illite, and minor amounts of kaolinite were identified by x-ray diffraction.

The soil core (borehole BH-77) from the surface to the 13.7-m (45-ft) depth showed considerable difference in properties with depth. The samples changed from sandy loam at the surface to dark-brown clay containing chert at the 13.7-m (45-ft) depth, which is just above the underlying red shale bedrock. The carbonate content increased consistently with depth. A uranium and radium profile for the BH-77 core clearly indicated contamination from the surface down to about 3.2 m

(10.5 ft); the highest values measured were 2600 μg/g for uranium and 8300 pCi/g for ^{226}Ra. Deeper samples revealed some contamination, which may have occurred, at least in part, during core removal. The samples showed no evidence of significant downward migration of uranium or radium during the 30-year history of the R-10 pile. Lateral migration could not be explored since only one vertical profile was available.

1.2 GROUNDWATER CHARACTERIZATION

Six well-water samples were received and characterized. In addition, 16 wells were sampled by pumping and, in some cases, by bailing techniques. These wells were cased to three depths: shallow soil [4.6 to 8.8 m (15 to 29 ft)], deep soil [10.7 to 16.8 m (35 to 55 ft)], and bedrock [15.2 to 30.5 m (50 to 100 ft)]. Chemical analysis of these samples showed characteristic differences in the water from the soil and that from the bedrock. Water from the various soil depths was primarily a dilute sodium/calcium/magnesium sulfate solution with lesser concentrations of bicarbonate and chloride at pH 7.3 to 7.9. Water from the bedrock was much more concentrated in sodium chloride/sulfate and lower in bicarbonate; the pH was 7.4 to 7.9. These analytical results suggested that there was not much vertical communication between the groundwater in the bedrock and that in the soil.

1.3 EXPERIMENTAL METHODOLOGY

Batch contact methodology was used to measure sorption ratios and to construct sorption isotherms for uranium in site soil/groundwater systems. Water from well W-1 was used in all of the tests. Various near-surface soil samples and BH-77 core samples were used. Soil and uranium-traced groundwater were usually contacted at a solution/solids ratio of 5 for 24 h. The solution was then recovered by centrifugation for analysis by alpha counting techniques. The sorption ratio was calculated from the solution uranium concentration, and the sorption isotherms were constructed by plotting the concentration in solution vs the concentration on the solids. The sorption isotherms were experimentally extended to high uranium concentrations in an attempt to measure apparent concentration limit values.

A few similar tests were carried out at one radium concentration to obtain radium sorption ratios. The radium activity was determined by allowing the samples to establish secular equilibrium and then counting the ^{214}Bi gamma peak.

1.4 SUMMARY OF URANIUM SORPTION AND APPARENT CONCENTRATION LIMIT INFORMATION

A summary of the uranium and radium sorption ratio and apparent concentration limit information is given in Table 1.

Since the uranium concentration present in groundwater at the site in groundwater migration events is expected to be relatively low, for the purposes of this summary table the sorption ratios obtained in contacts at an initial uranium concentration of 6 mg/L were tabulated for comparison. These are the highest sorption ratios measured; lower values were obtained

Table 1. Uranium and radium sorption ratios and apparent concentration limits

Soil sample	Uranium Rs[a] (L/kg)	Uranium Conc. limit (g/L)	Radium Rs[b] (L/kg)
Near-surface samples			
Gray clay[c]	5.6	>6.2	6,400
Brown-clay backfill[c]	8.7	>7.9	11,200
Lake clay[c]	6.1	>8.8	7,400
BH-77 core samples			
0.3 m	3.9	>4.0	d
2.1 m	7.0	>3.1	d
7.9 m	9.0	>7.2	d
12.2 m	7.0	>7.2	d
13.7 m	8400	>4.5	d

[a] Contacted at an initial uranium concentration of 6 mg/L.

[b] Contacted at an initial radium concentration of 10^5 pCi/mL.

[c] Name designated by Bechtel National.

[d] Not measured.

at higher uranium concentrations. The full sorption isotherms are included in the body of the report. The apparent concentration limit values obtained at the highest uranium concentrations tested are also given in the table. The sorption isotherms did not indicate approach to a concentration limit. The radium sorption ratio values were obtained in contacts at only one initial concentration, 10^5 pCi/mL.

Poor uranium sorption behavior was exhibited by nearly all the near-surface and core-profile soil samples. The uranium sorption ratios were similarly low for the three near-surface soils and for the BH-77 core samples, with the exception of the 13.7-m (45-ft) sample. The sorption ratios were highest, 3.9 to 9.0 L/kg, at the lowest uranium concentration and decreased to lower values (often near 1 L/kg) at the higher uranium concentrations. (Decreasing sorption ratios at higher solution concentrations are normally observed due to saturation of the sorption sites.) The only sample that had high uranium sorption characteristics was from the 13.7-m (45-ft) depth just above the bedrock. This sample was observed to contain appreciable amounts of an iron precipitate, which was probably responsible for the much higher uranium sorption ratios. In a separate series of tests, the NFSS soils showed greatly increased uranium sorption ratios when an iron oxyhydroxide precipitate was mixed with the soils. Very high radium sorption ratios were obtained for the three near-surface soils. This behavior is characteristic of radium in most geologic media.

Uranium was very soluble in all the soil/groundwater systems tested. The average uranium solubility limit was greater than 6 g/L; no apparent concentration limit was observed. The high solubility was shown spectrophotometrically to result from the formation of the soluble uranyltricarbonate anion. Formation of this anion would be expected because of the high carbonate content of the groundwater/soil systems. A much lower apparent concentration limit was observed in tests where the carbonate was artifically destroyed by acid pretreatment. The presence of anionic uranium species is also consistent with the low sorption ratios measured; anions normally are poorly adsorbed on geologic media.

The addition of 10% bentonite to the near-surface soils did not improve the uranium sorption behavior. Bentonite addition was evaluated because it is frequently used to help isolate hazardous wastes from groundwater due to its swelling properties when hydrated. The absence of any measurable effect was surprising since much higher uranium sorption ratios had been obtained in earlier tests with montmorillonite (a major component of bentonite) and uranium-traced water. Possibly, the high carbonate content of the NFSS soil/groundwater systems and the resulting formation of uranyltricarbonate anions prevented uranium sorption onto the bentonite.

1.5 IMPLICATIONS FOR URANIUM AND RADIUM RETENTION BY THE NIAGARA FALLS STORAGE SITE

While the geochemical information developed during this investigation may be quantitatively utilized in radioactivity migration calculations for the NFSS EIS, it is also possible to make some qualitative observations relative to expected uranium and radium behavior at the NFSS.

The NFSS soil/groundwater systems do not exhibit favorable aspects for uranium retardation; therefore, we may expect any uranium that is in solution in the site groundwater as a result of solubilization from solid wastes to be poorly retained by the soil. This conclusion is based on the high uranium solubility observed, in combination with the poor sorption behavior. Further, if readily leachable or soluble uranium-containing wastes are present, appreciable concentrations of uranium could be achieved in groundwater. Since the NFSS is in a high rainfall area, the release of solubilized uranium via groundwater migration may be a significant release pathway.

The one core profile analyzed in this study showed no evidence for significant uranium downward migration (as might be expected from the low sorption and high solubility) following emplacement of the R-10 pile. However, the vertical groundwater transmissivity is not known and may be low. Low transmissivity is suggested by the large difference in chemical analysis of the bedrock and soil groundwaters. Since only one core profile was analyzed, it is not possible to speculate on possible horizontal migration of uranium in groundwater over the 30-year history of the R-10 pile. Additional hydrologic information and analysis of a suite of soil cores would be needed in order to resolve this question.

The NFSS soil/groundwater systems appear very favorable for the retardation of radium. Radium solubilized in groundwater would be very strongly adsorbed by the soil as a result of the very high sorption ratios measured. Radium may be well retained by the NFSS.

The addition of bentonite to NFSS soil would not be expected to improve uranium retardation by sorption onto the soil since the addition of 10% bentonite did not increase the uranium sorption ratios observed. The use of bentonite could have favorable site retention aspects by aiding the restriction of groundwater access to wastes, but tests to explore this possibility were not included in our study.

The addition of iron oxyhydroxide to NFSS soils is a potential approach for greatly increasing uranium retention by the soil since much higher sorption ratios were observed with soils to which it had been added. Because iron oxyhydroxide is relatively inexpensive and easy to prepare, an admixture of it with soil to form a uranium migration barrier could be a potentially favorable remedial action option. Additional field testing would be desirable to confirm the laboratory findings.

2. INTRODUCTION

The most credible scenario for the release of emplaced radioactivity from a near-surface disposal site to the accessible environment involves groundwater intrusion of the waste, followed by dissolution and migration of radionuclides in groundwater to the surface or into a subsurface aquifer. The radionuclides may be transported either as dissolved species or as insoluble colloids or other suspended particulates. The geochemical processes or conditions that control migration as dissolved species are, in sequence of action: (1) groundwater intrusion, including both volume and migration rate; (2) dissolution of the emplaced radionuclide(s); (3) solubility product limitation of the radionuclide concentration in the groundwater; and (4) retardation of the dissolved radionuclide via sorption by soil and rock constituents of the site. In general, the quantity of any radionuclide that is released or is migrating from a disposal site can be no greater than its saturated solution concentration times the groundwater volume, and the radionuclide migration rate can be no faster than the groundwater migration rate. Migration of the radionuclide may be further retarded by sorption on surfaces of the soil and/or rock with which it comes in contact.

The degree of retardation of migrating radionuclides dissolved in groundwater by the geochemical sorption process is generally expressed as a retardation factor ($\underline{R}$), which is related to the experimentally measured distribution coefficient (Kd) by the relationship:

$$\underline{R} = 1 + Kd(\phi/\rho), \tag{1}$$

where

$\underline{R}$ = retardation factor, dimensionless;
Kd = distribution coefficient, L/kg;
ϕ = bulk density of geologic media, kg/L;
ρ = porosity of geologic media, dimensionless.

This expression is rigorously valid only when Kd is independent of radionuclide concentration, the sorption process is thermodynamically reversible, and a single solution species exists. These conditions are seldom met in most practical applications; therefore, judgment must be exercised in the design of the experiments employed to measure the distribution coefficients and in the extrapolation of small-scale laboratory tests to the prediction of geologic-scale behavior of the radionuclides.

The philosophy, or approach, taken in the experimental work reported here has been developed by taking into consideration the technical and theoretical aspects of the geochemical sorption process and solubility limit condition, as well as the programmatic information needs. Sorption values are frequently calculated as an empirical ratio (i.e., an effective distribution coefficent or sorption ratio). This 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 calculating the sorption ratio, the units are liters per kilogram (L/kg). In the past, there has been no generally accepted standard symbol(s) to represent sorption ratios or distribution coefficients. Recently, standard symbols and terminology have been recommended.[1] The symbol "Kd" should be reserved for distribution coefficients which have been obtained under steady-state or thermodynamic equilibrium conditions. Few practical experiments can be demonstrated as being at equilibrium; thus, most experimentally derived sorption values should be represented by the symbol "Rs" for sorption ratio or "Rd" for desorption ratio. We have chosen to express our short-term laboratory measurements of the distribution of radionuclides between groundwater and a solid adsorbent in terms of a sorption ratio (Rs), which does not imply ideal behavior. At thermodynamic equilibrium,

$$Rs = Rd = Kd, \tag{2}$$

where

Rs = sorption ratio, L/kg;
Rd = desorption ratio, L/kg;
Kd = equilibrium distribution coefficient, L/kg.

Only when sorption-desorption equilibrium has been established is the sorption ratio a true distribution coefficient (provided that multiple speciation does not exist).

The solubility product constant for a solid is expressed as:

$$Ksp = [A^{y+}]^x [B^{x-}]^y \tag{3}$$

for the hypothetical equilibrium:

$$AxBy = xA^{y+} + yB^{x-}. \tag{4}$$

The Ksp value is unique for each solid phase and solution species and is rigorously valid only for ionic activities. The activity coefficient for dilute species is often 1; thus, ionic concentrations are often substituted for activities. Since the solid phase, solution species, and activity coefficients are frequently difficult to determine or are not known for complex geologic soil/groundwater systems, an observed or apparent saturated solution concentration is usually obtained experimentally. This apparent saturated concentration is dependent on several experimental and site-specific parameters and, therefore, must be redetermined for each new soil/groundwater combination. In our work, we have designated this as an apparent concentration limit.

Many nuclear facilities (power plants, uranium production facilities, and disposal/storage sites) are located in areas of the eastern United States that experience high rainfall and have clay-rich soils derived from sedimentary rocks. A review of the published literature revealed little information describing sorption behavior for uranium species (one of the radionuclides of major concern at the NFSS) with water-saturated, clay-rich soils. Most of the reported values are for fission products or transuranic elements, are related to arid soils more typical of the western United States, or are for specific geological materials such as salt, granite, etc. Borovec[2] describes the uptake of UO_2^{2+} on kaolinite, illite, and montmorillonite from pure uranyl chloride solutions at pH 6. Natural waters, however, may contain low concentrations of many ionic species that can complex dissolved uranium, causing increased solubility.

Nonideal sorption behavior of U(VI) may be expected because of the multiple species possible under soil/groundwater conditions. Langmuir[3] has reviewed the stability data for uranium complex species, and Baes and Mesmer[4] have examined the solubility data for uranium species. The following conclusions for U(VI), the oxidation state to be expected in air-saturated groundwaters, have been reached: (1) below pH 5, the predominant ionic species in groundwaters would be UO_2^{2+}, or UO_2F^- if fluoride is present; (2) the complex $UO_2(HPO_4)_2^{2-}$ would be the principal species in the pH range from 5 to 7, but in the absence of PO_4^{3-} the hydrolyzed species would probably be $(UO_2)_2(OH)_2^{2+}$ or $UO_2(OH)^+$; and (3) above pH 7, the ion $UO_2(CO_3)_3^{4-}$ becomes the dominant form in the presence of dissolved CO_2, while insoluble uranium hydroxide and the ion $(UO_2)_3(OH)_5^+$ would be formed in the absence of CO_2. Lemire and Tremaine[5] have recently compiled the existing information on uranium solution equilibria and calculated equilibrium constants for pertinent reactions. According to Baes and Mesmer,[4] the U(VI) oxide solid phases in equilibrium with pure water (no complexing ions present) are not well characterized, and several modifications of $UO_2(2H_2O)$ and $UO_2(OH)_2$ are believed to exist; thus, an accurate value for the solubility of these forms has not been established.

Giblin et al.[6] evaluated the effects of carbonate and kaolinite in natural waters containing complexing ions on the mobility of U(VI). His studies showed that carbonate substantially increased mobility, while kaolinite reduced mobility, reportedly by immobilizing colloidal uranium oxides.

Single sorption ratio values obtained for a given adsorbent and test solution may be presented in data tables. Such tables often provide a comparison of various adsorbents under one set of fixed experimental conditions and, consequently, are useful for screening or gross comparison purposes. They are not recommended for a sorption behavior or retardation analysis, however, since single sorption ratio values give no hint of the relationship of sorption behavior to test conditions or geochemical parameters.

It is well known that the sorption ratio is frequently a function of the solute concentration, and plots of adsorbed concentration vs solution concentration have been employed as a method of presenting the data. Such plots are called isotherms, even though all the test parameters except concentration (and not just the test temperature) must be held constant. Adsorbed species, including many radionuclides on soils or rocks, sometimes display a nonlinear dependency of adsorbed concentration vs solution concentration (as saturation loading occurs) similar to that classically observed for the sorption of gases on solids. Such behavior was initially described by Freundlich in 1907, and his name has become a generic label for any sorption plot showing this type of curve. Similarly, in 1926, Langmuir developed an isotherm for the adsorption of gases on solids based on the assumption of a unimolecular surface; and for ideal systems, the plots that show the total gas pressure as a function of the adsorbed gas concentration yield a straight line. Some data for the adsorption of solutes from solution have been successfully graphed using a Langmuir isotherm approach.

Sorption of radionuclides on soil and rocks from groundwater is much more complex and involves many more factors than those dealt with by Freundlich or Langmuir isotherms; nonetheless, such data treatments have proved useful as a means of organizing and presenting sorption data. The Dubinin-Raduskevitch isotherm, which was developed as a means of obtaining linear plots of sorption values for gases on charcoal, has also proved useful for describing the sorption of sparingly soluble solutes since, unlike the Langmuir and Freundlich isotherms, it does not assume a homogeneous surface, constant adsorption potential, or monolayer surface saturation. In the Dubinin-Raduskevitch isotherm, the logarithm of the concentration of the adsorbed species is plotted vs a function of the sorption potential energy. This type of plot has been used to successfully linearize sorption data for uranium in geologic materials.[7]

The sorption data in this report are presented as isotherms, insofar as possible, supplemented by detailed tabulation of the test results in the appendixes. Single sorption ratio values may not reveal adequate information for accurate and defensible migration modeling. The apparent concentration limit or saturated solution concentration values are inferred from the sorption isotherm whenever possible. An example of this method of estimating apparent concentration limits is shown in Fig. 1. Here, uranium sorption isotherms that were determined at pH values ranging from 3 to 9 (and reported earlier[8]) exhibited a well-defined upward inflection at the point which we have defined as the apparent concentration limit. It can be seen that this point, which suggests a major species change, varies significantly with pH. Sorption rate experiments are shown to indicate the existence of, or proximity to, steady-state conditions. In some cases, desorption ratios (Rd) may be much larger than sorption ratios (Rs) due to chemisorption or other irreversible sorption processes. When this occurs, Rd will dominate the calculation of the retardation factor and should be used instead of Rs for accurate migration modeling calculations. Time limitations did not permit the determination of Rd values for the NFSS soils. If the radionuclide is present in more than one dissolved form (i.e., multiple speciation exists) during the test, then the measured sorption ratio will be an average of the sorption ratios for the various species, weighted by their relative abundance. Use of a single sorption value or single isotherm to model retardation for a system containing multiple speciation could lead to large errors and nonconservative migration predictions. Sorption column chromatography should be employed as a means of supplying experimental backup for the batch test determinations of sorption isotherms to explore the existence of multiple speciation.

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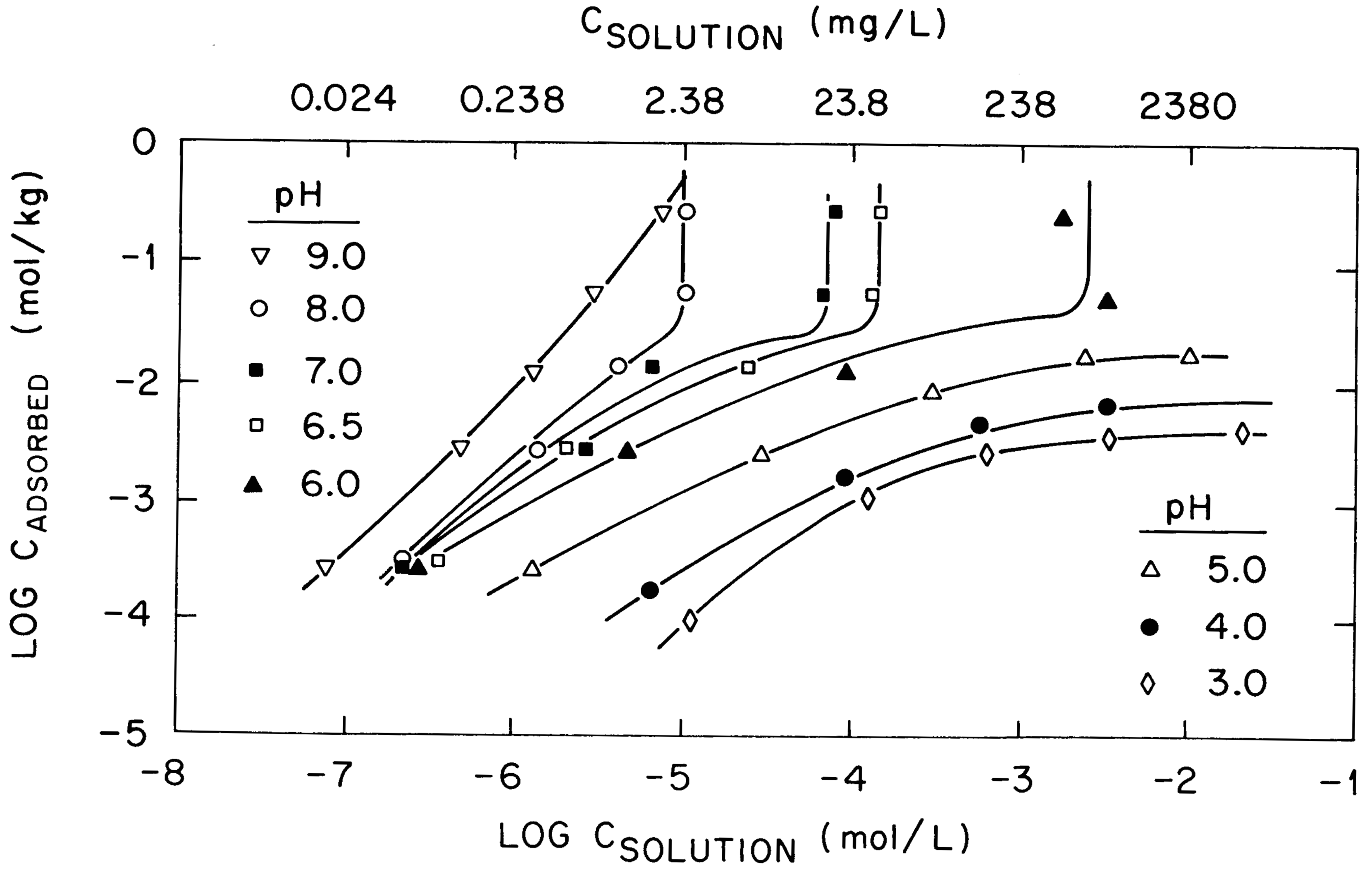

Fig. 1. Sorption of uranium on montmorillonite.

3. SITE CHARACTERIZATION

The Niagara Falls Storage Site, located in western New York within the town of Lewiston, Niagara County, is approximately 6.4 km (4 miles) south of Lake Ontario and 16 km (10 miles) north of Niagara Falls.[9] The site (see Fig. 2), which is owned by the DOE, is a portion of the original Lake Ontario Ordnance Works established by the U.S. Department of the Army early in World War II for the production of TNT. It is level, sloping slightly to the northwest, and ranges in altitude between 96 and 99 m (315 and 325 ft) above sea level. The site is underlain by Ordovician Queenston shale, which lies approximately 9 to 15 m (30 to 50 ft) below the surface and is approximately 366 m (1000 ft) deep at its maximum thickness. The soils above this shale stratum are primarily derived from lake bed sediments and are poorly drained silt loams with low organic matter. The site stratigraphy is presented in Table 2.

The climate at the NFSS is moderately humid, with a mean annual rainfall of 81 cm (32 in.) and a mean annual temperature of 9°C (48°F). Water is present in an aquifer which extends upward from the surface of the bedrock through numerous sand/gravel lenses and saturated clay zones 2 to 6 m (5 to 20 ft) in depth. Surface features at the site (Fig. 3) include several storage and service structures, most important of which are (1) a 50-m (164-ft) silo or tower (formerly used for water storage) currently being used as a repository for multicurie quantities of radium residues; (2) several abandoned buildings which have been used for contaminated waste storage; and (3) a mound of contaminated soil having a total volume of about 11,500 m^3 (15,000 yd^3) (designated as the R-10 pile) and containing an estimated 8.7×10^6 kg of dry solids with a uranium content of approximately 2300 ppm.[9]

The NFSS, designated as a SFMP site, has been the repository for most of the radioactive wastes generated by the pitchblende ore processing operations carried out during the 1940s and 1950s, as well as some other natural uranium wastes. During the time that radioactive wastes were being placed on the site, management practices did not provide accurate records concerning the characteristics, inventories, or detailed locations of the stored wastes. As a result, site characterization to

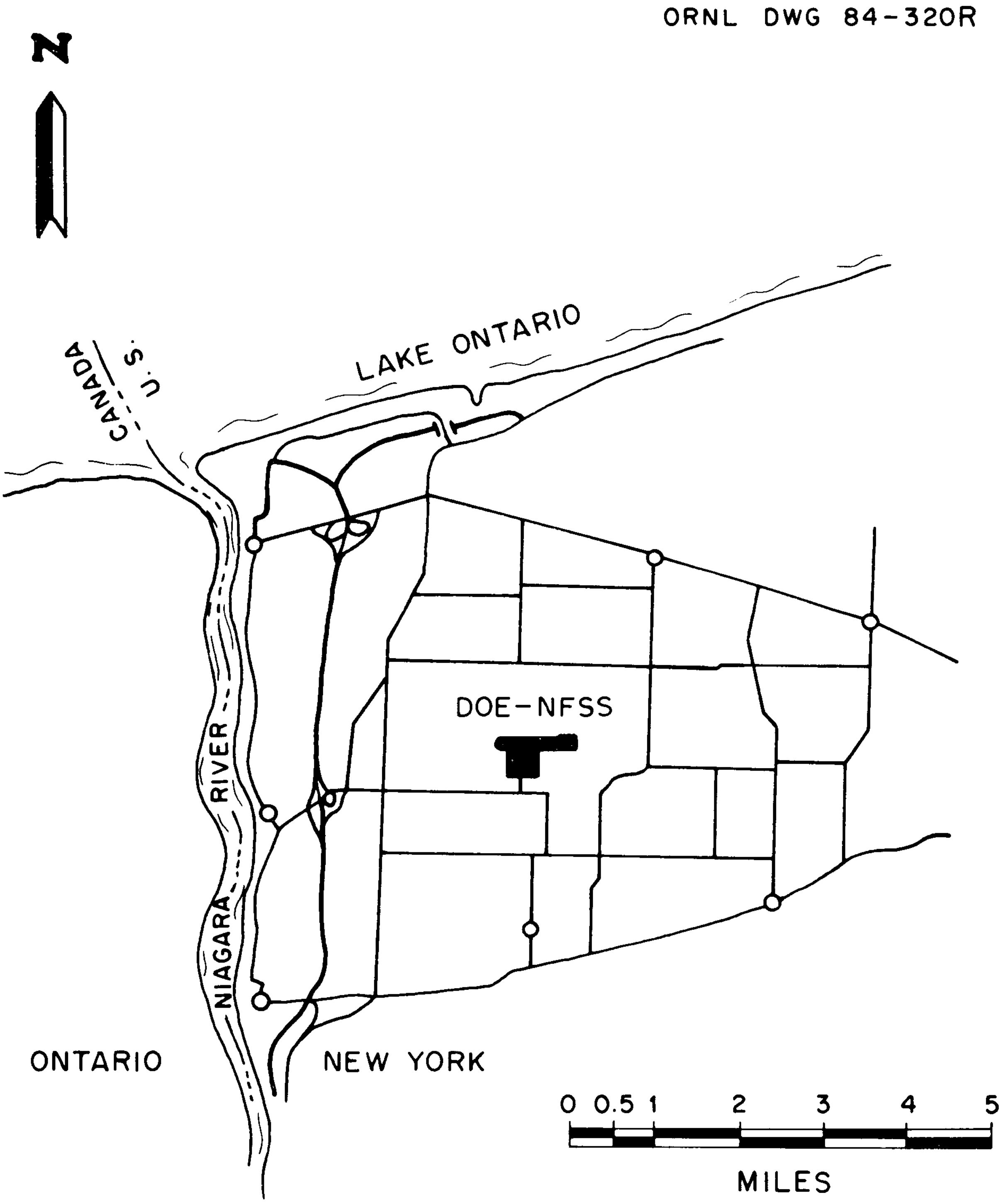

Fig. 2. Location of the Niagara Falls Storage Site (NFSS).

Table 2. Stratigraphy of the NFSS

Unit	Observed thickness (m)	Appearance	Description
A	Average, 0.6 to 0.9; maximum, 1.5	Brown silt and sand with mixtures of gravel and clay	Topsoil composed of weathered glacial-lacustrine sediments mixed with organic material; some localized disturbance and replacement with artificial fill[a]
B	Average. 3.0 to 4.6; maximum, 7.6	Brown silt containing clay areas of abundant gravel and sand	Unweathered glacial-lacustrine sediments, possibly from glacial Lake Iroquois
C	Average, 5.2 to 6.7; minimum, 3.0 to 4.6; maximum, 8.8	Brown-gray or gray-brown silt with a high clay content; contains small amounts of sand or gravel at scattered locations	Clay-rich glacial till
D	Average, 0.6 to 2.1	Brown silt with abundant sand in some locations	Glacial till
E	Average, 0.6 to 1.2; maximum, 2.1	Gravelly or sandy silt with fragments of bed rock; clayey in part	Basal glacial-lacustrine sequence member; glacial till of uncertain age
BR (bedrock)	--	Red-brown siltstone, mudstone, and shale	Upper Ordovician Queenston shale

[a] The localized disturbance and replacement may be as great as 15 to 25 ft in the R-10 pile area.

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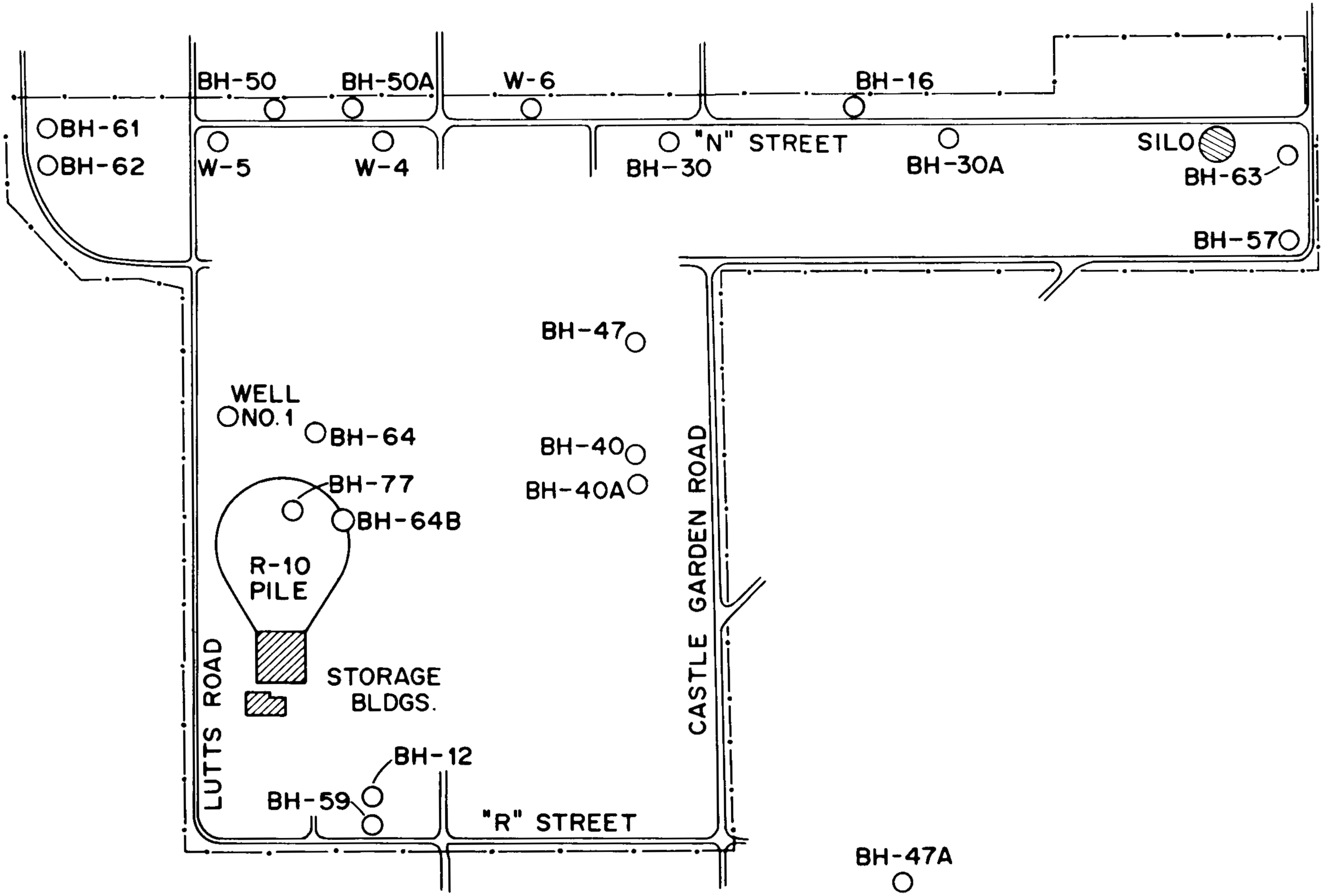

Fig. 3. Pertinent surface features at the NFSS.

determine current or potential future migration of radionuclides to the environment has become necessary. Bechtel National, the current DOE contractor for NFSS site management, has prepared several detailed scenarios for remedial action options of the NFSS.[10]

3.1 SOIL CHARACTERIZATION

3.1.1 Sample Description

Soil samples were obtained from the NFSS from two sources. Seven near-surface samples were supplied by Bechtel National. These included three samples of "gray clay," present at the bottom of trenches which were dug to provide containment for a slurry wall barrier against the migration of radionuclides from the R-10 pile; three samples of "brown-clay backfill," used for backfill operations around the trench and R-10 pile; and one sample of "lake clay," an off-site soil which had been hauled in for use in building a slurry wall around the R-10 pile to provide a barrier to the possible migration of radionuclides from the contaminated zone. Soil samples from a borehole (BH-77) drilled through the center of the R-10 pile to the Queenston red shale bedrock were obtained from the NFSS in 1981, during the time that National Lead of Ohio (NLO) was site manager and Acres American was the site geologic and hydrologic work subcontractor.

3.1.2 Sample Preparation and Physical Analysis

The seven samples were weighed, dried by lyophilization, reweighed, and lightly ground in a Braun mill to pass a 50-mesh screen (100% <297 μm). The moisture content of each is shown in Table 3. The moisture content was found to vary in the following order: gray clay (19 to 27%) > brown-clay backfill (17 to 19% > lake clay (10%). A summary of the particle size distribution analyses for these soil samples is presented in Table 4. Details of these particle size distribution analyses are shown in Figs. A-1 through A-7, Appendix A. All the samples have quite similar characteristics and contain major amounts of very fine particles. The 50% distribution point ranged from 4.5 to 7.2 μm. If a standard definition of soil types[2] is employed, all the NFSS near-surface soil samples can be categorized as clay or clayey silt.

Table 3. Weight loss on drying for the NFSS soil samples

Sample designation	Weight loss on drying (% of total)
GC-1	27
GC-2	23
GC-3	19
BCB-1	19
BCB-2	17
BCB-3	15
Lake clay	10

Table 4. Particle size distribution analysis of the NFSS soil samples

Soil sample	Density (g/cm^3)	Particle size distribution[a] (μm)		
		90%	50%	10%
GC-1	2.7	56	5	2
GC-2	2.7	73	5	2
GC-3	2.7	108	7	2
BCB-1	2.5	96	6	2
BCB-2	2.5	98	7	3
BCB-3	2.5	88	5	2
LC-1	2.3	17	5	2

[a] Particle size is less than the listed value.

Results of the surface area analysis of the seven soil samples by Brunauer, Emmett, and Teller (BET) nitrogen adsorption techniques are shown in Table 5. All the soils have relatively high surface areas, ranging from 19 to 29 m^2/g. These high surface areas are consistent with both the fine particle sizes and clay contents of the soils. The mean density (and standard deviation) was 2.56 (± 0.14) g/mL.

3.1.3 Chemical Analysis

The seven samples were analyzed for elemental contents by the inductively coupled plasma (ICP) spectrometric technique, and the resulting data are summarized in Table 6. The analyses are generally typical of those to be expected for clay soils. The relatively large amounts of calcium and magnesium present in the soils suggest that dolomite and/or calcite might be present. Carbonate analysis of these samples (see Table 7) indicated a moderately high concentration of this anion, supporting the possibility of the presence of dolomite and/or calcite. Concentrations of 8 to 11% CO_3^{2-} were found in the samples of gray clay and brown-clay backfill, while 15% CO_3^{2-} was found to be present in the sample of lake clay, which was chosen for slurry-wall construction material prior to the time that this analysis was made. Equivalent balances, shown in Table 8, further substantiate the possibility of the presence of dolomite or calcite.

X-ray diffraction (XRD) techniques were used to identify the crystalline phases present in the soil samples. The results of these analyses show that silica was the major crystalline component in all of the soils, and that illite, a typical soil clay mineral, was present at intermediate levels. Although these crystalline phases appear to be consistent with the elemental composition of the samples, there was no evidence of dolomite or calcite. These components, if present, may be below the detection limit of the XRD technique used in this system.

3.1.4 Core Profile Analysis

Core BH-77 was an auger-type core drilled through the highest surface radiation zone of the R-10 pile to the Queenston shale bedrock level, which was 13.7 m (45 ft) from the top of the R-10 pile and 10.7 m (35 ft) from the site surface. This core was selected for the determination of

Table 5. Surface area analysis of the NFSS soil samples

Sample No.	Surface area[a] (m^2/g)
LC-1	24.3
GC-1	29.3
GC-2	20.6
GC-3	18.3
BCB-1	19.0
BCB-2	23.6
BCB-3	22.4

[a] Surface area measured by BET nitrogen adsorption after samples were dried by lyophilization.

Table 6. Concentrations (mg/L) of elements in soil samples from the NFSS, as determined by ICP spectrometry

Element	Sample GC-1	 GC-2	 GC-3	 BCB-1	 BCB-2	 BCB-3	 LC-1
Ag	<4.5	<4.5	<4.5	<4.5	<4.5	<4.5	<4.5
Al	80,000.0	64,000.0	66,000.0	62,000.0	69,000.0	55,000.0	58,000.0
B	20.0	<19.0	<19.0	<19.0	<19.0	<19.0	26.0
Ba	460.0	380.0	440.0	410.0	450.0	410.0	370.0
Be	2.1	1.6	1.6	1.5	1.8	1.5	1.6
Ca	50,000.0	51,000.0	48,000.0	55,000.0	47,000.0	59,000.0	77,000.0
Cd	2.3	2.4	4.3	<2.3	2.7	<2.3	<2.3
Co	18.0	14.0	15.0	12.0	14.0	12.0	11.0
Cr	75.0	60.0	61.0	53.0	61.0	46.0	61.0
Cu	26.0	20.0	21.0	25.0	24.0	15.0	9.6
Fe	43,000.0	36,000.0	37,000.0	34,000.0	38,000.0	31,000.0	31,000.0
Ga	31.0	22.0	26.0	19.0	22.0	22.0	22.0
Hf	38.0	36.0	37.0	30.0	34.0	30.0	36.0
K	26,000.0	18,000.0	19,000.0	17,000.0	19,000.0	16,000.0	16,000.0
Mg	19,000.0	16,000.0	16,000.0	13,000.0	14,000.0	12,000.0	17,000.0
Mn	710.0	810.0	870.0	900.0	770.0	790.0	540.0
Mo	160.0	100.0	110.0	140.0	87.0	110.0	110.0
Na	7,900.0	9,900.0	11,000.0	10,000.0	9,700.0	8,400.0	8,400.0
Ni	31.0	<28.0	<28.0	<28.0	<28.0	<28.0	<28.0
Pb	<65.0	<65.0	<65.0	<65.0	<65.0	<65.0	<65.0
Sb	<35.0	<35.0	<35.0	<35.0	<35.0	<35.0	<35.0
Se	<110.0	<110.0	<110.0	<110.0	<110.0	<110.0	<110.0
Sr	190.0	180.0	170.0	170.0	160.0	150.0	200.0
Ti	4,600.0	4,500.0	4,300.0	3,900.0	4,000.0	3,600.0	3,600.0
V	100.0	80.0	81.0	72.0	84.0	71.0	73.0
Zn	160.0	89.0	100.0	74.0	85.0	68.0	170.0
Zr	110.0	110.0	120.0	110.0	110.0	92.0	90.0

Table 7. Carbonate analysis of NFSS soil samples

Soil sample	Carbonate content (%)
GC-1	9.20
GC-2	9.66
GC-3	8.65
BCB-1	9.58
BCB-2	7.79
BCB-3	11.40
LC-1	15.10

Table 8. Magnesium plus calcium equivalent balance to carbonate (In meq/g)

	Sample						
Element	GC-1	GC-2	GC-3	BCB-1	BCB-2	BCB-3	LC-1
Ca	2.50	2.50	2.40	2.74	2.35	2.94	3.84
Mg	1.56	1.32	1.32	1.07	1.15	0.99	1.40
Co	3.07	3.22	2.88	3.19	2.60	3.80	5.03
C/A[a]	1.32	1.19	1.29	1.19	1.35	1.03	1.04

[a] $C/A = \frac{(Ca + Mg)}{CO_3^{2-}}$.

its uranium and ^{226}Ra concentration profile in order to explore in situ migration of these radionuclides. Laboratory experiments designed to measure radionuclide sorption values can never exactly duplicate the waste site field parameters because of practical constraints on the experimental design. Therefore, in situ verification of experimentally predicted radionuclide retardation/migration is always desirable when evaluating a waste site. Field data for comparison with laboratory test results are often difficult to identify; thus, the NFSS R-10 pile is one of the few instances where significant quantities of radionuclides, particularly uranium and radium, have been deposited for an extended period of time (30 years) in a site with defined stratigraphy and geochemical evaluation. It seemed desirable, therefore, to investigate the downward migration of uranium and radium in the vicinity of the R-10 pile so that it could be compared with the uranium and radium sorption behavior and retardation factors derived from laboratory tests to be described later in this report.

The natural uranium analysis was accomplished by delayed-neutron activation of the natural ^{235}U content of the total uranium concentration in the soil. Radium-226 concentration in the soil samples was determined by integrating the counts in the 0.609-MeV ^{214}Bi gamma peak after establishing secular equilibrium. The concentration profile for selected core samples is given in Table 9. The maximum uranium and radium concentrations (2600 μg/g of uranium and 8300 pCi/g of ^{226}Ra) were in the 2.9- to 3.2-m (9.5- to 10.5-ft) samples. Below this level, the concentrations of both uranium and radium decreased sharply to about 4 μg/g and less than 5 pCi/g, respectively, in an approximately 0.3-m (1-ft) increase in depth. Parallel uranium and radium concentration spikes, which were observed in several samples at greater depths, were most likely due to contamination of these samples during boring recovery through the overlying contaminated zone. Therefore, it was concluded that the uranium and radium profile analysis did not show any evidence of significant downward migration beneath the R-10 pile for either radionuclide during the approximately 30-year waste emplacement. Lateral migration was not explored since only one core profile was analyzed.

Table 9. Radium and uranium concentration profile for the NFSS core BH-77

Sample No.	Core depth (m)	Core depth (ft)	Description[a]	^{226}Ra conc. (pCi/g)	Uranium conc. (μg/g)
S1-1	0.2	0.5	Sandy loam fill	16 ± 2.1	30 ± 0.9
S1-2	0.3	1.0	Sandy loam fill	19 ± 1.6	52.7 ± 1.6
S2-2	0.6	2.0	Sandy loam fill	21 ± 1.9	104.0 ± 3.1
S3-1	0.9	3.0	Sandy loam fill	27 ± 2.0	22.8 ± 0.7
S4-1	1.5	5.0	Dk.-brn. clay; roots; twigs	66 ± 3.4	57.3 ± 1.7
S5-1	2.1	7.0	Dk.-brn. clay; roots; straw	263 ± 6.6	84.3 ± 2.5
S6-1	2.9	9.5	Lt.-brn. sandy loam fill	7592 ± 38	2610.0 ± 78
S6-2	3.0	10.0	Lt.-brn. sandy loam fill	8318 ± 42	2660.0 ± 80
S6-3	3.2	10.5	Lt.-brn. sandy loam fill	6939 ± 35	1330.0 ± 40
S7-1	3.5	11.5	Dk.-brn. clay	<5	3.9 ± 0.1
S7-2	3.7	12.0	Dk.-brn. clay	6 ± 1.5	18.6 ± 0.6
S7-3	3.8	12.5	Dk.-brn. clay	295 ± 5.6	63.3 ± 1.9
S8-2	4.3	14.0	Med.-brn. clay and gravel	35 ± 2.6	13.1 ± 0.4
S9-3	5.0	16.5	Med.-brn. clay and gravel	<5	2.6 ± 0.1
S10-2	5.3	17.5	Med.-brn. clay and gravel	<5	1.5 ± 0.05
S10-4	5.6	18.5	Med.-brn. clay and gravel	20 ± 2.2	12.0 ± 0.4
S11-1	5.8	19.0	Med.-brn. sandy loam	<5	2.1 ± 0.1
S12-1	6.4	21.0	Dk.-brn. clay and gravel	6 ± 1.8	3.6 ± 0.1
S12-2	6.7	22.0	Dk.-brn. clay and gravel	7 ± 1.9	4.0 ± 0.1
S12-3	6.9	22.5	Dk.-brn. clay and gravel	77 ± 3.6	6.1 ± 0.2
S13-2	7.3	24.0	Dk.-brn. clay and gravel	5 ± 1.8	3.8 ± 0.1
S14-2	7.9	26.0	Dk.-brn. clay and gravel	<5	2.8 ± 0.1
S15-2	8.5	28.0	Dk.-brn. clay and gravel	7 ± 1.7	3.0 ± 0.1
S17-2	9.8	32.0	Dk.-brn. clay and gravel	<5	3.7 ± 0.1
S19-2	11.0	36.0	Dk.-brn. clay and gravel	77 ± 3.6	5.8 ± 0.2
S21-2	12.2	40.0	Dk.-brn. clay and gravel	14 ± 2.1	5.4 ± 0.2
S23-2	13.4	44.0	Dk.-red-brn. clay and gravel	5 ± 1.9	2.9 ± 0.1

[a] From field geologist's log sheet.

Elemental analyses of six selected samples [0.17 to 13.7 m (0.5 to 45 ft) in depth] were determined by the ICP technique. The data, shown in Table 10, were quite similar to the ICP results for the seven near-surface soil samples described in a previous section of this report. No major trends or differences in composition were noted with core depth. Chemical analysis of the carbonate content (Table 11) showed a steady increase in concentration with depth, ranging from 2.6% at the 0.2-m (0.5-ft) level to 10% at the 13.7-m (45-ft) level. Of the near-surface soil samples, only the lake clay had a significantly higher concentration of carbonate (15%).

3.2 GROUNDWATER CHARACTERIZATION

3.2.1 Source and Description of Samples

Two sets of groundwater samples were obtained from the NFSS. One set contained six samples, supplied by Bechtel National, which are of primary interest since a composite of four samples from one well (W-1, located a short distance north of the R-10 pile) was used in the sorption/solubility tests reported in this document. The second set consisted of 16 pumped and 5 bailed groundwater well samples that were obtained earlier (1981) when NLO was the site manager and Acres American was the site geologic and hydrologic subcontractor. These samples will serve as a basis for comparison with those furnished by Bechtel National. Approximate locations of the source wells for these samples are shown in Fig. 3.

3.2.2 Chemical Characterization

The six well-water samples supplied by Bechtel National were analyzed for elemental content by ICP spectrometry, a method which is capable of analyzing for as many as 30 elements simultaneously. The data for the well-water samples in as-received condition and after filtration through 0.8-μm filter media are shown in Tables 12 and 13, respectively. No major differences between the two types of samples are apparent, indicating that the as-received samples contained negligible amounts of suspended particulates. The major elements detected (which would be present as cations) were calcium, magnesium, and sodium. Only traces of potassium were detected in Well-1, but larger concentrations of this cation were present in Wells 5 and 6. Appreciable quantities of silicon, presumably

Table 10. Elemental concentrations (mg/L) of soil samples from the NFSS Core BH-77, as determined by ICP spectrometry

Element	Sample depth (ft)					
	0.5	8.5	10.5	18.5	26.0	45.0
Ag	<9.	<9.	<9.	<9.	<9.	<9.
Al	51,000.	46,000.	48,000.	57,000.	6,000.	40,000.
B	<500.	<500.	<500.	<500.	500.	<500.
Ba	390.	420.	1,300.	430.	370.	370.
Be	4.	4.	6.	4.	4.	4.
Ca	26,000.	35,000.	15,000.	4,200.	42,000.	49,000.
Cd	6.	6.	<5.	7.	7.	<5.
Co	<6.	130.	4,800.	23.	<7.	<7.
Cr	55.	81.	5.	49.	42.	49.
Cu	54.	92.	1,900.	38	44.	42.
Fe	30,000.	28,000.	14,000.	32,000.	32,000.	26,000.
Ga	<35.	<35.	<35.	<35.	<35.	<35.
Hf	59.	64.	99.	64.	56.	70.
K	19,000.	16,000.	75,000.	21,000.	39,000.	16,000.
Mg	8,500.	12,000.	56,000.	13,000.	15,000.	14,000.
Mn	920.	950.	1,000.	730.	780.	870.
Mo	63.	120.	290.	140.	120.	120.
Na	12,000.	12,000.	4,800.	11,000.	11,000.	14,000.
Ni	62.	460.	14,000.	120.	59.	75.
P	540.	520.	<170.	510.	360.	480.
Pb	220.	340.	4,200.	180.	160.	190.
Sb	230.	220.	190.	230.	240.	230.
Se	300.	350.	390.	310.	430.	350.
Sr	150.	160.	78.	180.	180.	180.
Ti	3,600.	3,200.	2,800.	3,800.	3,900.	3,100.
V	67.	78.	990.	82.	77.	47.
Zn	59.	88.	450.	48.	44.	27.
Zr	270.	290.	200.	190.	190.	280.

Table 11. Carbonate analysis of soil samples from the NFSS core BH-77

Sample depth (m)	Sample depth (ft)	Sample description	Carbonate content (%)
0.2	0.5	Sandy loam fill	2.59
2.6	8.5	Dark-brown clay; roots; straw	6.14
3.2	10.5	Light-brown sandy loam; some roots	4.35
5.6	18.5	Medium-brown clay; gravel	7.49
7.9	26.0	Dark-brown clay; gravel	8.78
13.7	45.0	Dark-brown clay; large gravel	10.07

Table 12. Elemental concentrations (mg/L) of NFSS well-water samples as received from Bechtel National

Element	W1-1	W1-2	W1-3	W1-4	W1-(AV)[a]	W1-5	W1-6
Ag	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030
Al	0.0206	<0.0150	<0.0150	<0.0150	0.0164	0.0651	0.2616
As	<0.0600	<0.0600	<0.0600	<0.0600	<0.0600	<0.0600	<0.0600
B	0.0345	0.0434	0.0336	0.0235	0.03375	0.257	0.2826
Ba	0.0209	0.0223	0.0223	0.0139	0.01985	0.0071	0.0071
Be	0.0006	<0.0002	0.0006	<0.0002	0.0004	<0.0002	<0.0002
Ca	36.49	33.60	37.39	31.73	34.80	6.40	12.94
Cd	<0.0041	<0.0041	<0.0041	<0.0041	<0.0041	<0.0041	<0.0041
Cr	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020
Cu	0.0034	0.0028	0.0037	0.0042	0.0035	0.0133	0.0047
Fe	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	0.280	0.1762
K	<0.10	0.530	0.534	0.937	0.525	2.55	2.64
Li	0.0260	0.0259	0.0275	0.0260	0.0264	0.0423	0.0491
Mg	48.95	49.12	50.00	56.87	51.24	56.91	88.63
Mn	0.0050	0.0050	<0.0050	<0.0050	0.0050	<0.0050	<0.0050
Mo	<0.0080	<0.0080	<0.0080	<0.0080	<0.0080	<0.0080	<0.0080
Na	19.03	18.87	18.85	123.6	45.09	84.71	72.28
Ni	<0.0040	<0.0040	<0.0040	<0.0040	<0.0040	<0.0040	<0.0040
P	<0.020	<0.020	<0.020	<0.020	<0.020	<0.0020	<0.0020
Pb	<0.020	<0.020	<0.020	<0.020	<0.020	<0.020	<0.020
Si	5.71	5.73	5.67	4.61	5.43	<0.010	<0.010
Sr	0.494	0.496	0.502	0.533	0.506	0.272	0.3224
Th	<0.050	<0.050	<0.050	<0.050	<0.050	<0.050	<0.050
Ti	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	0.0142	0.0207
V	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030
Zn	<0.002	<0.0020	<0.0020	<0.0020	<0.0020	0.4219	0.2625
Zr	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010

[a] W1-(AV) is an average of the four W1 well-water sample analyses. These values were employed for the W1 well-water composition used in the experimental work.

Table 13. Concentrations (mg/L) of elements in the NFSS well-water samples (obtained from Bechtel National) after filtration through 0.8-μm filter media

Element	Sample No. W1	W4	W5	W6
Ag	<0.0030	<0.0030	<0.0030	<0.0030
Al	<0.0150	<0.0150	<0.0150	0.0855
As	<0.0600	<0.0600	<0.0600	<0.0600
B	0.0343	0.5243	0.2564	0.2774
Ba	0.0243	0.0072	0.0028	0.0073
Be	0.0012	<0.0002	0.0011	0.0006
Ca	56.6	12.59	6.10	11.44
Cd	<0.0041	<0.0041	<0.0041	<0.0041
Cr	<0.0020	<0.0020	<0.0020	<0.0020
Cu	<0.0010	0.0039	0.0045	0.0048
Fe	<0.0050	0.0106	<0.0050	<0.0050
K	0.4380	3.14	2.36	2.19
Li	0.0272	0.0454	0.0421	0.0505
Mg	48.34	116.3	55.45	86.87
Mn	<0.0050	0.0140	<0.0050	<0.0050
Mo	<0.0080	0.0139	<0.0080	<0.0080
Na	18.72	109.6	80.29	70.96
Ni	<0.0040	0.0642	<0.0040	0.0122
P	<0.0020	<0.0020	<0.0020	<0.0020
Pb	<0.020	<0.020	<0.020	<0.020
Si	5.85	2.26	0.1110	0.2447
Sr	0.5352	0.2203	0.2667	0.2929
Th	<0.050	<0.050	<0.050	<0.050
Ti	<0.0010	<0.0010	<0.0010	<0.0010
V	<0.0030	0.0109	<0.0030	<0.0030
Zn	<0.0020	0.0088	0.0119	0.0100
Zr	<0.0010	<0.0010	<0.0010	<0.0010

as either one or more dissolved silicates or as colloidal silicates, were also present in Well 1.

The 16 pumped and 5 bailed groundwater well samples were analyzed at the time of their receipt in 1981. Analyses included both alkalinity (carbonate, bicarbonate, and hydroxide), shown in Table 14, and the major cation and anion concentrations, listed in Table 15. Cation and anion values (in meq/L), along with an equivalent balance of total cations/total anions, are shown in Table 16. The average cation/anion equivalent balance for the 16 samples is 0.94 ± 0.10, indicating that most major ionic species have been accounted for. Characteristic chemical compositional differences can be seen between the rock-well and the soil-well samples, which suggests that mixing of the aquifers at these depths is minimal. The radionuclide contents of these well waters were not determined because of time and funding constraints.

4. SORPTION/SOLUBILITY MEASUREMENTS

4.1 EXPERIMENTAL METHODS

The experimental protocol for determining ambient-pH sorption isotherms is summarized in Table 17. In essence, it involved the contact of site groundwater (spiked with the desired radionuclide and traced with an appropriate radioactive isotope) with samples of soil at a solution/solids ratio of 5 for 24 h in an argon atmosphere at room temperature (22°C), centrifugation at 5,000 x _g_ to recover a clear solution, and analysis of the solution for radionuclides. Details of this procedure are given in Appendix B. In these tests, the pH was allowed to equilibrate naturally with the soil and groundwater; no attempt was made to adjust or control it. Since the NFSS soil/groundwater system is not strongly buffered, some of the test pH values fell slightly below the pH of the initial groundwater, especially for increased cation loading of the clay minerals in the soil. However, this was considered to be a valid compromise between "real-world" conditions and artificial conditions in which a buffering agent would be added to the groundwater or the pH would be adjusted by the addition of acid or base, to arbitrarily establish fixed pH values.

Table 14. Alkalinity analysis of well-water samples from the NFSS

Well No.	Type of well	Initial pH	Alkalinity (mg/L)		
			Carbonate	Bicarbonate	Total[a]
		Pumped samples			
BH-30A	Shallow soil [4.6 to 6.1 m (15 to 20 ft)]	7.45	0	460	460
BH-40A		7.34	0	417	417
BH-47A		7.65	36	294	330
BH-50A		7.74	26	377	403
BH-64B		7.61	36	334	360
BH-16	Deep soil [10.7 to 16.8 m (35 to 55 ft)]	7.76	16	116	132
BH-40		7.71	10	90	100
BH-47		7.43	30	280	310
BH-50		7.91	14	64	78
BH-59		7.84	26	121	147
BH-61		7.83	14	105	119
BH-63		7.94	12	61	73
BH-64		7.71	26	311	337
BH-12	Rock [30.5 to 61 m (50 to 100 ft)]	7.89	10	33	43
BH-57		7.43	12	44	56
BH-62		7.93	10	34	44
		Bailed samples			
BH-30A	Shallow soil [4.6 to 6.1 m (15 to 20 ft)]	7.46	36	59	95
BH-40A		7.34	0	306	306
BH-47A		8.05	8	35	43
BH-50A		7.76	20	252	272
BH-64B		7.89	10	83	93

[a] No hydroxide was detected in any sample.

Table 15. Analysis of the NFSS well-water samples for alkali metals, alkaline earths, and anions

Well No.	Type of Well	Concentration (mg/L)								
		Na^+	K^+	Ca^{2+}	Mg^{2+}	$SO_4{}^{2-}$	$CO_3{}^{2-}$	$HCO_3{}^{2-}$	Cl^-	F^-
BH-30A	Shallow soil	76	1.8	72	94	270	0	460	17	0.6
BH-40A	[4.6 to 6.1 m	111	5.0	167	157	920	0	417	13	0.2
BH-47A	(15 to 20 ft)]	316	10.9	84	77	1,050	36	294	100	0.4
BH-64B		46	3.2	54	119	510	36	334	19	0.2
BH-16	Deep soil	195	6.9	130	124	1,070	16	116	50	0.5
BH-40	[10.7 to 16.8 m	135	8.4	193	90	980	10	90	72	0.3
BH-47	(35 to 55 ft)]	370	17.2	264	215	1,870	30	280	190	0.3
BH-50		169	9.1	165	86	1,120	14	64	75	0.4
BH-59		84	7.6	91	97	600	26	121	28	0.3
BH-61		185	11.8	168	115	1,130	14	105	66	0.3
BH-63		218	12.7	186	71	1,020	12	61	150	0.3
BH-64		72	8.5	85	115	480	26	311	21	0.3
BH-12	Rock	2,190	59	500	57	2,410	10	33	2,700	0.8
BH-57	[30.5 to 61 m	1,343	46	495	61	1,460	12	44	2,200	0.6
BH-62	(50 to 100 ft)]	470	35	349	48	2,360	10	34	180	0.7

Table 16. Equivalent balance analysis of alkali metals, alkaline earths, and anions in the NFSS well-water samples

Well No.	Type of Well	Concentration (mg/L)									Total cations / Total anions
		Na^+	K^+	Ca^{2+}	Mg^{2+}	$SO_4{}^{2-}$	$CO_3{}^{2-}$	$HCO_3{}^{2-}$	Cl^-	F^-	
BH-30A	Shallow soil	3.8	0.05	3.6	7.7	5.6	0	7.5	0.5	0.03	1.11
BH-40A	[4.5 to 6.1 m	4.8	0.13	8.3	2.9	19.2	0	6.8	0.4	0.01	0.99
BH-47A	(15 to 20 ft)]	14.0	0.28	4.2	6.3	21.9	1.2	4.8	0.5	0.01	0.81
BH-64B		2.0	0.08	2.7	9.8	10.6	1.2	5.5	0.5	0.01	0.82
BH-16	Deep soil	8.5	0.18	6.5	0.2	22.3	0.5	1.9	1.4	0.03	1.10
BH-40	[10.7 to 16.8 m	5.9	0.21	9.6	7.4	20.4	0.3	1.5	2.0	0.02	0.95
BH-47	(35 to 55 ft)]	16.0	0.44	13.2	7.7	39.0	1.0	4.6	5.4	0.02	0.95
BH-50		7.3	0.23	8.2	7.1	23.3	0.5	1.0	2.1	0.02	0.87
BH-59		3.7	0.19	4.5	8.0	12.5	0.9	2.0	0.8	0.02	1.01
BH-61		8.0	0.30	8.4	9.5	23.5	0.5	1.7	1.9	0.02	0.84
BH-63		9.5	10.32	9.3	5.8	21.3	0.4	1.0	4.2	0.02	0.93
BH-64		3.1	0.22	4.2	9.5	10.0	0.9	5.1	0.6	0.02	1.01
BH-12	Rock	95.0	1.50	25.0	4.7	50.2	0.3	0.5	76.1	0.04	0.99
BH-57	[30.5 to 61 m	62.0	1.18	24.7	5.0	30.4	0.4	0.7	62.0	0.03	0.99
BH-62	50 to 100 ft)]	20.0	0.90	17.4	3.9	49.2	0.3	0.6	6.1	0.04	0.76

Table 17. Batch contact ambient-pH method for determining radionuclide sorption and apparent concentration limit values[a]

- Contact (shake) 2.0 g of soil with 10 mL of radionuclide-traced groundwater for 24 h at room temperature under an argon atmosphere.
- Centrifuge sample to recover clear solution.
- Count radionuclide to determine concentration in solution, and calculate radionuclide concentration in soil.
- Calculate sorption ratio, Rs (L/kg), and plot sorption data as rate or isotherm graphs.[b]

[a] Details of the experimental protocol are presented in Appendix B.
[b] The program used for calculation is included in Appendix B.

Although many investigators contact groundwater and soil for relatively long periods (weeks or months), the amount of data required to fulfill our obligation to this project precluded the use of lengthy equilibration times.

4.2 URANIUM - SORPTION RATE

The experimental investigation of sorption behavior was conducted primarily with uranium since (1) this radionuclide is the major site contaminant in terms of quantity, (2) the well-known solubility of the uranyltricarbonate anion could affect uranium sorption behavior if it is complexed by the carbonate/bicarbonate in the NFSS soil/groundwater system, and (3) the analytical methods to be utilized allow prompt collection of the uranium data.

While many of the batch test parameters to be used in this study could be selected based on prior experience, the rate of uranium sorption by soils or rocks is sometimes slow and selection of the parameters requires a compromise between experiments designed to reach equilibrium and the exigency of data needs within the near term. Thus, scouting tests were carried out to measure the rate of uranium sorption by the three types of near-surface soils from the NFSS in contact with groundwater from

Well-1. The experimental procedure used in these tests was summarized in the previous section and is described more fully in Appendix B. The data for the sorption rate tests at two uranium concentrations (10 and 100 µg/mL) with soils GC-1, BCB-1, and LC-1 are shown, respectively, in Tables 18, 19, and 20. For all six test series, the uranium adsorbed after a 24-h contact was 93.5 ± 2.8% of the amount adsorbed after 72 h. Based on this information, a contact time of 24 h was selected for the uranium sorption isotherm tests. Uranium sorption ratio (Rs) values are also given in these tables so that comparison can be made with the values obtained from the sorption isotherms.

4.3 URANIUM - SORPTION ISOTHERMS

4.3.1 Sorption on Gray Clay

Uranium sorption isotherms were determined for the three samples of gray clay soil found at the bottom of the trench that was constructed around the R-10 pile in preparation for emplacement of a slurry-wall barrier. The experimental protocol for these sorption tests is summarized in Sect. 4.1; details are listed in Appendix B. The sorption behavior of uranium on samples GC-1, GC-2, and GC-3 (Fig. 4) was very similar; the data were defined by one curve. The sorption isotherm data are presented in Tables C-1 through C-3 in Appendix C.

The three soil samples exhibited only low Rs values; the extremes were 0.8 and 9.4 L/kg. Values of 1 to 2 L/kg may be typical of uranium sorption on these soil samples in contact with groundwater from W1 well at relatively high uranium concentration levels (4 to 40 mmol/L), while values of 2 to 6 L/kg may be typical of uranium sorption at relatively low uranium concentration levels (0.01 to 0.1 mmol/L).

The sorption isotherms for the three soil samples gave no definite indication of reaching a limiting solubility, even at the highest concentrations. However, a slight upward inflection of the isotherm does suggest a species change and perhaps the initiation of solubility limitation at a uranium concentration greater than 10 g/L. The lack of a definite indication of an apparent solubility limit could be the result of one or more of several scenarios: (1) the carbonate that is present in the

Table 18. Rate of uranium sorption on gray clay from the NFSS[a]

	Uranium concentration		
Time (h)	Solids (μg/g)	Aqueous (μg/mL)	Rs (L/g)
0	-	10.0	-
1	33.3	3.5	9.5
2	24.4	5.2	4.7
4	24.5	5.2	4.7
24	25.9	4.9	5.3
48	26.5	4.8	5.5
72	28.4	4.4	6.5
0	-	100.0	-
1	146.6	70.8	2.1
2	151.0	70.4	2.1
4	153.0	70.0	2.2
24	171.1	66.5	2.6
48	174.2	65.9	2.6
72	180.3	64.6	2.8

[a] Experimental conditions: 2.0 g of soil (GC-1) contacted with 10 mL of W1 well water spiked with ^{238}U + ^{233}U tracer. Final pH of contacted solutions was 7.5 to 7.8.

Table 19. Rate of uranium sorption on brown-clay backfill from the NFSS[a]

Time (h)	Uranium concentration Solids (µg/g)	Uranium concentration Aqueous (µg/mL)	Rs (L/kg)
0	-	10.0	-
1	25.9	4.9	5.3
2	21.6	5.7	3.9
4	21.9	5.7	3.8
24	24.3	5.2	4.7
48	28.0	4.5	6.2
72	26.0	4.9	5.3
0	-	100.0	-
1	148.4	70.9	2.1
2	152.6	70.1	2.2
4	167.7	67.2	2.5
24	176.8	65.3	2.7
48	180.6	64.6	2.8
72	189.0	62.9	3.0

[a] Experimental conditions: 2.0 g of soil (BCB-1) contacted with 10 mL of W1 well water spiked with $^{238}U + ^{233}U$ tracer. Final pH of contacted solutions was 7.5 to 7.8.

Table 20. Rate of uranium sorption on lake clay from the NFSS[a]

Time (h)	Uranium concentration: Solids (μg/g)	Uranium concentration: Aqueous (μg/mL)	Rs (L/kg)
0	-	10.0	-
1	21.3	5.8	3.7
2	20.9	5.9	3.6
4	21.3	5.8	3.7
24	24.8	5.1	4.8
48	24.5	5.2	4.7
72	25.2	5.1	5.0
0	-	100.0	-
1	147.5	71.1	2.1
2	149.8	71.5	2.1
4	152.9	70.0	2.2
24	172.7	66.1	2.6
48	176.7	65.4	2.7
72	192.8	62.2	3.1

[a] Experimental conditions: 2.0 g of soil (LC-1) contacted with 10 mL of W1 well water spiked with ^{238}U + ^{233}U tracer. Final pH of contacted solutions was 7.5 to 7.8.

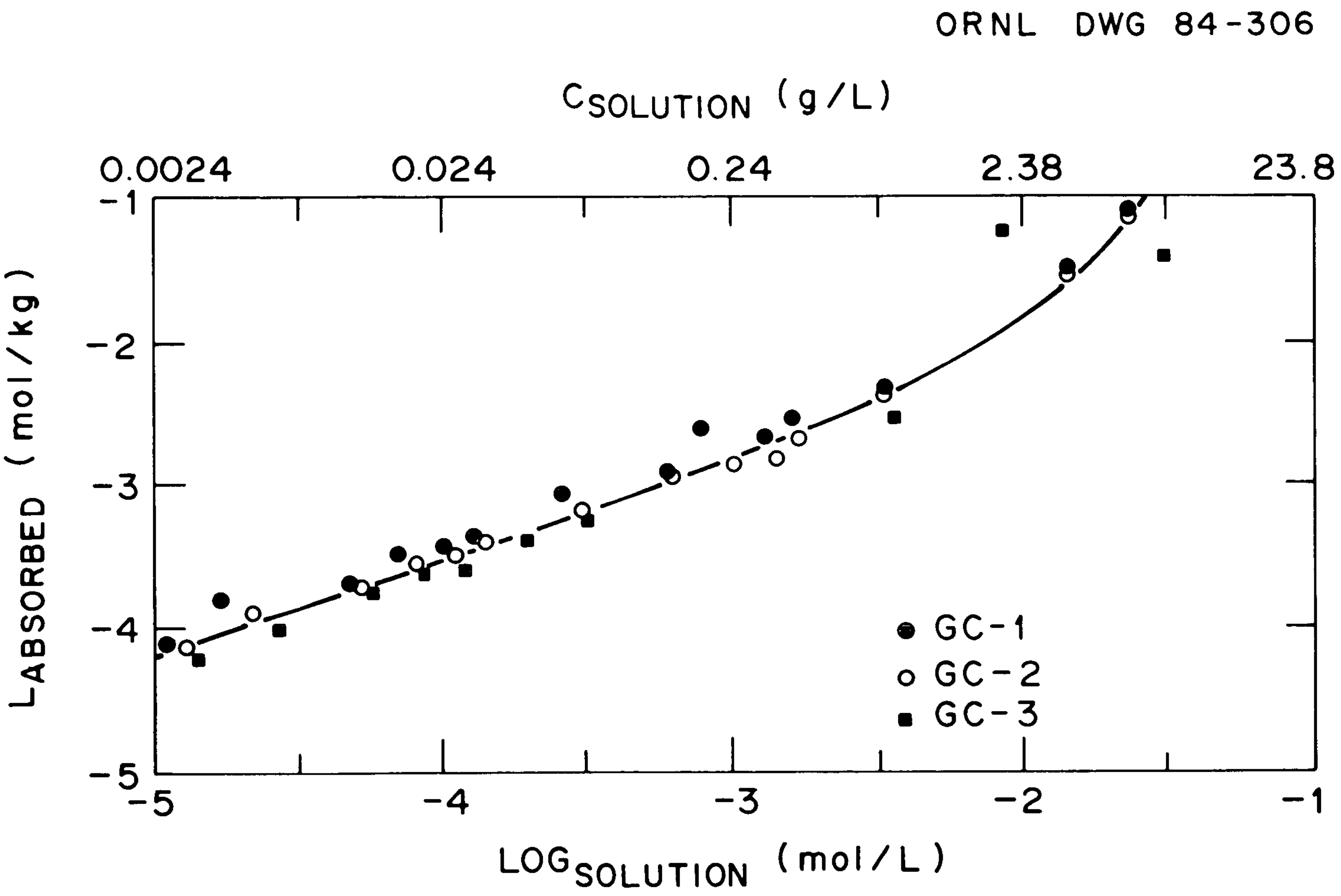

Fig. 4. Uranium sorption isotherms for samples of gray clay from the NFSS.

soil could cause increased solubility due to the formation of the carbonate complexes of uranium; (2) insoluble hydrolytic species of uranium may be present as colloidal dispersions; and (3) kinetics of the exchange reactions and, particularly, the hydrolytic reactions may be slow enough to allow a significant amount of the radionuclide to remain in the solution phase.

4.3.2 Sorption on Brown-Clay Backfill

Uranium sorption isotherms were determined for the three samples of brown-clay backfill. This material is a type of surface soil which was to be employed in backfill operations over and around the slurry wall in the vicinity of the R-10 pile. The experimental protocol for these sorption tests is summarized in Sect. 4.1; details are listed in Appendix B. As with the gray-clay soil samples, the sorption behavior of uranium on samples BCB-1, BCB-2, and BCB-3 (Fig. 5) was similar and the results were defined by a single curve. The sorption isotherm data are detailed in Tables C-4 through C-6 in Appendix C.

The three BCB soil samples exhibited only low Rs values; the extremes were 0.5 and 10.5 L/kg. Values of 1 to 3 L/kg may be typical of uranium sorption on these soil samples in contact with groundwater from W1 well at relatively high uranium concentration levels (4 to 40 mmol/L), while values of 4 to 10 L/kg may be typical of uranium sorption at relatively low uranium concentration levels (0.01 to 0.1 mmol/L).

The sorption isotherms for the three soil samples gave no definite indication of reaching a limiting solubility and showed only a very slight upward inflection, which might suggest some species change.

4.3.3 Sorption on Lake Clay

A uranium sorption isotherm was determined for the single sample, LC-1, of lake clay, a soil which was brought in from off-site for use in the construction of a slurry wall around the R-10 pile. The experimental protocol for these sorption tests is summarized in Sect. 4.1; details are listed in Appendix B. The sorption behavior of uranium (Fig. 6) was very similar to that found for the gray-clay and the brown-clay backfill samples. The sorption isotherm data are detailed in Table C-7 in Appendix C.

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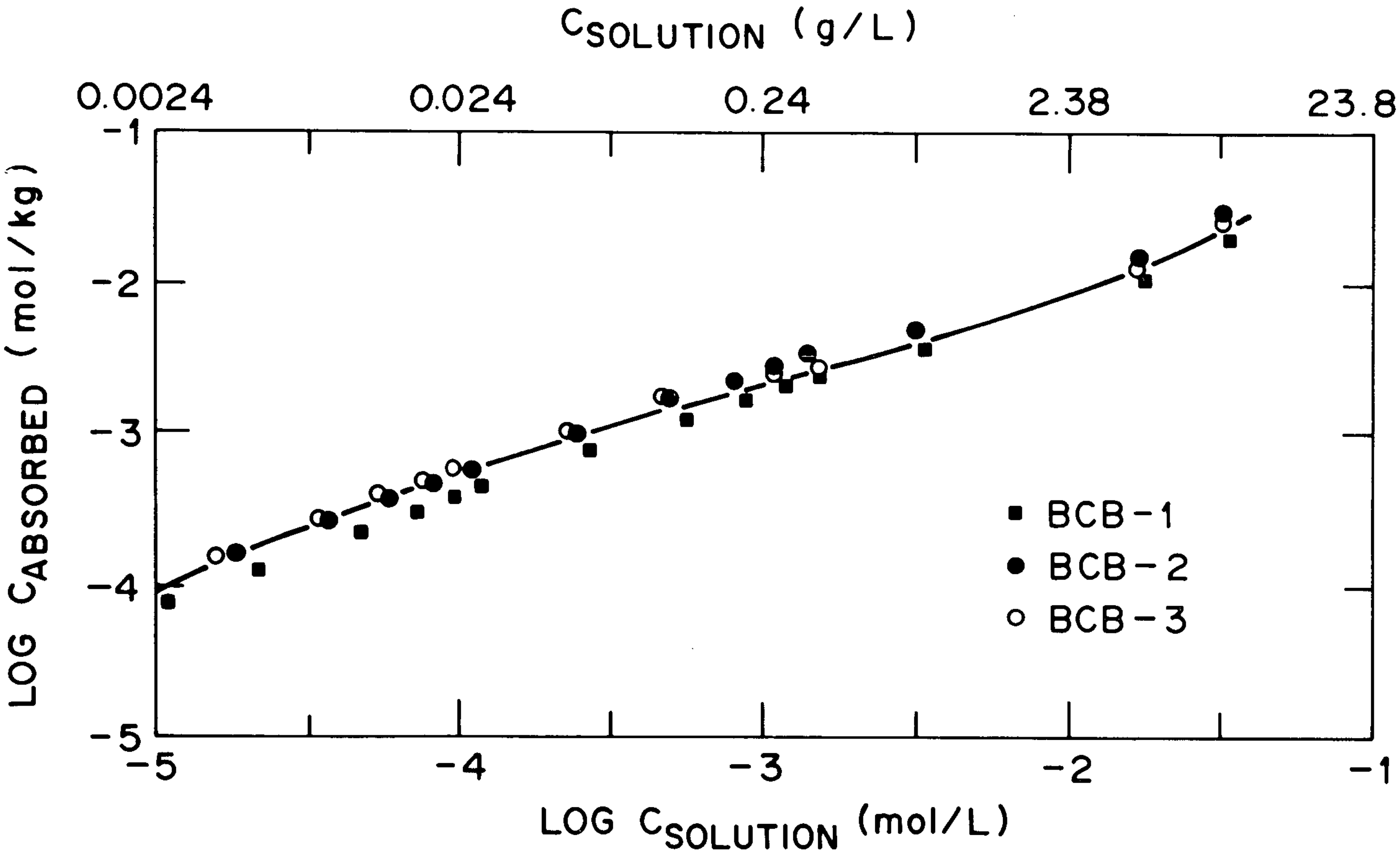

Fig. 5. Uranium sorption isotherms for samples of brown-clay backfill from the NFSS.

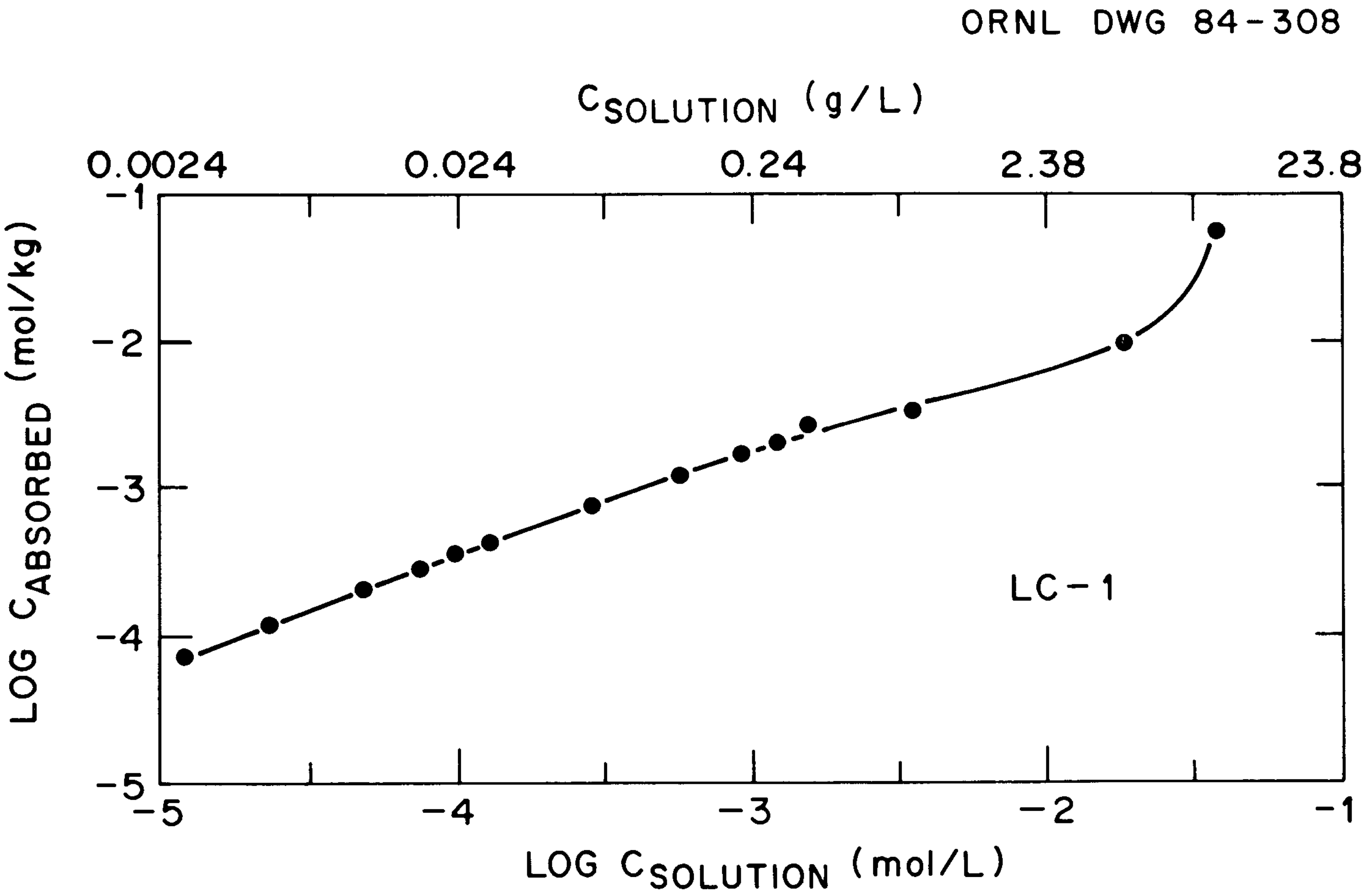

Fig. 6. Uranium sorption isotherm for a sample of lake clay from the NFSS.

Soil sample LC-1, like the others, exhibited low Rs values that ranged from 0.6 L/kg, obtained with a high initial uranium concentration (4 to 40 mmol/L), to 6 L/kg, obtained with a low initial uranium concentration (0.01 to 0.1 mmol/L).

LC-1 is the only one of the near-surface soil samples that exhibited a suggested approach to an apparent uranium solubility limit. The isotherm shows a definite upward inflection (based on one data point), indicating an apparent solubility limit of approximately 15 mg/L. At the contact pH of the system from which these particular data were derived (i.e., 6.5), this apparent solubility limit value may not be too far from the true solubility limit expected from classical solubility considerations (about 2 to 10 mg/L).

4.3.4 Sorption on Soil from Core BH-77

Uranium sorption isotherms were determined for seven soil samples from a core (BH-77) which was taken through the R-10 pile, at the point of maximum surface radioactivity, to bedrock. These samples were selected from the 0.3-, 2.1-, 3.2-, 5.6-, 7.9-, 12.2-, and 13.7-m (1-, 7-, 10.5-, 18.5-, 26-, 40-, and 45-ft) levels of the core. The procedure for determining the sorption isotherms is summarized in Sect. 4.1; details are given in Appendix B.

The uranium sorption isotherms for soil samples from the 0.3-, 2.1-, and 3.2-m (1-, 7-, and 10.5-ft) levels are shown in Fig. 7. Sorption isotherms for soil samples from the 5.6-, 7.9-, and 12.2-m (18.5-, 26-, and 40-ft) levels are shown in Fig. 8. For purposes of direct comparison, the sorption isotherm for the soil sample from the 12.2-m (40-ft) level is shown with duplicate isotherms for soil from the 13.7-m (45-ft) level in Fig. 9. The uranium sorption isotherms for all of the samples except those from the 3.2-m (10.5-ft) and the 13.7-m (45-ft) levels had similar characteristics and accompanying low Rs values (<10 L/kg). The sample from the 3.2-m (10.5-ft) level showed an intermediate range of uranium sorption (approximate Rs values, 7 to 70 L/kg), while the sample from the 13.7-m (45-ft) level showed much higher values (Rs values higher than 10,000 L/kg).

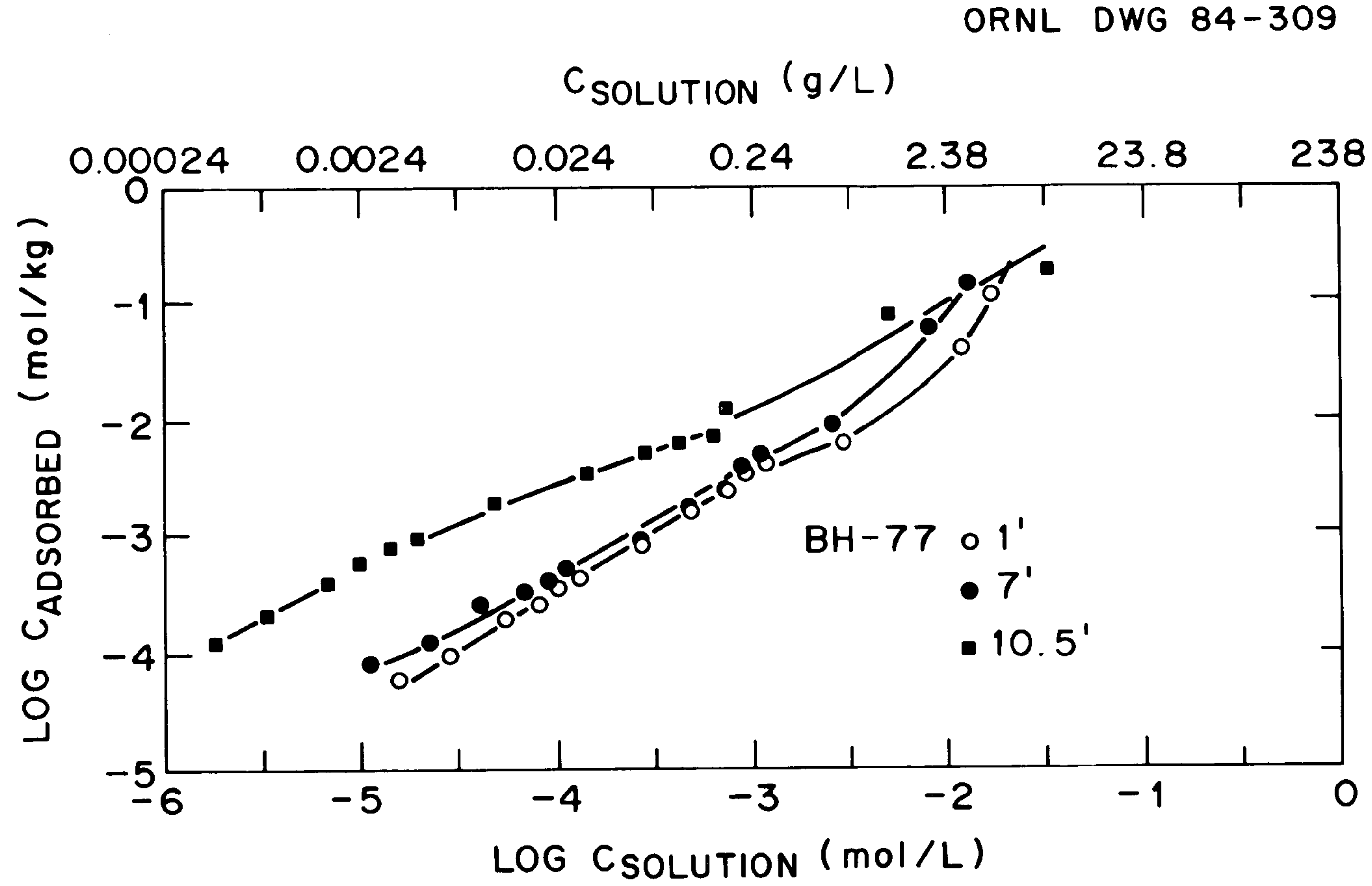

Fig. 7. Uranium sorption isotherms for soil samples from the 0.3-, 2.1-, and 3.2-m (1-, 7-, and 10.5-ft) levels of BH-77 from the NFSS.

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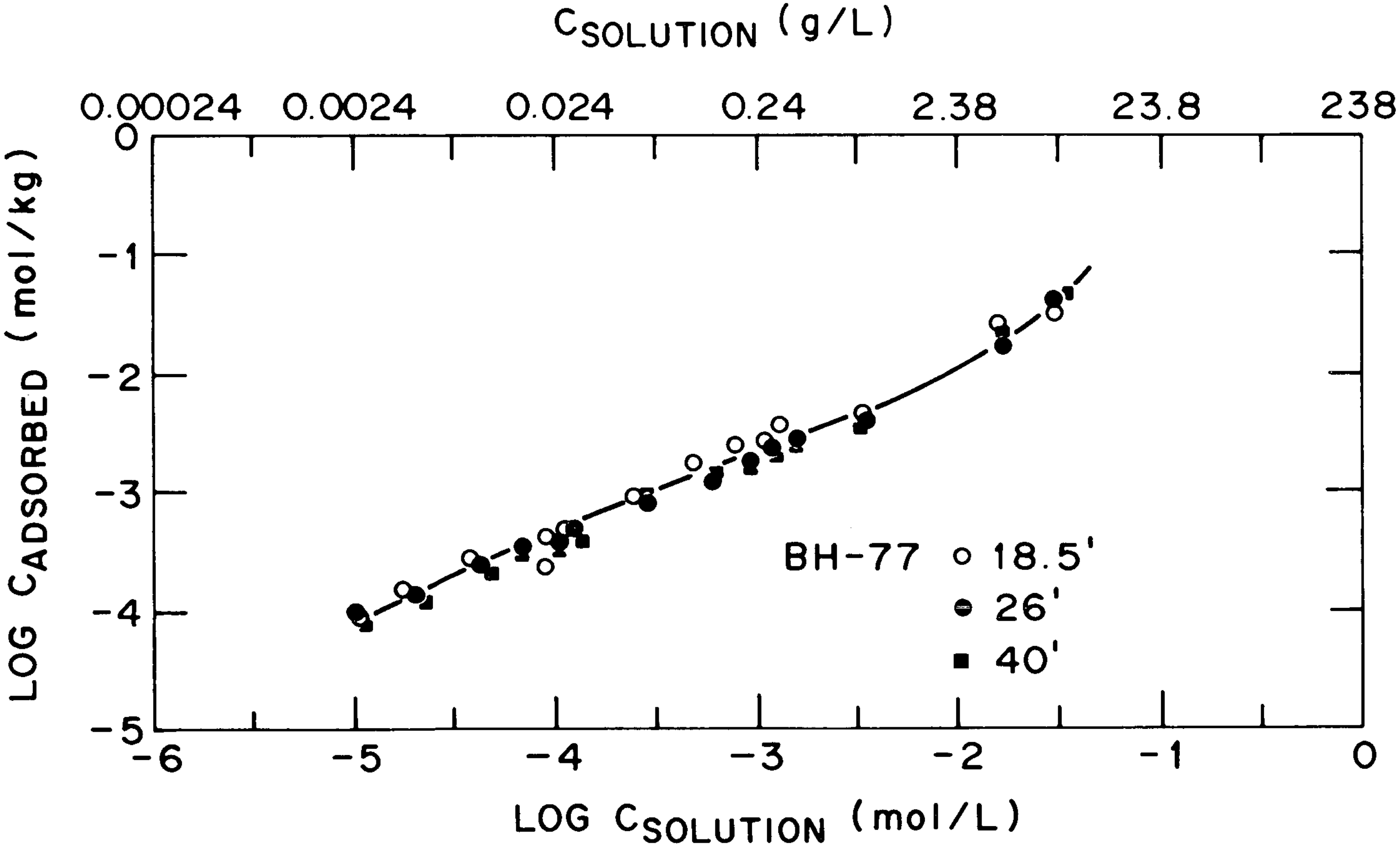

Fig. 8. Uranium sorption isotherms for soil samples from the 5.6-, 7.9-, and 12.2-m (18.5-, 26-, and 40-ft) levels of BH-77 from the NFSS.

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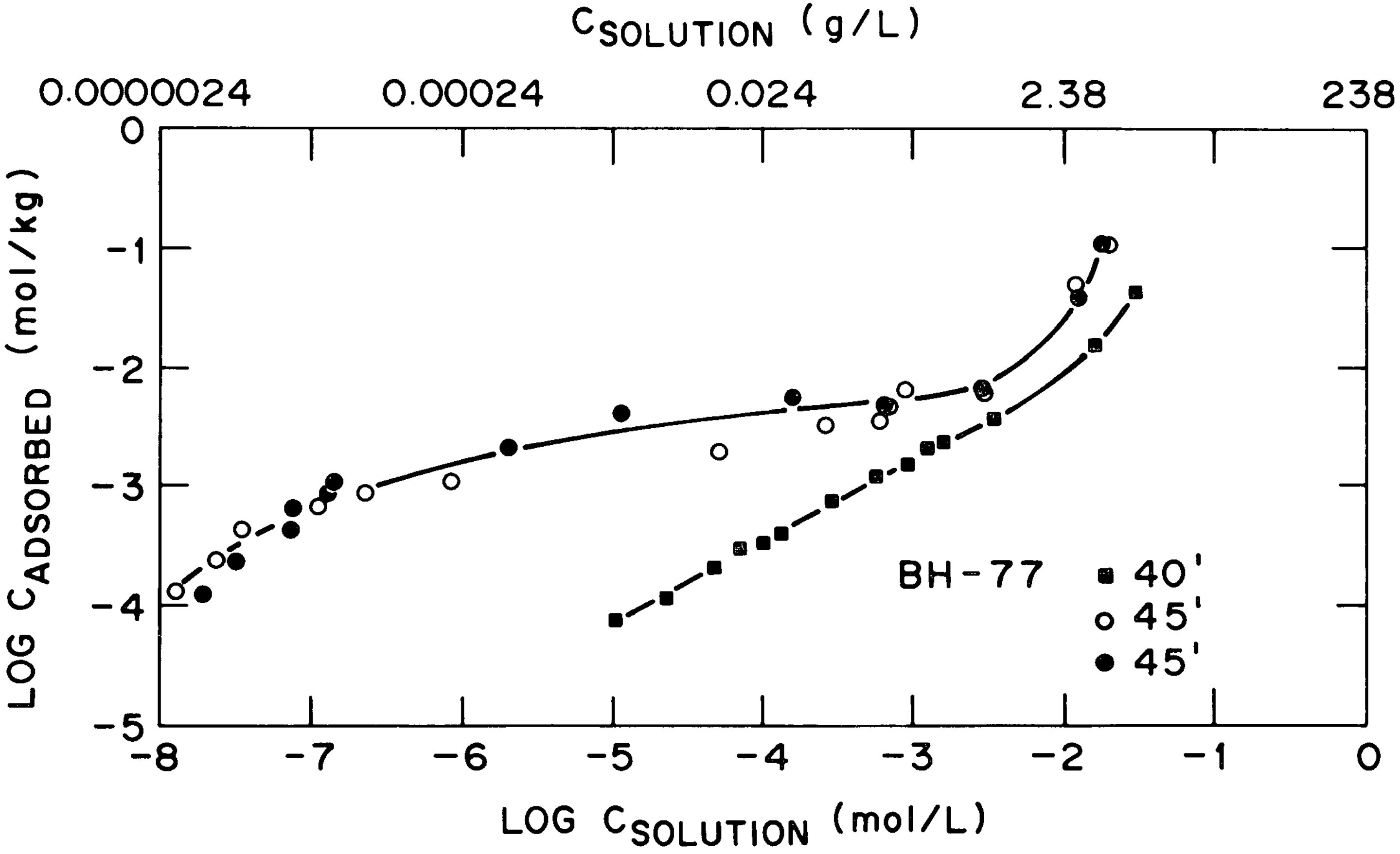

Fig. 9. Uranium sorption isotherms for soil samples from the 12.2- and 13.7-m (40- and 45-ft) levels of BH-77 from the NFSS.

4.4 EFFECT OF FERRIC OXYHYDROXIDE

During the sorption tests with soil from the 13.7-m (45-ft) level of core BH-77, a reddish-brown film was observed on the walls of the equilibration tube. Upon analysis, this film was shown to be an iron compound, possibly originating from the underlying Queenston shale. Several investigators have reported that the presence of hydrated ferric oxides or natural iron-containing minerals such as goethite in clay or rock samples can retard the migration of radionuclides via groundwater in contact with these formations.[2,3] In general, many hydrated metal oxides are known to have the ability to act as inorganic cation and anion exchangers.[4,5] It is interesting to consider the possibility that the poor uranium sorption characteristics of the NFSS near-surface soils might be improved by the use of this type of soil [i.e., the type found at the 13.7-m (45-ft) level] or by the addition of a ferric oxyhydroxide to the slurry wall materials.

The unique effect of hydrated ferric oxyhydroxide on the sorption of uranium(VI) from natural groundwater from the NFSS (Well 1) was determined by adding various concentrations of ferric nitrate (1×10^{-5} to 1×10^{-2} $\underline{M}$) to groundwater (no soil present) containing uranium at a concentration of 50 µg/mL, traced with ^{233}U, and then adjusting the pH to 7.5-8.5. The mixture was equilibrated for 16 h under argon with agitation provided by a magnetic stirrer. The samples were centrifuged, and the clear supernate was analyzed for ^{233}U activity. The data (see Fig. 10) indicate that the adsorption of uranium increased rapidly at Fe^{3+} concentrations above 1×10^{-4} $\underline{M}$, reaching 99.5% at 1×10^{-2} $\underline{M}$. The test was repeated in the presence of 20 g of lake clay per 100 mL of uranium-spiked groundwater. The resulting data, shown in Fig. 11 (details for both tests are listed in Table 21), indicate that the uranium Rs values are increased from 4 to 76 L/kg in the Fe^{3+} concentration range of 1×10^{-4} to 1×10^{-2} $\underline{M}$. The amount of ferric oxyhydroxide present in the clay at the highest Rs value would be approximately 4 kg (9 lb) per ton of lake clay.

Although the sorption mechanism has not been well characterized with respect to uranium and particular clay types, the addition of iron compounds or natural minerals such as goethite in combination with clay

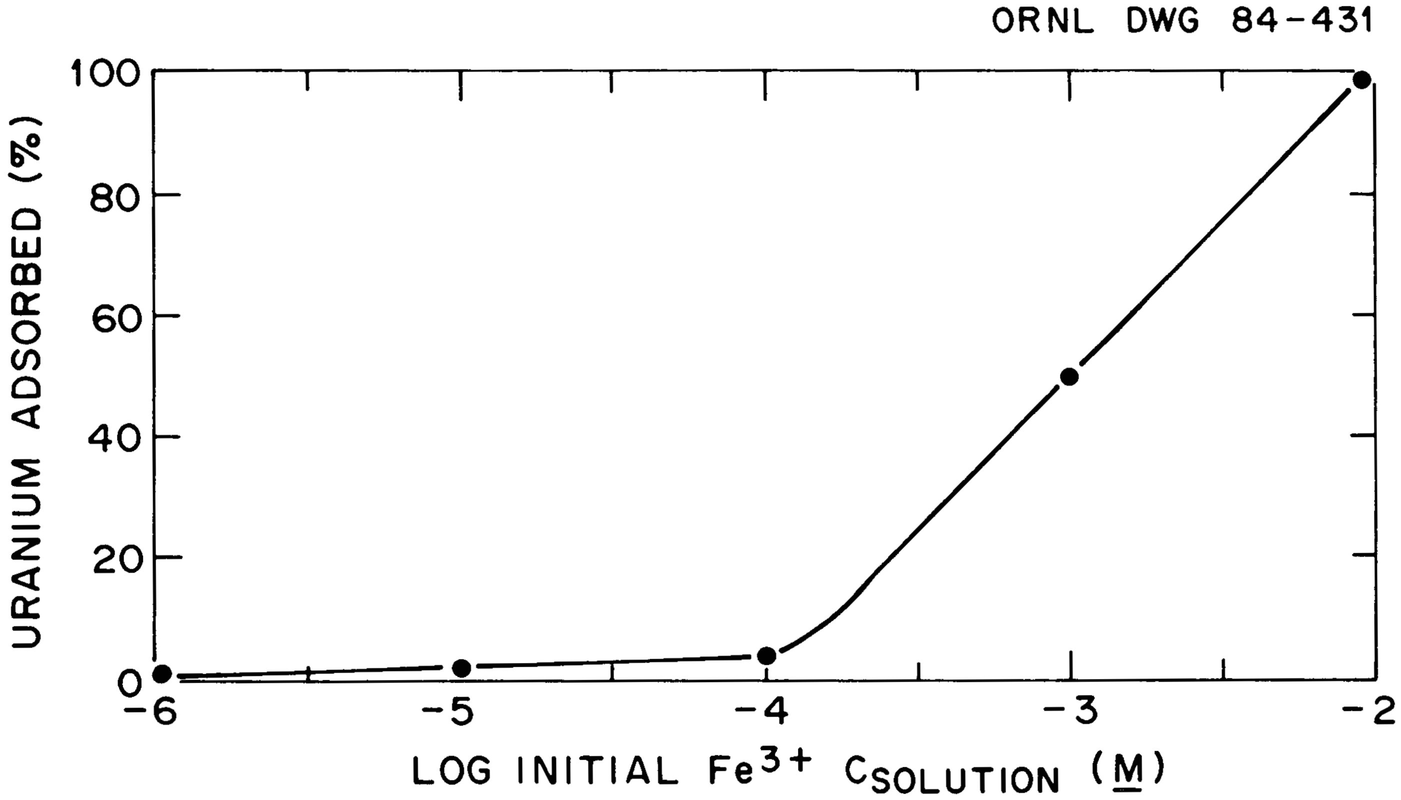

Fig. 10. Sorption of uranium on ferric oxyhydroxide (no soil present).

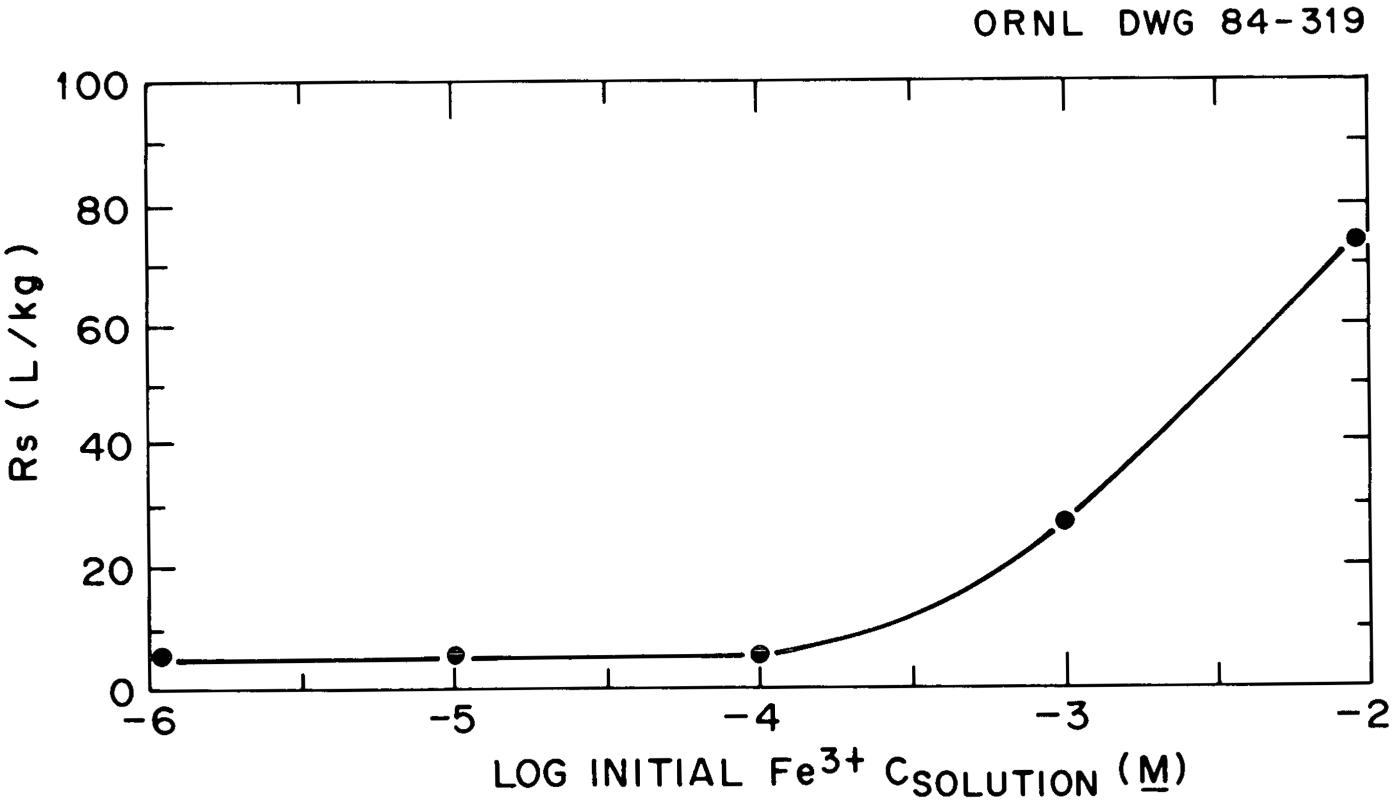

Fig. 11. Sorption of uranium on a sample of lake clay mixed with ferric oxyhydroxide [4 kg/ton (9 lb/ton)].

Table 21. Effect of the addition of hydrated ferric oxyhydroxide on the sorption of uranium on NFSS soil

Soil	Fe^{3+} conc. (M)	Final pH	Uranium adsorbed (%)	Rs (L/kg)
None	0.0	7.8	0.0	-
None	0.00001	7.9	2.5	-
None	0.0001	8.0	4.2	-
None	0.001	7.8	49.9	-
None	0.008	7.8	99.5	-
LC-1	0.0	8.0	40.0	4.3
LC-1	0.000009	8.5	48.4	5.8
LC-1	0.00009	8.1	38.7	4.0
LC-1	0.0009	8.9	51.7	26.1
LC-1	0.009	8.5	92.9	76.1

appears to be potentially attractive for remedial action use in the construction of slurry walls to prevent the migration of uranium from contaminated areas.

4.5 EFFECT OF THE ADDITION OF BENTONITE

Personnel at Bechtel National suggested that the use of bentonite as a soil additive might improve the retention of uranium at the NFSS. Uranium sorption isotherms were determined for three soil samples, with and without the addition of 10% bentonite: lake clay (LC-1), brown-clay backfill (BCB-1), and soil from the 7.9-m (26-ft) level of core BH-77. The data, shown in Figs. 12-14, indicate no apparent improvement as a result of the bentonite. This was surprising since the sorption of uranium on montmorillonite, a clay mineral which is a major component of bentonite, exhibited high uranium Rs values (described in the Introduction section of this report) and well-defined apparent solubility limits. Although there was no evidence of a uranium solubility limitation in these tests, the slight upward inflection of the isotherm suggested a possible species change at the higher solution concentrations of uranium. The data are detailed in Tables E-1 through E-6 in Appendix E.

4.6 EFFECT OF CARBONATE

Very high uranium solubility values (i.e., with no indication of an apparent solubility limit due to solution saturation) were encountered in all the isotherms determined for the soil samples obtained from the NFSS. Uranium remained in solution in the soil/well-water systems at concentrations as high as 3.4×10^{-5} mol/mL, or 8000 µg/mL. At the test pH levels of about 6.4 to 7.8, the uranium solubility of UO_3 or hydrated uranium(VI) oxides would be expected to be about 4×10^{-9} to 4×10^{-8} mol/mL, or approximately 1 to 10 µg/mL.[4] Since all of the soil samples from the NFSS contained significant quantities of carbonate and no definite apparent solubility limit had been identified in the uranium sorption isotherms, it seemed valid to consider that a uranyltricarbonate anionic complex was the likely source of the high uranium solubility in the NFSS soil/groundwater system.

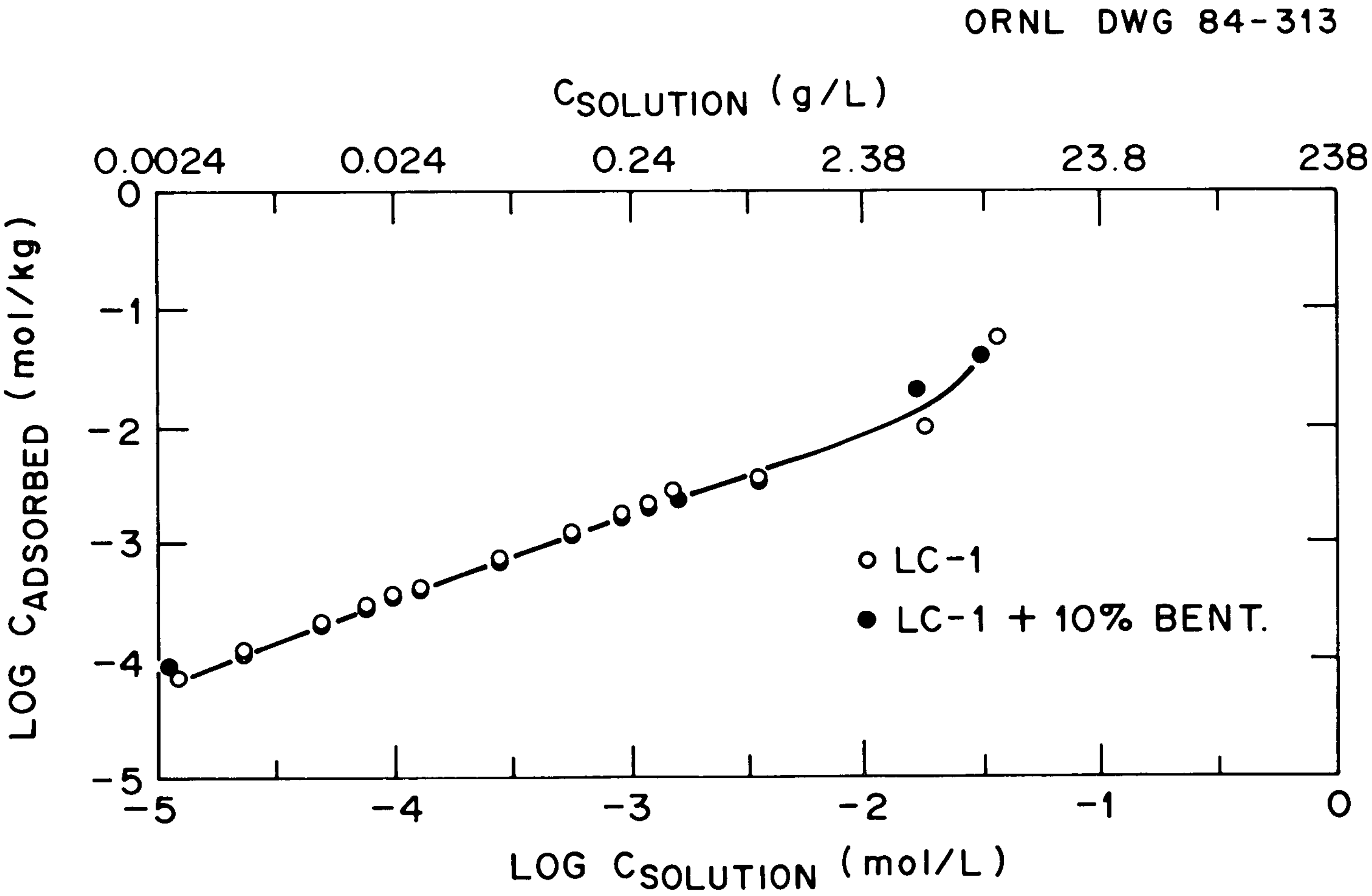

Fig. 12. Uranium sorption isotherms for samples of lake clay showing the effect of adding 10% bentonite.

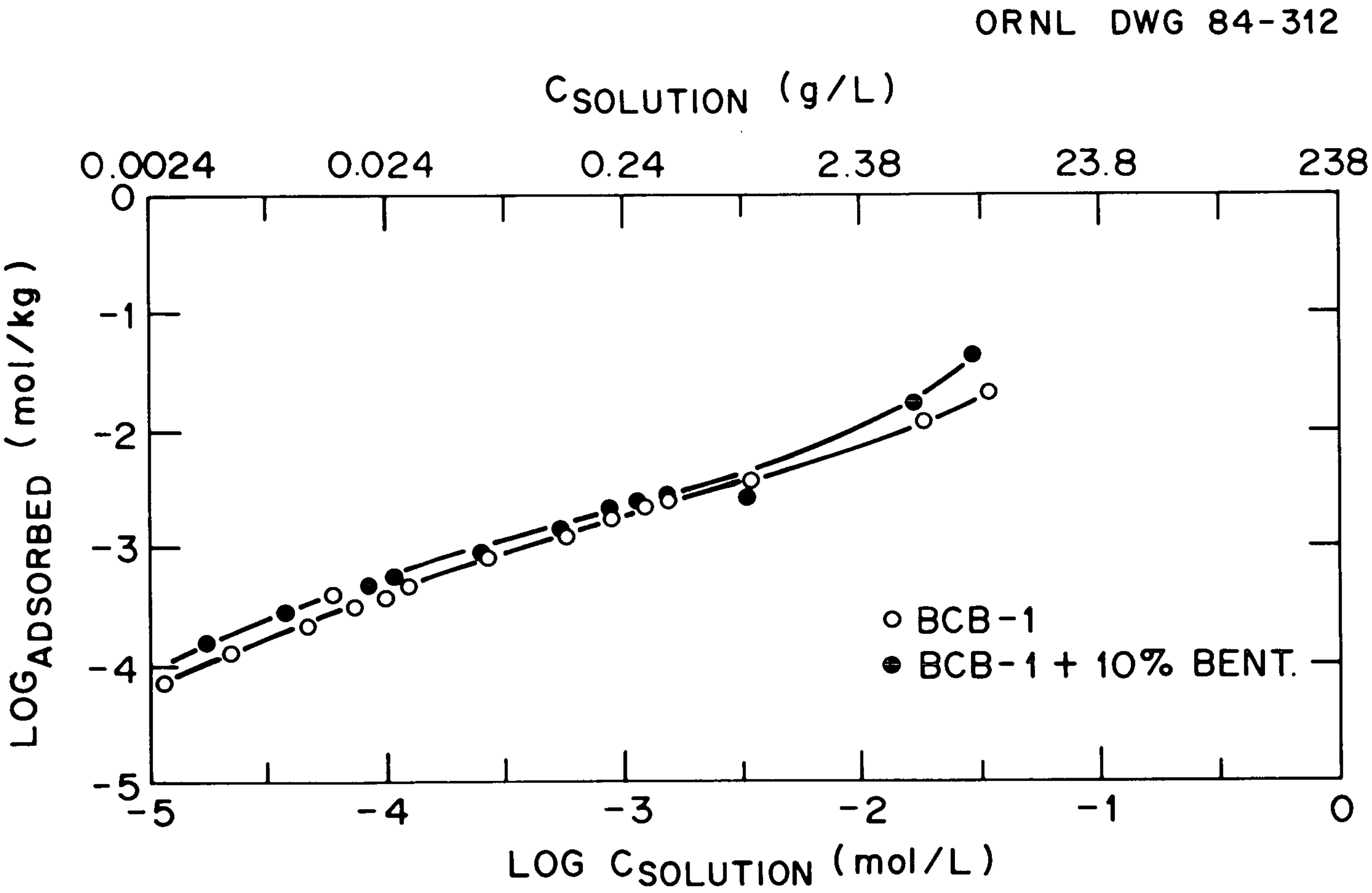

Fig. 13. Uranium sorption isotherms for samples of brown-clay backfill from the NFSS showing the effect of adding 10% bentonite.

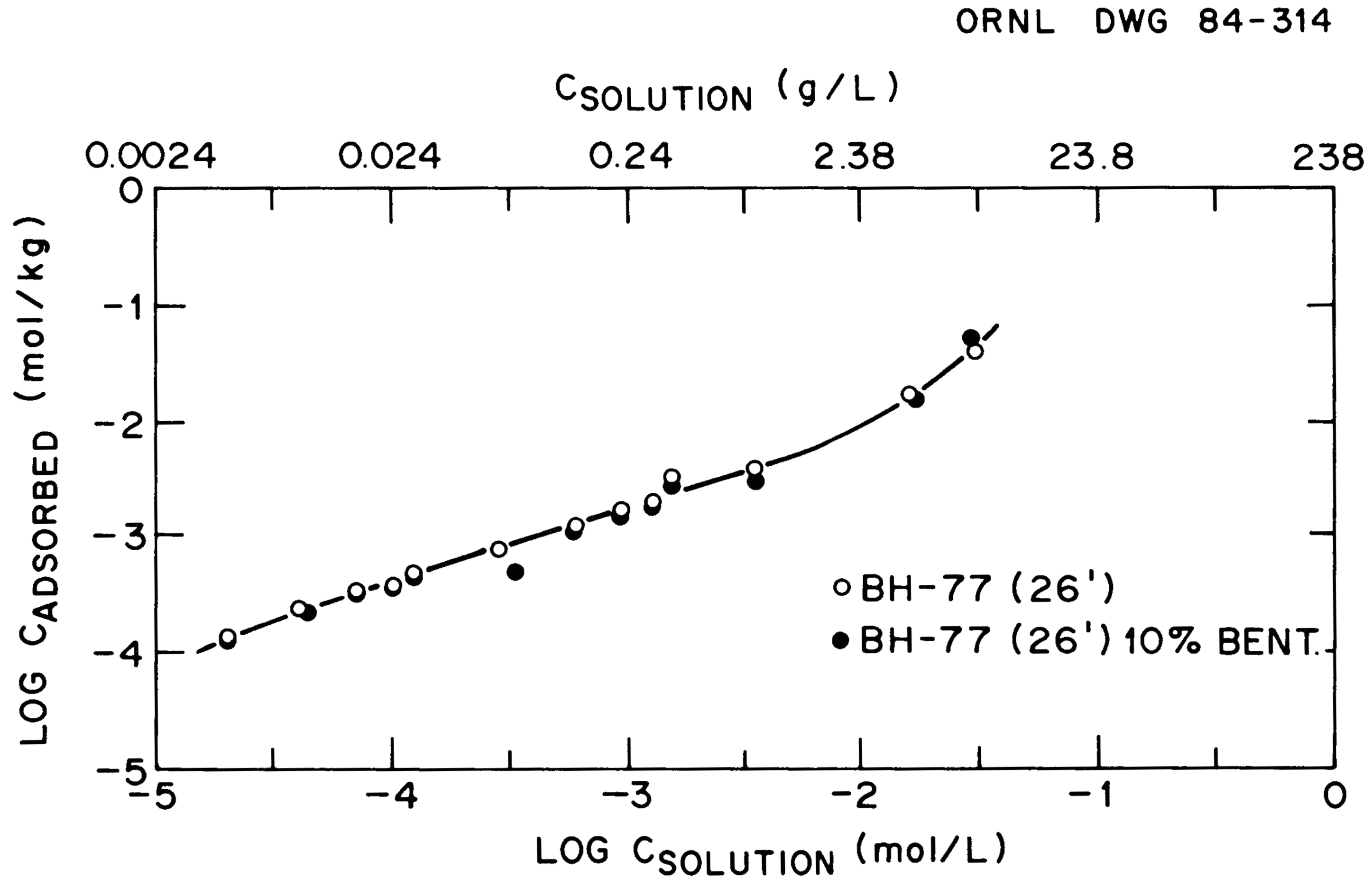

Fig. 14. Uranium sorption isotherms for soil from the 7.9-m (26-ft) level of BH-77 showing the effect of adding 10% bentonite.

The presence of the uranyltricarbonate complex in solutions recovered after soil/well-water contact was conclusively established by spectrophotometric comparison of the absorbance spectrum for the test solution with the known spectrum from sodium uranyltricarbonate.[4-6] The spectra (Fig. 15) show excellent agreement. Apparently, sufficient carbonate is present in these systems to solubilize appreciable amounts of uranium.

The low uranium sorption values and the lack of a uranium apparent solubility limit in these NFSS soil/well-water systems suggest that any uranium which becomes solubilized from waste emplaced in the NFSS soil would not be retarded for long periods of time. In other words, it would probably be readily available for migration.

In order to attain positive confirmation that the basis of the high uranium solubility is the carbonate found in the soil and groundwater system, several tests were conducted in which the pH of the soil/groundwater system was reduced to 3.0 and maintained at that level for 24 h. This treatment should destroy a significant amount of the available, or "active," carbonate that is found in the system. Following this initial acidification contact, the groundwater/solids slurry was spiked with uranium (50 mg/L) and the pH was readjusted to about 7.5; an additional contact period of 24 h was then provided. Uranium sorption isotherms were determined in this manner for the following soil samples: gray clay (GC-1), brown-clay backfill (BCB-1), lake clay (LC-1), and soil from the 7.9-m (26-ft) level of core BH-77. Data for the sorption isotherms for these tests are shown in Figs. 16-19; details are listed in Tables F-1 through F-8 in Appendix F.

A well-defined approach to an apparent uranium solubility limit, indicated by an upswing in the isotherm at approximately the 100-μg/mL concentration level, was noted in each of these tests. (The higher apparent Rs values recorded after the initiation of precipitation are not valid for modeling of sorption reactions.) The mean and standard deviation of the 22 apparent solubility limit values, which occur beyond the point of deviation from normal isotherm configurations in the four curves, was calculated to be $2.93 \times 10^{-7} \pm 2.30 \times 10^{-7}$ mol/mL, or 70 ± 55 μg/mL. These data definitely confirm that carbonate is the determining

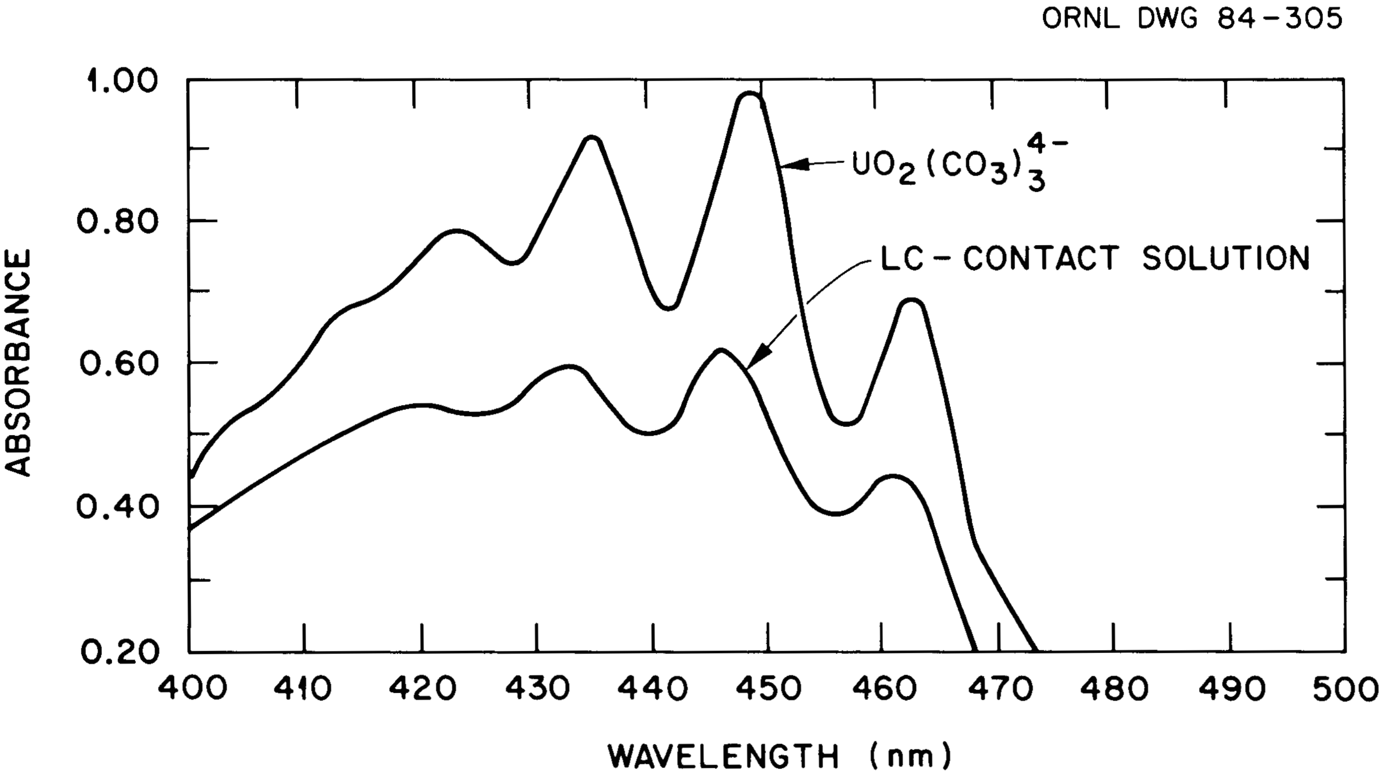

Fig. 15. Comparative visible spectra for $UO_2(CO_3)_3^{3-}$ and a U(VI)-groundwater sample contacted with lake clay from the NFSS.

ORNL DWG 84-315

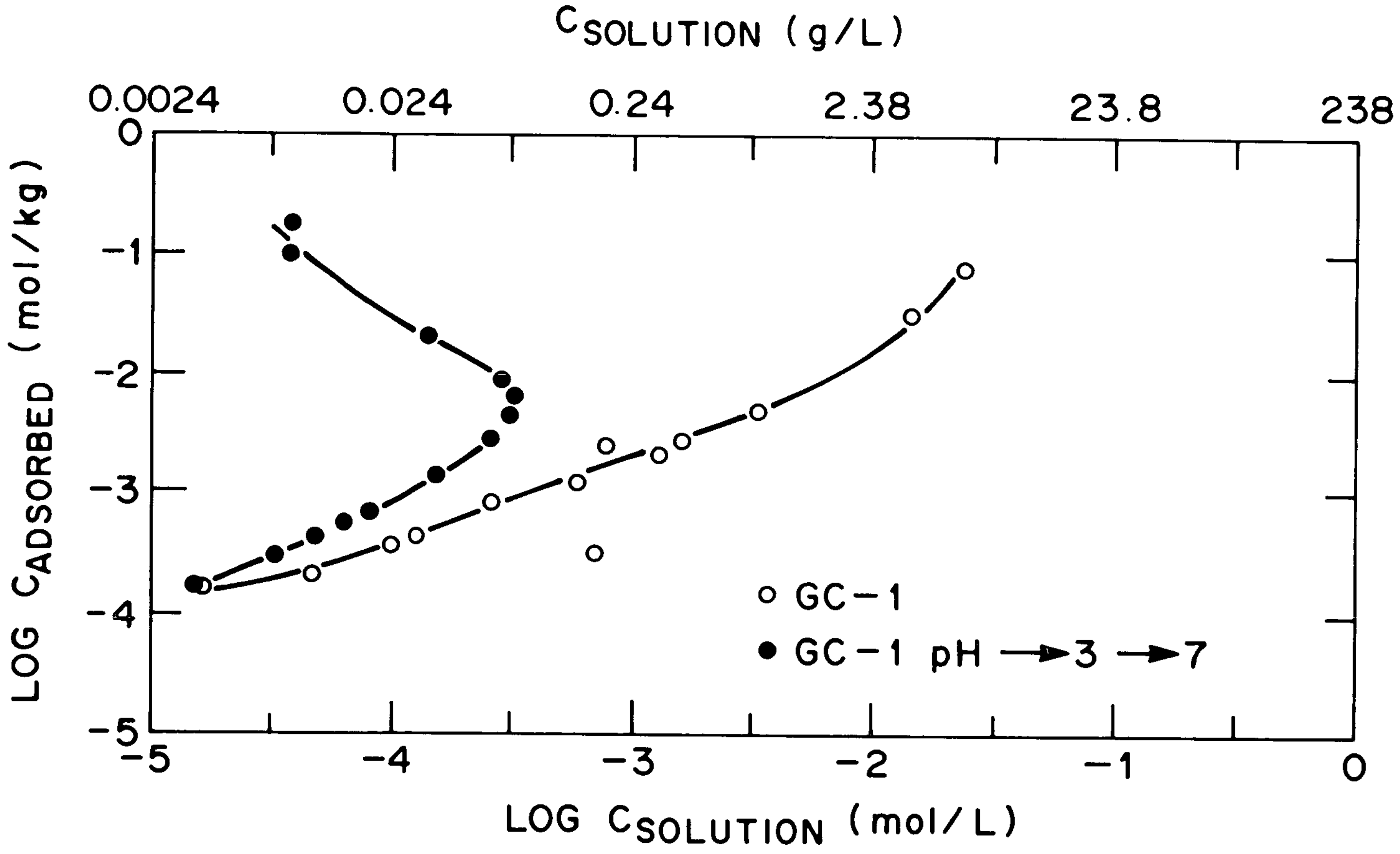

Fig. 16. Uranium sorption isotherms for a sample of gray clay from the NFSS: effect of initial contact of groundwater and soil at pH 3.0 prior to spiking with uranium and recontacting at pH 7.0 to 7.7.

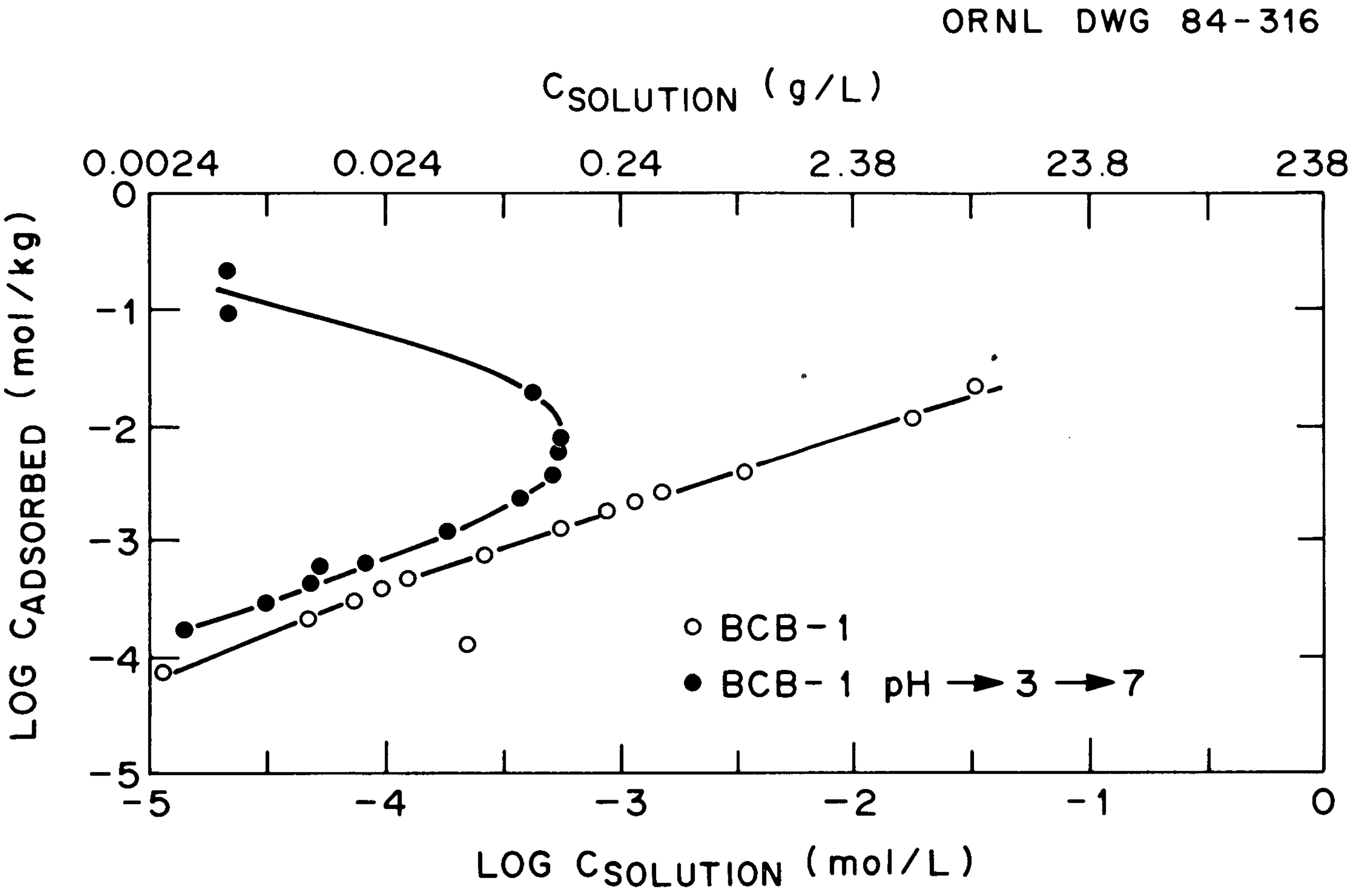

Fig. 17. Uranium sorption isotherms for a sample of brown-clay backfill from the NFSS: effect of initial contact of groundwater and soil at pH 3.0 prior to spiking with uranium and recontacting at pH 7.5 to 8.2.

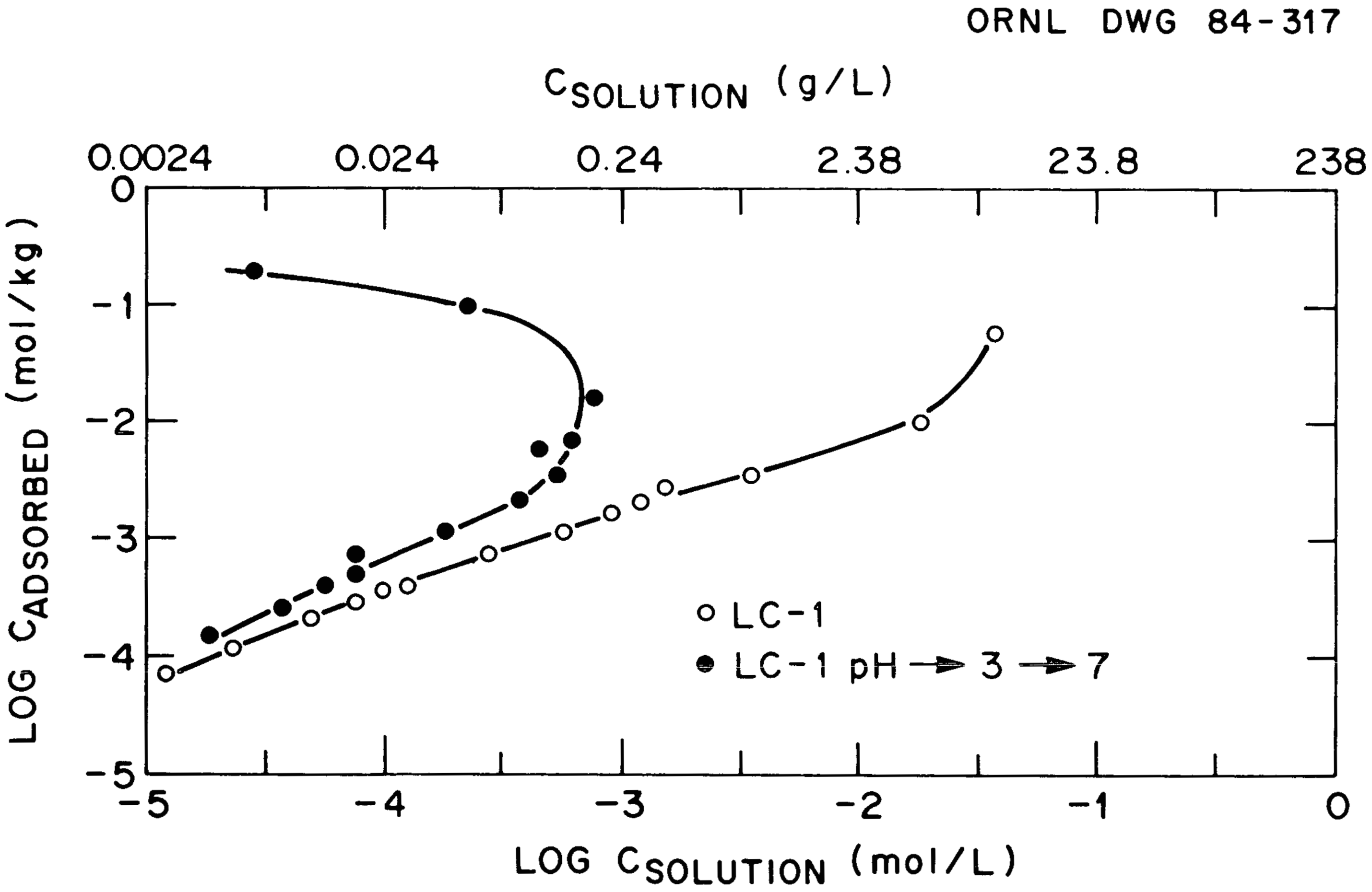

Fig. 18. Uranium sorption isotherms for a sample of lake clay from the NFSS area: effect of initial contact of groundwater and soil at pH 3.0 prior to spiking with uranium and recontacting at pH 7.4 to 7.8.

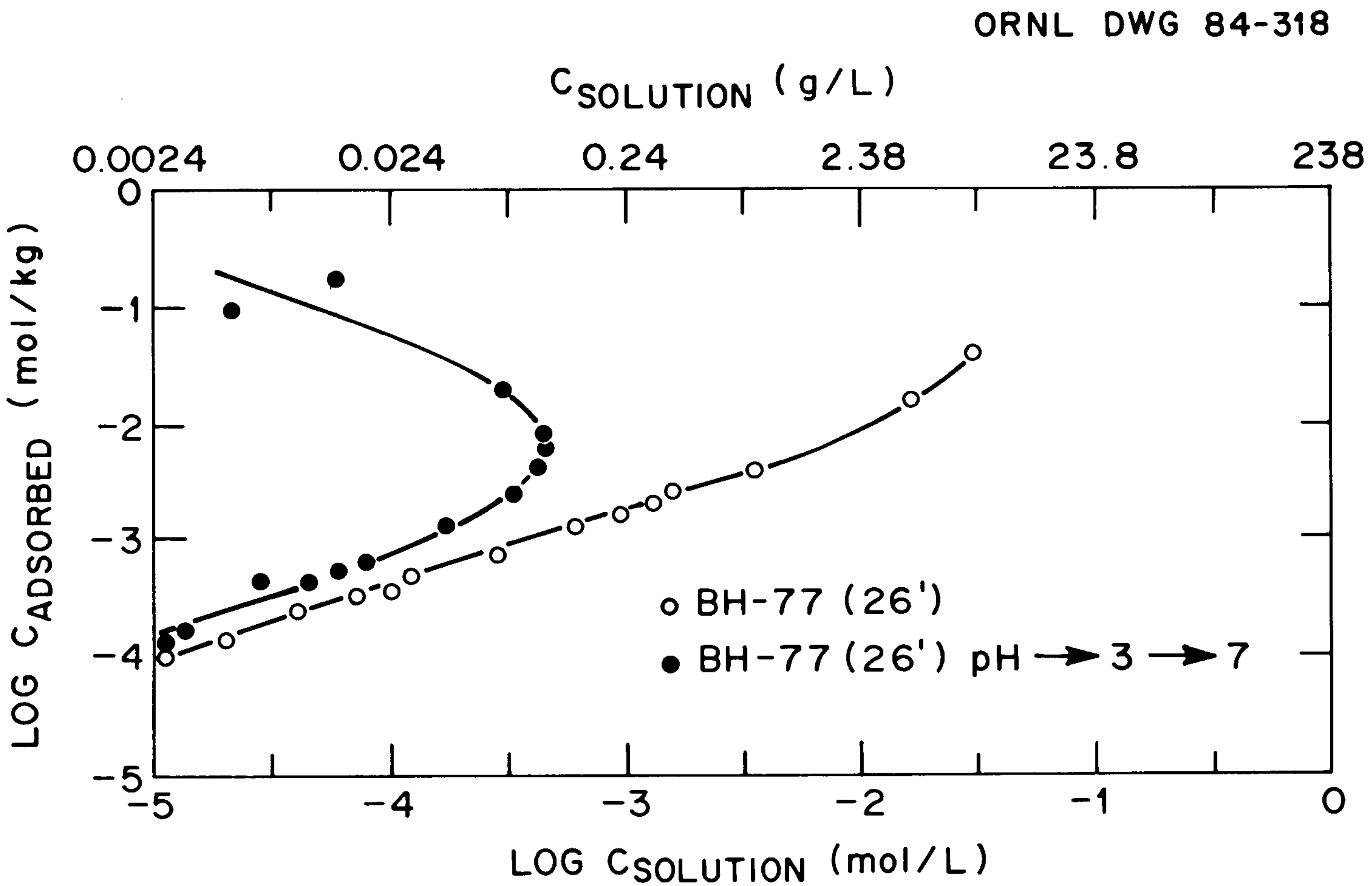

Fig. 19. Uranium sorption isotherms for soil from the 7.9-m (26-ft) level of BH-77 from the NFSS: effect of initial contact of groundwater and soil at pH 3.0 prior to spiking with uranium and recontacting at pH 7.0 to 7.5.

factor in the high uranium solubility and low uranium Rs values obtained in the NFSS soil/groundwater system.

4.7 RADIUM

Radium Rs values were determined for all seven near-surface soil samples from the NFSS. All of these tests used groundwater from Well 1 spiked with approximately 100,000 pCi ^{226}Ra/mL. The procedure for determining the sorption ratio values is the same as that used for sorption isotherms, details for which are listed in Appendix B. The analysis of ^{226}Ra was accomplished by integrating the gamma activity under the 0.609-MeV ^{214}Bi peak after a 30-d period in which the sample had been sealed in a counting tube to allow the decay products of ^{222}Rn to reach secular equilibrium. The sorption ratios obtained in these tests are presented in Table 22. The data are summarized in Table G-1 in Appendix G.

The radium sorption values listed in Table 22 represent an average material balance of approximately 79%, which is considered acceptable for radium analysis. The resulting Rs values, which ranged from 1,100 to 18,000 mL/g, indicate very effective adsorption of the radium by all of the NFSS soil samples. Although the initial concentration of radium was moderately high, the Rs values for these solutions are probably conservative since, in normal sorption isotherm determination, they tend to increase as the concentration of the radionuclide in solution decreases.

5. ACKNOWLEDGMENTS

The authors wish to acknowledge the excellent technical assistance of W. E. Shockley, G. C. Young, W. K. Alexander, S. M. Mrochek, H. J. Marquess, and K. W. Lennon, who conducted most of the laboratory tests reported in this document; the Materials and Chemistry Technology Department, ORGDP, for x-ray diffraction analyses of the clay soils; the Micro Meritics Group of the same department for surface area and particle-size distribution analyses; 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 Debbie Brown, Kim Kitts, Martha Dawson, and Martha Stewart for secretarial and editorial assistance in the preparation of this report.

Table 22. Summary of radium sorption behavior on soil from the NFSS

Soil sample	Initial radium concentration (pCi/mL)	After contact	
		pH	Rs (L/kg)
GC-1	100,000	8.0	1,600
GC-2	100,000	8.0	11,000
GC-3	100,000	8.0	6,700
BCB-1	100,000	8.0	3,500
BCB-2	100,000	8.0	18,000
BCB-3	100,000	8.0	12,000
LC-1	100,000	8.0	7,400

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7. APPENDIXES

Appendix A.

SITE CHARACTERIZATION DATA

ORNL DWG 84-298

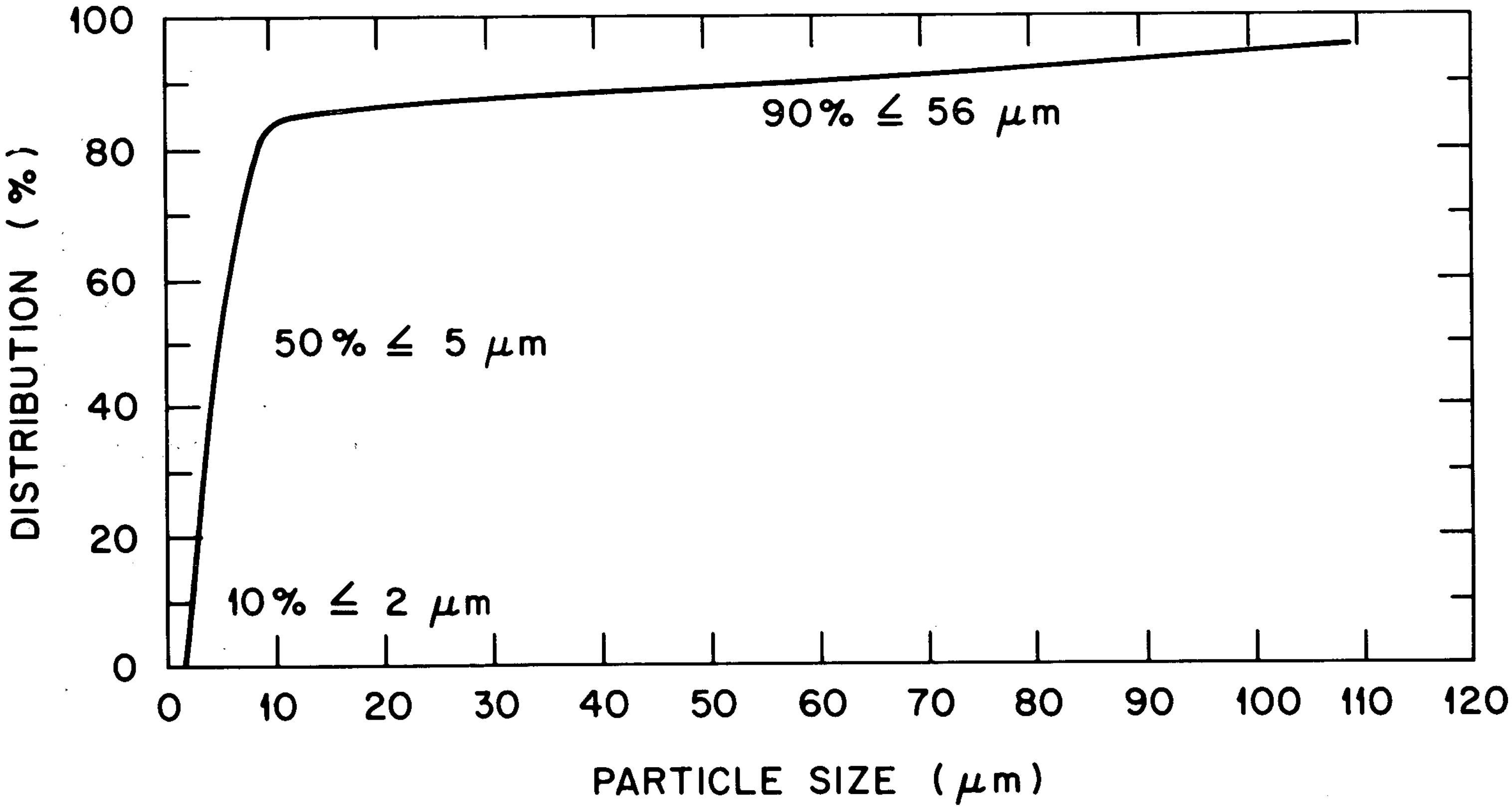

Fig. A-1. Particle size distribution analysis of soil sample GC-1 from the NFSS.

ORNL DWG 84-299

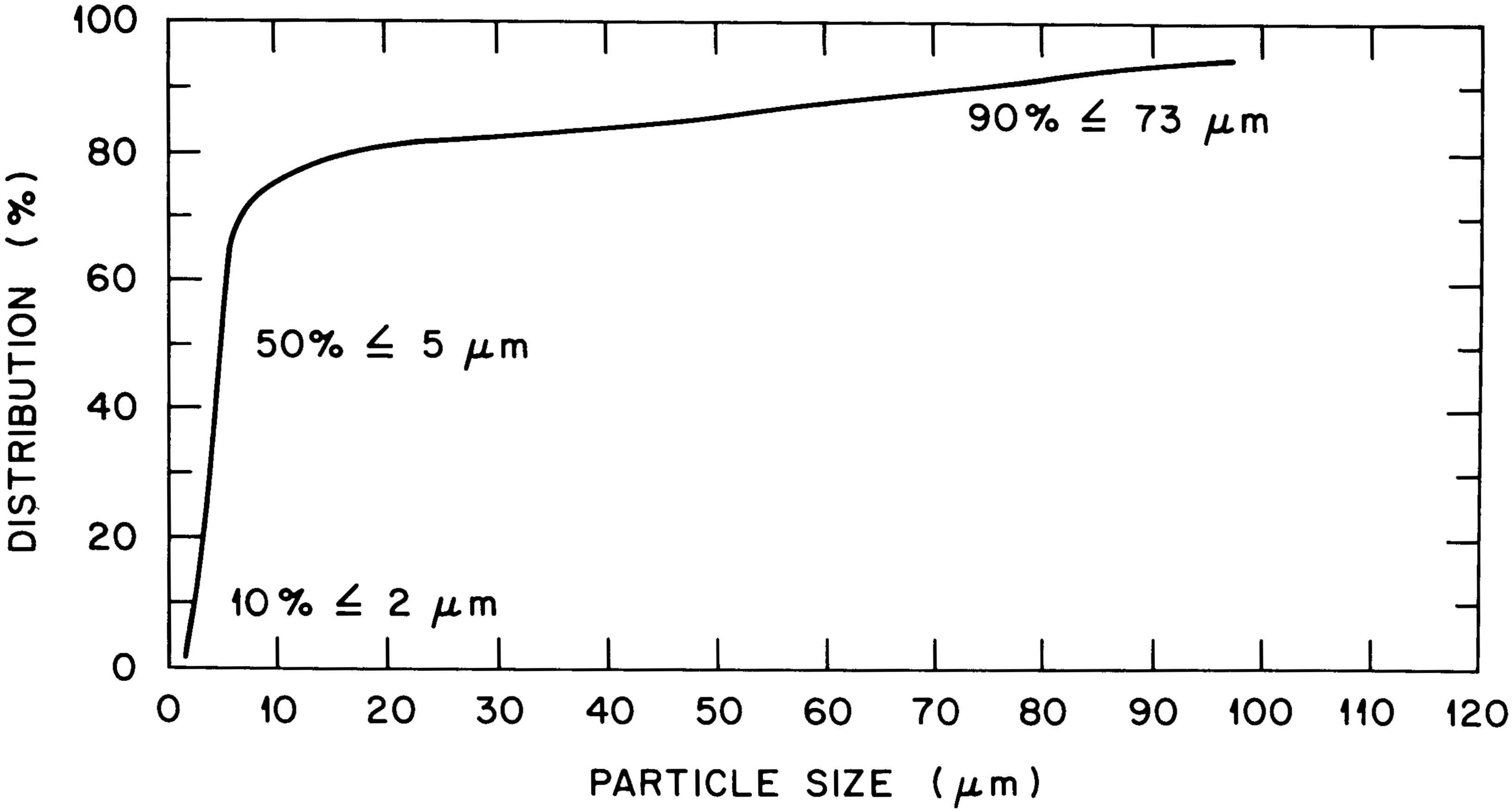

Fig. A-2. Particle size distribution analysis of soil sample GC-2 from the NFSS.

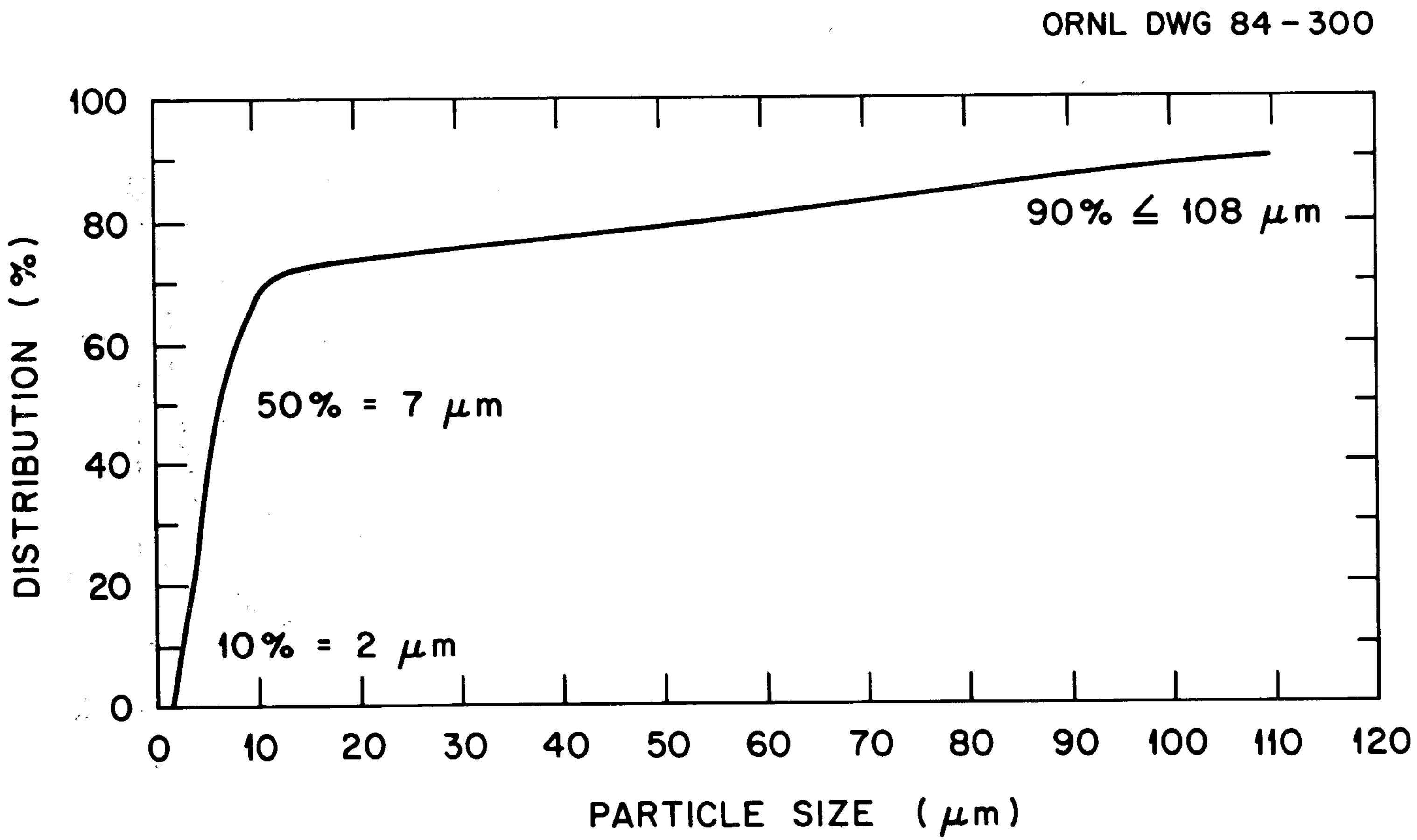

Fig. A-3. Particle size distribution analysis of soil sample GC-3 from the NFSS.

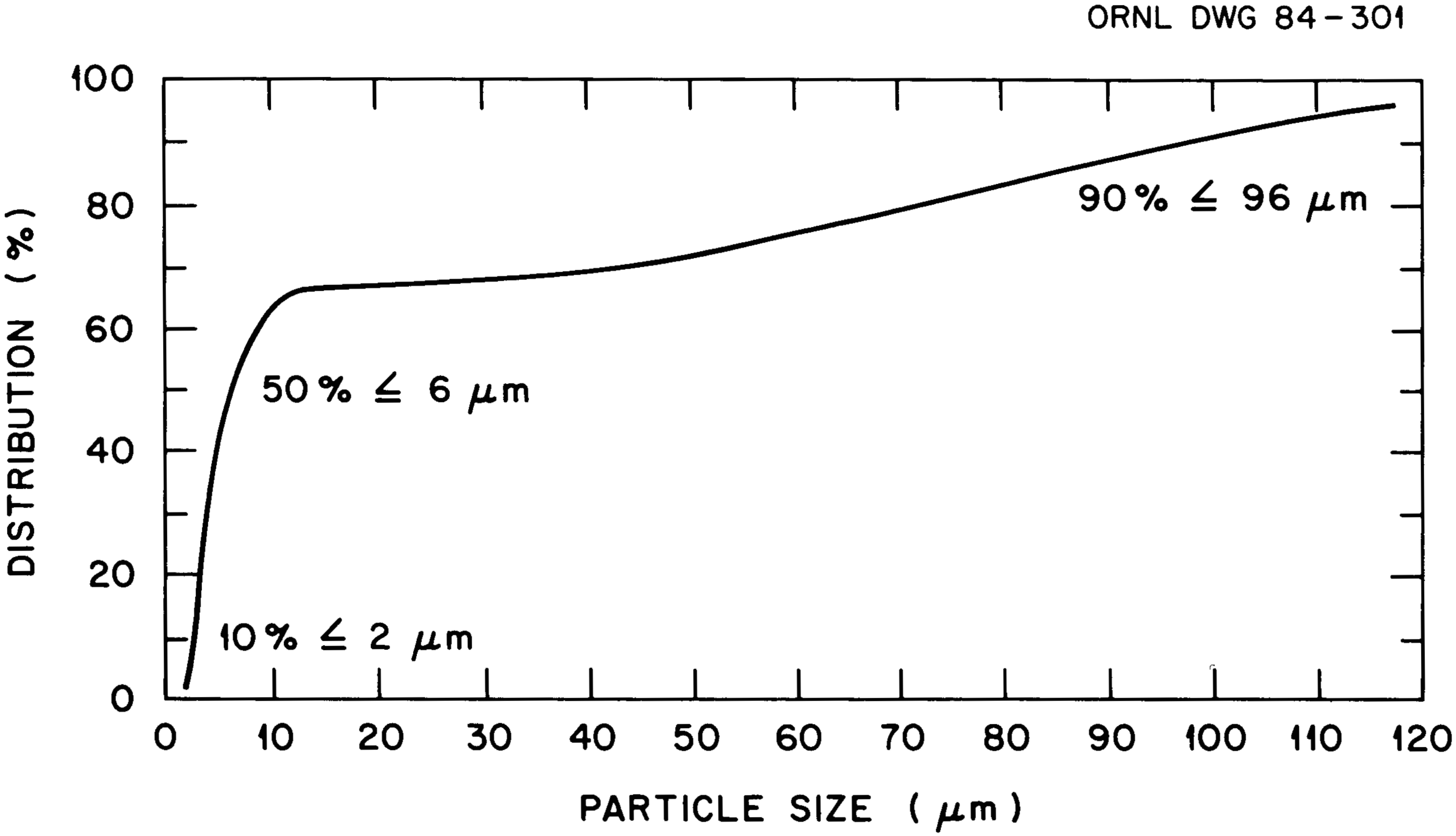

Fig. A-4. Particle size distribution analysis of soil sample BCB-1 from the NFSS.

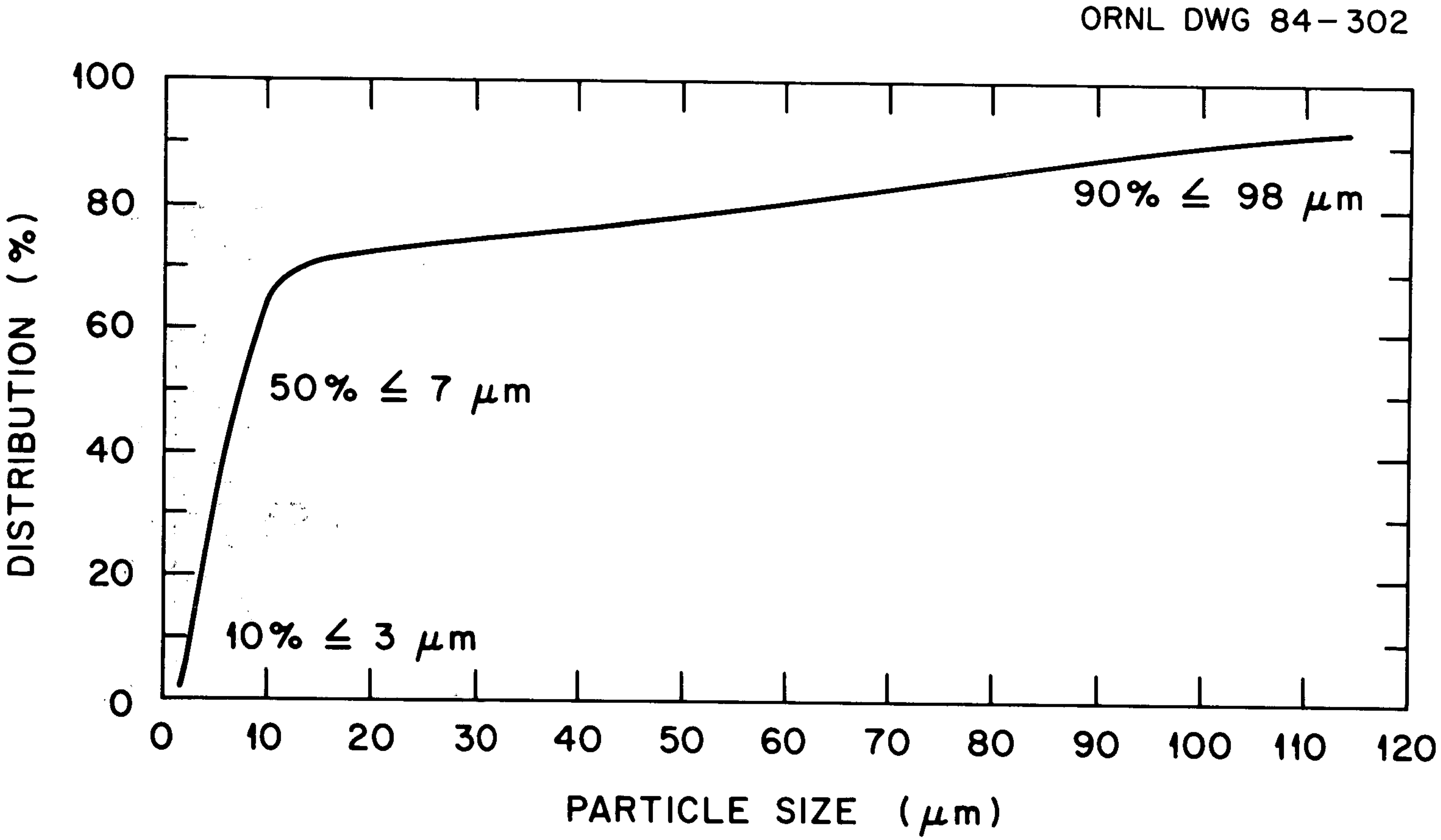

Fig. A-5. Particle size distribution analysis of soil sample BCB-2 from the NFSS.

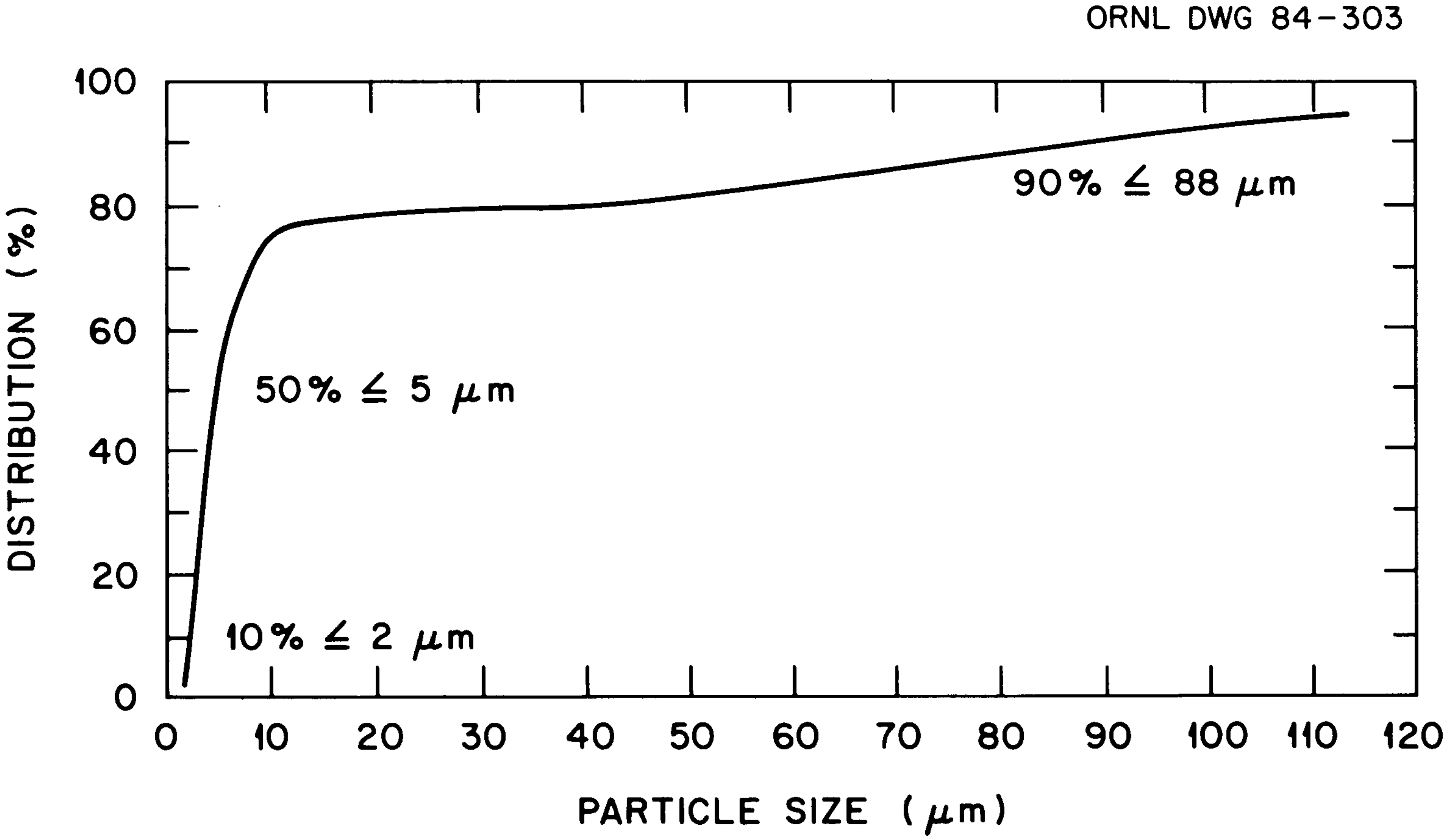

Fig. A-6. Particle size distribution analysis of soil sample BCB-3 from the NFSS.

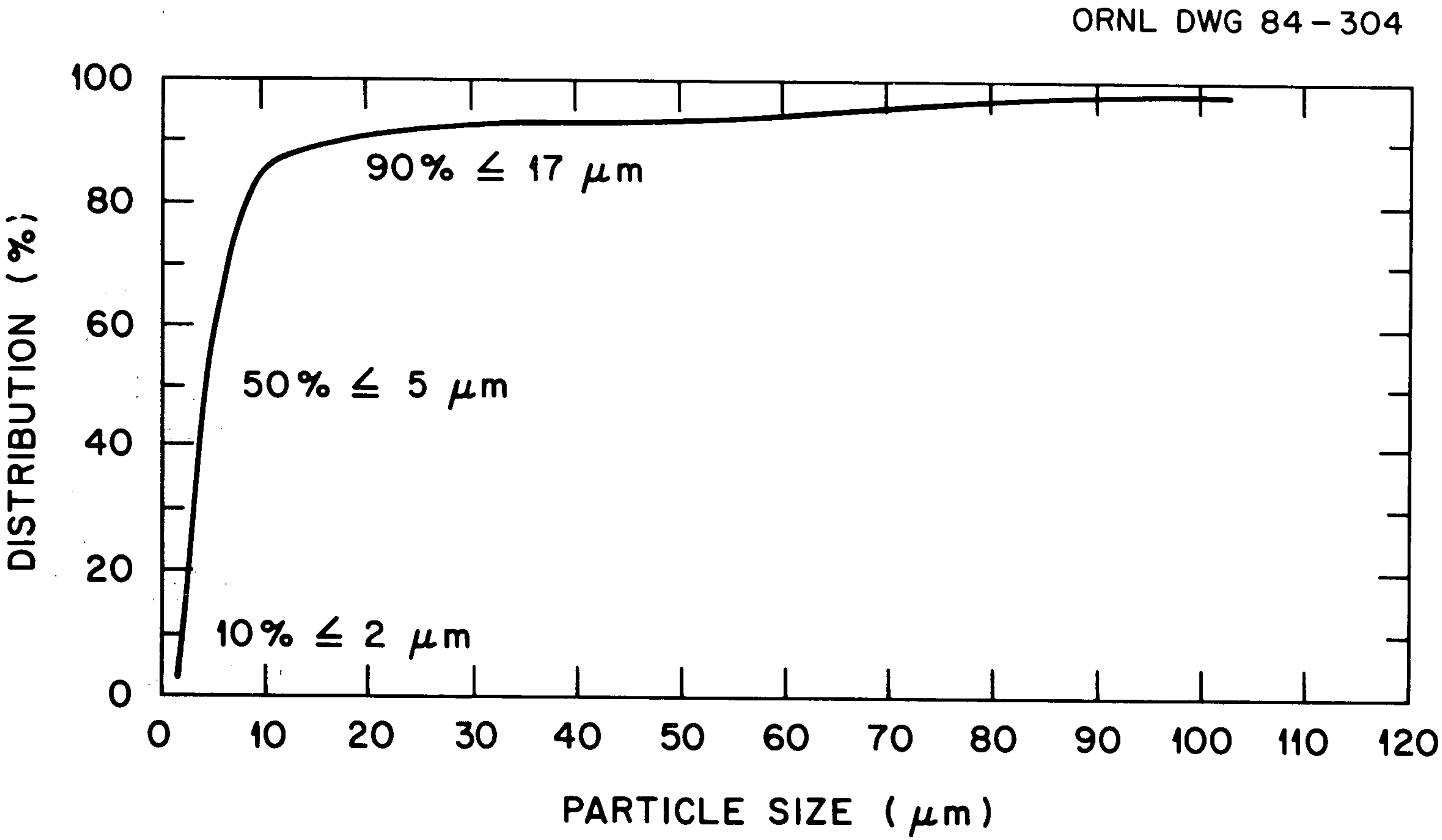

Fig. A-7. Particle size distribution analysis of soil sample LC-1 from the NFSS area.

Appendix B.

DETAILS OF EXPERIMENTAL METHOD USED FOR THE DETERMINATION OF RADIONUCLIDE SORPTION AND APPARENT CONCENTRATION LIMIT VALUES

B.1. EXPERIMENTAL PROCEDURE FOR THE DETERMINATION OF SORPTION ISOTHERMS (AMBIENT-pH METHOD)

To provide data for sorption isotherms, each test is conducted by contacting 2.0 g of soil with 10 mL of the synthetic groundwater in a polystyrene centrifuge tube for 3 h to allow the soil and the groundwater to reach steady-state conditions. An argon atmosphere is maintained above the slurry to minimize any additional source of carbonate from CO_2 in the air from being introduced into the sample and to prevent air oxidation of reduced components. The appropriate metal-ion spike is added to the slurry to obtain the desired initial concentration. The concentration of the metal-ion stock solutions is chosen such that the addition of 0.1 mL to the 10 mL of the groundwater will give the desired initial concentration. The radioactive tracer is then added to the sample. The activity of the appropriate 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 metal ion-spiked groundwater will give an initial counting level of approximately 100,000 cpm/mL. The sample is then placed on a shaker for 24 h. (Various contact times were 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.

After a stable pH level has been reached, the sample is placed in a Sorvall centrifuge and centrifuged at 5000 rpm for about 15 min. A 1.0-mL volume of the clear supernate is then removed, and the alpha and gamma activities are determined by counting in a Packard Tri-Carb scintillation spectrometer and a Packard automatic gamma spectrometer, respectively. Appropriate standards and blanks are also counted.

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

```
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   GOTO 490
00820   END
```

Appendix C.

URANIUM SORPTION ISOTHERM DATA FOR NEAR-SURFACE SOILS FROM THE NFSS

Table C-1. Uranium(VI) sorption isotherm data for NFSS soil sample GC-1

Calculated initial U conc.	After contact					
(µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.9	$1.1x10^{-8}$	$7.7x10^{-8}$	-7.96	-7.11	7.0
11	7.9	$1.7x10^{-8}$	$1.6x10^{-7}$	-7.77	-6.80	9.4
21	7.9	$4.7x10^{-8}$	$2.1x10^{-7}$	-7.33	-6.68	4.5
31	7.9	$6.9x10^{-8}$	$3.2x10^{-7}$	-7.16	-6.49	4.6
41	8.0	$9.9x10^{-8}$	$3.8x10^{-7}$	-7.00	-6.42	3.8
51	8.0	$1.3x10^{-7}$	$4.5x10^{-7}$	-6.89	-6.35	3.5
101	7.9	$2.6x10^{-7}$	$8.5x10^{-7}$	-6.59	-6.07	3.3
201	8.1	$5.9x10^{-7}$	$1.3x10^{-6}$	-6.23	-5.89	2.2
301	8.0	$7.8x10^{-7}$	$2.5x10^{-6}$	-6.11	-5.60	3.2
401	7.6	$1.3x10^{-6}$	$2.2x10^{-6}$	-5.89	-5.66	1.7
501	7.7	$1.6x10^{-6}$	$2.8x10^{-6}$	-5.80	-5.55	1.8
1001	7.7	$3.3x10^{-6}$	$4.9x10^{-6}$	-5.48	-5.31	1.4
4813	7.5	$1.4x10^{-5}$	$3.2x10^{-5}$	-4.85	-4.49	2.3
9191	7.3	$2.3x10^{-5}$	$8.1x10^{-5}$	-4.64	-4.09	3.5

Table C-2. Uranium(VI) sorption isotherm data for NFSS soil sample GC-2

Calculated initial U conc. (µg/mL)	pH	After contact Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.9	1.3×10^{-8}	7.0×10^{-8}	-7.89	-7.15	5.4
11	7.9	2.2×10^{-8}	1.3×10^{-7}	-7.66	-6.89	5.9
21	7.9	5.1×10^{-8}	1.9×10^{-7}	-7.28	-6.72	3.7
31	7.8	7.9×10^{-8}	2.7×10^{-7}	-7.10	-6.57	3.4
41	7.8	1.1×10^{-7}	3.1×10^{-7}	-6.96	-6.51	2.8
51	7.7	1.4×10^{-7}	3.9×10^{-7}	-6.85	-6.41	2.8
101	8.0	3.0×10^{-7}	6.4×10^{-7}	-6.52	-6.19	2.1
201	8.1	6.5×10^{-7}	1.0×10^{-6}	-6.19	-6.00	1.5
301	7.7	9.9×10^{-7}	1.4×10^{-6}	-6.00	-5.85	1.4
401	7.7	1.4×10^{-6}	1.5×10^{-6}	-5.85	-5.82	1.1
501	7.8	1.7×10^{-6}	2.1×10^{-6}	-5.77	-5.68	1.2
1001	7.9	3.4×10^{-6}	4.0×10^{-6}	-5.47	-5.40	1.2
4813	7.5	1.4×10^{-5}	2.8×10^{-5}	-4.84	-4.55	2.0
9191	7.1	2.5×10^{-5}	7.2×10^{-5}	-4.61	-4.14	2.9

Table C-3. Uranium(VI) sorption isotherm data for NFSS soil sample GC-3

Calculated initial U conc. (µg/mL)	After contact pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	1.4×10^{-8}	6.2×10^{-8}	-7.85	-7.21	4.4
11	7.7	2.7×10^{-8}	1.0×10^{-7}	-7.57	-7.00	3.7
21	7.7	5.5×10^{-8}	1.8×10^{-7}	-7.26	-6.74	3.2
31	7.8	8.5×10^{-8}	2.4×10^{-7}	-7.07	-6.62	2.8
41	7.7	1.2×10^{-7}	2.6×10^{-7}	-6.92	-6.59	2.1
51	-	-	-	-	-	-
101	7.7	3.1×10^{-7}	6.0×10^{-7}	-6.51	-6.22	1.9
201	7.6	6.5×10^{-7}	1.0×10^{-6}	-6.19	-6.00	1.6
301	7.6	9.9×10^{-7}	1.4×10^{-6}	-6.00	-5.85	1.4
401	7.6	1.4×10^{-6}	1.7×10^{-6}	-5.85	-5.77	1.2
501	7.6	1.7×10^{-6}	2.0×10^{-6}	-5.77	-5.70	1.2
1001	7.3	3.6×10^{-6}	3.0×10^{-6}	-5.44	-5.52	0.8
4813	6.6	8.4×10^{-6}	6.0×10^{-5}	-5.07	-4.22	7.1
9191	6.4	3.1×10^{-5}	3.9×10^{-5}	-4.51	-4.41	1.3

Table C-4. Uranium(VI) sorption isotherm data for NFSS soil sample BCB-1

Initial	After contact					
U conc. (µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.5	1.1×10^{-8}	7.4×10^{-8}	-7.96	-7.13	6.5
11	7.7	2.2×10^{-8}	1.3×10^{-7}	-7.66	-6.89	5.7
21	7.8	4.7×10^{-8}	2.2×10^{-7}	-7.33	-6.66	4.6
31	7.7	7.3×10^{-8}	3.0×10^{-7}	-7.14	-6.52	4.1
41	7.7	9.7×10^{-7}	3.8×10^{-7}	-7.01	-6.42	3.8
51	7.7	1.2×10^{-7}	4.6×10^{-7}	-6.92	-6.34	3.8
101	7.7	2.7×10^{-7}	7.7×10^{-7}	-6.57	-6.11	2.8
201	7.6	5.6×10^{-7}	1.3×10^{-6}	-6.25	-5.89	2.4
301	7.6	9.0×10^{-7}	1.7×10^{-6}	-6.05	-5.77	1.9
401	7.6	1.2×10^{-6}	2.2×10^{-6}	-5.92	-5.66	1.8
501	7.6	1.5×10^{-6}	2.6×10^{-6}	-5.82	-5.59	1.7
1001	7.3	3.4×10^{-6}	3.6×10^{-6}	-5.47	-5.44	1.1
4813	6.6	1.8×10^{-5}	1.1×10^{-5}	-4.74	-4.96	0.6
9191	6.4	3.4×10^{-5}	2.0×10^{-5}	-4.47	-4.70	0.6

Table C-5. Uranium(VI) sorption isotherm data for NFSS soil sample BCB-2

Initial U conc. (μg/mL)	After contact pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	9.4×10^{-9}	8.5×10^{-8}	-8.03	-7.07	9.0
11	7.6	1.8×10^{-8}	1.5×10^{-7}	-7.74	-6.82	8.3
21	7.7	3.7×10^{-8}	2.6×10^{-7}	-7.43	-6.59	7.0
31	7.6	5.9×10^{-8}	3.7×10^{-7}	-7.23	-6.43	6.3
41	7.6	8.2×10^{-8}	4.6×10^{-7}	-7.09	-6.34	5.6
51	7.6	1.1×10^{-7}	5.5×10^{-7}	-6.96	-6.26	5.2
101	7.6	2.4×10^{-7}	9.3×10^{-7}	-6.62	-6.03	3.9
201	7.8	4.9×10^{-7}	1.7×10^{-6}	-6.31	-5.77	3.4
301	7.5	8.0×10^{-7}	2.2×10^{-6}	-6.10	-5.66	2.8
401	7.5	1.1×10^{-6}	2.8×10^{-6}	-5.96	-5.55	2.4
501	7.5	1.4×10^{-6}	3.3×10^{-6}	-5.85	-5.48	2.4
1001	7.4	3.2×10^{-6}	4.4×10^{-6}	-5.49	-5.36	1.4
4813	6.7	1.7×10^{-5}	1.4×10^{-5}	-4.77	-4.85	0.8
9191	6.5	3.2×10^{-5}	3.0×10^{-5}	-4.49	-4.52	0.9

Table C-6. Uranium(VI) sorption isotherm data for NFSS soil sample BCB-3

Calculated initial U conc. (μg/mL)	After contact pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	$8.3\text{x}10^{-9}$	$9.0\text{x}10^{-8}$	-8.08	-7.05	10.5
11	7.6	$1.6\text{x}10^{-8}$	$1.5\text{x}10^{-7}$	-7.80	-6.82	9.4
21	7.6	$3.4\text{x}10^{-8}$	$2.8\text{x}10^{-7}$	-7.47	-6.55	8.1
31	7.6	$5.5\text{x}10^{-8}$	$3.9\text{x}10^{-7}$	-7.26	-6.41	7.1
41	7.6	$7.5\text{x}10^{-8}$	$4.9\text{x}10^{-7}$	-7.12	-6.31	6.4
51	7.6	$1.0\text{x}10^{-7}$	$5.6\text{x}10^{-7}$	-7.00	-6.25	5.6
101	7.6	$2.3\text{x}10^{-7}$	$9.9\text{x}10^{-7}$	-6.64	-6.00	4.3
201	7.5	$4.7\text{x}10^{-7}$	$1.8\text{x}10^{-6}$	-6.33	-5.74	3.9
301	7.6	$7.8\text{x}10^{-7}$	$2.3\text{x}10^{-6}$	-6.11	-5.64	3.0
401	7.5	$1.1\text{x}10^{-6}$	$2.5\text{x}10^{-6}$	-5.96	-5.60	2.2
501	7.5	$1.5\text{x}10^{-6}$	$2.8\text{x}10^{-6}$	-5.82	-5.55	1.9
1001	7.2	$3.2\text{x}10^{-6}$	$4.8\text{x}10^{-6}$	-5.49	-5.32	1.5
4813	6.2	$1.7\text{x}10^{-5}$	$1.3\text{x}10^{-5}$	-4.77	-4.89	0.8
9191	6.4	$3.4\text{x}10^{-6}$	$1.9\text{x}10^{-5}$	-4.47	-4.72	0.5

Table C-7. Uranium(VI) sorption isotherm data for NFSS soil sample LC-1

Calculated initial U conc. (µg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.8	1.2×10^{-8}	7.2×10^{-8}	-7.92	-7.14	6.1
11	7.7	2.3×10^{-8}	1.2×10^{-7}	-7.64	-6.92	5.3
21	7.8	4.8×10^{-8}	2.1×10^{-7}	-7.32	-6.68	4.4
31	7.8	7.5×10^{-8}	2.9×10^{-7}	-7.12	-6.54	3.9
41	7.8	9.8×10^{-8}	3.7×10^{-7}	-7.01	-6.43	3.8
51	7.8	1.3×10^{-7}	4.2×10^{-7}	-6.89	-6.38	3.3
101	7.8	2.8×10^{-7}	7.3×10^{-7}	-6.55	-6.14	2.6
201	7.8	5.8×10^{-7}	1.2×10^{-6}	-6.24	-5.92	2.1
301	7.8	9.1×10^{-7}	1.7×10^{-6}	-6.04	-5.77	1.8
401	7.7	1.2×10^{-6}	2.1×10^{-6}	-5.92	-5.68	1.7
501	7.7	1.5×10^{-6}	2.7×10^{-6}	-5.82	-5.57	1.8
1001	7.4	3.5×10^{-6}	3.4×10^{-6}	-5.46	-5.47	1.0
4813	6.8	1.8×10^{-5}	1.0×10^{-5}	-4.74	-5.00	0.6
9191	6.5	3.7×10^{-5}	5.6×10^{-5}	-4.43	-4.25	1.5

Appendix D.

URANIUM SORPTION ISOTHERM DATA FOR SOIL FROM CORE SAMPLE BH-77 FROM THE NFSS

Table D-1. Uranium(VI) sorption isotherm data for NFSS core sample from the 0.3-m (1-ft) level of BH-77

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)
6	7.9	1.5×10^{-8}	5.7×10^{-8}	-7.82	-7.24	3.9
11	7.1	2.8×10^{-8}	9.8×10^{-8}	-7.55	-7.01	3.6
21	7.2	5.3×10^{-8}	1.9×10^{-7}	-7.28	-6.72	3.5
31	7.1	8.1×10^{-8}	2.6×10^{-7}	-7.09	-6.58	3.2
41	8.0	9.9×10^{-8}	3.7×10^{-7}	-7.00	-6.43	3.7
50	7.1	1.3×10^{-7}	4.4×10^{-7}	-6.89	-6.36	3.5
101	7.7	2.6×10^{-7}	8.3×10^{-7}	-6.58	-6.08	3.2
195	7.6	4.8×10^{-7}	1.7×10^{-6}	-6.32	-5.77	3.6
294	8.0	7.3×10^{-7}	2.6×10^{-6}	-6.14	-5.58	3.6
392	7.1	9.9×10^{-7}	3.4×10^{-6}	-6.00	-5.47	3.4
486	8.1	1.2×10^{-6}	4.1×10^{-6}	-5.92	-5.39	3.3
979	7.0	2.9×10^{-6}	6.1×10^{-6}	-5.54	-5.21	2.1
4707	6.7	1.2×10^{-5}	4.0×10^{-5}	-4.92	-4.40	3.3
8989	6.1	1.7×10^{-5}	1.2×10^{-4}	-4.77	-3.92	6.8

Table D-2. Uranium(VI) sorption isotherm data for NFSS core sample from the 2.1-m (7-ft) level of BH-77

Initial U conc. (µg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	6.9	1.1×10^{-8}	7.7×10^{-8}	-7.96	-7.11	7.0
11	8.0	2.2×10^{-8}	1.2×10^{-7}	-7.66	-6.92	5.6
21	7.5	3.9×10^{-8}	2.6×10^{-7}	-7.41	-6.58	6.7
31	7.1	6.7×10^{-8}	3.3×10^{-7}	-7.17	-6.48	5.0
40	7.1	9.0×10^{-8}	4.1×10^{-7}	-7.05	-6.39	4.6
50	7.2	1.1×10^{-7}	5.0×10^{-7}	-6.96	-6.30	4.4
100	7.1	2.5×10^{-7}	8.8×10^{-7}	-6.60	-6.06	3.5
195	7.0	4.9×10^{-7}	1.7×10^{-6}	-6.31	-5.77	3.5
294	7.1	7.3×10^{-7}	2.6×10^{-6}	-6.14	-5.58	3.6
392	7.1	8.9×10^{-7}	3.9×10^{-6}	-6.05	-5.41	4.4
486	7.1	1.1×10^{-6}	4.8×10^{-6}	-5.96	-5.32	4.3
979	7.0	2.5×10^{-6}	8.3×10^{-6}	-5.60	-5.08	3.3
4707	6.6	8.1×10^{-6}	6.2×10^{-5}	-5.09	-4.21	7.6
8989	6.2	1.3×10^{-5}	1.4×10^{-4}	-4.89	-3.85	10.1

Table D-3. Uranium(VI) sorption isotherm data for NFSS core sample from the 3.2-m (10.5-ft) level of BH-77

Initial	After contact					
U conc. (µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	$1.8x10^{-9}$	$1.2x10^{-7}$	-8.74	-6.92	70.0
11	7.8	$3.3x10^{-8}$	$2.2x10^{-7}$	-8.48	-6.66	67.0
21	7.9	$6.7x10^{-9}$	$4.2x10^{-7}$	-8.17	-6.38	63.0
31	7.8	$1.0x10^{-8}$	$6.2x10^{-7}$	-8.00	-6.21	61.0
40	8.1	$1.4x10^{-8}$	$8.0x10^{-7}$	-7.85	-6.10	55.0
50	8.3	$1.9x10^{-8}$	$9.8x10^{-7}$	-7.72	-6.01	52.0
101	8.1	$4.8x10^{-8}$	$1.9x10^{-6}$	-7.32	-5.72	40.0
195	8.0	$1.4x10^{-8}$	$3.5x10^{-6}$	-6.85	-5.46	26.0
294	8.1	$2.8x10^{-7}$	$4.9x10^{-6}$	-6.55	-5.31	18.0
392	8.0	$4.3x10^{-7}$	$6.2x10^{-6}$	-6.37	-5.21	14.0
486	8.0	$6.3x10^{-7}$	$7.2x10^{-6}$	-6.20	-5.14	11.0
979	7.8	$1.8x10^{-7}$	$1.2x10^{-5}$	-6.74	-4.92	6.7
4707	6.8	$5.0x10^{-6}$	$7.8x10^{-5}$	-5.30	-4.11	16.0
8989	5.8	$3.2x10^{-6}$	$1.9x10^{-4}$	-4.49	-3.72	59.0

Table D-4. Uranium(VI) sorption isotherm data for NFSS core sample from the 5.6-m (18.5-ft) level of BH-77

Initial U conc. (µg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.7	1.1×10^{-8}	7.8×10^{-8}	-7.96	-7.11	7.1
11	7.7	1.7×10^{-8}	1.5×10^{-7}	-7.77	-6.82	8.8
21	7.7	3.7×10^{-8}	2.7×10^{-7}	-7.43	-6.57	7.2
31	7.6	8.7×10^{-8}	2.3×10^{-7}	-7.06	-6.64	2.6
41	7.7	9.0×10^{-8}	4.1×10^{-7}	-7.05	-6.39	4.6
50	7.7	1.1×10^{-7}	5.1×10^{-7}	-6.96	-6.29	4.6
101	7.6	2.4×10^{-7}	9.4×10^{-7}	-6.62	-6.03	4.0
195	7.7	4.9×10^{-7}	1.7×10^{-6}	-6.31	-5.77	3.4
294	7.7	7.7×10^{-7}	2.4×10^{-6}	-6.11	-5.62	3.1
392	7.8	1.1×10^{-6}	2.7×10^{-6}	-5.96	-5.57	2.4
486	7.7	1.3×10^{-6}	3.6×10^{-6}	-5.89	-5.44	2.8
979	7.6	3.2×10^{-6}	4.6×10^{-6}	-5.49	-5.34	1.4
4707	7.3	1.5×10^{-5}	2.6×10^{-5}	-4.82	-4.58	1.7
8989	6.6	3.0×10^{-5}	4.6×10^{-5}	-4.52	-4.34	1.6

Table D-5. Uranium(VI) sorption isotherm data for NFSS core sample from the 7.9-m (26-ft) level of BH-77

Calculated initial U conc. (µg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	1.0×10^{-8}	9.0×10^{-8}	-8.00	-7.05	9.0
11	7.7	2.0×10^{-8}	1.3×10^{-7}	-7.70	-6.87	6.5
21	7.8	4.0×10^{-8}	2.5×10^{-7}	-7.40	-6.60	6.3
31	7.7	7.0×10^{-8}	3.2×10^{-7}	-7.15	-6.49	4.6
41	7.7	1.0×10^{-7}	3.7×10^{-7}	-7.00	-6.43	3.7
50	7.7	1.2×10^{-7}	4.7×10^{-7}	-6.92	-6.33	3.9
101	7.7	2.8×10^{-7}	7.5×10^{-7}	-6.55	-6.12	2.7
195	7.6	6.0×10^{-7}	1.24×10^{-6}	-6.22	-5.91	2.1
294	7.7	9.4×10^{-7}	1.67×10^{-6}	-6.03	-5.78	1.8
392	7.6	1.29×10^{-6}	2.01×10^{-6}	-5.89	-5.70	1.6
486	7.6	1.58×10^{-6}	2.69×10^{-6}	-5.80	-5.57	1.7
979	7.2	3.44×10^{-6}	3.93×10^{-6}	-5.46	-5.41	1.1
4707	6.8	1.67×10^{-5}	1.64×10^{-5}	-4.78	-4.79	1.0
8989	6.4	3.03×10^{-5}	4.12×10^{-5}	-4.52	-4.39	1.4

Table D-6. Uranium(VI) sorption isotherm data for NFSS core sample from the 12.2-m (40-ft) level of BH-77

Initial	After contact					
U conc. (μg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.4	$1.1\text{x}10^{-8}$	$7.71\text{x}10^{-8}$	-7.96	-7.11	7.0
11	7.5	$2.31\text{x}10^{-7}$	$1.21\text{x}10^{-7}$	-7.64	-6.92	5.2
21	7.9	$4.76\text{x}10^{-8}$	$2.14\text{x}10^{-7}$	-7.32	-6.67	4.5
31	7.6	$7.19\text{x}10^{-8}$	$3.06\text{x}10^{-7}$	-7.14	-6.51	4.5
41	7.9	$1.00\text{x}10^{-7}$	$3.63\text{x}10^{-7}$	-7.00	-6.44	3.6
50	7.9	$1.33\text{x}10^{-7}$	$4.03\text{x}10^{-7}$	-6.88	-6.39	3.0
101	7.6	$2.79\text{x}10^{-7}$	$7.33\text{x}10^{-7}$	-6.55	-6.13	2.6
195	7.9	$5.75\text{x}10^{-7}$	$1.24\text{x}10^{-6}$	-6.24	-5.90	2.2
294	8.0	$9.16\text{x}10^{-7}$	$1.63\text{x}10^{-6}$	-6.04	-5.79	1.8
392	8.0	$1.23\text{x}10^{-6}$	$2.13\text{x}10^{-6}$	-5.91	-5.67	1.7
486	7.9	$1.57\text{x}10^{-6}$	$2.41\text{x}10^{-6}$	-5.80	-5.62	1.5
979	7.7	$3.39\text{x}10^{-6}$	$3.69\text{x}10^{-6}$	-5.47	-5.43	1.1
4707	6.8	$1.67\text{x}10^{-5}$	$1.64\text{x}10^{-5}$	-4.78	-4.79	1.0
8989	6.5	$3.00\text{x}10^{-5}$	$4.29\text{x}10^{-5}$	-4.52	-4.39	1.4

Table D-7. Uranium(VI) sorption isotherm data for NFSS core sample from the 13.7-m (45-ft) level of BH-77

Calculated initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	1.3×10^{-11}	1.3×10^{-7}	-10.89	-6.89	1.0×10^{4}
11	7.7	2.3×10^{-11}	2.4×10^{-7}	-10.64	-6.62	1.0×10^{4}
21	7.8	3.4×10^{-11}	4.5×10^{-7}	-10.47	-6.35	1.3×10^{4}
31	7.8	1.1×10^{-10}	6.7×10^{-7}	-9.96	-6.17	6.1×10^{3}
41	7.8	2.3×10^{-10}	8.8×10^{-7}	-9.64	-6.06	3.8×10^{3}
51	7.6	8.2×10^{-10}	1.1×10^{-6}	-9.09	-5.96	1.3×10^{3}
101	7.6	5.0×10^{-8}	1.9×10^{-6}	-7.30	-5.72	3.8×10^{1}
201	7.6	2.5×10^{-7}	3.1×10^{-6}	-6.60	-5.51	1.2×10^{1}
301	7.6	6.0×10^{-7}	3.4×10^{-6}	-6.22	-5.47	5.7×10^{0}
401	7.6	7.7×10^{-7}	4.7×10^{-6}	-6.11	-5.33	6.1×10^{0}
501	7.7	9.0×10^{-7}	6.2×10^{-6}	-6.05	-5.21	6.9×10^{0}
1001	7.8	2.9×10^{-6}	6.6×10^{-6}	-5.54	-5.18	2.3×10^{0}
4813	7.6	1.2×10^{-5}	4.8×10^{-5}	-4.92	-4.32	1.5×10^{0}
9191	8.4	2.1×10^{-5}	1.2×10^{-4}	-4.68	-3.92	5.7×10^{0}

Table D-8. Uranium(VI) sorption isotherm data for NFSS core sample from the 13.7-m (45-ft) level of BH-77: replicate data to that shown in Table D-7

Initial	After contact					
U conc. (µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.9	1.9x10^{-11}	1.3x10^{-7}	-10.73	-6.89	6.8x10^{3}
11	7.9	3.1x10^{-11}	2.4x10^{-7}	-10.51	-6.62	1.0x10^{3}
21	7.9	7.2x10^{-11}	4.6x10^{-7}	-10.14	-6.34	1.3x10^{3}
31	7.9	7.4x10^{-10}	6.7x10^{-7}	-10.13	-6.17	9.1x10^{3}
41	7.9	1.3x10^{-10}	8.7x10^{-7}	-9.90	-6.06	6.9x10^{3}
50	7.9	1.4x10^{-10}	1.1x10^{-6}	-9.85	-5.97	7.7x10^{3}
101	7.8	2.1x10^{-10}	2.2x10^{-6}	-8.69	-5.67	1.0x10^{4}
195	7.8	1.1x10^{-8}	4.1x10^{-6}	-7.95	-5.38	3.7x10^{2}
294	7.8	1.5x10^{-7}	5.5x10^{-6}	-6.81	-5.26	3.7x10^{2}
392	7.4	6.9x10^{-7}	4.9x10^{-6}	-6.16	-5.31	7.1x10^{0}
486	7.4	8.7x10^{-7}	6.0x10^{-6}	-6.06	-5.22	6.9x10^{0}
979	7.4	2.7x10^{-6}	7.0x10^{-6}	-5.56	-5.15	2.6x10^{0}
4707	6.1	1.3x10^{-5}	3.8x10^{-5}	-4.90	-4.42	2.9x10^{0}
8989	5.8	1.7x10^{-5}	1.1x10^{-4}	-4.76	-3.95	6.5x10^{0}

Appendix E.

URANIUM SORPTION ISOTHERM DATA SHOWING THE EFFECT OF ADDING BENTONITE TO SOIL SAMPLES FROM THE NFSS

Table E-1. Uranium(VI) sorption isotherm data for NFSS soil sample BCB-1

Calculated initial U conc.	After contact					
(μg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.5	$1.1\text{x}10^{-9}$	$7.4\text{x}10^{-8}$	-7.96	-7.13	6.5
11	7.7	$2.2\text{x}10^{-8}$	$1.3\text{x}10^{-8}$	-7.66	-6.89	5.7
21	7.8	$4.7\text{x}10^{-8}$	$2.2\text{x}10^{-7}$	-7.33	-6.66	4.6
31	7.7	$7.3\text{x}10^{-8}$	$3.0\text{x}10^{-7}$	-7.14	-6.52	4.1
41	7.7	$9.7\text{x}10^{-8}$	$3.8\text{x}10^{-7}$	-7.01	-6.42	3.8
51	7.7	$1.2\text{x}10^{-7}$	$4.6\text{x}10^{-7}$	-6.92	-6.34	3.8
101	7.7	$2.7\text{x}10^{-7}$	$7.7\text{x}10^{-7}$	-6.57	-6.11	2.8
201	7.6	$5.6\text{x}10^{-7}$	$1.3\text{x}10^{-6}$	-6.25	-5.89	2.4
301	7.6	$9.0\text{x}10^{-7}$	$1.7\text{x}10^{-6}$	-6.05	-5.77	1.9
401	7.6	$1.2\text{x}10^{-6}$	$1.1\text{x}10^{-6}$	-5.92	-5.66	1.8
501	7.6	$1.5\text{x}10^{-6}$	$2.6\text{x}10^{-6}$	-5.82	-5.59	1.7
1001	7.3	$3.6\text{x}10^{-6}$	$3.6\text{x}10^{-6}$	-5.47	-5.44	1.1
4813	6.6	$1.8\text{x}10^{-5}$	$1.1\text{x}10^{-5}$	-4.74	-4.96	0.6
9191	6.4	$3.4\text{x}10^{-5}$	$2.0\text{x}10^{-5}$	-4.47	-4.70	0.6

Table E-2. Uranium(VI) sorption isotherm data for NFSS soil sample BCB-1: effect of adding 10% bentonite

Calculated initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	8.0	9.21x10^{-9}	8.61x10^{-8}	-8.04	-7.07	9.3
11	7.9	1.75x10^{-8}	1.49x10^{-8}	-7.76	-6.83	8.6
21	8.2	3.70x10^{-8}	2.68x10^{-7}	-7.43	-6.57	7.3
31	8.2	5.87x10^{-8}	3.74x10^{-7}	-7.23	-6.43	6.4
41	8.0	8.20x10^{-8}	4.57x10^{-7}	-7.09	-6.34	5.6
50	8.2	1.08x10^{-7}	5.32x10^{-7}	-6.97	-6.27	4.9
101	8.0	2.52x10^{-7}	8.71x10^{-7}	-6.60	-6.06	3.5
195	8.0	5.39x10^{-7}	1.43x10^{-6}	-6.27	-5.84	2.7
294	8.2	8.59x10^{-7}	1.92x10^{-6}	-6.07	-5.72	2.2
392	8.1	1.19x10^{-6}	2.33x10^{-6}	-5.92	-5.63	2.0
486	8.0	1.53x10^{-6}	2.61x10^{-6}	-5.81	-5.58	1.7
979	7.5	3.32x10^{-6}	4.04x10^{-6}	-5.48	-5.59	1.2
4707	6.8	1.67x10^{-5}	1.62x10^{-5}	-4.78	-4.79	1.0
8989	6.5	3.00x10^{-5}	4.31x10^{-5}	-4.52	-4.37	1.4

Table E-3. Uranium(VI) sorption isotherm data for NFSS soil sample LC-1

Calculated initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.8	1.2×10^{-9}	7.2×10^{-8}	-7.92	-7.14	6.1
11	7.7	2.3×10^{-8}	1.2×10^{-8}	-7.64	-6.92	5.3
21	7.8	4.8×10^{-8}	2.1×10^{-7}	-7.32	-6.68	7.3
31	7.8	7.5×10^{-8}	2.9×10^{-7}	-7.12	-6.54	3.9
41	7.8	9.8×10^{-8}	3.7×10^{-7}	-7.01	-6.43	3.8
51	7.8	1.3×10^{-7}	4.2×10^{-7}	-6.89	-6.38	3.3
101	7.8	2.8×10^{-7}	7.3×10^{-7}	-6.55	-6.14	2.6
201	7.8	5.8×10^{-7}	1.2×10^{-6}	-6.24	-5.92	2.1
301	7.8	9.1×10^{-7}	1.7×10^{-6}	-6.04	-5.72	1.8
401	7.7	1.2×10^{-6}	2.1×10^{-6}	-5.92	-5.68	1.7
501	7.7	1.5×10^{-6}	2.7×10^{-6}	-5.82	-5.57	1.8
1001	7.4	3.5×10^{-6}	3.4×10^{-6}	-5.46	-5.47	1.0
4813	6.8	1.8×10^{-5}	1.0×10^{-5}	-4.74	-5.00	0.6
9191	6.5	3.7×10^{-5}	5.6×10^{-5}	-4.43	-4.25	1.5

Table E-4. Uranium sorption isotherm data for NFSS soil sample LC-1: effect of adding 10% bentonite

Calculated initial	After contact					
U conc. (μg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.8	$1.06\text{x}10^{-8}$	$7.90\text{x}10^{-8}$	-7.97	-7.10	7.4
11	7.9	$2.32\text{x}10^{-8}$	$1.20\text{x}10^{-7}$	-7.63	-6.92	5.1
21	8.1	$4.74\text{x}10^{-8}$	$2.15\text{x}10^{-7}$	-7.32	-6.67	4.5
31	8.1	$7.33\text{x}10^{-8}$	$2.99\text{x}10^{-7}$	-7.13	-6.52	4.1
41	8.0	$9.97\text{x}10^{-8}$	$3.66\text{x}10^{-7}$	-7.00	-6.44	3.7
50	7.8	$1.28\text{x}10^{-7}$	$4.27\text{x}10^{-7}$	-6.89	-6.37	3.3
101	7.8	$2.81\text{x}10^{-7}$	$7.21\text{x}10^{-7}$	-6.55	-6.14	2.6
195	7.9	$5.74\text{x}10^{-7}$	$2.25\text{x}10^{-6}$	-6.24	-5.90	2.2
294	7.8	$8.97\text{x}10^{-7}$	$1.72\text{x}10^{-6}$	-6.05	-5.76	1.9
392	7.8	$1.23\text{x}10^{-6}$	$2.13\text{x}10^{-6}$	-5.91	-5.67	1.7
486	7.6	$1.58\text{x}10^{-6}$	$2.34\text{x}10^{-6}$	-5.80	-5.63	1.5
979	7.5	$3.41\text{x}10^{-6}$	$3.59\text{x}10^{-6}$	-5.47	-5.44	1.1
4707	6.9	$1.66\text{x}10^{-5}$	$1.68\text{x}10^{-5}$	-4.78	-4.78	1.0
8989	6.6	$3.11\text{x}10^{-5}$	$3.68\text{x}10^{-5}$	-4.51	-4.43	1.2

Table E-5. Uranium(VI) sorption isotherm data for NFSS soil sample from core BH-77 [~7.9 m (~26 ft)]

Calculated initial	After contact					
U conc. (µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	1.0×10^{-8}	9.0×10^{-8}	-8.00	-7.05	9.0
11	7.7	2.0×10^{-8}	1.3×10^{-7}	-7.70	-6.87	6.5
21	7.8	4.0×10^{-8}	2.5×10^{-7}	-7.40	-6.60	6.3
31	7.7	7.0×10^{-8}	3.2×10^{-7}	-7.15	-6.49	4.6
41	7.7	1.0×10^{-7}	3.7×10^{-7}	-7.00	-6.43	3.7
50	7.7	1.2×10^{-7}	4.7×10^{-7}	-6.92	-6.33	3.9
101	7.7	2.8×10^{-7}	7.5×10^{-7}	-6.55	-6.12	2.7
195	7.6	6.0×10^{-7}	1.24×10^{-6}	-6.22	-5.91	2.0
294	7.7	9.4×10^{-7}	1.67×10^{-6}	-6.03	-5.78	1.8
392	7.6	1.3×10^{-6}	2.0×10^{-6}	-5.89	-5.70	1.5
486	7.6	1.6×10^{-6}	2.7×10^{-6}	-5.80	-5.57	1.7
979	7.2	3.4×10^{-6}	3.4×10^{-6}	-5.46	-5.41	1.0
4707	6.8	1.7×10^{-5}	1.6×10^{-5}	-4.78	-4.79	0.94
8989	6.4	3.0×10^{-5}	4.1×10^{-5}	-4.52	-4.39	1.4

Table E-6. Uranium(VI) sorption isotherm data for NFSS soil sample from core BH-77 [~7.9 m (~26 ft)] mixed with 10 wt % bentonite

Calculated initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.7	$1.0\text{x}10^{-8}$	$8.2\text{x}10^{-8}$	-8.00	-7.09	8.2
11	7.8	$2.0\text{x}10^{-8}$	$1.4\text{x}10^{-7}$	-7.70	-6.87	7.0
21	7.9	$4.4\text{x}10^{-8}$	$2.3\text{x}10^{-7}$	-7.35	-6.69	5.2
31	7.9	$7.0\text{x}10^{-8}$	$3.2\text{x}10^{-7}$	-7.16	-6.50	4.6
41	7.9	$1.0\text{x}10^{-7}$	$3.7\text{x}10^{-7}$	-7.00	-6.43	3.7
50	7.9	$1.2\text{x}10^{-7}$	$4.5\text{x}10^{-7}$	-6.91	-6.34	3.8
101	7.9	$3.3\text{x}10^{-7}$	$4.7\text{x}10^{-7}$	-6.48	-6.33	1.4
195	7.8	$5.9\text{x}10^{-7}$	$1.2\text{x}10^{-7}$	-6.23	-5.94	2.0
293	7.9	$9.4\text{x}10^{-7}$	$1.5\text{x}10^{-6}$	-6.03	-5.83	1.6
392	7.9	$1.3\text{x}10^{-6}$	$1.9\text{x}10^{-6}$	-5.90	-5.72	1.5
486	7.8	$1.5\text{x}10^{-6}$	$2.7\text{x}10^{-6}$	-5.82	-5.57	1.8
979	7.5	$3.4\text{x}10^{-6}$	$3.5\text{x}10^{-6}$	-5.46	-5.46	1.0
4707	6.7	$1.7\text{x}10^{-5}$	$1.5\text{x}10^{-5}$	-4.77	-4.83	0.9
8989	6.4	$2.9\text{x}10^{-5}$	$4.9\text{x}10^{-5}$	-4.54	-4.31	1.7

Appendix F.

URANIUM SORPTION ISOTHERM DATA FOR SOILS FROM THE NFSS SHOWING THE EFFECT OF pH ADJUSTMENT FOR CARBONATE CONTROL

Table F-1. Uranium(VI) sorption isotherm data for NFSS soil sample GC-1

Calculated initial U conc. (μg/mL)	After contact pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.9	$1.1\mathrm{x}10^{-8}$	$7.7\mathrm{x}10^{-8}$	-7.96	-7.11	7.0
11	7.9	$1.7\mathrm{x}10^{-8}$	$1.6\mathrm{x}10^{-7}$	-7.77	-6.80	9.4
21	7.9	$4.7\mathrm{x}10^{-8}$	$2.1\mathrm{x}10^{-7}$	-7.33	-6.68	4.5
31	7.9	$6.9\mathrm{x}10^{-8}$	$3.2\mathrm{x}10^{-7}$	-7.16	-6.49	4.6
41	8.0	$9.9\mathrm{x}10^{-8}$	$3.8\mathrm{x}10^{-7}$	-7.00	-6.42	3.8
51	8.0	$1.3\mathrm{x}10^{-7}$	$4.5\mathrm{x}10^{-7}$	-6.89	-6.35	3.5
101	7.9	$2.6\mathrm{x}10^{-7}$	$8.5\mathrm{x}10^{-7}$	-6.59	-6.07	3.3
201	8.1	$5.9\mathrm{x}10^{-7}$	$1.3\mathrm{x}10^{-6}$	-6.23	-5.89	2.2
301	8.0	$7.8\mathrm{x}10^{-7}$	$2.5\mathrm{x}10^{-6}$	-6.11	-5.60	3.2
401	7.6	$1.3\mathrm{x}10^{-6}$	$2.2\mathrm{x}10^{-6}$	-5.89	-5.66	1.7
501	7.7	$1.6\mathrm{x}10^{-6}$	$2.8\mathrm{x}10^{-6}$	-5.80	-5.55	1.8
1001	7.7	$3.3\mathrm{x}10^{-6}$	$4.9\mathrm{x}10^{-6}$	-5.48	-5.31	1.4
4813	7.5	$1.4\mathrm{x}10^{-5}$	$3.2\mathrm{x}10^{-5}$	-4.85	-4.49	2.3
9190	7.3	$2.3\mathrm{x}10^{-5}$	$8.1\mathrm{x}10^{-5}$	-4.64	-4.09	3.5

Table F-2. Uranium sorption isotherm data for NFSS soil sample GC-1: effect of reducing pH to 3.0 prior to contact with uranium-spiked groundwater at pH 7.1 to 7.7

Initial	After contact					
U conc. (µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.7	$8.6x10^{-9}$	$8.9x10^{-8}$	-8.06	-7.05	$1.0x10^{1}$
11	7.4	$1.6x10^{-8}$	$1.6x10^{-7}$	-7.81	-6.80	$1.0x10^{1}$
21	7.6	$3.2x10^{-8}$	$2.9x10^{-7}$	-7.49	-6.53	$9.0x10^{0}$
31	7.6	$4.8x10^{-8}$	$4.3x10^{-7}$	-7.32	-6.37	$9.1x10^{0}$
41	7.6	$6.5x10^{-8}$	$5.5x10^{-7}$	-7.19	-6.26	$7.5x10^{0}$
50	7.6	$8.0x10^{-8}$	$6.7x10^{-7}$	-7.10	-6.17	$7.4x10^{0}$
101	7.5	$1.6x10^{-9}$	$1.3x10^{-6}$	-6.80	-5.87	$7.4x10^{0}$
		Initiation of probable precipitation				
195	7.5	$2.6x10^{-7}$	$2.8x10^{-6}$	-6.58	-5.55	$1.1x10^{1}$
294	7.4	$3.1x10^{-7}$	$4.7x10^{-6}$	-6.50	-5.33	$1.5x10^{1}$
392	7.4	$3.2x10^{-7}$	$6.7x10^{-6}$	-6.49	-5.17	$2.1x10^{1}$
486	7.4	$2.9x10^{-7}$	$9.0x10^{-6}$	-6.54	-5.05	$3.1x10^{1}$
979	7.1	$1.5x10^{-7}$	$2.0x10^{-5}$	-6.84	-4.69	$1.4x10^{2}$
4706	7.5	$3.7x10^{-8}$	$9.9x10^{-5}$	-7.43	-4.00	$2.7x10^{3}$
8989	7.4	$3.8x10^{-8}$	$1.8x10^{-5}$	-7.42	-3.75	$4.7x10^{3}$

Table F-3. Uranium(VI) sorption isotherm data for NFSS soil sample BCB-1

Initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.5	$1.1\text{x}10^{-8}$	$7.4\text{x}10^{-8}$	-7.96	-7.13	6.5
11	7.7	$2.2\text{x}10^{-8}$	$1.3\text{x}10^{-7}$	-7.66	-6.89	5.7
21	7.8	$4.7\text{x}10^{-8}$	$2.2\text{x}10^{-7}$	-7.33	-6.66	4.6
31	7.7	$7.3\text{x}10^{-8}$	$3.0\text{x}10^{-7}$	-7.14	-6.52	4.1
41	7.7	$9.7\text{x}10^{-7}$	$3.8\text{x}10^{-7}$	-7.01	-6.42	3.8
51	7.7	$1.2\text{x}10^{-7}$	$4.6\text{x}10^{-7}$	-6.92	-6.34	3.8
101	7.7	$2.7\text{x}10^{-7}$	$7.7\text{x}10^{-7}$	-6.57	-6.11	2.8
201	7.6	$5.6\text{x}10^{-7}$	$1.3\text{x}10^{-6}$	-6.25	-5.89	2.4
301	7.6	$9.0\text{x}10^{-7}$	$1.7\text{x}10^{-6}$	-6.05	-5.77	1.9
401	7.6	$1.2\text{x}10^{-6}$	$2.2\text{x}10^{-6}$	-5.92	-5.66	1.8
501	7.6	$1.5\text{x}10^{-6}$	$2.6\text{x}10^{-6}$	-5.82	-5.59	1.7
1001	7.3	$3.4\text{x}10^{-6}$	$3.6\text{x}10^{-6}$	-5.47	-5.44	1.1
4813	6.6	$1.8\text{x}10^{-5}$	$1.1\text{x}10^{-5}$	-4.74	-4.96	0.6
9190	6.4	$3.4\text{x}10^{-5}$	$2.0\text{x}10^{-5}$	-4.47	-4.70	0.6

Table F-4. Uranium(VI) sorption isotherm data for NFSS soil sample BCB-1: effect of reducing initial pH to 3.0 prior to contact with uranium-spiked groundwater at pH 7.5 to 8.2

Initial U conc. (μg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	8.0	$8.1\text{x}10^{-9}$	$9.2\text{x}10^{-8}$	-8.09	-7.04	$1.1\text{x}10^{1}$
11	8.2	$1.4\text{x}10^{-8}$	$1.7\text{x}10^{-7}$	-8.85	-6.77	$1.2\text{x}10^{1}$
21	8.2	$3.2\text{x}10^{-8}$	$3.0\text{x}10^{-7}$	-7.50	-6.53	$9.4\text{x}10^{0}$
31	8.2	$4.9\text{x}10^{-8}$	$4.3\text{x}10^{-7}$	-7.31	-6.37	$8.8\text{x}10^{0}$
41	8.2	$5.3\text{x}10^{-8}$	$6.0\text{x}10^{-7}$	-7.27	-6.22	$1.1\text{x}10^{1}$
50	8.2	$8.6\text{x}10^{-8}$	$6.4\text{x}10^{-7}$	-7.07	-6.19	$7.6\text{x}10^{0}$
101	8.1	$1.9\text{x}10^{-7}$	$1.2\text{x}10^{-6}$	-6.73	-5.92	$6.5\text{x}10^{0}$
195	8.1	$3.8\text{x}10^{-7}$	$2.3\text{x}10^{-6}$	-6.42	-5.65	$6.0\text{x}10^{0}$
294	8.0	$5.2\text{x}10^{-7}$	$3.6\text{x}10^{-6}$	-6.28	-5.44	$7.0\text{x}10^{0}$
		Initiation of probable precipitation				
392	7.94	$5.5\text{x}10^{-7}$	$5.6\text{x}10^{-6}$	-6.26	-5.25	$1.0\text{x}10^{1}$
486	7.85	$5.6\text{x}10^{-7}$	$7.6\text{x}10^{-6}$	-6.25	-5.12	$1.3\text{x}10^{1}$
979	7.54	$4.4\text{x}10^{-7}$	$1.9\text{x}10^{-5}$	-6.36	-4.73	$4.2\text{x}10^{1}$
4707	7.50	$2.3\text{x}10^{-8}$	$9.4\text{x}10^{-5}$	-7.64	-4.03	$4.1\text{x}10^{3}$
8989	7.60	$2.2\text{x}10^{-8}$	$2.1\text{x}10^{-4}$	-7.66	-3.67	$9.5\text{x}10^{2}$

Table F-5. Uranium(VI) sorption isotherm data for NFSS soil sample LC-1

Calculated initial U conc.	After contact					
(µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.8	1.2×10^{-8}	7.2×10^{-8}	-7.92	-7.14	6.1
11	7.7	2.3×10^{-8}	1.2×10^{-7}	-7.64	-6.92	5.3
21	7.8	4.8×10^{-8}	2.1×10^{-7}	-7.32	-6.68	4.4
31	7.8	7.5×10^{-8}	2.9×10^{-7}	-7.12	-6.54	3.9
41	7.8	9.8×10^{-8}	3.7×10^{-7}	-7.01	-6.43	3.8
51	7.8	1.3×10^{-7}	4.2×10^{-7}	-6.89	-6.38	3.3
101	7.8	2.8×10^{-7}	7.3×10^{-7}	-6.55	-6.14	2.6
201	7.8	5.8×10^{-7}	1.2×10^{-6}	-6.24	-5.92	2.1
301	7.8	9.1×10^{-7}	1.7×10^{-6}	-6.04	-5.77	1.8
401	7.7	1.2×10^{-6}	2.1×10^{-6}	-5.92	-5.68	1.7
501	7.7	1.5×10^{-6}	2.7×10^{-6}	-5.82	-5.57	1.8
1001	7.4	3.5×10^{-6}	3.4×10^{-6}	-5.46	-5.47	1.0
4813	6.8	1.8×10^{-5}	1.0×10^{-5}	-4.74	-5.00	0.6
9190	6.5	3.7×10^{-5}	5.6×10^{-5}	-4.43	-4.25	1.5

Table F-6. Uranium sorption isotherm data for NFSS soil sample LC-1: effect of initial reduction of pH to 3.0 prior to contact with uranium-spiked groundwater at pH 7.4 to 7.8

Initial U conc. (µg/mL)	After contact					
	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.4	$9.8x10^{-9}$	$8.3x10^{-8}$	-8.01	-7.08	8.5
11	7.5	$1.8x10^{-8}$	$1.5x10^{-7}$	-7.73	-6.83	7.9
21	7.5	$3.8x10^{-8}$	$2.6x10^{-7}$	-7.42	-6.58	6.9
31	7.6	$5.7x10^{-8}$	$3.9x10^{-7}$	-7.25	-6.41	6.8
41	7.7	$7.6x10^{-8}$	$4.9x10^{-7}$	-7.12	-6.31	6.4
50	7.8	$7.7x10^{-9}$	$6.9x10^{-7}$	-7.11	-6.16	8.9
101	7.8	$1.9x10^{-7}$	$1.2x10^{-6}$	-6.73	-5.92	6.4
195	7.8	$3.9x10^{-7}$	$2.2x10^{-6}$	-6.41	-5.66	5.5
294	7.8	$5.5x10^{-7}$	$3.5x10^{-6}$	-6.26	-5.46	6.4
		Initiation of probable precipitation				
392	7.8	$4.7x10^{-7}$	$6.0x10^{-6}$	-6.33	-5.22	$1.3x10^{1}$
486	7.8	$6.4x10^{-7}$	$7.2x10^{-6}$	-6.20	-5.14	$1.1x10^{1}$
979	7.7	$7.9x10^{-9}$	$1.7x10^{-5}$	-6.10	-4.77	$2.2x10^{1}$
4707	7.4	$2.3x10^{-7}$	$9.8x10^{-5}$	-6.63	-4.01	$4.3x10^{2}$
8989	7.5	$2.8x10^{-8}$	$1.8x10^{-5}$	-7.55	-3.74	$6.4x10^{2}$

Table F-7. Uranium(VI) sorption isotherm data for NFSS core sample from the 7.9-m (26-ft) level of BH-77

Calculated initial U conc.		After contact				
(µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	7.6	1.0×10^{-8}	9.0×10^{-8}	-8.00	-7.05	9.0
11	7.7	2.0×10^{-8}	1.3×10^{-7}	-7.70	-6.87	6.5
21	7.8	4.0×10^{-8}	2.5×10^{-7}	-7.40	-6.60	6.3
31	7.7	7.0×10^{-8}	3.2×10^{-7}	-7.15	-6.49	4.6
41	7.7	1.0×10^{-7}	3.7×10^{-7}	-7.00	-6.43	3.7
50	7.7	1.2×10^{-7}	4.7×10^{-7}	-6.92	-6.33	3.9
101	7.7	2.8×10^{-7}	7.5×10^{-7}	-6.55	-6.12	2.7
195	7.6	6.0×10^{-7}	1.24×10^{-6}	-6.22	-5.91	2.1
294	7.7	9.4×10^{-7}	1.67×10^{-6}	-6.03	-5.78	1.8
392	7.6	1.29×10^{-6}	2.01×10^{-6}	-5.89	-5.70	1.6
486	7.6	1.58×10^{-6}	2.69×10^{-6}	-5.80	-5.57	1.7
979	7.2	3.44×10^{-6}	3.93×10^{-6}	-5.46	-5.41	1.1
4707	6.8	1.67×10^{-5}	1.64×10^{-5}	-4.78	-4.79	1.0
8989	6.4	3.03×10^{-5}	4.12×10^{-5}	-4.52	-4.39	1.4

Table F-8. Uranium sorption isotherm data for NFSS core sample from the 7.9-m (26-ft) level of BH-77: effect of initial reduction of pH to 3.0 prior to contact with uranium-spiked groundwater at pH 7.0 to 8.1

Initial	After contact					
U conc. (µg/mL)	pH	Solution (mol/mL)	Adsorbed (mol/g)	Solution (log mol/mL)	Adsorbed (log mol/g)	Rs (L/kg)
6	8.1	7.1×10^{-9}	9.7×10^{-8}	-8.15	-7.01	1.4×10^{1}
11	7.8	1.4×10^{-8}	1.7×10^{-7}	-7.86	-6.78	1.2×10^{1}
21	8.0	2.9×10^{-8}	4.4×10^{-7}	-7.54	-6.35	1.5×10^{1}
31	8.1	4.6×10^{-8}	4.4×10^{-7}	-7.34	-6.36	9.6×10^{0}
41	8.1	6.1×10^{-8}	5.6×10^{-7}	-7.21	-6.25	9.2×10^{0}
50	8.1	8.0×10^{-8}	6.7×10^{-7}	-7.10	-6.17	8.5×10^{0}
101	8.0	1.8×10^{-7}	1.3×10^{-6}	-6.76	-5.90	7.2×10^{0}
195	7.9	3.4×10^{-7}	2.4×10^{-6}	-6.47	-5.61	7.1×10^{0}
294	7.9	4.3×10^{-7}	4.1×10^{-6}	-6.36	-5.39	9.4×10^{0}
		Initiation of probable precipitation				
392	7.8	4.6×10^{-7}	6.1×10^{-6}	-6.34	-5.22	1.3×10^{1}
486	7.7	4.5×10^{-7}	8.1×10^{-6}	-6.35	-5.09	1.8×10^{1}
979	7.4	3.0×10^{-7}	1.9×10^{-5}	-6.52	-4.71	6.5×10^{1}
4707	7.0	2.2×10^{-8}	9.5×10^{-5}	-7.66	-4.04	4.1×10^{3}
8989	7.0	5.7×10^{-8}	1.8×10^{-5}	-7.23	-3.76	3.2×10^{2}

Appendix G.

RADIUM SORPTION DATA FOR SOILS FROM THE NFSS

Table G-1. Radium sorption data for soil samples from the NFSS

Soil sample	Initial solution (pCi/mL)	After contact					
		pH	Solution (pCi/mL)	Adsorbed (pCi/g)	Solution (log pCi/mL)	Adsorbed (log pCi/g)	Rs (L/kg)
GC-1	1.0E5	8.0	2.3E2	3.6E5	2.36	5.56	1,600
GC-2	1.0E5	8.0	3.4E1	3.7E5	1.53	5.57	1,100
GC-3	1.0E5	8.0	6.3E1	4.2E5	1.80	5.62	6,700
BCB-1	1.0E5	8.0	1.2E2	4.2E5	2.08	5.62	3,500
BCB-2	1.0E5	8.0	2.4E1	4.2E5	1.38	5.62	18,000
BCB-3	1.0E5	8.0	3.4E1	3.9E5	1.53	5.59	12,000
LC-1	1.0E5	8.0	5.3E1	3.9E5	1.72	5.59	7,000

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