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Geochemical Information for Sites Contaminated with Low-Level Radioactive Wastes:
II – St. Louis Airport Storage Site

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GEOCHEMICAL INFORMATION FOR SITES CONTAMINATED WITH LOW-LEVEL RADIOACTIVE WASTES: II - ST. LOUIS AIRPORT STORAGE SITE

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PREFACE

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

SFMP Site	Report Number
Niagara Falls Storage Site	ORNL-6083

GEOCHEMICAL INFORMATION FOR SITES CONTAMINATED WITH LOW-LEVEL RADIOACTIVE WASTES: II - ST. LOUIS AIRPORT STORAGE SITE

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ABSTRACT

The St. Louis Airport Storage Site (SLASS) became radioactively contaminated as a result of wastes that were being stored from operations to recover uranium from pitchblende ores in the 1940s and 1950s. The U.S. Department of Energy is considering various remedial action options for the SLASS under the Formerly Utilized Site Remedial Action Program (FUSRAP). This report describes the results of geochemical investigations carried out at Oak Ridge National Laboratory (ORNL) to support the FUSRAP activities and to aid in quantifying various remedial action options. Soil and groundwater samples from the site were characterized, and sorption ratios for uranium and radium and apparent concentration limit values for uranium were measured in soil/groundwater systems by batch contact methodology.

Soil samples from the surface to bedrock were found to be typical silt-clay materials. Illite was the major clay identified by x-ray diffraction analysis. The uranium and radium concentrations were significantly above background near the old contaminated surface horizon (now at the 0.3- to 0.9-m depth); the maximum values were 1566 μg/g and 101 pCi/g, respectively. Below about the 6-m depth, the concentrations appeared to be typical of those naturally present in soils of this area (3.8 ± 1.2 μg/g and 3.1 ± 0.6 pCi/g).

Uranium sorption ratios showed stratigraphic trends but were generally moderate to high (100 to 1000 L/kg). The sorption isotherm suggested an apparent uranium concentration limit of about 200 mg/L. This relatively high solubility can probably be correlated with the carbonate content of the soil/groundwater systems; that is, a solubility of this magnitude is assumed to result from formation of the soluble uranyltricarbonate complex. The lower sorption ratio values obtained from the sorption isotherm may have resulted from changes in the experimental procedure or the groundwater used. The SLASS appears to exhibit generally favorable behavior for the retardation of uranium solubilized from waste in the site.

Parametric tests were conducted to estimate the sensitivity of uranium sorption and solubility to the pH and carbonate content of the system. The potential of several soil additives for improving uranium retention at the site was explored in a few

experiments. The very high sorption ratios (10,000 L/kg) obtained for fly ash from coal combustion indicate that this material could be a possible remedial action candidate.

Sorption tests with radium-spiked SLASS well water gave moderately high sorption ratios (800 to 940 L/kg). Thus, the SLASS may also exhibit favorable behavior for retarding radium solubilized from waste emplaced at the site.

1. EXECUTIVE SUMMARY

The St. Louis Airport Site (SLASS) is one of the radioactively contaminated sites being evaluated by the U.S. Department of Energy (DOE) Formerly Utilized Site Remedial Action Program (FUSRAP). 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. Under the auspices of the FUSRAP activities, DOE is considering various remedial action alternatives for these contaminated sites.

The SLASS was used during 1946-1966 for storing waste residues and scrap from other process facilities in the area around St. Louis, Missouri. In 1966 and 1967, most of the residues were removed to other locations (primarily the Latty Avenue site in nearby Hazelwood, Missouri, and the Niagara Falls Storage Site at Niagara Falls, New York). However, some contaminated material was reportedly buried at the SLASS and some surface contamination remained. The ownership of SLASS was subsequently transferred to the St. Louis Airport Authority.

Determination of geochemical information for the SLASS is one of the FUSRAP activities. This report describes the results of an investigation, at ORNL, of two geochemical aspects: (1) characterization of SLASS soils and groundwater, and (2) laboratory measurement of uranium and radium sorption behavior and uranium apparent concentration limit values in SLASS soil/groundwater systems. In this report, the apparent concentration limit is defined as that point on a sorption isotherm at which an abrupt change in slope occurs, as indicated by a sharp upward inversion of a normal isotherm, due to precipitation. This is differentiated from true thermodynamic solubility since equilibrium may not be established in these tests. The resulting information, particularly the radioactive element sorption ratios and the solubility data, are needed for the Environmental Impact Statement (EIS) in order to quantify radioactive element mobility predictions for various remedial action options.

1.1 SOIL CHARACTERIZATION

Three complete core profiles (four cores) from the surface to the limestone bedrock were obtained from the SLASS. The properties of the soil samples taken from various depths for these profiles were found to be similar; all were characteristic of silt-clay soils. X-ray diffraction analysis showed primarily patterns for illite. Elemental analysis confirmed the presence of typical clay elements (Si, Al, Fe, etc.), and only minor differences in trace element concentrations were observed.

Profiles of the ^{238}U and ^{226}Ra concentrations were determined for the three core locations. Uranium concentrations were significantly above background at depths of 0.3 to 2 m; the highest value measured was 1566 μg/g. The average concentration at core depths of greater than 6 m was 3.8 ± 1.3 μg/g, which is probably the natural uranium content of soils in this area. Maximum radium concentrations were also observed at the 0.3- to 2-m depths; the highest value measured was 101 pCi Ra/g, while the mean concentration below 6 m was 2.8 ± 0.5 pCi/g. Both the uranium and the radium data suggest that the old contaminated site horizon is now at a depth of approximately 0.3 to 0.9 m. Uranium concentrations significantly above background extended to somewhat greater depths than those for radium and thus may be evidence for greater mobility of uranium than radium at the SLASS.

1.2 GROUNDWATER CHARACTERIZATION

Anionic and elemental analyses of samples of groundwater obtained from two shallow wells at the SLASS showed considerable differences in the concentrations of dissolved components. Such results suggest that these wells could be sampling different minor perched water tables. The pH of each groundwater sample was 7.4. The major cationic and anionic species present were calcium/magnesium/sodium and bicarbonate/sulfate/chloride/nitrate, in order of decreasing concentration.

1.3 EXPERIMENTAL METHODOLOGY

Batch contact methodology was used to measure sorption ratios and to construct a sorption isotherm for uranium in site soil/groundwater systems. Both synthetic groundwater and well water (from well D) were used in the tests; the core soil samples were taken from various depths. Soil and uranium-traced groundwater were contacted at a solution/solids ratio of 10 for 10 to 20 h. The solution was then recovered by centrifugation for analysis by alpha-counting techniques. The sorption ratio was calculated from the uranium concentration of the solution, and the sorption isotherm was constructed by plotting the concentration in solution vs the concentration on the solids. The sorption isotherm was extended to high uranium concentrations in order to measure the apparent concentration limit value.

A few batch contact tests were also carried out to obtain radium sorption ratios. Only one radium concentration was used. Radium measurements were made by allowing the samples to establish secular equilibrium and then counting the 0.609-MeV ^{214}Bi gamma peak.

1.4 SUMMARY OF URANIUM SORPTION AND APPARENT CONCENTRATION LIMIT INFORMATION

Uranium sorption ratio profiles were completed for the soil cores using spiked synthetic groundwater. Similar results were obtained for each core. The uranium sorption ratios showed stratigraphic differences. Low to moderate sorption ratios (10 to 100 L/kg) were measured for the near-surface samples, and moderate to high sorption ratios (100 to >1000 L/kg) were obtained with the 6- to 12-m samples. Lower sorption ratios were generally obtained near the 15-m depth, while moderate to high sorption ratios were again measured for samples taken below the 15-m depth.

An apparent uranium concentration limit of approximately 200 mg/L was estimated from a sorption isotherm plot using soil from one core sample and spiked groundwater from well D. This relatively high solubility is probably consistent with the carbonate content of the groundwater since elevated solubility levels were presumed to result from formation of the soluble uranyltricarbonate complex. The sorption isotherm yielded much lower uranium sorption ratios (1 to 38 L/kg) than those measured with synthetic groundwater. The reason for this difference in sorption behavior was not explored further due to completion of project support for this geochemical activity. It could have resulted from changes in the test procedure or the groundwater used.

In general, the SLASS site appears to have favorable properties for retarding any uranium that is solubilized from waste in the soil. However, additional work would be needed in order to quantify the uranium retardation behavior more accurately.

A parametric investigation of uranium sorption on typical clay materials, and the resulting apparent uranium concentration limit, was undertaken to determine the influence of pH and carbonate concentration. The results could generally be correlated with existing information on uranium speciation. Maximum sorption was observed near neutral pH. Increased solubility was observed in the presence of additional CO_3^{2-}. Such information may be useful for estimating the sensitivity of site performance to changes in various geochemical parameters.

A few tests were performed to evaluate several soil additives for possible use in improving uranium retardation at the site. The results showed that fly ash from coal combustion gives much higher uranium sorption ratios (10,000 L/kg) than does SLASS soil or standard clays. Thus, fly ash is a potential candidate for remedial action purposes.

1.5 RADIUM SORPTION INFORMATION

A few batch contact tests were completed to explore radium sorption behavior with SLASS soil samples. The moderate sorption ratios obtained (880 to 940 L/kg) suggest that the SLASS site could provide significant retardation of any radium which may become solubilized from waste currently present at the site.

2. INTRODUCTION

SLASS, which is located approximately 15 miles northwest of downtown St. Louis, Missouri, lies directly north of the Lambert—St. Louis International Airport. This 21.7-acre site is bounded by Brown Road on the north and east, the Norfork and Western Railroad on the south, and Coldwater Creek and adjacent McDonnell-Douglas Corporation property on the west. A map of the area is presented in Fig. 1. Only the details pertinent to the location of boreholes and/or groundwater sampling wells employed to obtain samples used in the study reported in this document are included in the map.

During the period between 1946 and 1966, this site was used for storing waste residues from the processing of natural uranium and some other activities involving radioactive materials. In addition to the storage of leached pitchblende ore and uranium waste residues, approximately 60 truckloads of contaminated scrap metal and a contaminated vehicle were reportedly buried at the site. Some of the stored residues were sold for their mineral content during 1966 and 1967. At that time, some of the residues were removed to the Latty Avenue site in nearby Hazelwood, and ownership of the site was then transferred to the Airport Authority. In accordance with the terms of the transfer agreement, the barium sulfate residue (which contained the radium recovered during the uranium processing) was removed from the site and transported to the NFSS, at Niagara Falls, New York. These activities and other details of the site's radiological and remedial action history are described in reports and documents prepared by ORNL and Roy F. Weston, Inc.[1-3]

The philosophy, or approach, used in the experimental work reported here has been developed by taking into consideration the technical and theoretical aspects of reported geochemical sorption processes and apparent solubility limit conditions that might be encountered in the near-surface disposal of low-level radioactive wastes.[4] Programmatic information and funding needs were major controlling factors in the collection of the data described in this report. Two separate, relatively short-term funding periods separated by approximately 2 years of program inactivity caused a significant discontinuity in the collection and interpretation of the geochemical information reported here.

Since the use of sorption distribution coefficient (Kd) values infers thermodynamic equilibrium, we have chosen to report radionuclide sorption data as sorption ratio (Rs) values, which are calculated as empirical ratios. The Rs value is simply the ratio of the concentration of the

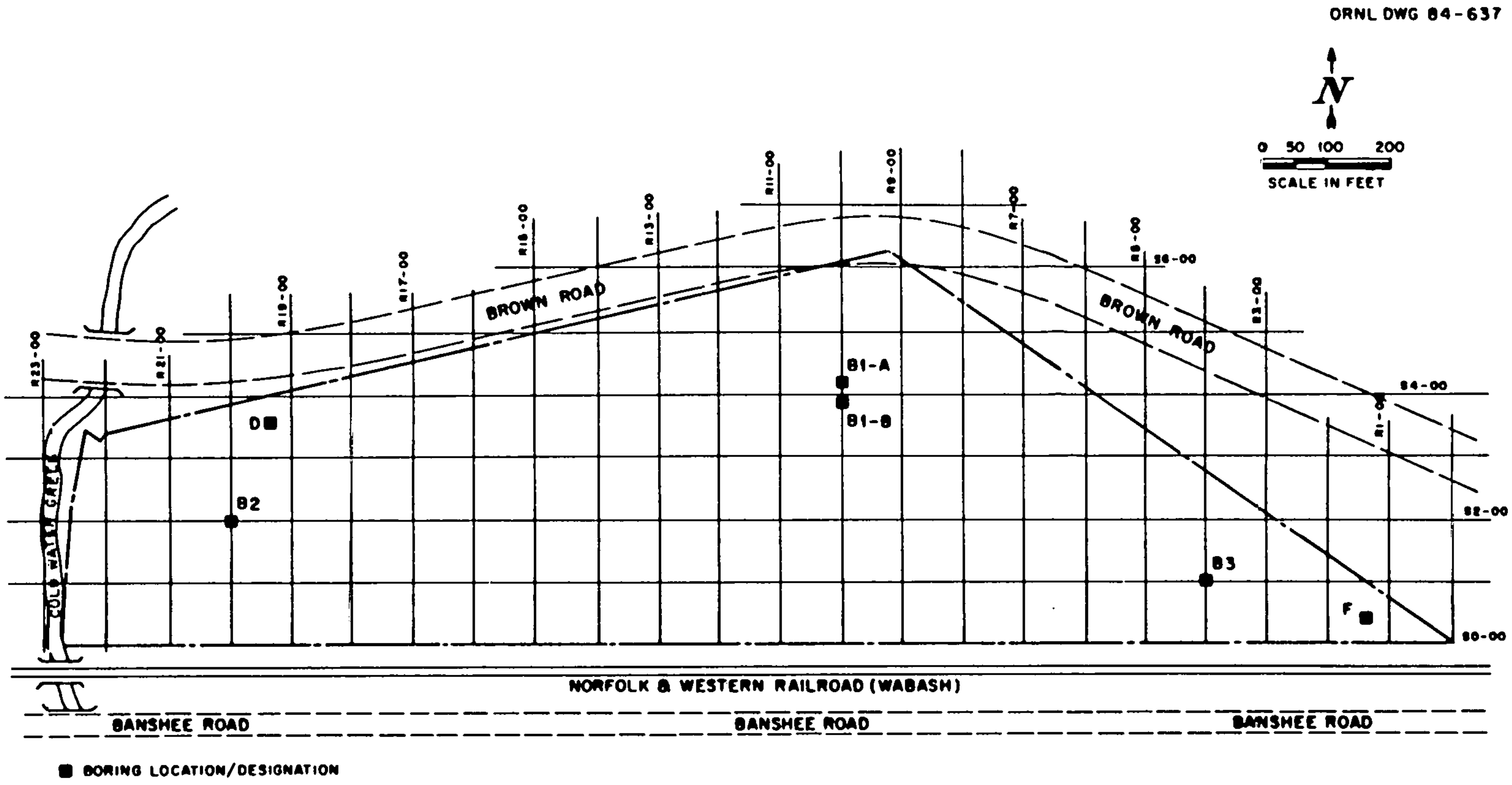

Fig. 1. Pertinent surface features of the St. Louis Airport Storage Site.

species on the solid adsorbent divided by the concentration of the solute in the solution after contact. Since volume is being divided by mass in the calculation of the sorption ratio, the Rs units are liters per kilogram (L/kg).

The information provided in this report is designed to characterize the soil at the SLASS with respect to its current radionuclide burden and also to determine its ability to retain these radionuclides in the event that they should become solubilized. The effects of pH and carbonate concentration are discussed, and comparisons of the site soil with standard generic clays are made. The apparent concentration limit, or saturated solution concentration, is inferred from sorption isotherms.

3. SITE CHARACTERIZATION

3.1 SOIL CHARACTERIZATION

3.1.1 Material Preparation

In the initial phase of this investigation, four cores from the SLASS area were provided by a subcontractor, Roy F. Weston, Inc. These cores (location shown in Fig. 1) comprised complete sequences from the surface to limestone bedrock (ranging from 14 to 27 m) for three locations. Two of the cores, B1-A and B1-B, were taken approximately 3 m apart. The first 2.7 m of core B1-A was somehow lost; therefore, the second core, B1-B, was drilled to supply the first 3-m and additional samples from the 15- to 21-m levels. The cores were received in 76.2-cm Shelby tubes that had been separated into halves at the site by sawing longitudinally. The tubes were sealed with wax and tape and then numbered sequentially. After the seal had been removed and the split tubes opened, the core material, which usually consisted of about 60 cm of compacted soil, was sectioned into approximately 15-cm samples and placed in jars. One portion was dried, while the other was stored in an as-received condition in a sealed jar. Drying was accomplished either by heating at 110°C or by lyophilization. The dried samples were lightly ground in a Braun mill to -60 mesh prior to further testing. No differences in clay sorption characteristics were noted between the two methods of drying.

The arithmetic mean values for the loss on drying for the core samples were: B1-A, 20 ± 2%; B1-B, 17 ± 5%; B2, 17 ± 7%, and B3, 15 ± 4%.

3.1.2 Core Profile Analyses

Profile analyses for ^{238}U and ^{226}Ra are shown for the four cores in Tables 1-4. The ^{238}U values were determined by the ORNL Analytical Chemistry Division via delayed neutron activation analysis of the ^{235}U isotopic content. Since only normal-uranium wastes were stored at the site, these values could be related directly to the ^{238}U contents of the soil samples. Uranium values significantly above background levels were obtained at depths of about 0.3 to 2 m in different cores and probably represent residual contamination of the prefill surface. The highest value seen, 1566 mg/kg at 1.07 m in core B3, was probably due to the presence of a single chunk of "hot" concrete observed in the 15-cm sample taken at this level. The average ^{238}U concentration at core depths of greater than 6.1 m is 3.8 ± 1.2 mg/kg, which is probably the natural background concentration for soil in this area.[1]

The ^{226}Ra contents of selected samples from the four cores were analyzed by sealing 5-g samples of the dried and ground material into counting tubes, waiting 30 d to ensure secular equilibrium in the portion of the ^{238}U decay chain between ^{226}Ra and ^{214}Bi (the 3.8-d half-life of ^{222}Rn is the limiting factor), and then determining the ^{226}Ra content by integrating the gamma activity of the 0.609-MeV ^{214}Bi peak between the 0.55- and 0.67-MeV energy levels during 100-min counting periods. The net arithmetic mean and standard deviation were calculated from three replicate samples and three blanks. Counter efficiency calibrations for ^{226}Ra were made by frequent counting of New Brunswick Laboratories (NBL) uranium standards in which the entire ^{238}U decay chain was in secular equilibrium. Additional details of this analytical technique are given in Appendix A.

The radionuclide profiles suggest that uranium may be more mobile than radium and may be penetrating the soil at the site to greater depths. Both the uranium and the radium data (Tables 1-4) indicate that the contaminated prefill horizon is at a depth of approximately 0.3 to 0.9 m. This agrees well with the historical record that reports that 0.3 to 0.9 m of clean fill was placed on the site in 1970 following the cleanup during

Table 1. Radionuclide characterization of soil samples from core B1-A from the St. Louis Airport Storage Site

Sample	Core depth (m)	Soil description and unit[a]	^{238}U[b] (µg/g)	^{226}Ra[c] (pCi/g)
S1-1A	2.74[d]	Brown clay-silt loam, unit 3	27.7	63.3
S1-2A	2.89	Brown clay-silt loam, unit 3	9.4	3.1
S1-3A	3.05	Brown clay-silt loam, unit 3	14.0	2.9
S2-1A	3.36	Brown clay-silt loam, unit 3	19.6	12.3
S2-2A	3.51	Brown clay-silt loam, unit 3	17.5	4.1
S2-3A	3.75	Gray/brown silty clay, unit 3	11.4	2.7
S3-1A	3.96	Gray/brown silty clay, unit 3	12.9	7.9
S3-2A	4.12	Gray/brown silty clay, unit 3	8.3	2.5
S4-1A	4.57	Gray/brown silty clay, unit 3	7.5	3.6
S5-1A	5.18	Gray/brown silty clay, unit 3	5.7	4.1
S7-1A	6.40	Gray/brown silty clay, unit 3	5.8	2.3
S9-1A	7.62	Gray silty clay/organic material, unit 4	4.2	2.6
S11-1A	8.84	Brown/gray silty clay, unit 4	3.0	2.6
S12-2A	9.60	Brown/gray silty clay, unit 4	2.8	2.3
S15-1A	11.58	Gray silty clay, unit 4	2.7	2.8
S20-1A	14.63	Blue/gray plastic clay, unit 5	6.0	3.6
S23-4A	17.37	Moist olive/gray silt, unit 6	2.5	2.5

[a] From *Formerly Utilized Sites Remedial Action Program: St. Louis Airport Storage Site (SLAPSS)*, Draft Report, Appendix D, Roy F. Weston, Inc., West Chester, Pa., June 1981.

[b] Average standard deviation, ±3%.

[c] Average standard deviation, ±7%.

[d] No samples were taken at levels above 2.7 m.

Table 2. Radionuclide characterization of soil samples from core B1-B from the St. Louis Airport Storage Site

Sample	Core depth (m)	Soil description and unit[a]	^{238}U[b] (μg/g)	^{226}Ra[c] (pCi/g)
S24-1A	0.31	Soil fill/concrete rubble, unit 1	2.8	3.1
S24-2A	0.61	Soil fill/concrete rubble, unit 1	3.5	2.7
S25-1A	0.91	Brown silt loam, unit 2	164.0	101.0
S25-2A	1.22	Brown silt loam, unit 2	96.6	4.8
S26-1A	1.52	Brown silt loam, unit 2	175.0	42.3
S26-2A	1.74	Brown silt loam, unit 2	107.0	22.4
S26-3A	1.92	Brown clay/silt loam, unit 3	17.4	3.4
S27-1A	2.13	Brown clay/silt loam, unit 3	9.6	3.1
S27-2A	2.35	Brown clay/silt loam, unit 3	9.1	3.6
S27-3A	2.53	Brown clay/silt loam, unit 3	6.9	2.9
S28-1A	2.74	Brown clay/silt loam, unit 3	6.1	3.1
S28-2A	2.90	Brown clay/silt loam, unit 3	4.8	2.3
S28-3A	3.05	Brown clay/silt loam, unit 3	4.4	2.3
S28-4A	3.20	Brown clay/silt loam, unit 3	6.0	2.7
S29-1A	15.24[d]	Blue/gray plastic clay, unit 5	3.9	3.7
S30-4A	16.31	Moist olive/gray silt, unit 6	3.0	2.3
S35-1A	18.59	Moist olive/gray silt, unit 6	2.9	3.1
S38-2A	21.03	Residuum in blue/gray clay, unit 8	5.4	3.7

[a] From *Formerly Utilized Sites Remedial Action Program: St. Louis Airport Storage Site (SLAPSS)*, Draft Report, Appendix D, Roy F. Weston, Inc., West Chester, Pa., June 1981.

[b] Average standard deviation, ±3%.

[c] Average standard deviation, ±7%.

[d] No samples were taken at depths between 3 and 15 m.

Table 3. Radionuclide characterization of soil samples from core B2 from the St. Louis Airport Storage Site

Sample	Core depth (m)	Soil description and unit[a]	^{238}U[b] (μg/g)	^{226}Ra[c] (pCi/g)
S1-1A	0.0	Soil fill, unit 1	5.1	2.7
S1-2A	0.15	Soil fill, unit 1	5.7	2.5
S1-3A	0.31	Soil fill, unit 1	578.0	94.2
S3-1A	1.83[d]	Building material, misc. fill, unit 1	228.0	27.1
S3-2A	1.98	Brown silt loam, unit 2	240.0	6.3
S3-3A	2.13	Brown silt loam, unit 2	249.0	6.1
S3-4A	2.29	Brown silt loam, unit 2	104.0	5.9
S4-1A	2.44	Gray/brown silt loam, unit 3	151.0	5.4
S4-2A	2.59	Gray/brown silt loam, unit 3	59.5	4.0
S4-3A	2.74	Gray/brown silt loam, unit 3	17.8	3.0
S5-1A	3.05	Gray/brown silt loam, unit 3	25.8	4.1
S5-2A	3.14	Gray/brown silt loam, unit 3	11.5	2.8
S6-1A	3.35	Gray/brown silt loam, unit 3	21.3	3.5
S7-1A	4.42	Light-gray silt loam, unit 3	55.0	3.7
S8-1A	5.18	Light-gray silt loam, unit 3	27.3	3.1
S9-1A	5.94	Light-gray silt loam, unit 3	7.7	2.0
S10-1A	6.71	Dark-gray silty clay/organic, unit 4	6.3	2.4
S15-1A	10.52	Blue/gray silty clay, unit 4	3.2	2.8
S20-1A	14.33	Very plastic blue/gray clay, unit 5	3.0	3.1
S25-1A	18.14	Very plastic blue/gray clay, unit 5	3.9	3.5
S30-1A	21.95	Moist brown/gray silt, unit 6	2.7	2.6
S34-4A	25.45	Residuum, unit 8	4.7	3.1

[a] From Formerly Utilized Sites Remedial Action Program: St. Louis Airport Storage Site (SLAPSS), Draft Report, Appendix D, Roy F. Weston, Inc., West Chester, Pa., June 1981.

[b] Average standard deviation, ±3%.

[c] Average standard deviation, ±7%.

[d] No samples obtained at depths between 7 and 10.5 m.

Table 4. Radionuclide characterization of soil samples from core B3 from the St. Louis Airport Storage Site

Sample	Core depth (m)	Soil description and unit[a]	^{238}U[b] (μg/g)	^{226}Ra[c] (pCi/g)
S1-1A	0.0	Soil fill, unit 1	6.5	3.1
S2-1A	0.76	Soil fill, unit 1	26.2	3.4
S2-3A	0.91	Brown silt loam, unit 2	125.0	6.5
S2-4A	1.07	Brown silt loam, unit 2	1566.0	71.9
S3-1A	1.22	Brown silt loam, unit 2	193.0	9.3
S3-2A	1.37	Brown silt loam, unit 2	164.0	5.0
S3-3A	1.52	Brown silt loam, unit 2	167.0	4.4
S3-4A	1.68	Brown silt loam, unit 2	168.0	4.0
S4-1A	1.83	Brown silt loam, unit 2	95.9	5.0
S5-1A	2.44	Gray/brown silt loam, unit 3	37.4	3.6
S6-1A	3.20	Gray/brown silt loam, unit 3	6.6	2.9
S8-1A	4.72	Gray/brown silt loam, unit 3	3.4	2.1
S10-1A	6.25	Dark-brown silty clay/organics, unit 4	3.2	2.4
S15-1A	10.06	Black/gray plastic clay, unit 5	3.8	2.0
S20-2A	13.87	Residuum, unit 8	3.0	3.2

[a] From Formerly Utilized Sites Remedial Action Program: St. Louis Airport Storage Site (SLAPSS), Draft Report, Appendix D, Roy F. Weston, Inc., West Chester, Pa., June 1981.

[b] Average standard deviation, ±3%.

[c] Average standard deviation, ±7%.

1966-1969. In samples taken below the old prefill surface, the radium content rapidly decreased to background values of 2 to 4 pCi/g. However, uranium concentrations significantly above background can be seen to approximately 3 to 4.6 m in depth, suggesting downward migration during the site history. Surface water is in equilibrium with the CO_2 content of the atmosphere and thus would be expected to contain some bicarbonate anions (at higher groundwater pH values, carbonate may also be present). For this reason, it seems plausible to suggest that mobile $UO_2(CO_3)_3^{4-}$ anions are formed due to the downward percolation of surface water. Radium does not form similar carbonate complexes; therefore, it would not be expected to migrate as an anionic complex species under these conditions.

It is interesting to note that the 2.7- to 3.4-m-depth sections are quite dissimilar, even though cores B1-A and B1-B were drilled close together (about 3 m apart). Both ^{238}U and ^{226}Ra were essentially background at this level in core B1-B, while significant concentrations of these nuclides were found at corresponding levels in B1-A. There are indications that the first sample in several core sections may have been contaminated by "knockdown" during the drilling operations. Even if knockdown was not experienced at this particular location, it is apparent that little lateral migration of uranium has occurred.

3.1.3 Physicochemical Characterization of the Core Material

Selected samples from all four cores, representing the entire sequence from surface to bedrock, were analyzed by the Technical Services Division, Microanalysis Group, at the Oak Ridge Gaseous Diffusion (ORGDP) facility by three techniques: x-ray diffraction (XRD), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). All of the XRD patterns were similar and in agreement with the standard pattern for illite, except for a few additional diffraction lines attributed to montmorillonite and quartz. These results were confirmed by using SEM/XRF techniques, which showed only small differences in the concentrations of several of the trace elements between core samples.

Although the TEM method is not as positive as XRD for determining various types of clays, it is capable of making a distinction between some of the major types. Based on a comparison with known standards, the TEM analysis was in agreement with both the XRD and the SEM results. TEM micrographs, considered to be representative of the sample matrix, showed a trend toward smaller particle size with increase in depth of the core sample.

These findings indicate that the large volume of clay soil underlying the site is relatively uniform in its mineralogical makeup.

3.2 GROUNDWATER CHARACTERIZATION

Groundwater used in the initial phase of the investigation of the sorption characteristics of SLASS soil was prepared by equilibrating the soil with distilled water prior to adding the desired radionuclide. Although this type of groundwater was not analyzed, previous experience has shown that synthetic groundwater prepared in this manner usually varies only to a slight degree from that obtained from site wells.

In the second phase of this investigation, groundwater samples from two wells at the SLASS, D and F (see Fig. 1), were provided by Bechtel National. Anionic concentrations of duplicate samples of groundwater from each well are shown in Table 5. The concentration levels of the anions that were present in significant amounts (HCO_3^-, SO_4^{2-}, NO_3^-, and Cl^-) were considerably lower in Well F groundwater than in Well D groundwater. Since both wells were shallow, these differences suggest that separate minor perched water tables may have been sampled. Groundwater from Well D was used in the uranium and radium sorption investigations reported in Sect. 4.

Table 6 summarizes the elemental concentrations of the groundwater samples from Wells D and F, which were analyzed by inductively coupled plasma (ICP) spectrometric techniques. Four elements were present in significant concentrations: Ca, Mg, Na, and Si. Good agreement was obtained in the duplicate samples; and, consistent with the anion analyses, the concentration levels in Well D were higher than in Well F. Significant concentrations of calcium and magnesium were expected because of the presence of bicarbonate.

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Table 5. Anionic concentrations of groundwater samples from the St. Louis Airport Storage Site

Analysis[a]	Well No. D-1[b]	D-2[b]	F-1[c]	F-2[c]
pH	7.3	7.5	7.4	7.4
HCO_3^-	842	747	145	145
CO_3^{2-}	0	0	0	0
SO_4^{2-}	277	276	144	144
NO_3^-	118	123	<4	<4
NO_2^-	<2	<2	<2	<2
F^-	1	1	<1	<1
Cl^-	174	160	24	24
Br^-	<5	<5	<5	<5
PO_4^{3-}	<4	<4	<4	<4

[a] All analyses, except pH, are given in mg/L.

[b] Location of Well D: S3-60, R18-20.

[c] Location of Well F: S0-40, R1-40.

Table 6. Elemental analysis of groundwater from the St. Louis Airport Storage Site

	Well No.			
Analysis[a]	D-1[b]	D-2[b]	F-1[c]	F-2[c]
Ag	<0.11	<0.11	<0.02	<0.02
Al	<0.35	<0.35	<0.06	<0.06
B	0.59	<0.05	<0.08	<0.08
Ba	<0.03	0.03	0.06	0.06
Be	<0.008	<0.008	<0.0013	<0.0013
Ca	88.	96.	60.	60.
Cd	<0.06	<0.06	<0.009	<0.009
Co	<0.08	<0.08	<0.013	<0.013
Cr	<0.14	<<0.14	<0.023	<0.023
Cu	<0.19	<0.19	<0.032	<0.032
Fe	<0.12	<0.12	<0.02	<0.02
Ga	<0.42	<0.42	<0.07	<0.07
Hf	<0.24	<0.24	<0.043	<0.043
K	<3.9	<3.9	<0.65	<0.65
Mg	110.	110.	30.	30.
Mn	0.014	0.014	0.0018	0.0021
Mo	<0.16	<0.16	<0.027	<0.027
Na	130.	130.	22.	22.
Ni	<0.66	<0.66	<0.11	<0.11
Pb	<1.6	<1.6	<0.26	<0.26
Sb	<0.84	<0.84	<0.14	<0.14
Se	<2.6	<2.6	<0.43	<0.43
Si	6.5	6.7	4.6	4.6
Sr	0.68	0.73	0.11	0.11
Ti	0.027	<0.025	0.0041	0.0043
V	<0.09	<0.09	<0.015	<0.015
Zn	<0.11	<0.11	<0.018	<0.018
Zr	<0.11	<0.11	<0.018	<0.018

[a] All analyses are given in mg/L.

[b] Location of Well D: S3-60, R18-20.

[c] Location of Well F: S0-40, R1-40.

4. RADIONUCLIDE SORPTION MEASUREMENTS - SOIL/GROUNDWATER SYSTEMS

4.1 URANIUM SORPTION CHARACTERISTICS OF CORE SOIL

During the initial phase of this investigation, results were obtained from a series of experiments designed to determine the relative uranium mobility/adsorption characteristics of the SLASS core material. These data are shown graphically in Figs. 2-5. In these experiments, soil samples from selected levels of the four cores were contacted with an unbuffered uranyl sulfate solution which had a uranium concentration of 10 mg/L and was spiked with ^{233}U. Each sample was first contacted with distilled water at a solution/solids ratio of 10 for a period of 20 h to form a synthetic groundwater. Then uranium was added from a stock uranyl sulfate solution having a pH of 4.5 to ensure the presence of dissolved cationic uranyl species upon initial contact with the soil samples. The concentration of uranium in the stock solution was such that only 0.1 mL had to be added to the 10 mL of synthetic groundwater to obtain the desired contact concentration and count rate. The sorption test was made at the same solution/solids ratio (10) and contact time (20 h) as had been used in the preparation of the synthetic groundwater. The uranium concentrations, from which these data were calculated, were determined by alpha liquid scintillation spectrometric counting of the ^{233}U-spiked ^{238}U solutions before and after contact with the core material. In general, the uranium sorption ratio values for the core samples showed considerable variation: low to moderate (Rs = 10 to 100 L/kg) for the near-surface soil (<6-m) samples, moderate to high (Rs = 100 to greater than 1000 L/kg) for the 6- to 12-m levels, low at the 15-m level, and moderate to high at depths greater than 15 m. The data are summarized in Tables B-1 through B-4, Appendix B. These four cores are considered to be representative of the entire site.

It should be emphasized that no attempt was made to adjust the contact pH to a constant value; the solution was allowed to adjust to the natural pH of the soil with which it was in contact. Although the final pH of the contact solutions varied only from pH 6.6 to 8.0, this variation might account for some of the observed differences in the sorption ratio values. The role that pH variations may play will be discussed in more

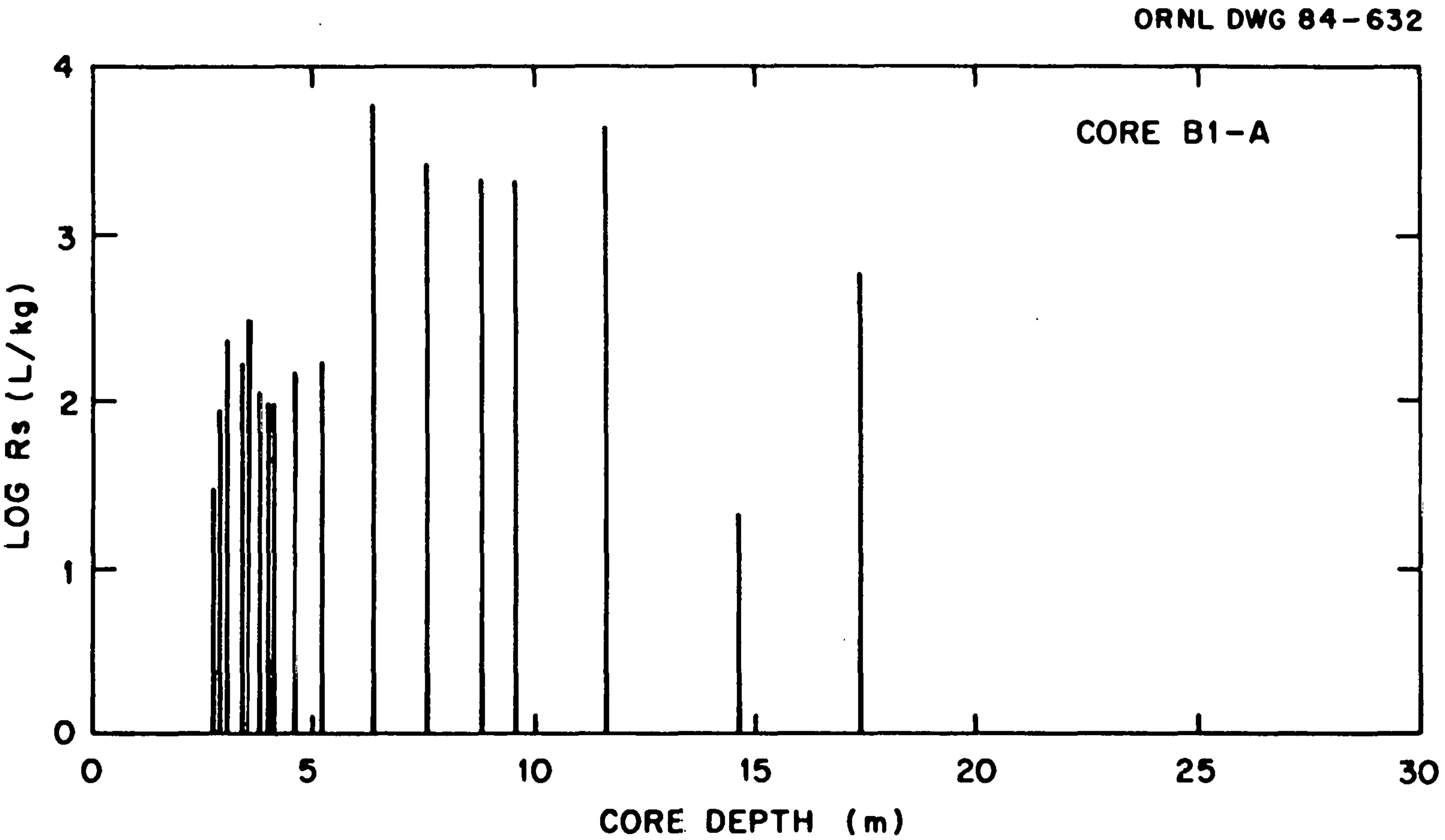

Fig. 2. Uranium sorption profile for SLASS core B1-A.

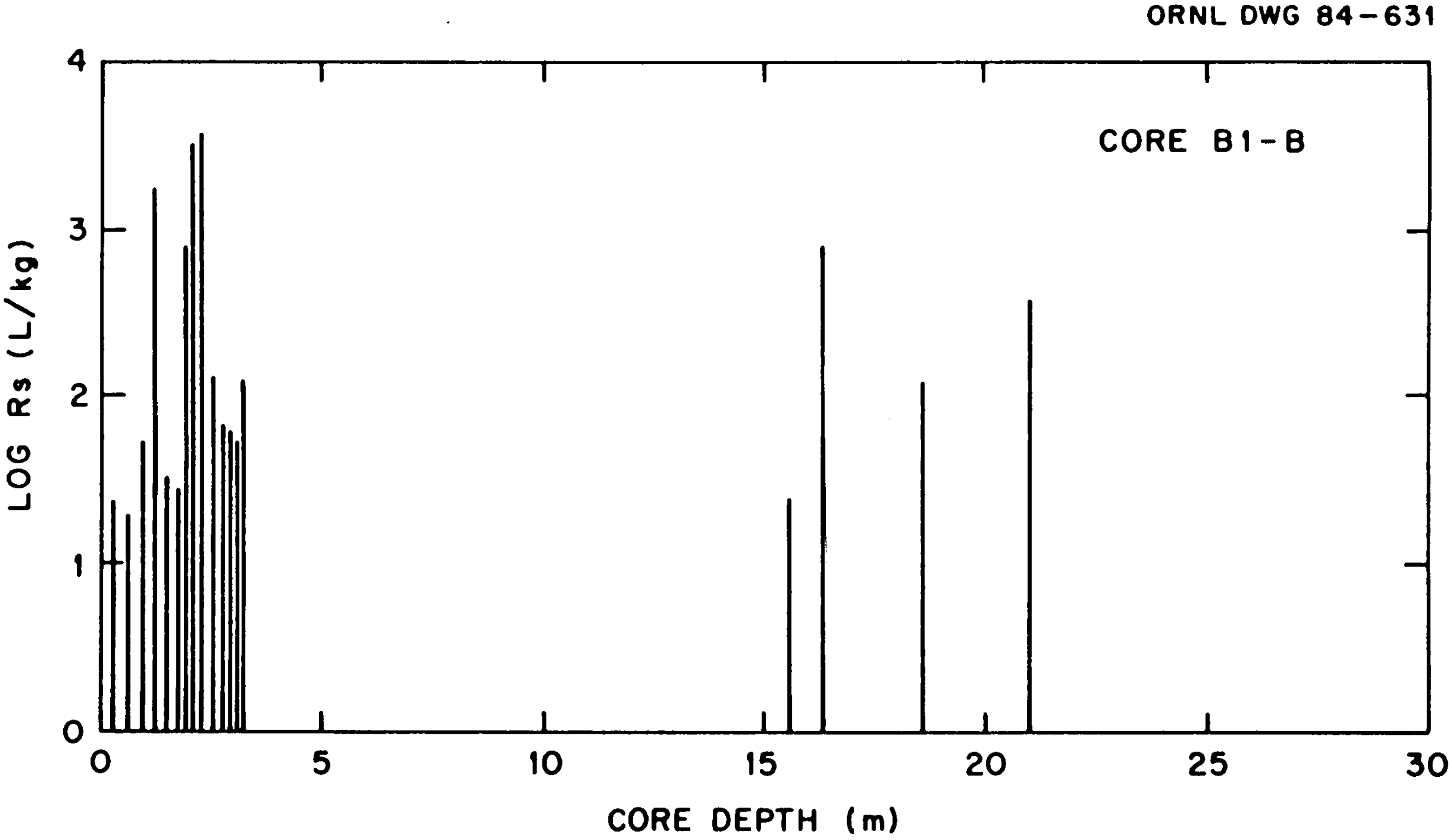

Fig. 3. Uranium sorption profile for SLASS core B1-B.

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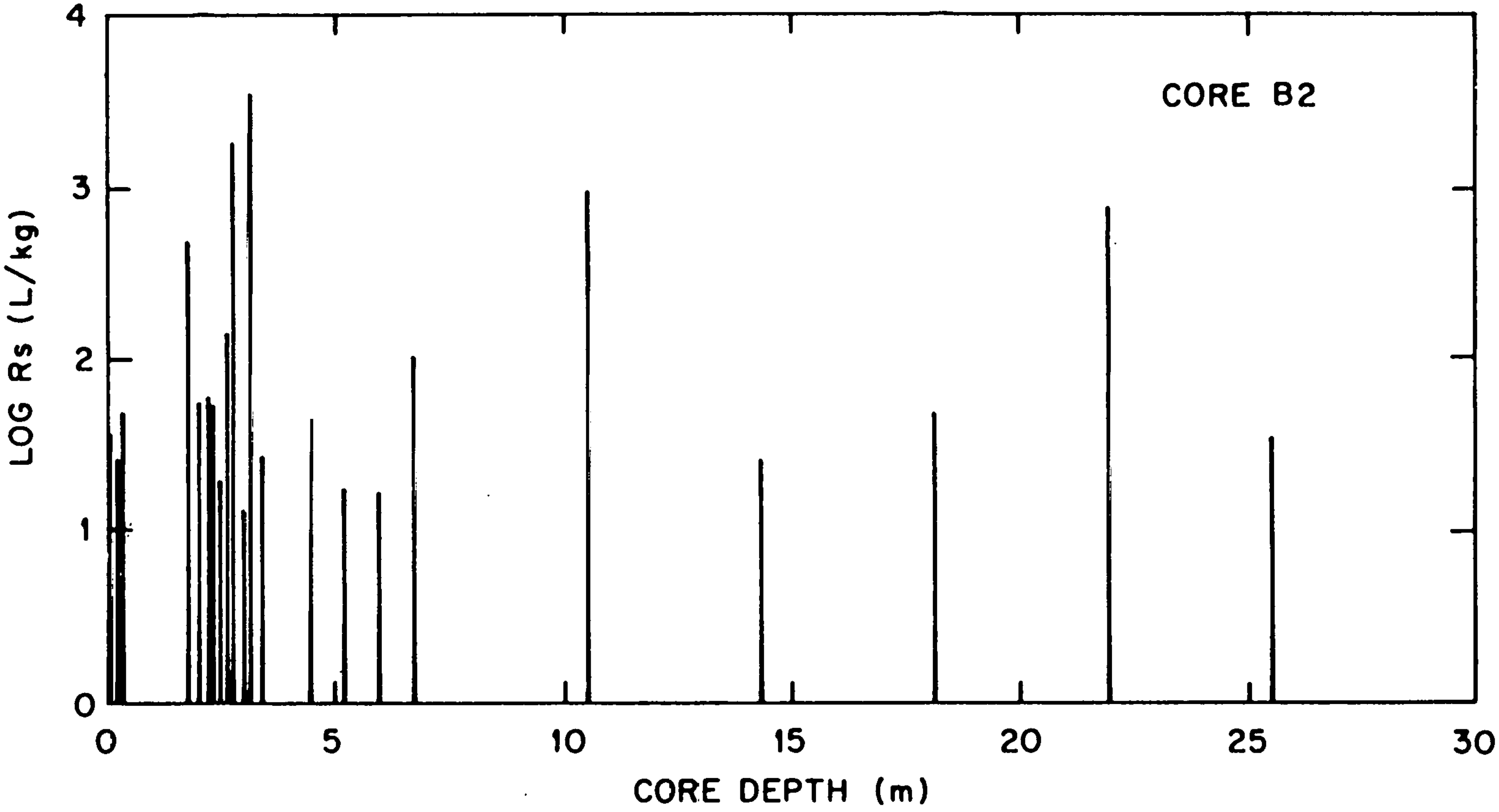

Fig. 4. Uranium sorption profile for SLASS core B2.

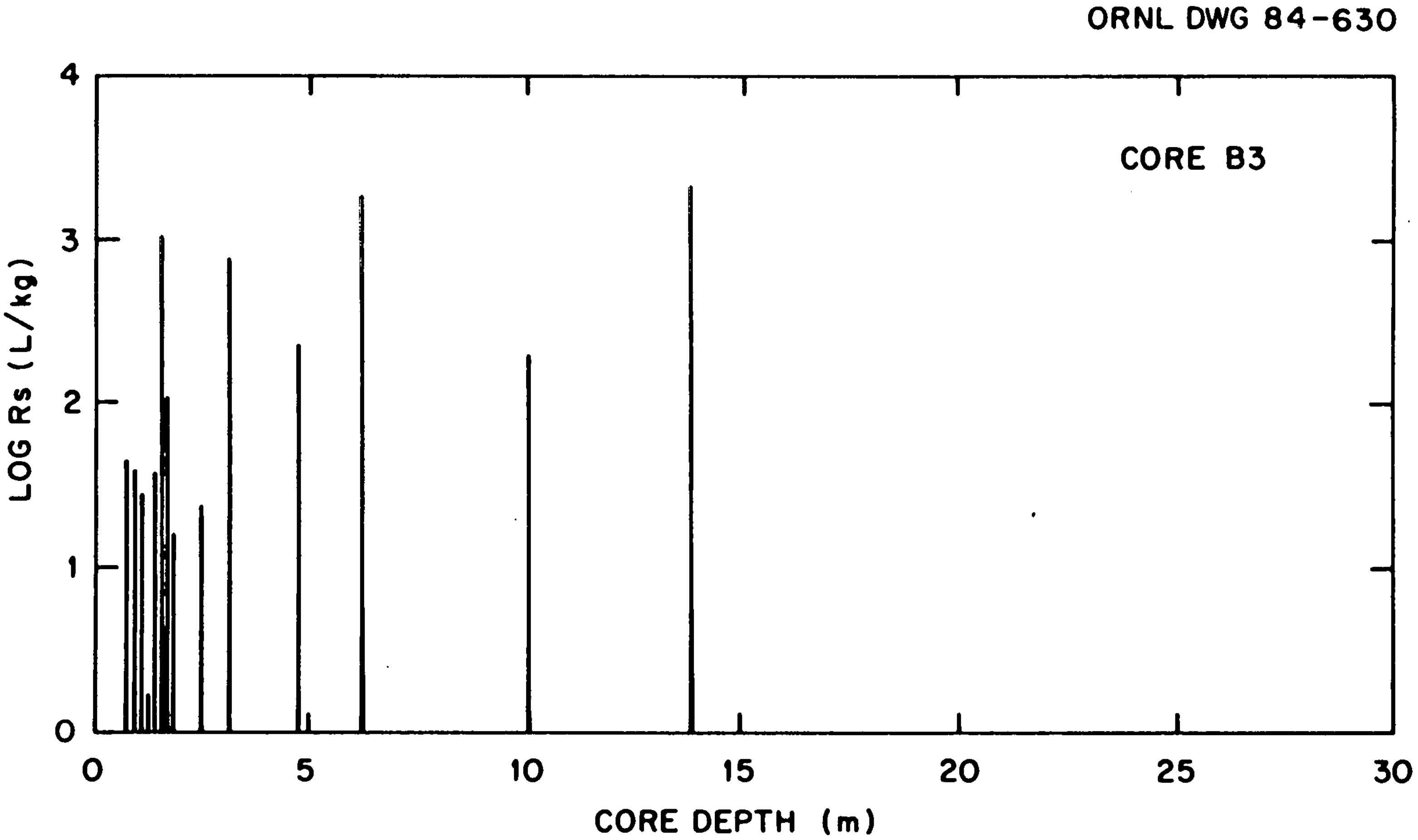

Fig. 5. Uranium sorption profile for SLASS core B3.

detail in Sect. 5, which deals with a comparison of the core material with generic clays.

The relatively high uranium Rs values obtained in these sorption tests indicate that the native material in place at the SLASS may be effectively retarding the uranium migration, either through or from the site. Such tests involve only a single-stage contact, while actual migration of a uranium-bearing solution in the soil would be a multistage phenomenon, thus improving the probability that migration of uranium would be minimal.

4.2 URANIUM SORPTION ISOTHERM

A uranium(VI) sorption isotherm was determined in a test using a soil sample from core B1-A (9.6-m depth) and a groundwater sample from Well D. The experimental procedure is described in detail in Appendix A. Briefly, 2 g of soil is contacted with 10 mL of groundwater spiked with the desired concentration of uranium, traced with ^{233}U, in an argon atmosphere (to prevent intrusion of CO_2 from the air) for a period of 20 h at room temperature. After contact, the slurry is immediately centrifuged at approximately 5000 × g; then the supernate is sampled and counted by liquid scintillation spectrometric techniques. This method is described in detail in Appendix A.

The uranium sorption behavior for this soil is summarized in Table 7, and the corresponding sorption isotherm is shown in Fig. 6. The sorption data are summarized in Table C-1 in Appendix C. The sorption ratio was 2.3 L/kg at low initial uranium concentration (5 mg/L), 38.9 L/kg at medium initial uranium concentration (1000 mg/L), and 0.8 L/kg at high initial uranium concentration (10,000 mg/L). This increase in Rs value from low to medium initial uranium concentration is apparently due to precipitation at a uranium solubility limit of approximately 200 mg/L. The decrease at high initial uranium concentration can probably be attributed to the lower pH levels for these samples, which would encourage redissolution of any precipitated uranium. The low pH values obtained at relatively high uranium concentration are the result of allowing the pH to equilibrate naturally. A small amount of acid is required to keep the uranium in the stock solutions solubilized prior to addition to the groundwater and soil. This, along with the ion exchange reaction, tends

Table 7. Summary of uranium(VI) sorption behavior on soil from the St. Louis Airport Storage Site[a]

Sample No.	Sample location		Initial uranium conc.[b]	After contact	
	Groundwater	Soil		pH	Rs (mL/g)
B1A-S12-2B	S3-60, R18-20	S4-00, R10-00	Low	7.3	2.3
			Medium	5.7	38.9
			High	3.3	0.8

[a] Stability Concept and Method Testing Project, St. Louis Airport Storage Site, St. Louis, Missouri - Progress Report, Appendix E - Plates, Draft Report, Roy F. Weston, Inc., West Chester, Pa., June 1981.

[b] Low = 5 μg/mL; medium = 1000 μg/mL; high = 10,000 μg/mL.

to decrease the pH to a greater extent than would be expected in a natural environment. However, if the pH is adjusted in each sample, the variable concentration of sodium ion in the test solutions would present a problem. It became apparent at this point that the effect of pH should be studied more thoroughly. The results of an investigation of the effect of pH are discussed in Sect. 5.3.

4.3 RADIUM SORPTION ON SLASS SOIL

Radium sorption data were determined for one soil sample from SLASS and two shallow well groundwater samples (Well D and Well F), each of which was spiked with an initial radium concentration of approximately 100,000 pCi/mL. The procedure for determining these single data points was the same as that outlined for determining sorption isotherms (see Appendix A). 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 values obtained in these tests (see Table 8) represent an average activity balance of approximately 65%, which is considered marginal, but acceptable, for radium analysis. The resulting Rs values ranged from 880 to 940 L/kg, indicating moderate, but effective, sorption of the radium by the soil samples from the SLASS.

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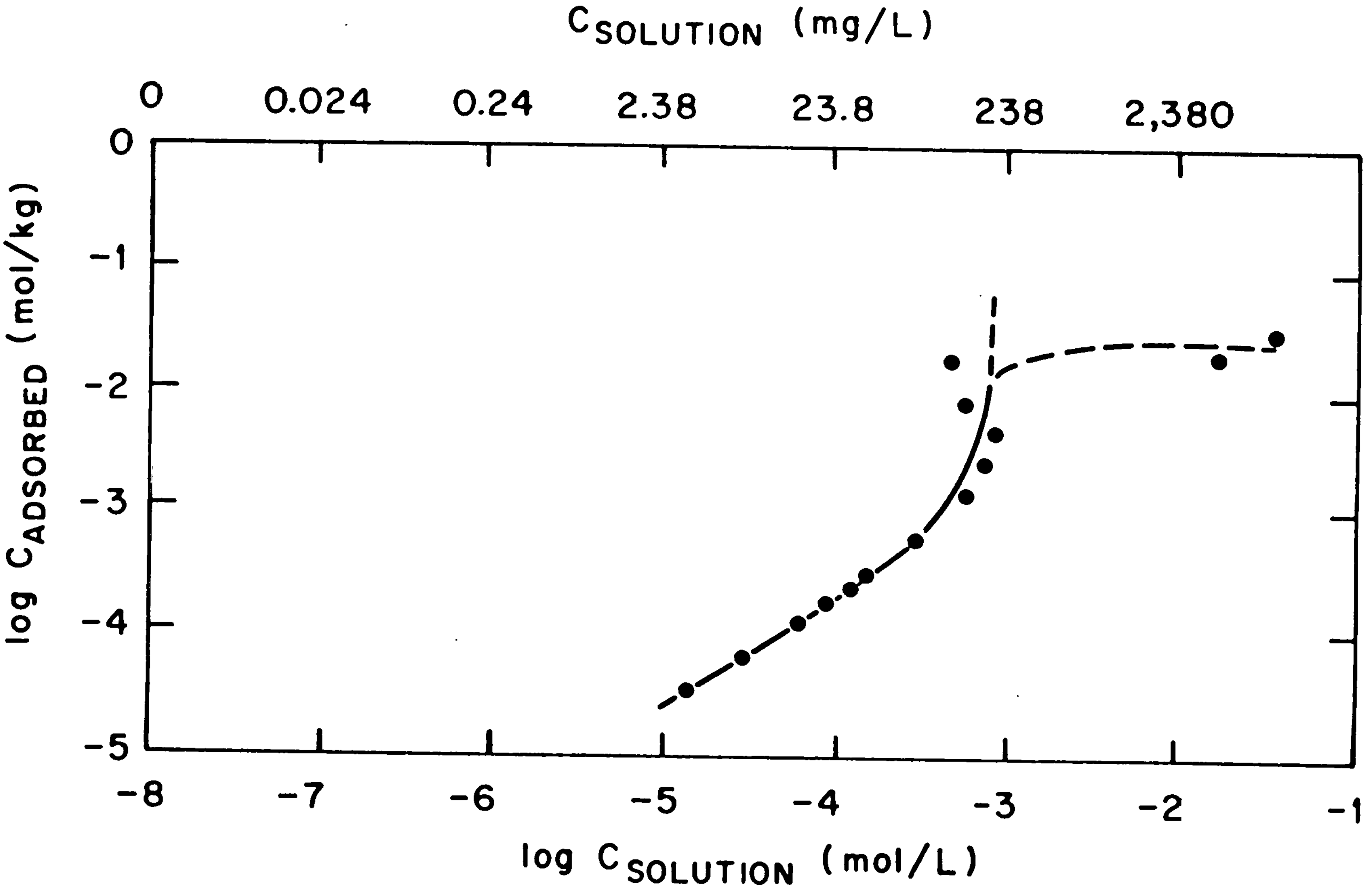

Fig. 6. Uranium sorption isotherm for soil from the St. Louis Airport Storage Site.

Table 8. Summary of radium sorption behavior on soil from the St. Louis Airport Storage Site

Sample No.	Location of groundwater sampling station	Initial radium conc. (pCi/mL)	After contact pH	After contact Rs (mL/g)
B1A-S12-2B[a]	S3-60, R18-20[b]	100,000	7.4	8.8E2
B1A-S12-2B[a]	S0-40, R1-40[c]	100,000	7.6	9.4E2

[a] Core location: S4-00, R10-00; sample depth: 31.5 ft. (Information obtained from Roy F. Weston, Inc.)

[b] Well D.

[c] Well F.

5. COMPARISON OF URANIUM SORPTION ON STANDARD CLAYS AND SLASS SOIL

5.1 EXPERIMENTAL METHODS AND DESCRIPTION OF STANDARD CLAYS

The objective of the investigations described here was to make measurements that would permit the distribution of uranium to the clay-rich soils from the SLASS to be compared with that observed for standard clays and fly ash. One of the problems encountered in such an effort is controlling the experimental variables so that the results will be truly comparative. For instance, the experimental method used for a soil containing a residuum of dolomite or some other source of carbonate must either take into account, or eliminate, the active carbonate that is present.

A simple procedure was developed to provide rapid determination of Rs values in clay and soil samples. In this procedure, the effect of active carbonate, which may be present in varying degrees in soil samples, was minimized by initially adjusting the pH of the contact system to 3.0. The pH of the clay solids/groundwater test system was then systematically increased by titration with NaOH to successive values in the range of 3 to 9, with an equilibration period and sample removal at each target pH value. An argon atmosphere was maintained over the slurry to exclude CO_2

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from the air. A back-titration of the system with HCl, with associated equilibration periods and sample removal at target pH values, was also performed to provide some insight into system reversibility and proximity to steady-state conditions. This method of testing was designed to yield uranium Rs values that could be utilized in mobility modeling. Values obtained in this manner may be particularly useful in estimating upper and lower mobility limits, especially in cases involving complex soil stratigraphy, because large numbers of samples can be tested in a relatively short period of time. It must be emphasized, however, that the effect of carbonate must be assessed separately; this method merely allows one to compare various types of soils with standard clays being used as a convenient reference.

A description of the types and sources of the standard clays and clay soils used in these investigations is given in Table 9.

Table 9. Sources of clays and clay-rich soils

Type of clay	Source location	Supplier
Halloysite No. 13	Dragon Iron Mine, Eureka, Utah	Ward's Natural Science Establishment, Inc.
Halloysite No. 29	Wagon Wheel Gap, Colorado	Ward's Natural Science Establishment, Inc.
Montmorillonite	Wyoming	Clay Source Repository, Department of Geology, University of Missouri
Kaolin	Georgia	Clay Source Repository, Department of Geology, University of Missouri
Illite	Beaverbend, Oklahoma	Oklahoma Geologic Survey
Illite soil (3-m depth)	SLAPSS Core B1-A, St. Louis, Missouri	Roy F. Weston, Inc.
Illite soil (10-m depth)	SLAPSS Core B1-A, St. Louis, Missouri	Roy F. Weston, Inc.
Illite soil (17-m depth)	SLAPSS Core B1-A, St. Louis, Missouri	Roy F. Weston, Inc.

5.2 RATE OF URANIUM SORPTION ON STANDARD CLAYS

In the study of the adsorption of metal ions on clays, some experimenters have employed equilibrium times of 1 week or more,[5] while others have allowed no equilibration time at all.[6] Such major differences in experimental methods may make comparison of data difficult. To ensure that test results will be comparable, it is first important to consider the rate at which the uranium sorption reactions occur and to determine whether there are any differences in rate with the various clay types.

Tests were carried out with four standard clays to determine the change in uranium adsorption with time, while maintaining a constant pH of 6.0. Only a relatively minor, gradual increase in uranium Rs values was observed (Fig. 7) for contact times ranging from 15 min to 120 h. These data indicate that the initial uranium sorption reaction was rapid. This is typical behavior for ion exchange reactions. Although the sorption rate behavior for the four clays was generally quite similar, the Rs values varied by more than two orders of magnitude. A wide variation of sorption behavior is possible for the same type of clay obtained from two locations. For example, the data for the two halloysite samples showed the highest and lowest values for uranium sorption. From these test data it was apparent that, for our purposes, a period of 15 min was adequate to achieve a stable, near-steady-state sorption value.

5.3 EFFECT OF pH ON URANIUM SORPTION

Although groundwater pH is usually in the range of 6 to 8, higher or lower levels could be encountered in some waste site situations. Because pH can influence both the ionic speciation and the solubility of dissolved uranium, the uranium mobility or its reciprocal, Rs, could be expected to be sensitive to pH. Uranium(VI) sorption ratios were measured for five standard clays over the pH range of 3 to 9. In these tests, a synthetic groundwater spiked with the desired concentration of uranium (100 mg/L), as UO_2^{2+} and ^{233}U tracer, was initially prepared by contacting distilled water with each sample of clay, or clay soil, prior to spiking and pH adjustment. Details of the experimental procedure used in these sorption/titration tests are listed in Appendix A.

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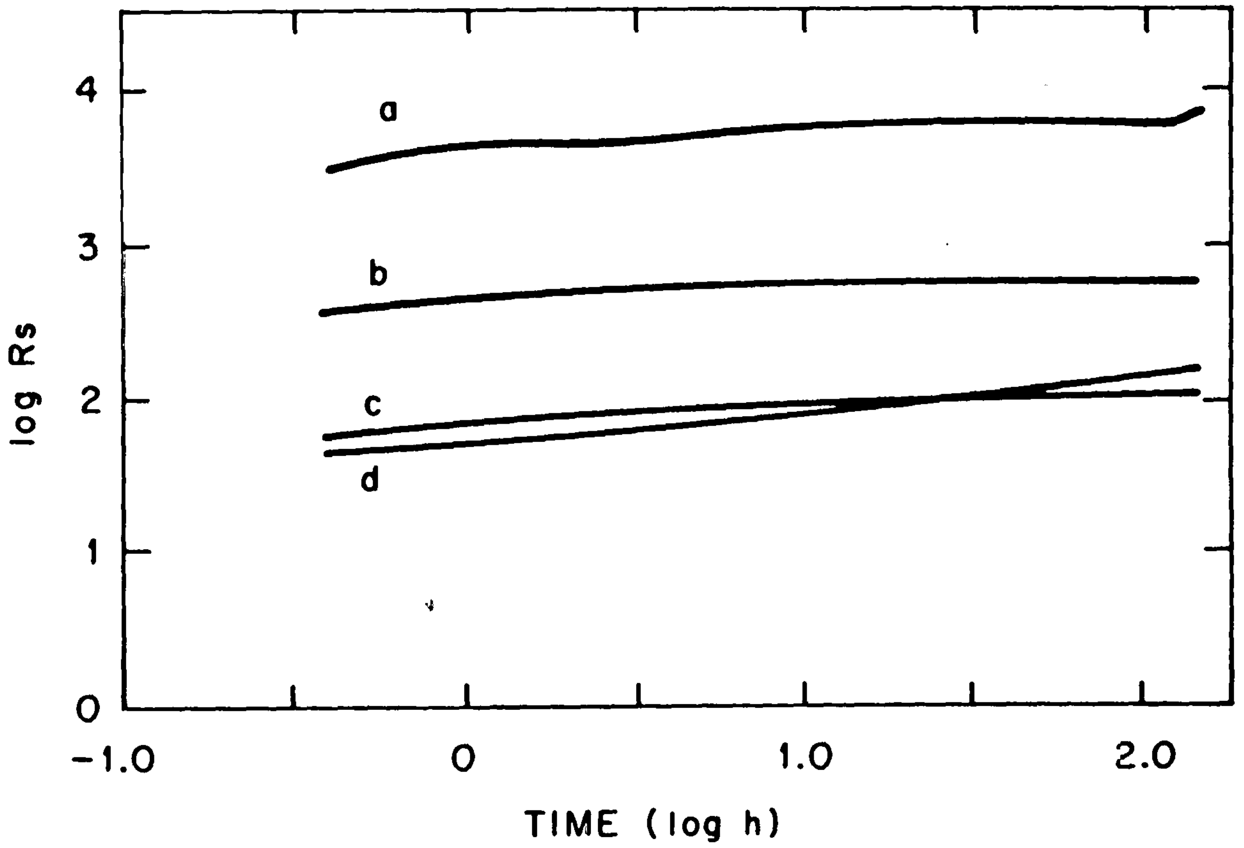

Fig. 7. Uranium sorption on standard clays as a function of time. Clay identification: (a) halloysite-13, (b) kaolin, (c) illite, and (d) halloysite-29. Test conditions: liquid/solids ratio = 10; uranium concentration of solution = 100 mg/L; pH = 6.0 (monitored and maintained throughout test); cover gas = argon. Samples for analysis were withdrawn at seven time periods between 15 min and 120 h.

Data for these tests, all of which were made under argon, are plotted in Fig. 8. At low pH values (3 to 5), Rs values of about 1 to 10 L/kg were obtained, while at higher pH levels (greater than 6), values of approximately 1000 to 10,000 L/kg were obtained. In three cases (illite, montmorillonite, and halloysite-29), a general increase in Rs was observed over the entire pH range of 3 to 9. In two cases (kaolin and halloysite-13), however, the Rs value reached a maximum in the pH range 5 to 7 and then decreased significantly in the 7 to 9 range.

Hysteresis between the forward- and back-titration curves was minimal in every test except one. This behavior is typical of systems in which reaction rates are rapid relative to the experimental time. For example, such behavior is exhibited by non-diffusion-limited ion exchange reactions and by rapid adsorption or precipitation-dissolution phenomena. The halloysite-13 sample showed substantial hysteresis, which could be caused by slow precipitation-dissolution reactions or by slow diffusion of an ionic species in restricted pores.

According to Langmuir,[7] the predominant ionic species in a uranyl sulfate solution at low uranium and sulfate concentrations should be $UO_2{}^{2+}$ up to a pH of about 5.5, UO_2OH^+ from pH 5.5 to 6.5, and $(UO_2)_3(OH)_3{}^+$ above pH 6.5. Ion exchange behavior might be expected to predominate in these pH regions. Although uranium(VI) solubility at neutral pH is not well established, it is low above pH 7 (possibly less than 10 mg/L). Thus, in these tests most of the uranium should be in a precipitated form at the higher pH levels. However, the insoluble hydrous oxide might be present as a dispersed sol, rather than as a discrete solid phase, and thus could remain mobile and/or undergo physical adsorption. In this latter case, the higher the surface area of the adsorbing media, the lower the apparent uranium mobility. Surface adsorption-desorption phenomena involving solids or dispersions might be expected to be relatively slow. Therefore, it was surprising that greater hysteresis effects were not encountered, especially at the high pH levels.

Samples that exhibited a significant decrease in Rs values at the higher pH levels (halloysite-13 and kaolin) may have contained a small amount of carbonate which was not removed during the initial equilibration

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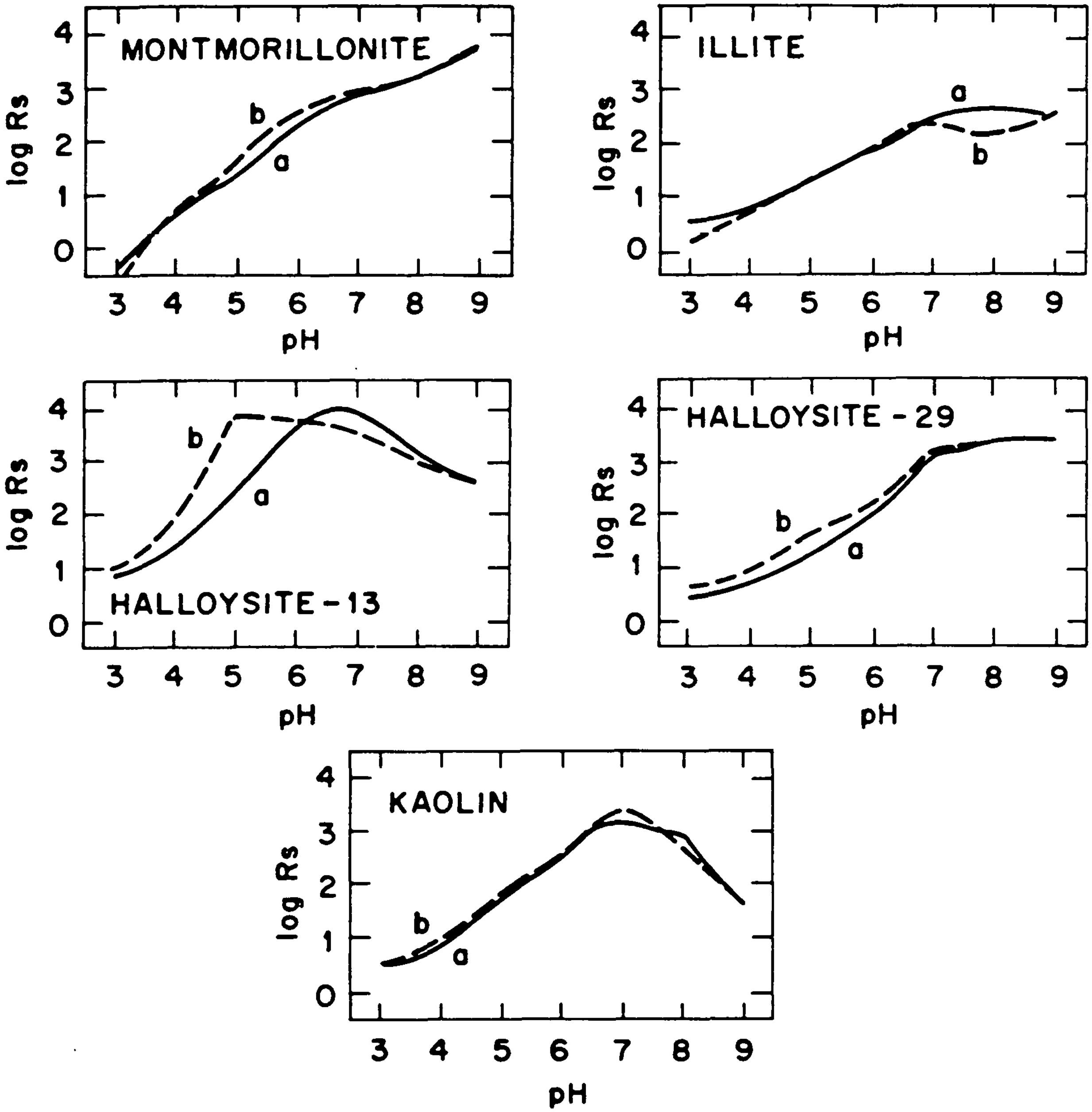

Fig. 8. Uranium sorption on standard clays as a function of pH. Test conditions: liquid/solids ratio = 10; uranium concentration of solution = 100 mg/L; cover gas = argon; forward titration (curve a) and back titration (curve b) were performed as described in Appendix A. Rs values were determined at pH 3, 4, 5, 6, 6.5, 7, 8, and 9 after 15-min equilibration at each pH.

at pH 3.0. It will be shown in Sect. 5.5 that a small amount of carbonate is capable of causing this effect.

Titration/sorption tests were made with SLASS core material to determine the effect of pH on the adsorption of U(VI) for comparison with that of the standard clays. Samples of Core B1-A were taken from the 3-, 10-, and 17-m levels of the boring. The results of the titrations, shown in Fig. 9, indicate that all the sorption maxima are in the pH range of 6.0 to 6.5. The small amount of hysteresis in the low-pH range indicates the existence of a reversible cation exchange reaction. The decrease in Rs at the higher-pH level suggests the possibility of the presence of a small amount of carbonate in the clay soil, since these tests were conducted under argon.

Comparison of the Rs values obtained in these tests with those from sorption profile tests at equivalent pH values (see Sect. 4.1) shows remarkable agreement, suggesting that the initial treatment at pH 3.0 in the sorption/titration tests apparently did not alter the sorption properties of the clay component of the soil to a significant degree. If, however, these data are compared with those obtained in the determination of the single sorption isotherm for soil from the 10-m depth of core B1-A, the Rs values appear to be significantly lower. The maximum Rs value obtained in the determination of the sorption isotherm was only 38 L/kg at pH 5.7, while the value for the same soil sample at this pH level obtained in the sorption/titration test was approximately 600 L/kg. The only variable not common to both tests was the groundwater. Synthetic groundwater was used in the sorption/titration test, while actual groundwater (Well D) was employed for the determination of the sorption isotherm. Unfortunately, no analysis was made of the organic carbon in this groundwater. The lower Rs values may be due to the presence of dissolved CO_2 or carbonate in the well water. Additionally, the initial pH adjustment in the sorption titration tests probably removed some active carbonate from the system and could have slightly affected the surface characteristics of the clay particles.

5.4 SORPTION OF URANIUM ON FLY ASH

The sorption of uranium on two samples of fly ash from coal combustion, obtained from the Appalachian and Midwestern supply regions, was

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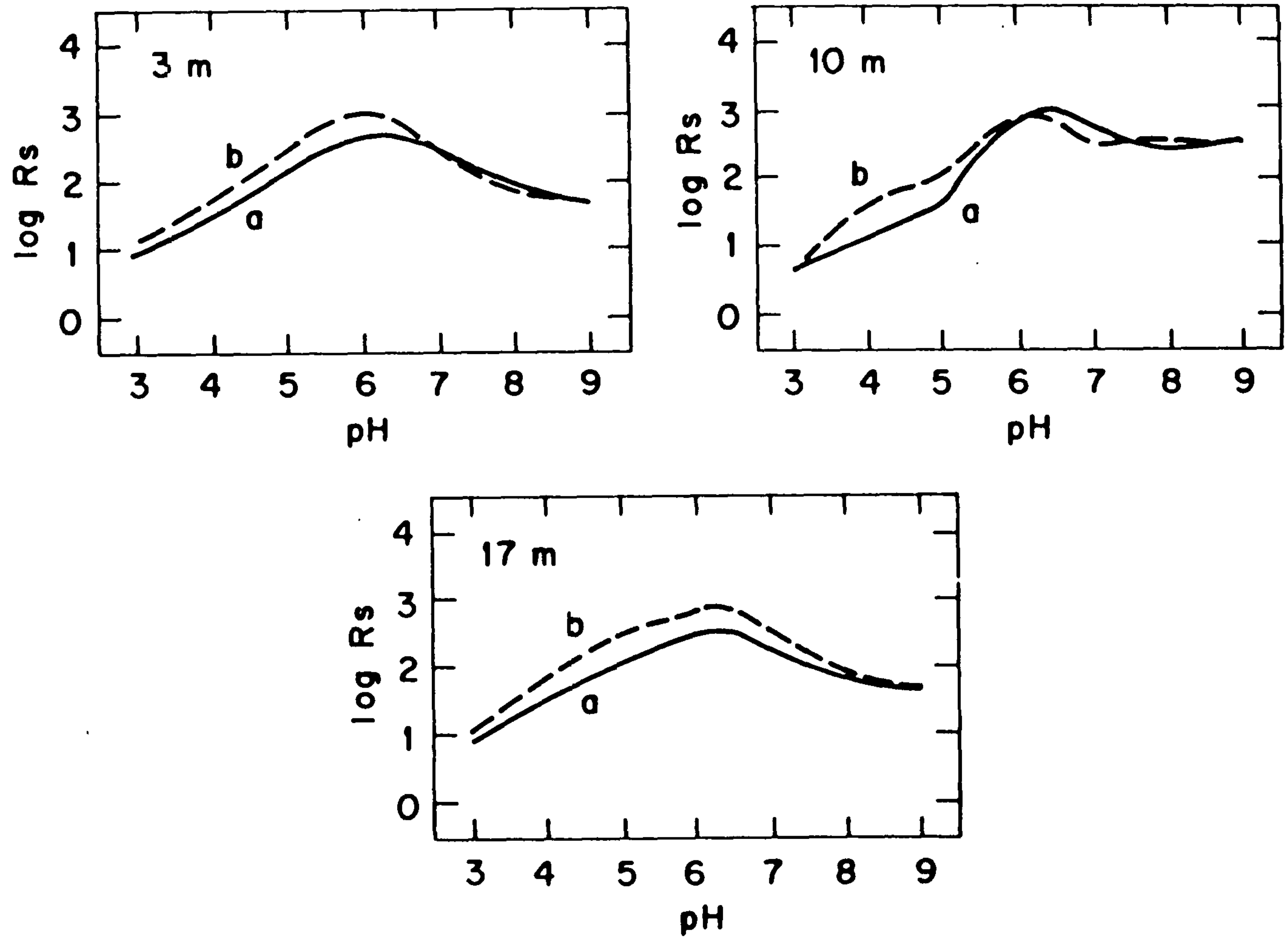

Fig. 9. Uranium sorption on illite-bearing soil samples from SLASS core B1-A as a function of pH and depth. Test conditions were the same as those given for Fig. 8.

determined as a function of pH. This study was part of a cursory search for readily available nonclay materials that might be utilized to reduce uranium mobility in groundwaters near low-level waste storage sites. The Rs values (see Fig. 10) were surprisingly high, exceeding 10,000 L/kg at pH values above 7. In general, they are higher than those found for the standard clays. Some hysteresis was observed in the back-titration, indicating probable formation of an insoluble species of uranium which was retained on the surface of the fly-ash particles.

Clays are often used, or considered for use, in waste site stabilization because they are readily available low-cost materials having high distribution coefficients for heavy metals. These test results suggest that fly ash, a significant waste in its own right, may also be a potentially useful, inexpensive stabilizing material for uranium-containing radioactive waste sites.

5.5 EFFECT OF DISSOLVED CARBON DIOXIDE ON URANIUM SORPTION

Dementryev and Syromyatnikov[8] reported sorption-vs-pH curves for the sorption of uranium onto materials such as coal, peat, and ion exchange resins. All of the curves showed maxima near pH 5 to 6. Because these tests were not protected from the atmosphere, dissolved CO_2 could have been responsible for the decrease in sorption at pH levels above 6. No reports describing the effect of pH on the binding of U(VI) to clay-soils were identified in our literature search.

Soluble uranylcarbonate anion complexes are well known.[7,9] Groundwaters that are equilibrated with air (about 330 ppm CO_2) or with alkaline minerals (e.g., calcite or dolomite) could contain carbonate anions, which could increase uranium solubility through the formation of uranylcarbonate anions. Spectrophotometric evidence of such anionic species was observed in soil/groundwater systems investigations of the Niagara Falls Storage Site.[4]

In uranium sorption tests designed to show the effect of CO_2 dissolved in the groundwater, a 5% CO_2 atmosphere was selected to provide sufficient $CO_3{}^{2-}$ (from the dissolved CO_2) to ensure that uranyl complexing would occur during the 15-min test equilibration period. The uranium

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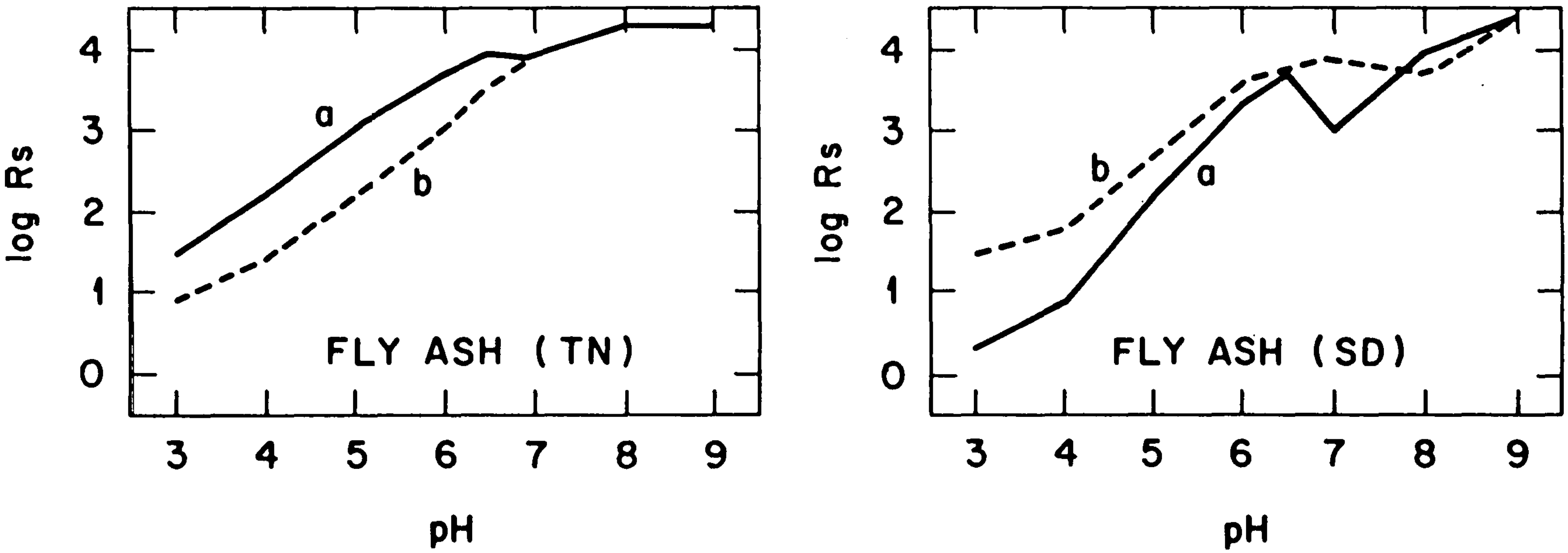

Fig. 10. Uranium sorption on fly ash as a function of pH. Test conditions were the same as those given for Fig. 8.

Rs values for three standard clays and one fly ash (Fig. 11) show a marked reduction above pH 5 compared with the results obtained in tests in which an argon atmosphere was maintained (Figs. 8 and 10).

Parallel uranium sorption-vs-pH tests were made with SLASS soil from the 10-m level of Core B1-A with argon and CO_2 atmospheres at three uranium concentrations: 5, 100, and 5000 mg/L. The resulting data (Fig. 12) again indicated a drastic reduction of the Rs values (from one to two orders of magnitude at pH levels from 6 to 9) in the presence of dissolved carbon dioxide.

6. ACKNOWLEDGMENTS

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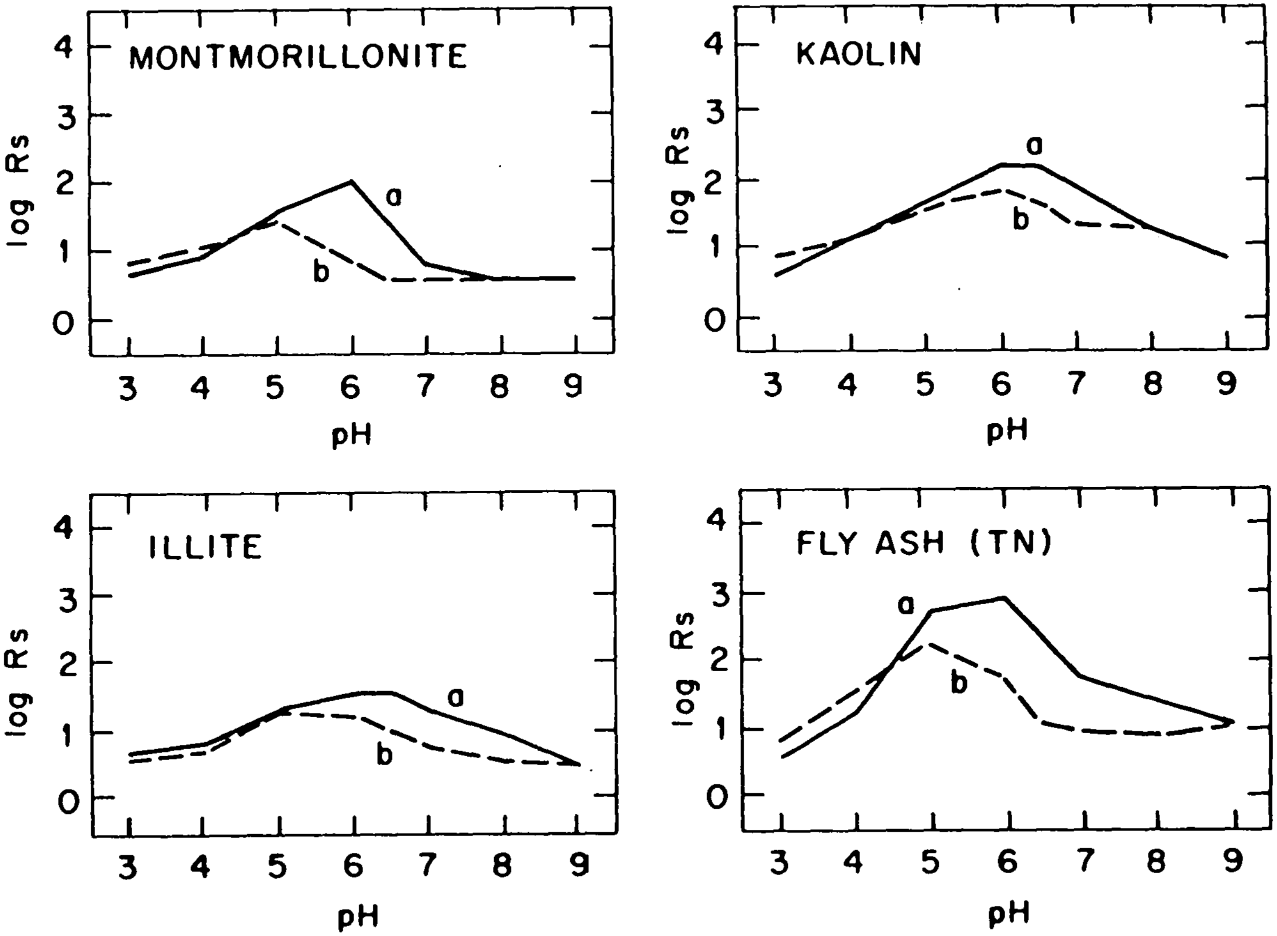

Fig. 11. Effect of dissolved CO_2 on the sorption of uranium by standard clays and fly ash as a function of pH. Test conditions were the same as those given for Fig. 8, except that the cover gas was air—5% CO_2.

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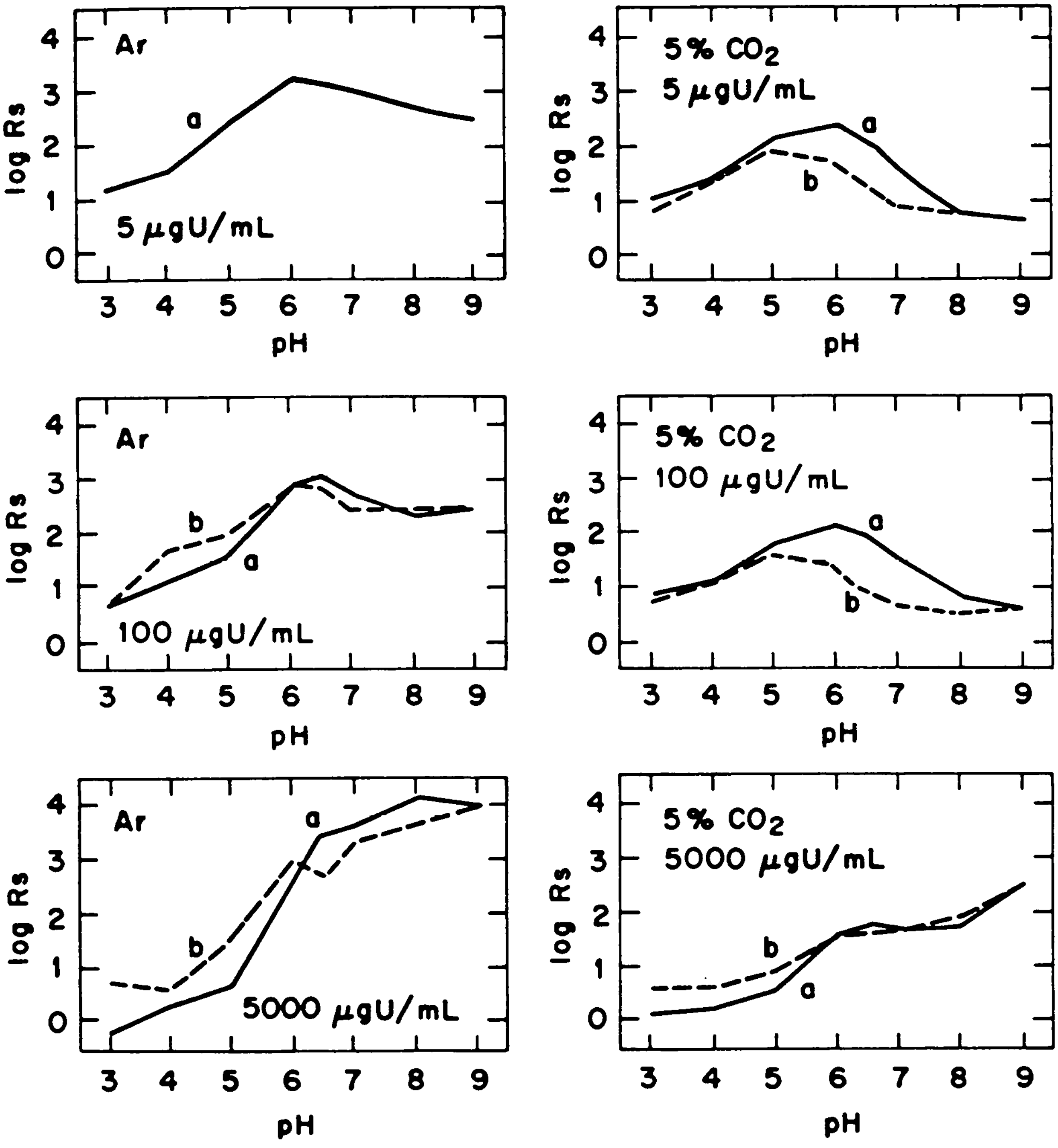

Fig. 12. Effect of dissolved CO_2 on uranium sorption on illite-bearing soil from the 10-m level of SLASS core B1-A as a function of pH at three uranium concentrations. Test conditions were the same as those given for Fig. 8, except for the change in uranium concentration and the use of an air—5% CO_2 cover gas in three of the graphs.

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8. APPENDIXES

Appendix A.

EXPERIMENTAL METHODS

A.1. EXPERIMENTAL PROCEDURE FOR THE DETERMINATION OF SINGLE SORPTION VALUES AND SORPTION ISOTHERMS (AMBIENT-pH METHOD)

To provide data for sorption isotherms, each batch contact test is conducted by contacting 1.0 g of soil with 10 mL of the synthetic groundwater in a polystyrene centrifuge tube for an initial period of 3 h to allow the soil and the groundwater to reach steady-state conditions. An argon atmosphere is maintained above the slurry to minimize the introduction of additional carbonate from CO_2 in the air 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 each metal-ion stock solution 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 activity 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 4 h. (Various contact times can be used in sorption rate studies.) After the sample has been removed from the shaker, the solids are allowed to settle and the pH of the supernate is measured. If there is any significant departure from the predetermined experimental pH (the natural pH established between the soil and the well water) being investigated, an adjustment is made to bring it to the desired level. A 4-h period of recontacting is employed for any sample requiring pH adjustment.

After the pH of a sample has become stabilized, the sample is placed in a Sorvall centrifuge and centrifuged at 5000 rpm for about 15 min. The radionuclide concentration is then determined by removing 1.0 mL of the clear supernate for counting. If the sample is alpha-active, it is placed in 10 mL of a liquid scintillation fluid and counted on a Packed Tri-Carb scintillation spectrometer; if the sample is gamma-active, it is counted directly on a Packard automatic gamma spectrometer that is interfaced with a Nuclear Data ND-60 multichannel analyzer. Appropriate standards and blanks are also included. A computer program (see Sect. A.4) was used to calculate the counting statistics. This program determines the mean count rate and standard deviation for samples counted one to ten times.

A.2. TITRATION METHOD FOR DETERMINING THE ADSORPTION OF URANIUM ON CLAYS AND CLAY SUBSTITUTES

The large number of determinations necessary to ensure that results are representative of the total site environment require a relatively rapid experimental routine for measuring sorption distribution coefficients. The titration adsorption method described here strikes a very favorable compromise between practical and absolute equilibrium values. As shown in the adsorption rate studies in Sect. 5.2, the values obtained for those distribution coefficients that were higher than 100 L/kg after 15-min equilibrations were greater than 90 to 95% of those values obtained after several days of contact. This magnitude of deviation is probably well within experimental error.

A weighed sample (usually 10.0 g) of the clay to be tested is placed in a 250-mL titration vessel equipped with a cover with ports for the introduction of cover gas, combination pH electrode, burette tip, and sampling. The solid sample is then contacted with 100 mL of a uranyl sulfate solution, of known uranium concentration, which has been spiked with approximately 10^5 dpm/mL of high-purity, high-specific-activity ^{233}U. The U(VI) solution always has an initial pH of 3.0 to ensure the existence of uranyl species. An atmosphere of argon is maintained as the cover gas unless otherwise specified.

The slurry was titrated with standardized 0.1 $\underline{N}$ NaOH solution to pH levels of 4.0, 5.0, 6.0, 6.5, 7.0, 8.0, and 9.0, with a 15-min equilibration period at each step (while maintaining a constant pH). This procedure was then reversed, and the slurry was titrated with standardized 0.1 $\underline{N}$ HCl to the same pH levels in reverse order. The slurry was agitated with a magnetic stirring bar. At each of the pH levels, a 5-mL sample of the slurry was removed following the 15-min equilibration period and quickly centrifuged. Then, 1.0 mL of the supernate was removed and added to 10 mL of Insta-Gel (a liquid scintillation cocktail obtained from Packard Instrument Co.). The alpha activity was subsequently determined in a Packard Tri-Carb liquid scintillation spectrometer and compared with that of the head solution.

The uranium distribution coefficient (D_U), in units of L/kg (or mL/g), was calculated as follows:

$$D_U = \frac{\dfrac{A_i - A_e}{W_s}}{\dfrac{A_e}{V_L}} ,$$

where

A_i = disintegrations per minute per mL of initial solution,

A_e = disintegrations per minute per mL of equilibrated solution,

W_s = weight of the solid sample (g), and

V_L = volume of the solution (mL).

A.3. COMPUTER PROGRAM FOR CALCULATING SORPTION OR DESORPTION RATIOS

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

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

A.4. PROGRAM FOR THE CALCULATION OF COUNT RATE AND STANDARD DEVIATION

```
PROGRAM NAME: CSIG2.BAS
00005 PRINT "THIS PROGRAM CALCULATES STANDARD DEVIATION FOR COUNTING"
00007 PRINT "FOR SAMPLES COUNTED 1 TO 10 TIMES"
00010 PRINT "TO TERMINATE PROGRAM TYPE 0 FOR TIMES COUNTED"
00020 PRINT "HOW MANY TIMES WAS SAMPLE COUNTED";
00030 INPUT N
00035 IF N = 0 THEN 410
00040 PRINT "ENTER TOTAL COUNT (INCL. BKG.) AND COUNTING TIME"
00041 PRINT "IN PAIRS, ONE PAIR PER LINE"
00042 S1 = 0
00043 S2 = 0
00044 S3 = 0
00050 FOR I=1 TO N
00060 INPUT C1(I),T1(I)
00070 R(I) = C1(I)/(T1(I)**2)
00100 S1 = S1 + R(I)
00110 S2 = S2 + C1(I)
00115 S3 = S3 + T1(I)
00116 R1 = S2/S3
00125 NEXT I
00126 G1 = (SQRT(S1))/N
00130 PRINT "HOW MANY TIMES WAS BKG COUNTED";
00140 INPUT M
00150 PRINT "ENTER BKG COUNT AND COUNTING TIME IN PAIRS"
00155 PRINT "ONE PAIR PER LINE"
00156 S4 = 0
00157 S5 = 0
00158 S6 = 0
00160 FOR J = 1 TO M
00170 INPUT B(J),T2(J)
00180 D(J) = B(J)/(T2(J)**2)
00199 S4 = S4 + T2(J)
00200 S5 = S5 + D(J)
00210 S6 = S6 + B(J)
00215 NEXT J
00220 G2 = (SQRT(S5))/M
00230 R2 = S6/S4
00240 D2 = R1-R2
00241 X = S2/(S3**2)
00242 Y = S6/(S4**2)
00243 G = SQRT(X+Y)
00244 P = G*100/D2
00250 IF D2 <= 0 THEN 370
00251 F1 = G1**2
00252 F2 = G2**2
00260 G3 = SQRT(F1+F2)
00262 A = S2/N
00264 P2 = G3*100/A
```

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

Appendix B.

URANIUM SORPTION DATA FOR CORE SOIL SAMPLES

Table B-1. Sorption characterization of soil from core B1-A from the St. Louis Airport Storage Site

Core depth	After contact					
(m)	pH	Solution (mol/L)	Adsorbed (mol/kg)	Solution (log mol/L)	Adsorbed (log mol/kg)	Rs (L/kg)
2.74	8.0	8.7E-6	2.5E-4	-5.06	-3.61	2.9E1
2.90	7.9	4.1E-6	3.4E-4	-5.39	-3.47	8.3E1
3.05	7.8	1.8E-6	4.0E-4	-5.75	-3.40	2.3E2
3.35	7.9	2.6E-6	3.7E-4	-5.58	-3.43	1.4E2
3.51	7.8	1.3E-6	3.9E-4	-5.88	-3.40	3.0E2
3.75	7.9	3.3E-6	3.5E-4	-5.48	-3.45	1.1E2
3.96	7.8	3.6E-6	3.5E-4	-5.44	-3.46	9.6E1
4.11	7.8	3.5E-6	3.5E-4	-5.46	-3.45	1.0E2
4.57	7.7	2.5E-6	3.7E-4	-5.60	-3.43	1.5E2
5.18	7.7	2.2E-6	3.8E-4	-5.65	-3.43	1.7E2
6.40	6.8	7.1E-8	4.2E-4	-7.15	-3.38	5.9E3
7.62	6.9	1.6E-7	4.2E-4	-6.80	-3.38	2.6E3
8.83	6.9	2.1E-7	4.2E-4	-6.69	-3.38	2.0E3
9.60	6.6	2.0E-7	4.2E-4	-6.70	-3.38	2.1E3
11.58	6.7	9.7E-8	4.2E-4	-7.01	-3.38	4.3E3
14.63	7.9	1.0E-5	2.2E-4	-4.99	-3.67	2.1E1
17.37	7.5	7.3E-7	4.1E-4	-6.14	-3.39	5.6E2

Table B-2. Sorption characterization of soil from core B1-B from the St. Louis Airport Storage Site

	After contact					
Core depth (m)	pH	Solution (mol/L)	Adsorbed (mol/kg)	Solution (log mol/L)	Adsorbed (log mol/kg)	Rs (L/kg)
0.30	7.9	9.6E-6	2.3E-4	-5.02	-3.64	2.4E1
0.61	7.8	1.1E-5	2.0E-4	-4.97	-3.69	1.9E1
0.91	8.1	5.8E-6	3.0E-4	-5.23	-3.52	5.2E1
1.22	7.5	2.4E-7	4.2E-4	-6.63	-3.38	1.8E3
1.52	7.9	8.1E-6	2.6E-4	-5.09	-3.59	3.2E1
1.74	7.9	8.7E-6	2.5E-4	-5.06	-3.61	2.8E1
1.92	7.4	5.5E-7	4.1E-4	-6.26	-3.39	7.5E2
2.13	7.2	1.3E-7	4.2E-4	-6.89	-3.38	3.2E3
2.35	7.2	1.2E-7	4.2E-4	-6.93	-3.38	3.6E3
2.53	7.8	2.8E-6	3.6E-4	-5.55	-3.44	1.3E2
2.74	7.9	4.9E-6	3.2E-4	-5.31	-3.49	6.5E1
2.90	7.9	5.1E-6	E.2E-4	-5.29	-3.50	6.3E1
3.05	7.8	5.8E-6	3.0E-4	-5.24	-3.52	5.3E1
3.20	7.8	3.0E-6	3.6E-4	-5.53	-3.44	1.2E2
15.54	7.9	9.4E-6	2.3E-4	-5.03	-3.63	2.5E1
16.31	7.7	5.3E-7	4.1E-4	-6.28	-3.39	7.8E2
18.59	7.7	3.1E-6	3.6E-4	-5.51	-3.45	1.2E2
21.03	7.0	1.1E-6	4.0E-4	-5.97	-3.40	3.8E2

Table B-3. Sorption characterization of soil from core B2 from the St. Louis Airport Storage Site

	After contact					
Core depth (m)	**pH**	**Solution (mol/L)**	**Adsorbed (mol/kg)**	**Solution (log mol/L)**	**Adsorbed (log mol/kg)**	**Rs (L/kg)**
0.00	8.8	7.3E-6	2.7E-4	-5.14	-3.56	3.8E1
0.15	7.9	9.3E-6	2.4E-4	-5.03	-3.63	2.5E1
0.31	8.4	6.1E-6	3.0E-4	-5.22	-3.53	4.9E1
1.83	9.6	8.7E-7	4.0E-4	-6.06	-3.40	4.6E2
1.98	7.7	5.8E-6	3.0E-4	-5.24	-3.52	5.3E1
2.13	7.8	5.2E-6	3.2E-4	-5.28	-3.50	6.1E1
2.29	7.9	5.7E-6	3.1E-4	-5.25	-3.51	5.4E1
2.44	7.9	1.1E-5	2.1E-4	-4.97	-3.68	1.9E1
2.59	7.5	2.5E-6	3.7E-4	-5.60	-3.43	1.5E2
2.74	7.2	2.3E-7	4.2E-4	-6.64	-3.38	1.8E3
3.05	7.8	1.3E-5	1.6E-4	-4.89	-3.80	1.2E1
3.14	6.6	1.2E-7	4.2E-4	-6.92	-3.38	3.5E3
3.35	7.7	9.2E-6	2.4E-4	-5.04	-3.63	2.6E1
4.42	7.4	6.7E-6	2.9E-4	-5.17	-3.54	4.3E1
5.18	8.1	1.1E-5	2.0E-4	-4.96	-3.70	1.8E1
5.94	8.2	1.2E-5	1.9E-4	-4.93	-3.73	1.6E1
6.71	7.9	3.5E-6	3.5E-4	-5.45	-3.46	9.9E1
10.52	6.6	4.4E-7	4.1E-4	-6.36	-3.39	9.5E2
14.33	7.9	1.1E-5	2.0E-4	-4.95	-3.71	1.7E1
18.14	7.9	7.4E-6	2.7E-4	-5.13	-3.57	3.7E1
21.95	7.0	5.7E-7	4.1E-4	-6.24	-3.39	7.2E2
25.45	7.7	7.9E-6	2.6E-4	-5.10	-3.58	3.3E1

Table B-4. Sorption characterization of soil from core B3 from the St. Louis Airport Storage Site

Core depth (m)	After contact					
	pH	Solution (mol/L)	Adsorbed (mol/kg)	Solution (log mol/L)	Adsorbed (log mol/kg)	Rs (L/kg)
0.00	7.9	7.3E-6	2.7E-4	-5.14	-3.56	3.8E1
0.76	8.0	6.5E-6	2.9E-4	-5.19	-3.54	4.4E1
0.91	7.9	7.2E-6	2.8E-4	-5.14	-3.56	3.8E1
1.07	9.3	8.7E-6	2.5E-4	-5.06	-3.61	2.8E1
1.22	8.9	2.1E-5	3.8E-6	-4.68	-5.42	0.2E-1
1.37	8.1	7.6E-6	2.7E-4	-5.12	-3.57	3.5E1
1.52	7.5	3.8E-7	4.1E-4	-6.42	-3.38	1.1E3
1.68	7.7	3.3E-6	3.5E-4	-5.48	-3.45	1.1E2
1.83	8.6	1.2E-5	1.8E-4	-4.92	-3.74	1.5E1
2.44	8.1	9.9E-6	2.2E-4	-5.00	-3.66	2.2E1
3.20	7.3	5.4E-7	4.1E-4	-6.27	-3.39	7.5E2
4.72	8.0	1.8E-6	3.8E-4	-5.75	-3.42	2.2E2
6.25	7.0	2.3E-7	4.2E-4	-6.65	-3.38	1.8E3
10.06	7.7	2.1E-6	3.8E-4	-5.67	-3.42	1.8E2
13.87	6.7	2.0E-7	4.2E-4	-6.71	-3.38	2.1E3

Appendix C.

URANIUM SORPTION ISOTHERM DATA

Table C-1. Uranium(VI) sorption isotherm data for soil from the St. Louis Airport Storage Site[a]

Initial uranium conc.	After contact					
(μg/mL)	pH	Solution (mol/L)	Adsorbed (mol/kg)	Solution (log mol/L)	Adsorbed (log mol/kg)	Rs (L/kg)
5	7.3	1.5E-5	3.3E-5	-4.84	-4.48	2.3
10	7.2	2.9E-5	6.5E-5	-4.53	-4.19	2.2
20	7.2	6.0E-5	1.2E-4	-4.22	-3.92	2.0
30	7.2	9.1E-5	1.8E-4	-4.04	-3.75	1.9
40	7.2	1.2E-4	2.3E-4	-3.91	-3.63	1.9
50	7.2	1.5E-4	3.0E-4	-3.82	-3.53	2.0
100	7.1	3.0E-4	6.1E-4	-3.52	-3.22	2.0
200	6.9	5.6E-4	1.4E-3	-3.25	-2.85	2.5
300	6.7	7.3E-4	2.7E-3	-3.13	-2.57	3.7
400	6.6	8.0E-4	4.5E-3	-3.09	-2.35	5.6
500	6.4	5.6E-4	7.9E-3	-3.25	-2.10	14.0
1,000	5.7	4.9E-4	1.9E-2	-3.31	-1.72	38.9
5,000	4.1	1.7E-2	2.2E-2	-1.77	-1.67	1.3
10,000	3.3	3.7E-2	3.1E-2	-1.44	-1.52	0.8

[a] Soil sample from core B1-A (31.5-ft depth); groundwater from Well D.

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