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Uptakes of Cs and Sr on San Joaquin Soil Measured Following ASTM Method C1733

Chemical Sciences and Engineering Division

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by
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ACRONYMS

ANL	Argonne National Laboratory
ASTM	American Society for Testing and Materials (ASTM–International)
DOE	U.S. Department of Energy
ICP-MS	Inductively coupled plasma-mass spectrometry
ILS	Inter-laboratory study
K_d	Distribution coefficient
NIST	National Institute of Standards and Technology
SEM	Scanning electron microscope (or microscopy)
SRM	Standard reference material
XRD	X-ray diffraction

CONTENTS

LIST OF FIGURES

LIST OF TABLES

Abstract

Series of tests were conducted following ASTM Standard Procedure C1733 to evaluate the repeatability of the test and the effects of several test parameters, including the solution-to-soil mass ratio, test duration, pH, and the concentrations of contaminants in the solution. This standard procedure is recommended for measuring the distribution coefficient (K_d) of a contaminant in a specific soil/groundwater system. One objective of the current tests was to identify experimental conditions that can be used in future inter-laboratory studies to determine the reproducibility of the test method. This includes the recommendation of a standard soil, the range of contaminant concentrations and solution matrix, and various test parameters. Quantifying the uncertainty in the distribution coefficient that can be attributed to the test procedure itself allows the differences in measured values to be associated with differences in the natural systems being studied.

Tests were conducted to measure the uptake of Cs and Sr dissolved as CsCl and $Sr(NO_3)_2$ in a dilute $NaHCO_3/SiO_2$ solution (representing contaminants in a silicate groundwater) by a NIST standard reference material of San Joaquin soil (SRM 2709a). Tests were run to measure the repeatability of the method and the sensitivity of the test response to the reaction time, the mass of soil used (at a constant soil-to-solution ratio), the solution pH, and the contaminant concentration. All tests were conducted in screw-top Teflon vessels at 30 °C in an oven. All solutions were passed through a 0.45-µm pore size cellulose acetate membrane filter and stabilized with nitric acid prior to analysis with inductively-coupled plasma mass spectrometry (ICP-MS). Scoping tests with soil in demineralized water resulted in a solution pH of about 8.0 and the release of small amounts of Sr from the soil. Solutions were made with targeted concentrations of 1×10^{-6} $\underline{m}$, 1×10^{-5} $\underline{m}$, 2.5×10^{-5} $\underline{m}$, 5×10^{-5} $\underline{m}$, 1×10^{-4} $\underline{m}$, and 5×10^{-4} $\underline{m}$ to measure the effects of the Cs and Sr concentrations on their uptake by the soil. The pH values of all solutions were adjusted to about pH 8.5 so that the effects of pH and concentration could be measured separately. The 1×10^{-4} $\underline{m}$ solutions were used to measure the repeatability of the test and the effects of duration, scale, and imposed pH on the test response.

The following reference conditions are recommended to measure the precision and bias of the test method and establishing a reference test response that can be used for direct comparisons of test execution and the effects of other test parameter and environmental variables:

Soil:	NIST SRM 2709a San Joaquin soil without further treatment
Solution:	1.5×10^{-4} $\underline{m}$ CsCl (0.0253 g CsCl/kg solution) in ASTM C1220 synthetic silicate groundwater
Solution pH:	8.5
Mass Ratio:	1.0 g soil in 25.0 g solution
Temperature:	30 °C
Duration:	2.0 days

1.   INTRODUCTION AND BACKGROUND

The ASTM standard procedure C1733 was recently issued as "a measurement technique for determining the degree of partitioning between liquid and solid, under a certain set of conditions, for the species of interest" (ASTM 2010). ASTM C1733 represents a significant revision of ASTM standard procedure D4319, which was withdrawn by ASTM in due, in part, to disbanding of the subcommittee D18.14 on Geotechnics of Waste Management that was responsible for maintaining and updating the standard. That method was revised and reissued by subcommittee C26.07 on Waste Materials (part of Nuclear Fuel Cycle committee C26), which made modifications to the test method and data analysis, provided a standard format for documenting the test results, and refined the recommended usage of the test results. The work described in this report was done to (1) evaluate the sensitivity of the ASTM C1733 test responses to several test parameters and provide measures of intra-laboratory precision and bias, and (2) identify a material and test conditions that can be used in a future study to measure inter-laboratory precision and bias.

The ASTM C1733 test procedure is a static method that can be used to measure the distribution coefficient (K_d) value of a particular system. (The terms distribution coefficient and partition coefficient are used interchangeably in the literature.) The distribution coefficient is commonly used to quantify the amount of a solute that has been removed from a solution due to interactions with a solid, and is defined as:

$$K_d = \frac{\text{mass of solute on the solid phase per unit mass of solid phase}}{\text{mass of solute in solution per unit volume of the liquid phase}} . \tag{1}$$

This definition stems from the thermodynamic basis of distribution as it was developed to quantify equilibrium conditions for ion exchange reactions. The use of K_d has expanded to empirical use in contamination transport models, where the distribution coefficient is used to quantify the collective effects of several processes in addition to ion exchange, which may include sorption, absorption, precipitation, coprecipitation, size exclusion, complexation, and oxidation-reduction reactions. Most of these processes are expected to be sensitive to temperature, solution pH and Eh, ionic strength, etc., and perhaps most importantly, to the solution concentration of the solute of interest. In such usage, the value of K_d represents the behavior of a particular solid and solution under a specific set of conditions, and may or may not be considered an equilibrium value.

While K_d values can be determined for any pure solid material, the focus of ASTM C1733 is measuring values for use in the modeling of contaminant transport through geological media. For application to soils, the texture, porosity, water and organic matter content of the soil, and the presence of microbes can also affect K_d. In the procedure, it is stated that collected soils can be evaluated in the natural aggregated state or be disaggregated, and may be used in the natural state with retained pore water or may be dried. Note that K_d is calculated based on the dry mass of the soil, so a separate sample must be dried to determine the dry mass even if K_d is to be measured using a sample that is not dried. The state of the soil will affect the test result. The US Environmental Protection Agency has issued three reports that provide detailed discussion of the thermodynamic basis of K_d and the empirical use in contaminant transport models (EPA 1999a, EPA 1999b, EPA 2004). The reader is referred to these documents for more detailed and thorough descriptions of the processes that are summarized in this report.

1.1   Measuring K_d with Laboratory Tests

The distribution coefficient (K_d) is the ratio of the concentration of a substance in (on) a solid (in this case soil) per unit dry mass to the concentration in solution. The uncertainty in the measured value of K_d and the sensitivity to environmental conditions contribute inherently high uncertainties to source term models. The value of K_d can be measured in the laboratory using dynamic column tests or static batch tests. These methods can yield different values due to the effects of non-equilibrium, convection, and diffusion in the column method and failure to reach equilibrium in the batch method. In general, the batch method is used to provide values under hydrodynamically saturated and equilibrium conditions, whereas the column method is used to provide values for a more representative unsaturated soil condition. In column tests, the 50% breakthrough curves for the solvent (usually water) and solute are compared as

$$\frac{V_{solute}}{V_{water}} = \frac{\theta}{1 + \rho K_d / \theta}, \tag{2}$$

where V_{solute} and V_{water} are the volumes at 50% solute breakthrough and water breakthrough, respectively, ρ is the density of the soil and θ its volumetric water content. The denominator in this equation is often referred to as the retardation factor R

$$R = 1 + \rho K_d / \theta. \tag{3}$$

The retardation factor is commonly used in the advection-dispersion equations included in contaminant transport calculations and occurs in the denominator of a dispersion term. Higher values of K_d indicate stronger interactions between the contaminant and soil and slower transport. The effect of K_d on the amount of material released and transported increases with the reaction time. The distribution coefficient can be written as

$$K_d = \frac{C_{sorbed} (g_{contaminant} / g_{soil})}{C_{test\ solution} (g_{contaminant} / mL_{test\ solution})}, \tag{4}$$

where C_{sorbed} is the concentration of the contaminant that has been taken up by the soil and $C_{solution}$ is the concentration of the contaminant remaining in solution. In the batch tests, soil and a solution containing a known amount of contaminant are shaken together and left to react for a period of time sufficient for the system to equilibrate. In this report, the initial solution is referred to as the leachant to distinguish it from the solution recovered at the end of the test, which is referred to as the test solution. The value of K_d is determined from the initial contaminant concentration (mass) in the leachant and concentration (mass) of the contaminant in the test solution. Equation 4 can be rearranged to give Equation 5:

$$K_d = \frac{C_{leachant} \bullet V_{leachant}}{C_{test\ solution} \bullet M_{soil}} - \frac{V_{test\ solution}}{M_{soil}}. \tag{5}$$

Since K_d is the ratio of two measured concentrations ($C_{leachant}$ and $C_{test\ solution}$), small errors in measurement can lead to a large error in the ratio. Values of K_d can also be inferred from field tests and field modeling studies using inverse modeling methods.

2

1.2   ASTM C1733 Batch Test Method

Due to the many process that affect the distribution coefficient, the use of generic K_d values for particular contaminants and soil types is strongly discouraged in the procedure, as inappropriate values can greatly impact contaminant migration and site-remediation analyses (e.g., Yu *et al* 2001). In current practice, bounding values of K_d are often used to provide conservative estimates to evaluate remediation needs. Conservatively low values of K_d are used when groundwater contamination is of concern and conservatively high values are used when soil contamination is of concern. Nevertheless, measurements made under site-specific conditions are considered to be essential for defensible assessments. The ASTM C1733 recommends careful collection of groundwater and soils, stabilization during transport to the laboratory and storage, and detailed characterization of the groundwater and soil mineralogy. While the test method itself is simple to perform, the sensitivity of test response to condition of the solid and the groundwater chemistry requires the utmost care during sample collection, handling, and storage for representative results.

Likewise, the usefulness of the measured values is greatly increased if dependencies of the K_d value on key environmental variables can be quantified, probably most importantly the contaminant concentration in the groundwater. For example, the method recommends that measurements be made for a range of contamination concentrations that can be used to develop an analytical relationship between the contaminant concentration and the K_d value. This is usually quantified in the form of an adsorption isotherm, although the contributions of other processes besides adsorption are taken into account in the (empirical) measured value. While the dependence on the contaminant concentration is probably the most important relationship for remediation analyses, additional relationships can be determined between K_d values and other environmental factors, such as pH, T, and Eh (groundwater redox), the presence of complexants and competing species, ionic strength, etc. Other than stressing the importance of simulating the natural conditions, ASTM C1733 does not address these other dependencies. Though it is not addressed in ASTM C1733, the reversibility of contaminant uptake can be measured following the same procedure to provide additional insight into the dominant process.

The ASTM C1733 method recommends that tests be conducted at reference pH and temperature values and at a reference water-to-soil mass ratio to facilitate comparisons of other contaminants and soils, but only provides the recommended water-to-soil mass ratio. Since K_d is usually considered to be an equilibrium value, the method calls for the use of scoping tests to determine the duration required to reach a constant solution concentration indicating the system has reached equilibrium (or steady state). Since this will vary with the system being evaluated, it is left to the user to determine the appropriate test duration. One objective of the research presented in this report is to provide a set of reference test conditions that can be included in ASTM C1733 for direct comparison of results obtained at different laboratories.

Because the purpose of this study was to evaluate repeatability of the test method and sensitivity to test parameters exclusive of the uncertainties inherent in collected soil and groundwater, a sample of San Joaquin soil was purchased from the National Institute for Standards and Technology (NIST) and used as the geologic medium. This is a dried soil that has been crushed, sized, and well mixed. The gross chemical composition has been certified as uniform in the 50-g subsamples provided as SRM 2709a (see NIST 2011 and Appendix A). An ASTM synthetic silicate groundwater spiked with small amounts of either CsCl or $Sr(NO_3)_2$ was used to represent contaminated groundwater.

2.   EXPERIMENTAL APPROACH

2.1   Soil Sample

San Joaquin Soil supplied by the National Institute of Standards and Technology was used as the solid: Standard Reference Material 2709a (NIST 2009). The gross composition of SRM 2709a has been standardized, with the concentrations of 19 constituent elements being certified, reference values for 15 elements, and information values provided for 10 constituents. (The elemental compositions provided with SRM 2709a are included in Appendix A.) The soil is provided as a finely divided powder that passes through a 200 mesh sieve (75 μm openings). This material was selected, in part, based on its availability and the intent of using this SRM in a future inter-laboratory study to further evaluate the ASTM C1733 method. Although the mineralogy of the soil is not standardized, it is assumed that the consistency of the composition reflects consistency in the mineral constituents. Soil from two 50-g bottles of SRM 2709a was used in the study. These are referred to as Jar 1 and Jar 2. The source for each test is indicated in the prefix of the test number as SJ1 and SJ2 for soil taken from Jar 1 and Jar 2, respectively.

2.2   Solution Preparation

To mimic a generic groundwater, a silicate solution containing sodium bicarbonate and silicic acid was used to prepare all of the cesium and strontium solutions. The silicate solution was prepared following the method outlined by ASTM Standard Procedure C1220 in step 7.4. All of the solid reagents were dried at about 150 °C prior to use, and freshly demineralized water (17.8 MΩ cm) was used. Four liters of the silicate solution was prepared in two 2-liter Nalgene bottles. In each bottle, 0.358 g $SiO_2 \cdot 2H_2O$ and 0.192 g $NaHCO_3$ were mixed with enough demineralized water to make 2 kg solution. Small amounts of dilute HNO_3 and NaOH solutions were then added to adjust the solutions in the two bottles to pH 8.99 and pH 8.98, respectively. The solution pH was measured with a combination electrode that was calibrated with pH 3, 7, and 10 buffers before and after the solutions were analyzed.

An approximately 0.02 $\underline{m}$ CsCl stock solution was prepared by adding 0.3351 g CsCl to the silicate solution to make 100.02 g of solution. [Molality ($\underline{m}$) is defined as moles solute per kg solution.] This solution was diluted to prepare two other solutions:
- 9.99 g of the 0.02 $\underline{m}$ CsCl solution and 990.00 g of sodium silicate solution were mixed to make 999.99 g of the 1×10^{-4} $\underline{m}$ CsCl solution, which is referred to as Cs solution A.
- 5.00 g of the 0.02 $\underline{m}$ CsCl solution and 97.97 g of silicate solution were mixed to make 102.97 g of a 5×10^{-4} $\underline{m}$ CsCl solution. This is referred to as Cs solution B.

Aliquots of solutions A and B were diluted to make other CsCl solutions for use in the series of tests to study the effects of the Cs concentration:

- 50.00 g of the 1×10^{-4} $\underline{m}$ solution and 51.80 g of the sodium silicate solution were mixed to make 101.80 g of the 5×10^{-5} $\underline{m}$ CsCl solution. This is referred to as Cs solution C.
- 10.02 g of the 1×10^{-4} $\underline{m}$ solution and 90.11 g of the sodium silicate solution were mixed to make 100.13 g of the 1×10^{-5} $\underline{m}$ CsCl solution. This is referred to as Cs solution D.
- 1.00 g of the 1×10^{-4} $\underline{m}$ solution and 99.00 g of the sodium silicate solution were mixed to make 100.00 g of the 1×10^{-6} $\underline{m}$ CsCl solution. This is referred to as Cs solution E.
- 5.01 g of the 5×10^{-4} $\underline{m}$ solution and 94.97 g of the sodium silicate solution were mixed to make 100.01 g of the 2.5×10^{-5} $\underline{m}$ CsCl solution. This is referred to as Cs solution F.

The CsCl solutions C, D, E, and F were then adjusted to pH 8.5 by the addition of small amounts of HNO_3 or NaOH to separate the effects of solution pH from Cs concentration. A portion of the A solution was also adjusted to pH 8.5 by adding a small amount of HNO_3; the pH-adjusted solution is referred to as A2.

The strontium solutions were prepared following the same procedure. An approximately 0.02 $\underline{m}$ $Sr(NO_3)_2$ solution was prepared by adding 0.4279 g $Sr(NO_3)_2$ to the silicate solution to make 100.03 g final solution. This solution was diluted to prepare the other Sr solutions:

- 10.60 g of the 0.02 $\underline{m}$ $Sr(NO_3)_2$ solution and 990.07 g of sodium silicate solution were mixed to make 1000.67 g of the 1×10^{-4} $\underline{m}$ $Sr(NO_3)_2$ solution, which is referred to as Sr solution A.
- 5.01 g of the 0.02 $\underline{m}$ $Sr(NO_3)_2$ solution and 95.04 g of silicate solution were mixed to make 100.05 g of a 5×10^{-4} $\underline{m}$ $Sr(NO_3)_2$ solution. This is referred to as Sr solution B.

Aliquots of solutions A and B were diluted to make other $Sr(NO_3)_2$ solutions:

- 50.01 g of the 1×10^{-4} $\underline{m}$ solution and 50.02 g of the sodium silicate solution were mixed to make 100.03 g of the 5×10^{-5} $\underline{m}$ $Sr(NO_3)_2$ solution. This is referred to as Sr solution C.
- 10.05 g of the 1×10^{-4} $\underline{m}$ solution and 89.95 g of the sodium silicate solution were mixed to make 100.00 g of the 1×10^{-5} $\underline{m}$ $Sr(NO_3)_2$ solution. This is referred to as Sr solution D.
- 1.02 g of the 1×10^{-4} $\underline{m}$ solution and 100.48 g of the sodium silicate solution were mixed to make 101.50 g of the 1×10^{-6} $\underline{m}$ $Sr(NO_3)_2$ solution. This is referred to as Sr solution E.
- 5.00 g of the 5×10^{-4} $\underline{m}$ solution and 95.02 g of the sodium silicate solution were mixed to make 100.02 g of the 2.5×10^{-5} $\underline{m}$ $Sr(NO_3)_2$ solution. This is referred to as Sr solution F.

The $Sr(NO_3)_2$ solutions C, D, E, and F were adjusted to pH 8.5 by the addition of small amounts of HNO_3 or NaOH to separate the effects of solution pH from Sr concentration. A portion of the A solution was also adjusted to pH 8.5 by adding a small amount of HNO_3; the pH-adjusted solution is referred to as A2. The Cs and Sr concentrations in all of the final solutions were measured directly in the leachant blank series.

The densities of Cs solution A and Sr solution A were measured using a 100 mL volumetric flask as follows. The volume of the flask was measured first by filling to just below the line with demineralized water, equilibrating at 30.0 °C, and then adding enough water to raise the meniscus to line. The filled volumetric was weighed to determine the mass of water, and the volume calculated by using the known density of air-saturated water at 30.0 °C (0.99565 g/cm^3). The masses of Cs solution A and Sr solution A required to fill the volumetric were then measured at ambient temperature and divided by the known volume of the flask to calculate the density. The density of Cs solution A was determined to be 0.9946 g/cm^3 and that of Sr solution A 0.9966 g/cm^3. These values were used as the densities of the other Cs and Sr solutions, as well.

2.3   Vessel Preparation

Thirty 45-mL Teflon screw top vessels were cleaned for use in these tests. The vessels had been used in previous tests and had been cleaned with dilute NaOH solutions when new and then with dilute nitric acid solutions and water after use in other tests. The vessels were cleaned prior to use in these tests as follows. The vessels were subjected to three washes with demineralized water and then filled with demineralized water and 5 drops (about 1 mL) of concentrated nitric acid, capped, and shaken to help mix the acid and rinse the cap. The containers were then placed in a 70 °C oven overnight. The acid solution was

discarded the following morning and the vessels and caps rinsed three times with demineralized water. The vessels were then filled with demineralized water, capped, and placed in a 70 °C oven overnight. The water was discarded the following morning and the vessels were dried in the oven then capped for storage. The Teflon containers were labeled 1-30.

2.4   Test Method

Simple batch experiments were performed to determine how much cesium and strontium was taken up by the soil. Tests were conducted by placing a weighed amount of SRM 2709a soil in a clean Teflon vessel and adding a known mass of solution. The vessels were tightly sealed with screw-top lids, shaken to suspend the soil in the solution, and then placed in a convection oven that was set to 30.0 °C using a NIST-traceable thermometer. The test was considered to be initiated at the time it was placed in the oven, even though about 30 minutes were required to heat the vessel from ambient to 30.0 °C. The vessels were shaken to suspend the solids at the beginning and end of the work day. The termination time was taken to be the time at which the solution was removed from the vessel, even though the vessel had been removed from the oven several minutes earlier. Vessels were shaken to suspend the solids when they were removed from the oven, then allowed to settle for several minutes before the vessels were opened. The leachates were decanted from the test vessel into a syringe fitted with a 0.45 μm pore size polycarbonate syringe filter. A small amount of soil was entrained in the solution poured into the syringe. The solution was forced through the filter with a plunger and collected in a pre-weighed solution bottle. A small sample of each filtrate (about 1 mL) was removed from the solution bottle with a pipette for pH analysis. The solution pH was measured at ambient temperature using a combination microelectrode that had been calibrated with three commercial buffer solutions (pH 3, pH 7, and pH 10) prior to the measurements. The remaining filtrate was acidified with about 0.05 mL concentrated nitric acid and stored several days prior to analysis with inductively coupled plasma-mass spectrometry (ICP-MS). Blank tests conducted without soil were treated the same as tests with soil, including the filtration step. Solutions from each test series, including blank tests, were analyzed as a set to eliminate day-to-day variance in the performance of the ICP-MS. Solutions from some tests with the Cs and Sr solutions of similar concentrations were combined for analyses to lower the analytical costs.

2.5   Solution Analysis

All solutions were analyzed for Cs and Sr using inductively coupled plasma-mass spectrometry (ICP-MS). Some solutions were also analyzed for Na and Si. To reduce analytical costs, the solutions from some tests conducted with the Cs leachant were mixed with the solution from a corresponding test conducted with the Sr leachant and analyzed as a single sample. Each solution was stabilized with concentrated HNO_3 before mixing and the amounts of each solution were measured. Each leachant provides the same matrix (a dilute silicate solution near pH 8.7) and Cs and Sr do not have interferences in the ICP-MS analysis. Demineralized water was added to a few solutions (prior to acidification) to provide sufficient solution for analysis (at least 10 mL).

The pH was measured for samples of most solutions using a combination electrode. The pH values of commercial buffer standards were measured to calibrate the meter response before and after the test samples were analyzed to account for drift. The initially-measured pH values of the buffers were recorded but the meter was not adjusted. Instead, the as-measured values of the unknown solutions were measured and recorded, and then the buffers were reanalyzed. A calibration curve was determined later using the two sets of measured buffer pH values (i.e., before and after measuring the test solutions) and

this was used to calculate the pH values of the test solutions. This approach takes into account any drift in the pH meter that occurred during the measurements.

2.6   Test Series

Several series of tests were conducted to evaluate various aspects of the ASTM C1733 procedure. All tests were conducted at 30 °C. These are summarized below:

Soil Blank Tests—A series of tests was run with 12.5 g demineralized water and 0.5 g soil to measure the amounts of Cs and Sr released from the San Joaquin soil into solution during 1-, 3-, and 7-day test durations.

Leachant Blank Tests—A series of tests was run with 12.5 g of each leachant without soil for 1, 2 and 7 days to evaluate the stabilities of the solutions and any interactions with the Teflon vessels. The measured concentrations were used as background values for other tests.

Repeatability Tests—Series of 5 replicate tests were conducted for 1- and 2-day durations with the 1×10^{-4} m Cs A solution and for 1 day with the 1×10^{-4} m Sr A solution, all with 1 g soil and 25 g solution.

Effects of Scale Tests—A series of tests was conducted using the 1×10^{-4} m Cs A solution to study the effect of scale. Tests were conducted for 1 day durations with 0.2, 0.5, 1, and 1.5 g soil and the appropriate amount of solution to attain a mass ratio of 25 g solution-to-1 g solid. These tests address the homogeneity of the soil and test uncertainty associated with handling different amounts of solid and solution.

Effects of Duration Tests—A series of tests was conducted with the 1×10^{-4} m Cs A solution and the 1×10^{-4} m Sr A solution for durations between 15 minutes and 7days to track the approach to equilibrium (steady state). Tests were conducted at a mass ratio of 12.5 g solution and 0.5 g soil.

Effects of pH Tests—A series of tests was conducted with samples of the 1×10^{-4} m Cs A2 solution and the 1×10^{-4} m Sr A2 solution that were adjusted to pH values of 6.5, 7.0, 8.0, and 8.5. Tests were conducted for 1 and 2 days at a mass ratio of 12.5 g solution and 0.5 g soil.

Effects of Concentration Tests—Series of tests were conducted using six different Cs concentrations and six different Sr concentrations (i.e., the A2, B, C, D, E, and F solutions) for 1, 2, and 3 days at a mass ratio of 12.5 g solution and 0.5 g soil to quantify the effect of the solution concentration on K_d in the form of sorption isotherms.

2.7   Soil Characterization by XRD

Powder x-ray diffraction (XRD) was performed on a small sample of soil from Jar 1. Sections of the resulting spectrum are shown in Figure 1, and the measured values of two theta and intensity values for the major peaks are given in Table 1 with the calculated *d*-spacings, estimated background intensities, and calculated relative intensities. The *d*-spacings of the peaks were calculated using the Bragg diffraction equation (calculations are discussed in Appendix B) with an X-ray wavelength of $\lambda = 0.15406$ nm:

(a)

(b)

$I_o = 2314$
$2\theta = 26.63°$

(c)

(d)

(e)

(f)
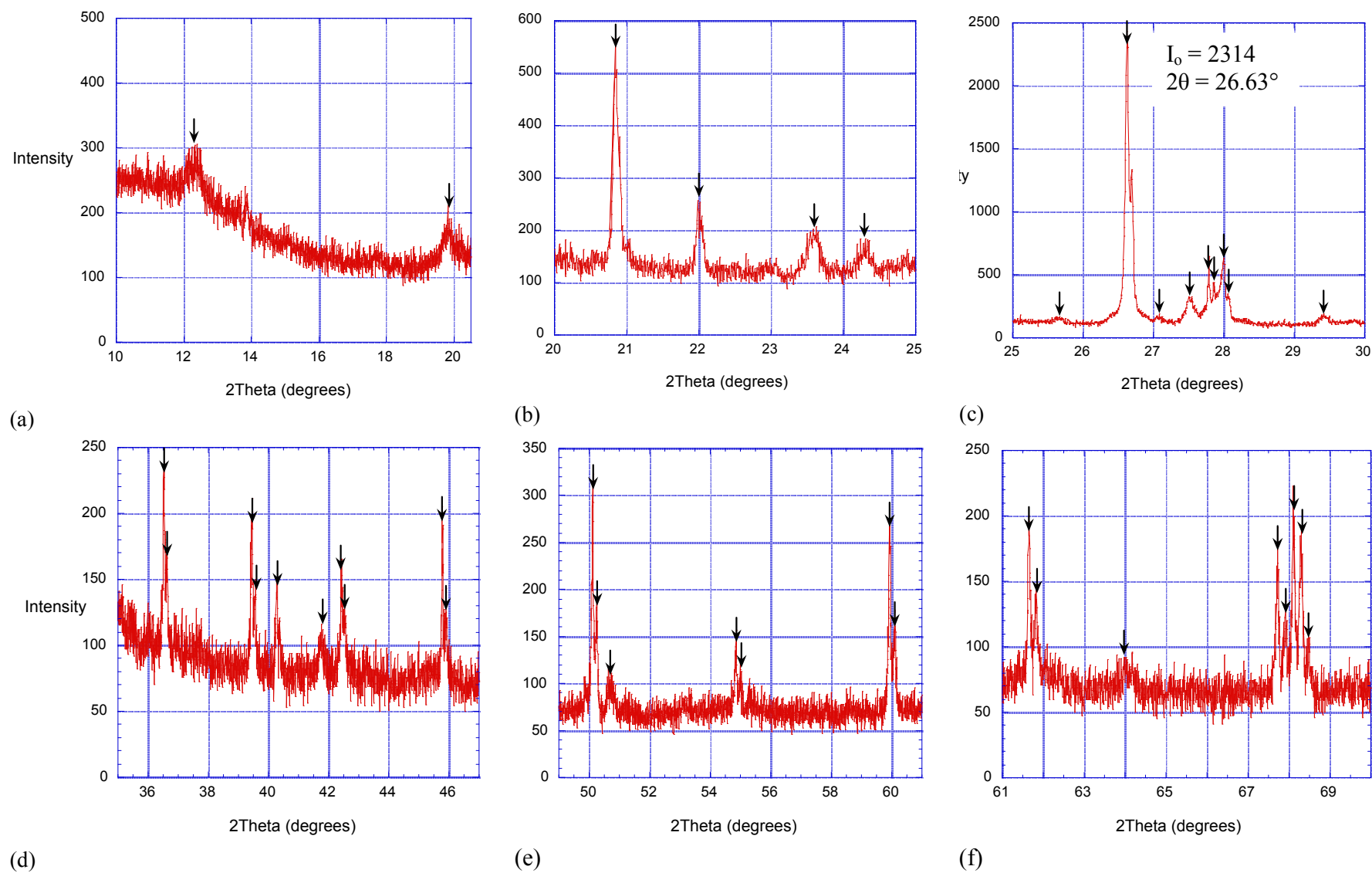
Figure 1. XRD results of the San Joaquin soil sample from Jar 1 showing major peaks. The arrows locate the peaks that are included in Table 1.

8

Table 1. Summary of peaks in XRD spectrum of NIST SRM 2709a

2 Theta	d-spacing, Å	I, relative	I, background	I/Io
12.317	7.180	302	229	3.2
19.819	4.476	211	124	3.8
20.842	4.259	567	131	18.8
21.983	4.040	257	122	5.8
23.580	3.770	201	116	3.7
24.312	3.658	184	123	2.6
25.627	3.473	169	121	2.1
26.630	3.345	2429	115	100
27.085	3.290	186	124	2.7
27.535	3.237	315	136	7.7
27.786	3.208	647	169	20.7
27.853	3.201	438	164	11.8
27.996	3.185	634	154	20.7
28.073	3.176	328	147	7.8
29.419	3.034	183	145	1.6
36.531	2.458	234	99	5.8
36.639	2.451	170	98	3.1
39.458	2.282	211	81	5.6
39.561	2.276	138	81	2.5
40.287	2.237	147	78	3.0
41.884	2.155	108	79	1.3
42.416	2.129	158	76	3.5
42.539	2.123	121	76	1.9
45.788	1.980	191	75	5.0
45.901	1.975	132	74	2.5
50.127	1.818	323	67	11.1
50.255	1.814	194	66	5.5
50.762	1.797	108	66	1.8
54.866	1.672	157	69	3.8
55.004	1.668	115	69	2.0
59.942	1.542	272	69	8.8
60.116	1.538	163	69	4.1
61.646	1.503	196	69	5.5
61.820	1.500	135	69	2.9
63.974	1.454	92	66	1.1
67.699	1.383	161	65	4.1
67.919	1.379	135	65	3.0
68.109	1.376	223	64	6.9
68.298	1.372	189	64	5.4
68.482	1.369	106	64	1.8

$$\lambda = 2d \sin\theta. \tag{6}$$

The most intense peak occurred at a two theta value of 26.63 degrees and had an intensity of 2429 with an estimated background of 115. The mineral identifications provided by the commercial software were not plausible (based on the known composition and absence of strong peaks expected for the suggested minerals) and no further analysis of the spectrum was done. Documentation of these results is presented for future comparisons with other samples of NIST SRM 2709a San Joaquin soil.

2.8   Soil Characterization by SEM

A small amount of soil from Jar 1 was examined using a scanning electron microscope (SEM) to document the range of particle sizes in the soil samples. The photomicrographs in Figure 3 show a few larger particles, but a preponderance of 1-micrometer-sized particles. Clearly, the small particles provide the vast majority of surface area in the tests. While sieving the as-received soil to better constrain the particle size was considered initially, this was not done for the following reasons. The ASTM C1733 procedure does not specify a constrained size range; rather, it identifies a typical upper size limit of 2 mm to exclude gravel. It is reasoned that the majority of the surface area is provided by smaller particles and excluding particles larger than 2 mm simplifies test performance without contributing significant error. Since the test is conducted on a mass-soil basis, the size fraction will affect the test response. Therefore, it is crucial that a representative sample be used to ensure that the test response is representative of the material of interest. For the present purpose, the samples used in the tests are assumed to represent all of the material in the jar. The validity of this assumption is determined, in part, by the measured effect of the sample size on the test response, which is discussed in Section 3.4. In addition, the range of particle sizes of soil in the jars used in these tests is assumed to be the same as that of soils in other jars of SRM 2709a. Although performing a sieve analysis to quantify the size fractions was considered, this was not done due to the limited amount of soil that was available and the primary focus of this project being evaluation of the test method.
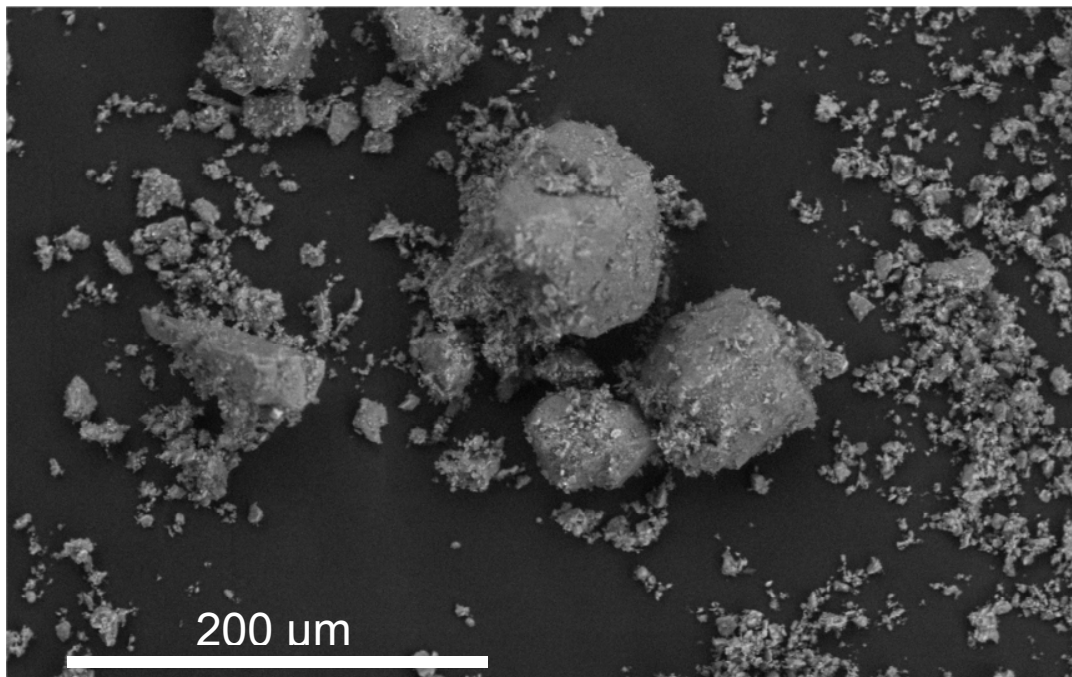
(a)

Figure 2. SEM photomicrographs of the San Joaquin soil from Jar 1 at (a) low, (b) moderate, and (c) high magnification.
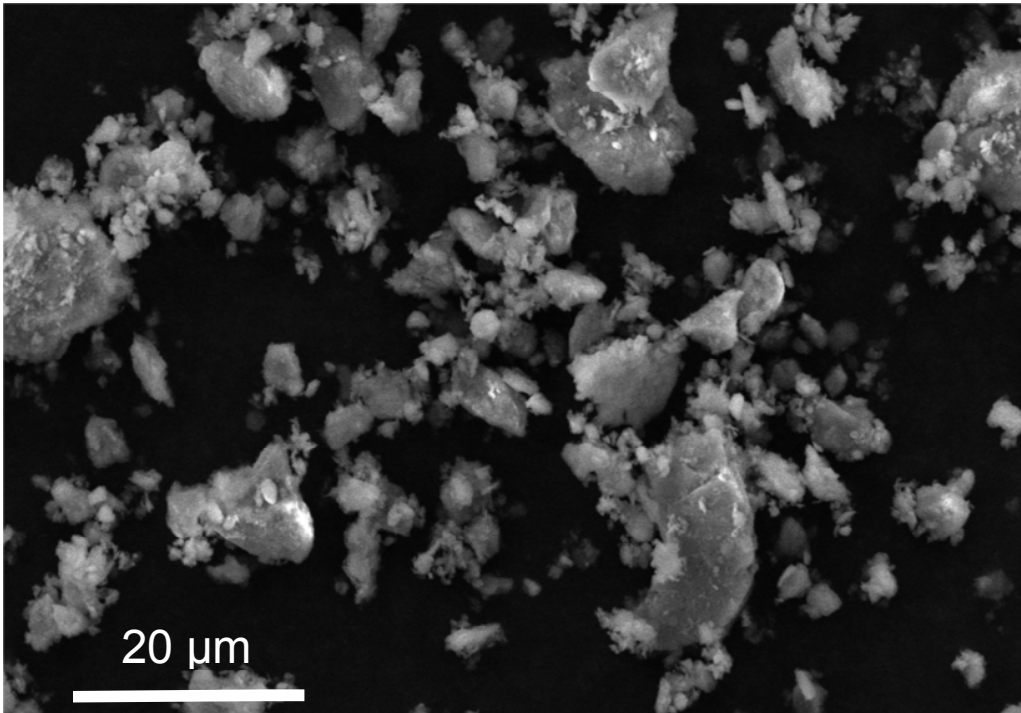
(b)
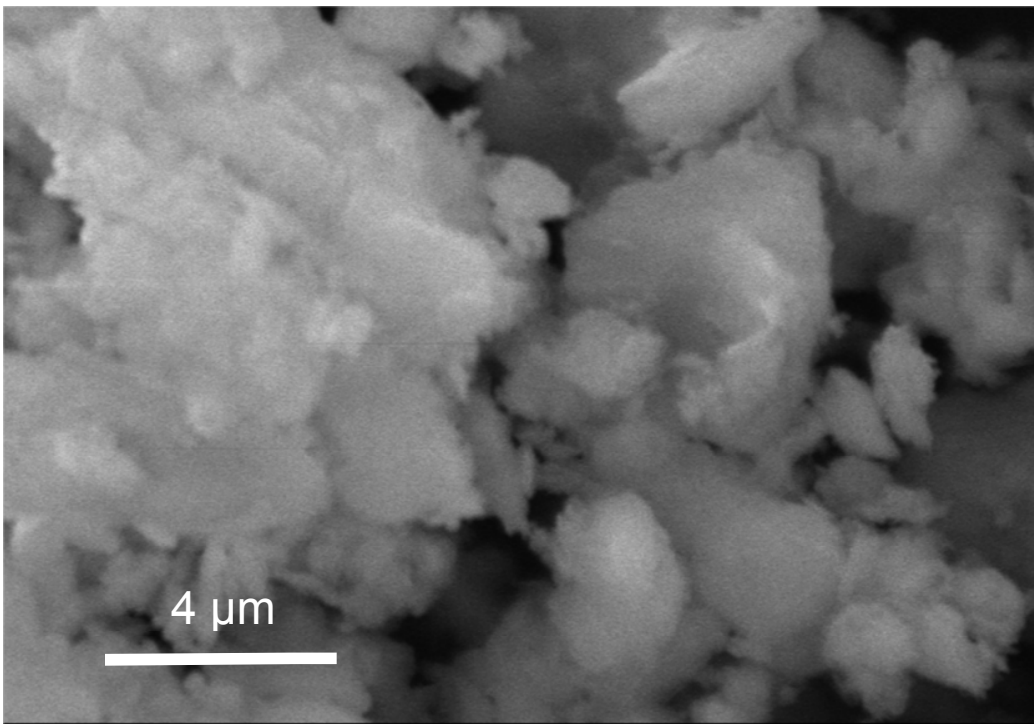
(c)

Figure 2. (cont.)

3.   TEST RESULTS

3.1   Soil Blank Tests

A series of blank tests were run to measure the amounts of Cs and Sr released from the soil into solution. Three tests were conducted with 0.50 g soil in 12.5 g demineralized water for about 1, 3, and 7 days. Soil from Jar 1 was used in these tests. The test data and results are given in Table 2. Calculations of final pH, dilution factors, and dilution-corrected concentrations are discussed in Appendix B. The average solution concentrations for the three soil blank tests are 0.2×10^{-9} molal Cs and 1.1×10^{-6} molal Sr. From these measured concentrations, averages of 0.77 ng Cs and 2.47 µg Sr were released per gram soil in the three soil blank tests. The release of Cs is negligible, but enough Sr is released to affect the test response.

Table 2. Data and results for soil blank tests

Test Number	Duration, h	Mass Soil, g	Mass DIW, g	Final pH
SJ1-Blank-0	71.6	0.50	12.59	8.09
SJ1-Blank-1	23.6	0.50	12.55	8.04
SJ1-Blank-2	167.6	0.50	12.50	8.02

Test Number	Solution Bottle, g	+ test solution, g	+ DIW + HNO_3, g	Dilution Factor
SJ1-Blank-0	11.19	22.89	27.25	1.37265
SJ1-Blank-1	11.11	22.37	22.42	1.00444
SJ1-Blank-2	11.19	22.42	22.48	1.00534

Test Number	Measured Cs, µg/L	Measured Sr, µg/L	Dil.-corrected Cs, µg/L	Dil.-corrected Sr, µg/L
SJ1-Blank-0	0.0200	59.4	0.0275	81.5
SJ1-Blank-1	0.0242	89.0	0.0244	89.4
SJ1-Blank-2	0.0395	124	0.0397	125

3.2   Leachant Blank Tests

Three solution blank tests were conducted with the nominal 1×10^{-4} m Cs A solution and 1×10^{-4} m Sr A solution for 1, 2, and 7 days to evaluate the stability of the solutions. Single blank tests were later conducted with the leachants made with different Cs and Sr concentrations for duration of about 3 days. Those solutions were adjusted to a common pH value before use by adding small amounts of nitric acid or NaOH solutions. This was done to distinguish between the effects of time, concentration, and pH. The test data are provided in Table 3. The dilution factors listed in Table 3 are due to dilution of the test solution by adding nitric acid. The Cs and Sr test solutions from tests run for the same duration were combined for analysis. Table 4 lists the solutions that were combined and the dilution factors that resulted from mixing the solutions. Calculations of final pH, dilution factors, and dilution-corrected concentrations are discussed in Appendix B.

Table 3. Data and results for leachant blank tests

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Final pH
Cs-A-Blank-1	24.62	0	12.53	not measured
Cs-A-Blank-2	48.20	0	12.49	8.61
Cs-A-Blank-3	168.37	0	12.63	8.46
Sr-A-Blank-1	24.62	0	12.56	not measured
Sr-A-Blank-2	48.20	0	12.51	8.36
Sr-A-Blank-3	168.37	0	12.50	8.39
SJ1-Cs-A2-0	74.87	0	12.51	8.56
SJ1-Cs-B-0	71.63	0	12.50	8.53
SJ1-Cs-C-0	71.63	0	12.51	8.40
SJ1-Cs-D-0	71.63	0	12.50	8.47
SJ1-Cs-E-0	71.63	0	2.50	8.44
SJ1-Cs-F-0	71.63	0	12.50	8.48
SJ1-Sr-A2-0	74.87	0	12.51	8.56
SJ1-Sr-B-0	78.93	0	12.51	8.10
SJ1-Sr-C-0	78.93	0	12.50	8.52
SJ1-Sr-D-0	78.93	0	12.50	8.52
SJ1-Sr-E-0	78.93	0	12.51	8.50
SJ1-Sr-F-0	78.93	0	12.50	8.50
SJ1-Sr-F-0*	47.88	0	12.50	8.62

Test Number	Solution Bottle, g	Bottle + test solution, g	Bottle + test solution + DIW + HNO$_3$, g	Dilution Factor
Cs-A-Blank-1	11.09	23.32	23.36	1.003
Cs-A-Blank-2	11.16	22.99	23.03	1.003
Cs-A-Blank-3	11.12	23.10	23.16	1.005
Sr-A-Blank-1	11.10	23.35	23.40	1.004
Sr-A-Blank-2	11.09	22.94	22.98	1.003
Sr-A-Blank-3	11.12	23.02	23.09	1.006
SJ1-Cs-A2-0	11.16	22.97	23.03	1.005
SJ1-Cs-B-0	11.17	22.82	22.88	1.005
SJ1-Cs-C-0	11.14	23.03	23.11	1.007
SJ1-Cs-D-0	11.16	22.9	22.93	1.003
SJ1-Cs-E-0	11.15	22.89	22.93	1.003
SJ1-Cs-F-0	11.19	22.94	23.00	1.005
SJ1-Sr-A2-0	11.16	22.97	23.03	1.005
SJ1-Sr-B-0	11.1	22.73	22.78	1.004
SJ1-Sr-C-0	11.21	22.87	22.94	1.006
SJ1-Sr-D-0	11.19	22.87	22.94	1.006
SJ1-Sr-E-0	11.17	22.91	22.94	1.003
SJ1-Sr-F-0	11.24	22.98	23.02	1.003
SJ1-Sr-F-0*	11.11	22.82	22.92	1.009

The solution results for the leachant blank tests are summarized in Table 5. These results show the Cs concentration to remain constant in blanks conducted through 7 days, but the Sr concentrations to decrease with time. The Cs solutions made for the concentration study were about 50% higher than the target concentrations and the Sr solutions were about 30% lower. The reason for this is not known, but the measured concentrations were used to calculate the K_d values in tests with soil.

Table 4. Combination of leachant blank test solutions for analysis

Solution Number	Solution 1	Solution 2
Cs/Sr-A-Blank-1	Cs-A-Blank-1	Sr-A-Blank-1
Cs/Sr-A-Blank-2	Cs-A-Blank-2	Sr-A-Blank-2
Cs/Sr-A-Blank-3	Cs-A-Blank-3	Sr-A-Blank-3
SJ-A2-0	SJ1-Cs-A2-0	SJ1-Sr-A2-0
SJ-B-0	SJ1-Cs-B-0	SJ1-Sr-B-0
SJ-C-0	SJ1-Cs-C-0	SJ1-Sr-C-0
SJ-D-0	SJ1-Cs-D-0	SJ1-Sr-D-0
SJ-E-0	SJ1-Cs-E-0	SJ1-Sr-E-0
SJ-F-0	SJ1-Cs-F-0	SJ1-Sr-F-0

Solution Number	Mixed Solutions Bottle, g	Bottle + Cs test solution, g	Bottle + Cs + Sr test solution, g	Cs Dilution Factor	Sr Dilution Factor
Cs/Sr-A-Blank-1	11.09	23.24	35.46	2.006	1.994
Cs/Sr-A-Blank-2	11.11	22.87	34.68	2.004	1.996
Cs/Sr-A-Blank-3	11.19	23.16	35.09	1.997	2.003
SJ-A2-0	11.12	22.98	34.80	1.997	2.003
SJ-B-0	11.16	22.79	34.34	1.993	2.007
SJ-C-0	11.18	23.11	34.77	1.977	2.023
SJ-D-0	11.19	22.86	34.57	2.003	1.997
SJ-E-0	11.19	22.88	34.6	2.003	1.997
SJ-F-0	11.23	22.98	34.66	1.994	2.006
SJ-F-0*	11.11	—	22.92	—	1.003

3.3   Repeatability Tests

Replicate tests were run using the 1.5×10^{-4} $\underline{m}$ Cs A solution and 0.50×10^{-4} $\underline{m}$ Sr A solution to measure the precision of the procedure for determining the K_d values following the ASTM C1733 procedure. Tests were conducted using 1.0 g soil and 25.0 g of solution. Five replicate tests were conducted with the Cs solution for both 1 and 2 days, and five replicate tests were conducted with the Sr solution for 1 day. All of the tests in the replicate test series were run with soil from the second container of San Joaquin soil and labeled with the prefix SJ2. Table 6 provides the test data. Note that the dilution factors due to the addition of concentrated HNO_3 are neglected in subsequent calculations.

The test solutions from 1-day tests with the Cs solution and with the Sr solution with the same test indices were combined for analyses. The test solutions from the 2-day tests with the Cs solution were analyzed without mixing. Data for the combined solutions are given in Table 7. The mass of the empty solution bottle is given and then the accumulated masses of solution 1 and solution 2 are given. The solution masses were calculated by difference. The dilution factors due to mixing the solutions were calculated and are provided in the table. Calculations are discussed in Appendix B.

The analytical results for the combined and single solutions are summarized in Table 8. These results indicate that an error occurred when the tests were initiated, namely, that the Cs A solution was used in the 1-day Sr tests and the Sr A solution was used in the 2-day Cs tests. (The Cs A solution was used correctly in the 1-day Cs tests.) The evidence is clear: First, no Cs was detected in SJ2-A-Y-6 through -10 but a significant concentration of Sr was measured. The Sr concentration is similar to that measured in other tests conducted with the same Sr solution used in this test series. Second, the Sr concentrations

Table 5. Results for leachant blank tests

Test Number	Measured Cs, µg/L	Dil.-corrected Cs, µg/L	Measured Sr, µg/L	Dil.-corrected Sr, µg/L
Cs/Sr-A-Blank-1	12500	25070	5450	10900
Cs/Sr-A-Blank-2	12600	25250	3810	7600
Cs/Sr-A-Blank-3	12500	24960	2950	5910
SJ-A2-0	12300	24600	7360	14745
SJ-B-0	63200	125965	18000	36125
SJ-C-0	6140	12141	3860	7809
SJ-D-0	1260	2524	772	1541
SJ-E-0	135	270	82.8	165
SJ-F-0	3140	6261	—	—
SJ-F-0*	—	—	3830	3840

Test Number	Mass Cs in test solution, µg	Measured Cs concentration, molal	Targeted Cs concentration, molal
Cs-A-Blank-1	316	1.49×10^{-4}	1.0×10^{-4}
Cs-A-Blank-2	317	1.50×10^{-4}	1.0×10^{-4}
Cs-A-Blank-3	317	1.48×10^{-4}	1.0×10^{-4}
SJ1-Cs-A2-0	309	1.46×10^{-4}	1.0×10^{-4}
SJ1-Cs-B-0	1583	7.48×10^{-4}	5.0×10^{-4}
SJ1-Cs-C-0	152.7	7.21×10^{-5}	5.0×10^{-5}
SJ1-Cs-D-0	31.7	1.50×10^{-5}	1.0×10^{-5}
SJ1-Cs-E-0	3.40	1.60×10^{-6}	1.0×10^{-6}
SJ1-Cs-F-0	78.69	3.72×10^{-5}	2.50×10^{-5}

Test Number	Mass Sr in test solution, µg	Measured Sr concentration, molal	Targeted Sr concentration, molal
Sr-A-Blank-1	137	5.14×10^{-5}	1.0×10^{-4}
Sr-A-Blank-2	95.5	3.59×10^{-5}	1.0×10^{-4}
Sr-A-Blank-3	74.1	2.79×10^{-5}	1.0×10^{-4}
SJ1-Sr-A2-0	186.24	6.97×10^{-5}	1.0×10^{-4}
SJ1-Sr-B-0	454.62	1.71×10^{-4}	5.0×10^{-4}
SJ1-Sr-C-0	99.09	3.69×10^{-5}	5.0×10^{-5}
SJ1-Sr-D-0	20.47	7.28×10^{-6}	1.0×10^{-5}
SJ1-Sr-E-0	3.22	7.80×10^{-7}	1.0×10^{-6}
SJ1-Sr-F-0	—	—	2.50×10^{-5}
SJ1-Sr-F-0*	49.18	1.81×10^{-5}	2.50×10^{-5}

measured in SJ2-A-Y-1 are consistent with the Sr concentrations measured in the soil blank tests. The results for tests SJ2-Cs-A-Y-6 through -10 are interpreted as being conducted with the Sr solution. The test numbers were retained to maintain traceability of test results. The measured Cs concentrations in the combined solutions SJ2-A-Y-1 through -5 reflect the combined results of tests conducted for 1 and 2 days. Although amounts cannot be attributed to each test solution, the volumes of each solution in the mixture are nearly equal and the test conditions are the same except for the reaction time. Other tests discussed below indicate that the test response is not sensitive to time between 1 and 2 days, so the combined solution provides a reliable measure of the repeatability of the test. Therefore, the results for the combined 1-day and 2-day test solutions are used with the added leachant and solid masses to calculate the distribution coefficient for Cs. Calculations are discussed in Appendix B. Propagation of uncertainties used to estimate uncertainty for plotted results is discussed in Appendix C.

Table 6. Data for Replicate Tests

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Final pH
SJ2-Cs-A-Y-1	24.02	1.00	25.00	8.61
SJ2-Cs-A-Y-2	24.02	1.00	25.00	8.48
SJ2-Cs-A-Y-3	24.02	1.00	25.00	8.45
SJ2-Cs-A-Y-4	24.02	1.00	25.02	8.47
SJ2-Cs-A-Y-5	24.02	1.00	25.00	8.49
SJ2-Sr-A-Y-1[a]	24.02	1.00	25.00	8.45
SJ2-Sr-A-Y-2[a]	24.02	1.00	25.00	8.44
SJ2-Sr-A-Y-3[a]	24.02	1.00	25.02	8.43
SJ2-Sr-A-Y-4[a]	24.02	1.00	25.00	8.43
SJ2-Sr-A-Y-5[a]	24.02	1.00	25.04	8.44
SJ2-Cs-A-Y-6[b]	48.00	1.00	25.04	8.27
SJ2-Cs-A-Y-7[b]	48.00	1.00	25.00	8.29
SJ2-Cs-A-Y-8[b]	48.00	1.00	25.00	8.29
SJ2-Cs-A-Y-9[b]	48.00	1.00	25.01	8.29
SJ2-Cs-A-Y-10[b]	48.00	1.00	25.00	8.42

Test Number	Solution Bottle, g	Bottle + test solution, g	Bottle + test solution + DIW + HNO$_3$, g	Dilution Factor
SJ2-Cs-A-Y-1	11.12	34.31	34.35	1.002
SJ2-Cs-A-Y-2	11.26	34.41	34.46	1.002
SJ2-Cs-A-Y-3	11.18	34.26	34.33	1.003
SJ2-Cs-A-Y-4	11.21	34.36	34.45	1.004
SJ2-Cs-A-Y-5	11.17	34.28	34.36	1.003
SJ2-Sr-A-Y-1[a]	11.19	34.19	34.26	1.003
SJ2-Sr-A-Y-2[a]	11.20	34.24	34.34	1.004
SJ2-Sr-A-Y-3[a]	11.18	34.15	34.22	1.003
SJ2-Sr-A-Y-4[a]	11.19	34.07	34.14	1.003
SJ2-Sr-A-Y-5[a]	11.17	34.28	34.33	1.002
SJ2-Cs-A-Y-6[b]	11.17	19.96	20.01	1.006
SJ2-Cs-A-Y-7[b]	11.20	24.02	24.08	1.005
SJ2-Cs-A-Y-8[b]	11.16	34.10	34.17	1.003
SJ2-Cs-A-Y-9[b]	11.19	34.19	34.26	1.003
SJ2-Cs-A-Y-10[b]	11.19	34.28	34.33	1.002

[a] Test was inadvertently conducted using Cs A solution.
[b] Test was inadvertently conducted using Sr A solution.

Mean values are given for the amounts of Cs and Sr associated with the soil and in solution and for K_d values calculated from the replicate tests. Standard deviation values and relative standard deviation values are given of the solution concentration and K_d values, but not for the amounts on the soil. This is because the amounts taken up by the soil are calculated as the difference between the masses calculated from the concentrations in the initial solution (for which single values for Cs and Sr were used in all calculations) and each test solution. The average values from three blank test analyses are used to calculate the initial Cs and Sr contents in the initial solution (leachant). The relative standard deviations for the contaminant concentrations in the test solutions are less than the 10% uncertainty that is assumed for each measurement. Likewise, the precision of the replicate K_d measurements are within the propagated analytical uncertainty of about 15% for each value.

Table 7. Combination of replicate test solutions for analysis

Solution Number	Solution 1	Solution 2
SJ2-A-Y-1	SJ2-Cs-A-Y-1	SJ2-Sr-A-Y-1[a]
SJ2-A-Y-2	SJ2-Cs-A-Y-2	SJ2-Sr-A-Y-2[a]
SJ2-A-Y-3	SJ2-Cs-A-Y-3	SJ2-Sr-A-Y-3[a]
SJ2-A-Y-4	SJ2-Cs-A-Y-4	SJ2-Sr-A-Y-4[a]
SJ2-A-Y-5	SJ2-Cs-A-Y-5	SJ2-Sr-A-Y-5[a]
SJ2-A-Y-6	SJ2-Cs-A-Y-6[b]	—
SJ2-A-Y-7	SJ2-Cs-A-Y-7[b]	—
SJ2-A-Y-8	SJ2-Cs-A-Y-8[b]	—
SJ2-A-Y-9	SJ2-Cs-A-Y-9[b]	—
SJ2-A-Y-10	SJ2-Cs-A-Y-10[b]	—

[a] Test was inadvertently conducted using Cs A solution.

[b] Test was inadvertently conducted using Sr A solution.

Solution Number	Mixed Solution Bottle, g	Bottle + Solution 1, g	Bottle + 1 + Solution 2, g	Solution 1 Dilution Factor	Solution 2 Dilution Factor
SJ2-A-Y-1	11.19	22.68	34.35	2.016	1.985
SJ2-A-Y-2	11.17	22.50	33.95	2.011	1.990
SJ2-A-Y-3	11.16	22.88	35.41	2.069	1.935
SJ2-A-Y-4	11.21	22.94	34.27	1.966	2.035
SJ2-A-Y-5	11.21	22.43	33.71	2.008	1.992
SJ2-A-Y-6	11.17	19.96	—	1.006	—
SJ2-A-Y-7	11.20	24.02	—	1.005	—
SJ2-A-Y-8	11.16	34.10	—	1.003	—
SJ2-A-Y-9	11.19	34.19	—	1.003	—
SJ2-A-Y-10	11.19	34.28	—	1.002	—

Figures 3a and 4a compare the amounts of Cs and Sr in the leachant with the masses taken up by the soil and remaining in solution, respectively, and Figures 3b and 4b compare the concentrations taken up by the soil and remaining in the solution, and the calculated K_d values. The variance and uncertainties in the amounts of Cs or Sr taken up by the soil and the K_d values are due to uncertainties in the leachant concentrations and the solution concentrations. As emphasized by the bar graphs, the calculated uptakes of both elements are the differences between large and small numbers. K_d is the ratio of a large number divided by a small number. The propagated uncertainties in the calculation of K_d is discussed in Appendix C. As displayed in Figures 3a and 4a, the uncertainties in the masses of Cs and Sr on the soil and in the solution in each test are greater than the variance between tests. Likewise, the variance in the calculated concentrations on the soil and K_d values for the replicate tests lie within the uncertainties of the values calculated for the individual tests.

The mean value of K_d(Cs) is 521.60 mL/g with a standard deviation (1 σ) of 16.45 mL/g, which gives a relative standard deviation of 3.13%. The mean value of K_d(Sr) is 162.34 mL/g with a standard deviation (1 σ) of 8.34 mL/g, which gives a relative standard deviation of 5.13%. The repeatability of the tests with a range defined by ± 2σ lies within the propagated uncertainties for both Cs and Sr.

Table 8. Results for replicate tests

Solution Number	Measured Cs, μg/L	Measured Sr, μg/L	Dil.-corrected Cs in test solution, μg/L	Dil.-corrected Sr in test solution, μg/L
SJ2-A-Y-1	1200	79.2	1200	157
SJ2-A-Y-2	1140	77.1	1140	153
SJ2-A-Y-3	1170	82.6	1170	160
SJ2-A-Y-4	1120	82.0	1120	167
SJ2-A-Y-5	1120	79.6	1120	159
SJ2-A-Y-6[b]	<0.2	1010	0	1013
SJ2-A-Y-7[b]	<0.2	1090	0	1095
SJ2-A-Y-8[b]	<0.2	1070	0	1073
SJ2-A-Y-9[b]	<0.2	1090	0	1093
SJ2-A-Y-10	<0.2	1140	0	1142

Solution Number	Mass Cs in test solution, μg	Mass Sr in test solution, μg	Leachant Volume, mL	Mass Cs in Leachant, μg	Mass Sr in Leachant, μg
SJ2-A-Y-1	60.06	—	50.048	1256	—
SJ2-A-Y-2	57.06	—	50.048	1256	—
SJ2-A-Y-3	58.58	—	50.068	1256	—
SJ2-A-Y-4	56.08	—	50.069	1256	—
SJ2-A-Y-5	56.10	—	50.088	1257	—
SJ2-A-Y-6	—	25.28	24.954	—	202.8
SJ2-A-Y-7	—	27.27	24.914	—	202.5
SJ2-A-Y-8	—	26.74	24.914	—	202.5
SJ2-A-Y-9	—	27.25	24.924	—	202.6
SJ2-A-Y-10	—	28.46	24.914	—	202.5

Test Numbers	Mass Cs on soil, μg	μg Cs/g soil	Cs in test solution, μg/mL	K_d (Cs), mL/g
SJ2-Cs-A-Y-1 + SJ2-Sr-A-Y-1[a]	1196	597.9	1.20	498.3
SJ2-Cs-A-Y-2 + SJ2-Sr-A-Y-2[a]	1199	599.4	1.14	525.8
SJ2-Cs-A-Y-3 + SJ2-Sr-A-Y-3[a]	1198	598.9	1.17	511.9
SJ2-Cs-A-Y-4 + SJ2-Sr-A-Y-4[a]	1200	600.2	1.12	535.9
SJ2-Cs-A-Y-5 + SJ2-Sr-A-Y-5[a]	1201	600.4	1.12	536.1
Mean		*599.42*	*1.150*	*521.60*
Standard Deviation		—	*0.035*	*16.45*
Relative Standard Deviation		—	*3.01%*	*3.13%*

[a] Test was inadvertently conducted using Cs A solution instead of Sr A solution.

Test Number	Mass Sr on soil, μg	μg Sr/g soil	Sr in test solution, μg/mL	K_d (Sr), mL/g
SJ2-Cs-A-Y-6[b]	178	178	1.01	175.2
SJ2-Cs-A-Y-7[b]	175	175	1.09	160.1
SJ2-Cs-A-Y-8[b]	176	176	1.07	163.8
SJ2-Cs-A-Y-9[b]	175	175	1.09	160.4
SJ2-Cs-A-Y-10[b]	174	174	1.14	152.3
Mean	*175.4*		*1.083*	*162.34*
Standard Deviation	—		*0.047*	*8.34*
Relative Standard Deviation	—		*4.32%*	*5.13%*

[b] Test was inadvertently conducted using Sr A solution instead of Cs A solution.
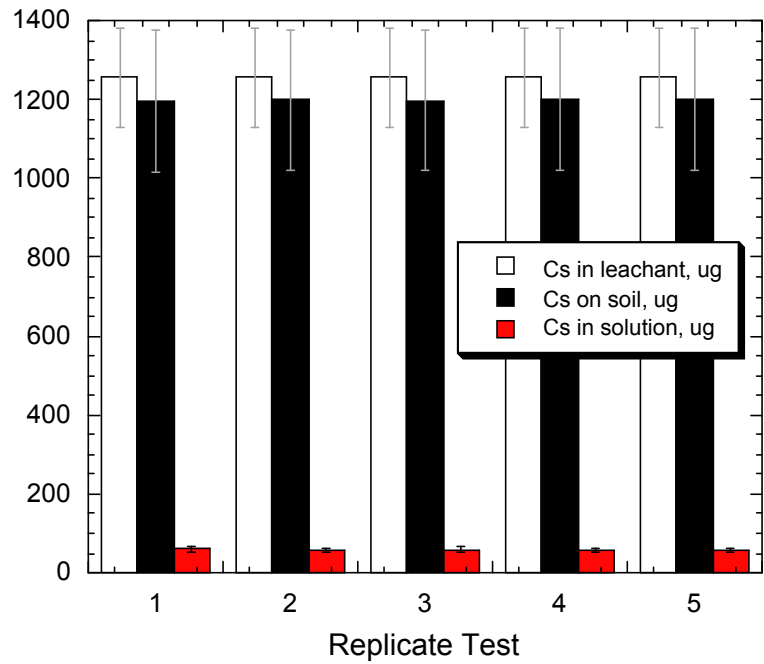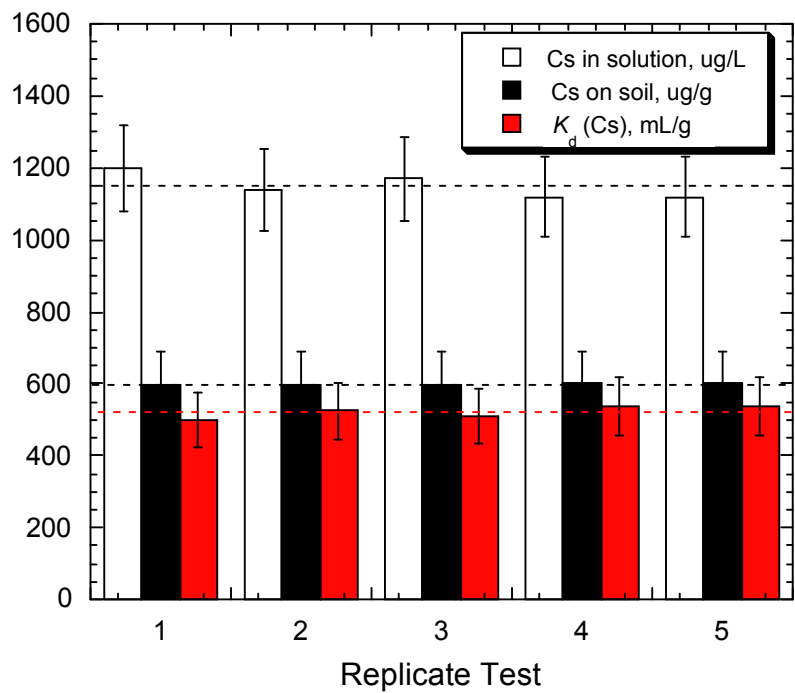
Figure 3. Results of replicate tests with Cs solution A: (a) masses of Cs in leachant, test solution, and taken up by soil, and (b) Cs concentrations in solution and on soil and K_d values. Dashed lines show mean values.
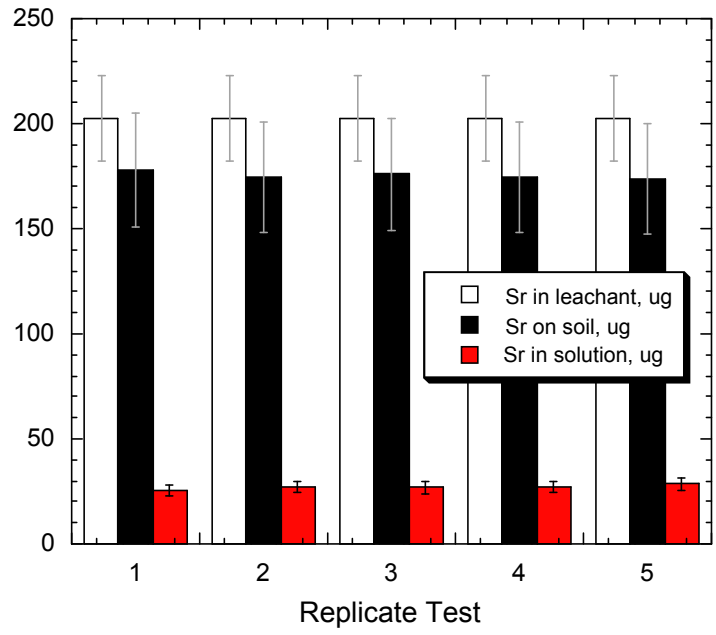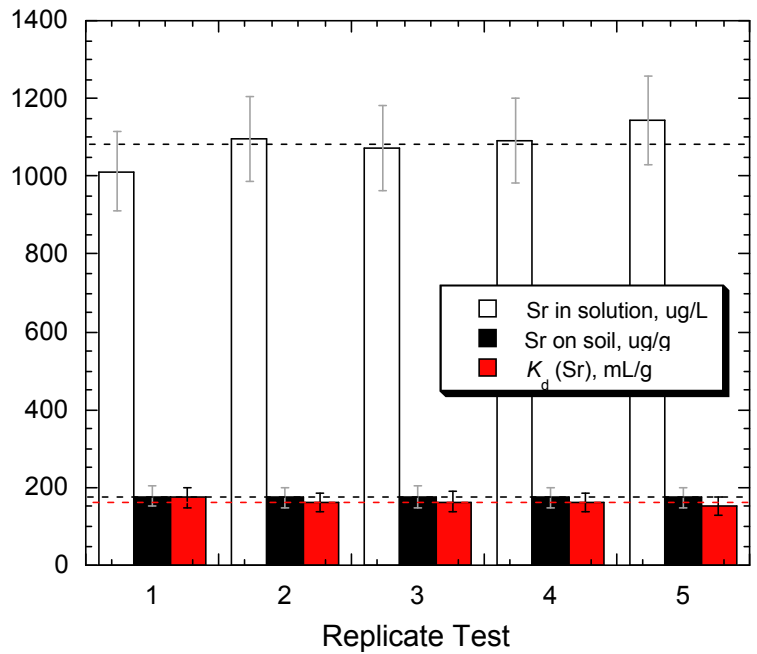
Figure 4. Results of replicate tests with Sr solution A: (a) masses of Sr in leachant, test solution, and taken up by soil, and (b) Sr concentrations in solution and on soil and K_d values. Dashed lines show mean values.

3.4 Effects of Scale Tests

A series of tests was conducted in which the total amounts of soil and solution varied but the ratio was constant at the ASTM C1733-specified ratio of 1 g soil and 25 g groundwater. Four different masses of soil were used: 0.20, 0.50, 1.00, and 1.50 g. Tests were conducted with the Cs A solution. Table 9 provides the test data and Table 10 gives the results. Calculations are discussed in Appendix B. The mean value of K_d(Cs) is 568.2.58 mL/g with a standard deviation of 34.64 mL/g. The standard deviation is 6.16% of the mean. (Excluding the test with 0.20 g soil changes the mean to 547.73 mL/g and reduces the standard deviation to 21.87 mL/g.) Figure 5 shows the K_d(Cs) values for all the soil masses with uncertainty bars drawn at 15% for the individual tests. The uncertainty in K_d(Cs) for the repeatability of the test based on the replicate tests is represented by the size of the diamonds. These results show a negative correlation between K_d(Cs) and the mass of soil. This is outside the uncertainty in the repeatability of the test method but within the uncertainties of the individual measurements.

Table 9. Data for tests conducted with different amounts of soil and solution

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Soil/Leachant mass ratio	Final pH
SJ2-Cs-A-Y-11	24.02	0.20	5.01	0.0399	8.48
SJ2-Cs-A-Y-12	24.02	0.50	12.50	0.0400	8.43
SJ2-Cs-A-Y-13	24.02	1.00	25.02	0.0400	8.67
SJ2-Cs-A-Y-14	24.02	1.50	37.50	0.0400	8.48

Test Number	Solution Bottle, g	Bottle + Cs solution, g	Bottle + Cs solution + DIW + HNO$_3$, g	Cs Dilution Factor
SJ2-Cs-A-Y-11	11.16	15.32	22.84	2.808
SJ2-Cs-A-Y-12	11.20	22.51	22.55	1.004
SJ2-Cs-A-Y-13	11.22	34.47	34.51	1.002
SJ2-Cs-A-Y-14	11.17	45.82	45.87	1.001

Table 10. Results for tests conducted with different amounts of soil and solution

Test Number	Measured Cs, µg/L	Dil.-corrected Cs in test solution, µg/L	Mass Cs in test solution, µg
SJ2-Cs-A-Y-11	356	999.5	5.035
SJ2-Cs-A-Y-12	1080	1085	13.63
SJ2-Cs-A-Y-13	1070	1073	27.00
SJ2-Cs-A-Y-14	1150	1154	43.49

Test Number	Leachant Volume, mL	Mass Cs in Leachant, µg	Mass Cs on soil, µg	µg Cs/g soil	K_d (Cs) mL/g
SJ2-Cs-A-Y-11	5.037	126.40	121.4	606.83	607.1
SJ2-Cs-A-Y-12	12.567	315.37	301.7	603.48	556.3
SJ2-Cs-A-Y-13	25.155	631.25	604.2	604.25	563.0
SJ2-Cs-A-Y-14	37.702	946.11	902.6	601.75	521.6
				Mean	*562.01*
				Standard Deviation	*34.12*
				Relative Standard Deviation	*6.07%*

The differences in the values for tests with different amounts of soil are all from differences in the measured solution concentration. The same value for the mass of Cs in the leachant is used in all calculations, so differences in the mass sorbed and K_d are due solely to differences in the measured solution concentrations. The uncertainty is slightly higher for the concentration measured in the 0.20 g test because that solution was diluted prior to analysis to provide sufficient solution. The data are plotted in Figure 5a to show almost all of the Cs is taken up by the soil and that the amount remaining in solution increases linearly with the mass of soil (and leachant) used in the test. The concentrations plotted in Figure 5b show a slight increase in Cs in the solution, whereas the concentration on the soil, which is calculated from the difference in Cs contents in the leachant and test solution, remains essentially constant. The decrease in the value of K_d(Cs) with increasing soil masses reflects the increase in the

(a)

(b)
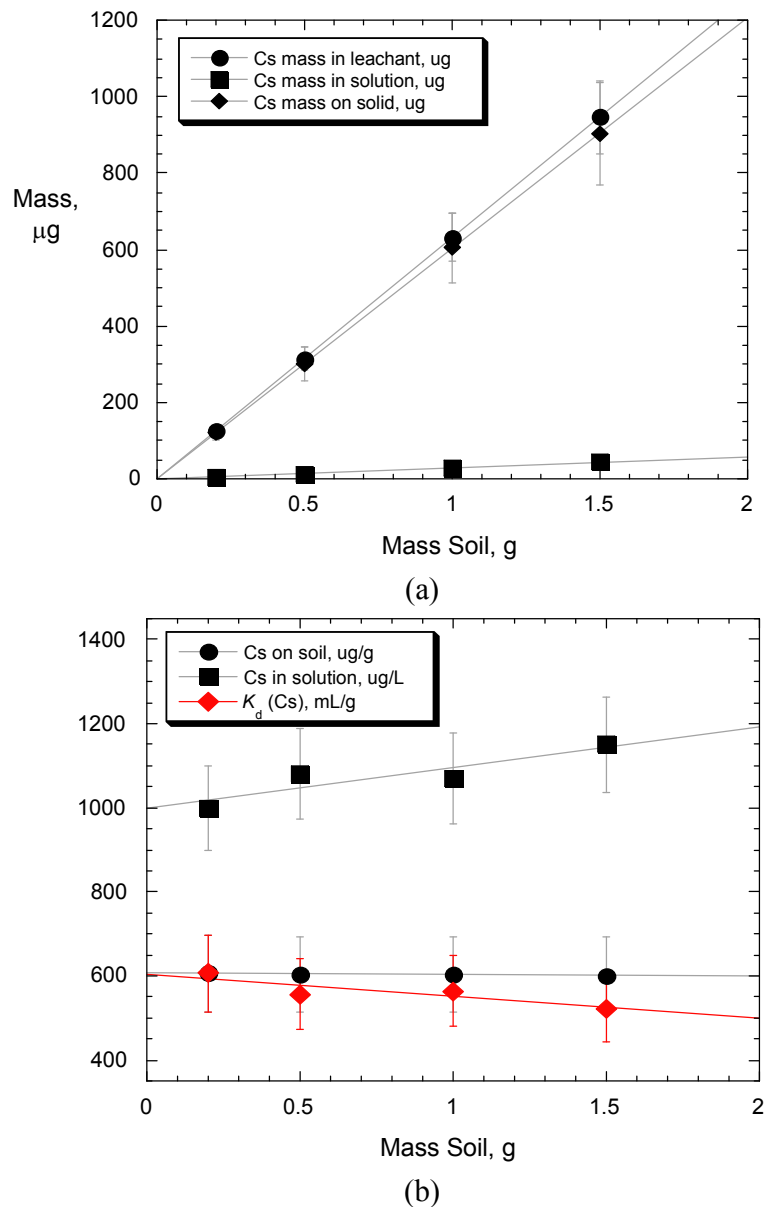
Figure 5. Results of tests with different amounts of soil and solution: (a) mass of Cs in leachant and in test solution or on soil vs. mass soil and (b) concentrations of Cs on soil and in solution, and K_d value, vs. mass soil.

solution concentrations. Figure 6 presents the results of the replicate tests on a bar graph. Note the 1000-fold difference in scale between the concentrations on the soil and in the test solution.
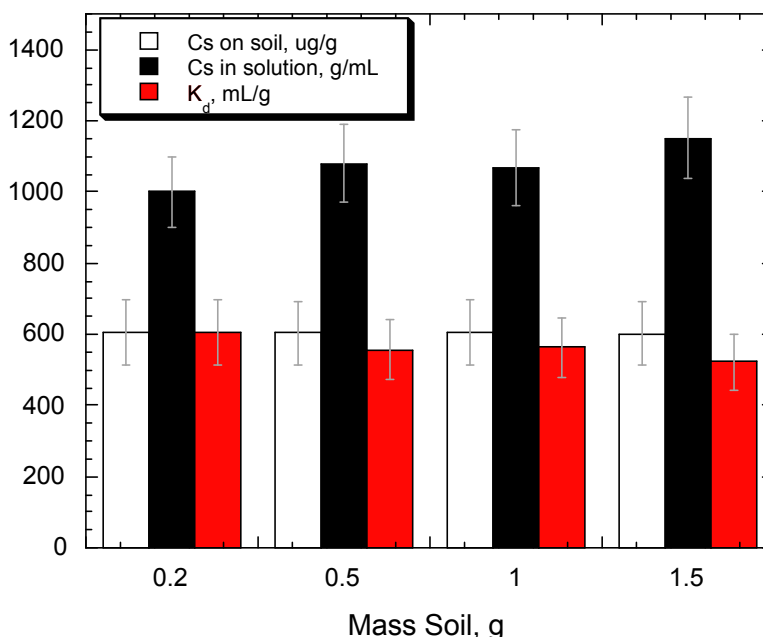
Figure 6. Results of tests with different amounts of soil and solution: comparison of concentrations of Cs on soil and in solution, and K_d values for different masses of soil

3.5   Effects of Duration Tests

A series of tests having durations between 15 minutes and about 7 days (167 hours and 46 minutes) was run to characterize the approach to equilibrium over time. The test data are provided in Table 11. Separate tests were conducted with about 0.5 g soil and 12.5 g of either the 1.4×10^{-4} m Cs A solution or the 0.50×10^{-4} m Sr A solution. Tests SJ1-Cs-A-1 through SJ1-Cs-A-10 were conducted using soil from Jar 1 and tests SJ2-Cs-A-11 through SJ2-Cs-A-14 were conducted about 3 weeks later after it was determined that equilibrium had been reached within 7 days. The latter tests were conducted using soil from Jar 2. The solutions from tests run with Cs and Sr solutions for the same duration were combined for analysis. The data for the combined solutions are given in Table 12. The results are given in Table 13. Calculations are discussed in Appendix A.

Figures 7 and 8 show the amounts of cesium and strontium sorbed per gram of soil and remaining in solution as a function of time, along with the K_d values calculated for each test. Uncertainty bars are drawn at 10% for the solution concentrations and at 15% for the sorbed concentrations and at 15% for the K_d values: 10% uncertainty is conservative for solution analyses, the sorbed concentrations are calculated as the difference of two measured numbers and K_d values are calculated from the difference and quotient of two measured numbers. These plots show equilibrium was reached in less than two hours. In Figures 6b and 7b, the uncertainty bars for K_d(Cs) and K_d(Sr) are drawn at 3% and 5%, respectively, to represent the repeatability measured with the replicate tests. This shows the variance in K_d(Cs) to be about twice the measured testing repeatability, although there is no correlation with the duration. The variance in K_d(Sr) was within the measured testing repeatability for tests longer than 10 hours.

Table 11. Data for tests conducted for different durations

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Soil/Leachant mass ratio	Final pH
SJ1-Cs-A-1	6.90	0.50	12.52	0.0399	not measured
SJ1-Cs-A-2	30.85	0.50	12.50	0.0400	8.32
SJ1-Cs-A-3	54.12	0.50	12.51	0.0400	8.27
SJ1-Cs-A-4	24.02	0.50	12.50	0.0400	not measured
SJ1-Cs-A-5	47.97	0.50	12.50	0.0400	8.32
SJ1-Cs-A-6	71.63	0.50	12.50	0.0400	8.37
SJ1-Cs-A-7	143.70	0.50	12.51	0.0400	8.30
SJ1-Cs-A-8	167.77	0.50	12.51	0.0400	8.44
SJ1-Cs-A-9	95.82	0.50	12.50	0.0400	8.47
SJ1-Cs-A-10	119.90	0.50	12.50	0.0400	8.52
SJ2-Cs-A-11	0.27	0.50	12.50	0.0400	8.69
SJ2-Cs-A-12	0.52	0.50	12.51	0.0400	8.54
SJ2-Cs-A-13	1.00	0.50	12.50	0.0400	8.43
SJ2-Cs-A-14	2.00	0.50	12.53	0.0399	8.47

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Soil/Leachant mass ratio	Final pH
SJ1-Sr-A-1	6.62	0.50	12.53	0.0399	not measured
SJ1-Sr-A-2	30.57	0.50	12.54	0.0399	8.27
SJ1-Sr-A-3	53.83	0.50	12.50	0.0400	8.26
SJ1-Sr-A-4	23.78	0.50	12.51	0.0400	not measured
SJ1-Sr-A-5	47.73	0.50	12.50	0.0400	8.31
SJ1-Sr-A-6	71.00	0.50	12.50	0.0400	8.23
SJ1-Sr-A-7	143.47	0.50	12.52	0.0399	8.37
SJ1-Sr-A-8	167.63	0.50	12.51	0.0400	8.37
SJ1-Sr-A-9	95.82	0.50	12.50	0.0400	8.42
SJ1-Sr-A-10	119.90	0.50	12.50	0.0400	8.46
SJ2-Sr-A-11	0.25	0.50	12.50	0.0400	8.42
SJ2-Sr-A-12	0.50	0.50	12.50	0.0400	8.31
SJ2-Sr-A-13	1.00	0.50	12.50	0.0400	8.35
SJ2-Sr-A-14	2.00	0.50	12.50	0.0400	8.38

Test Number	Solution Bottle, g	Bottle + Cs test solution, g	Bottle + Cs test solution + DIW + HNO_3, g	Dilution Factor
SJ1-Cs-A-1	11.20	22.64	22.68	1.003
SJ1-Cs-A-2	11.16	22.39	22.44	1.004
SJ1-Cs-A-3	11.20	22.61	22.66	1.004
SJ1-Cs-A-4	11.14	22.50	22.54	1.004
SJ1-Cs-A-5	11.10	22.42	22.48	1.005
SJ1-Cs-A-6	11.14	22.48	22.52	1.004
SJ1-Cs-A-7	11.13	22.30	22.34	1.004
SJ1-Cs-A-8	11.23	22.49	22.55	1.005
SJ1-Cs-A-9	11.13	22.28	22.32	1.004
SJ1-Cs-A-10	11.18	22.43	22.48	1.004
SJ2-Cs-A-11	11.12	22.54	22.61	1.006
SJ2-Cs-A-12	11.20	22.55	22.64	1.008
SJ2-Cs-A-13	11.22	22.63	22.69	1.005
SJ2-Cs-A-14	11.16	22.55	22.63	1.007

Test Number	Solution Bottle, g	Bottle + Sr test solution, g	Bottle + Sr solution + DIW + HNO₃, g	Dilution Factor
SJ1-Sr-A-1	11.17	22.70	22.74	1.003
SJ1-Sr-A-2	11.16	22.55	22.58	1.003
SJ1-Sr-A-3	11.13	22.43	22.48	1.004
SJ1-Sr-A-4	11.15	22.68	22.73	1.004
SJ1-Sr-A-5	11.12	22.36	22.42	1.005
SJ1-Sr-A-6	11.11	22.37	22.42	1.004
SJ1-Sr-A-7	11.08	22.29	22.33	1.004
SJ1-Sr-A-8	11.11	22.36	22.41	1.004
SJ1-Sr-A-9	11.17	22.34	22.38	1.004
SJ1-Sr-A-10	11.11	22.30	22.35	1.004
SJ2-Sr-A-11	11.17	22.45	22.51	1.005
SJ2-Sr-A-12	11.16	22.56	22.63	1.006
SJ2-Sr-A-13	11.24	22.54	22.60	1.005
SJ2-Sr-A-14	11.13	22.46	22.54	1.007

Table 12. Combination of test solutions for tests conducted for different durations

Solution Number	Solution 1	Solution 2	Solution Number	Solution 1	Solution 2
SJ-A-1	SJ1-Cs-A-1	SJ1-Sr-A-1	SJ-A-8	SJ1-Cs-A-8	SJ1-Sr-A-8
SJ-A-2	SJ1-Cs-A-2	SJ1-Sr-A-2	SJ-A-9	SJ1-Cs-A-9	SJ1-Sr-A-9
SJ-A-3	SJ1-Cs-A-3	SJ1-Sr-A-3	SJ-A-10	SJ1-Cs-A-10	SJ1-Sr-A-10
SJ-A-4	SJ1-Cs-A-4	SJ1-Sr-A-4	SJ-A-11	SJ2-Cs-A-11	SJ2-Sr-A-11
SJ-A-5	SJ1-Cs-A-5	SJ1-Sr-A-5	SJ-A-12	SJ2-Cs-A-12	SJ2-Sr-A-12
SJ-A-6	SJ1-Cs-A-6	SJ1-Sr-A-6	SJ-A-13	SJ2-Cs-A-13	SJ2-Sr-A-13
SJ-A-7	SJ1-Cs-A-7	SJ1-Sr-A-7	SJ-A-14	SJ2-Cs-A-14	SJ2-Sr-A-14

Test Number	Mixed Solution Bottle, g	Bottle + Cs test solution, g	Bottle + Cs + Sr test solution, g	Cs Dilution Factor	Sr Dilution Factor
SJ-A-1	11.16	22.48	33.71	1.992	2.008
SJ-A-2	11.08	22.17	33.44	2.016	1.984
SJ-A-3	11.12	22.36	33.49	1.990	2.010
SJ-A-4	11.14	22.37	33.82	2.020	1.981
SJ-A-5	11.17	22.27	33.34	1.997	2.003
SJ-A-6	11.17	22.36	33.49	1.995	2.005
SJ-A-7	11.16	22.18	33.18	1.998	2.002
SJ-A-8	11.07	22.12	33.27	2.009	1.991
SJ-A-9	11.09	22.02	33.04	2.008	1.992
SJ-A-10	11.10	22.10	33.08	1.998	2.002
SJ-A-11	11.18	22.55	33.80	1.989	2.011
SJ-A-12	11.25	22.58	33.96	2.004	1.996
SJ-A-13	11.19	22.54	33.76	1.989	2.012
SJ-A-14	11.18	22.47	33.79	2.003	1.997

Table 13. Results for tests conducted for different durations

Test Number	Measured Cs, µg/L	Dil.-corrected Cs, µg/L	Cs in test solution, µg	Cs in test solution, µg/mL
SJ-A-1	510	1016	12.79	1.016
SJ-A-2	518	1044	13.13	1.044
SJ-A-3	565	1124	14.14	1.124
SJ-A-4	519	1048	13.17	1.048
SJ-A-5	516	1031	12.95	1.031
SJ-A-6	528	1053	13.24	1.053
SJ-A-7	552	1103	13.87	1.103
SJ-A-8	469	942.2	11.85	0.9422
SJ-A-9	542	1088	13.68	1.088
SJ-A-10	583	1165	14.64	1.165
SJ-A-11	507	1009	12.68	1.009
SJ-A-12	530	1062	13.36	1.062
SJ-A-13	585	1163	14.62	1.163
SJ-A-14	560	1121	14.13	1.121

Test Number	Measured Sr, µg/L	Dil.-corrected Sr, µg/L	Sr in test solution, µg	Sr in test solution, µg/mL
SJ-A-1	465	933.7	11.74	0.9337
SJ-A-2	492	976.1	12.28	0.9761
SJ-A-3	489	982.8	12.33	0.9828
SJ-A-4	513	1016	12.76	1.016
SJ-A-5	480	961.3	12.06	0.9613
SJ-A-6	488	978.6	12.28	0.9786
SJ-A-7	506	1013	12.73	1.013
SJ-A-8	482	959.7	12.05	0.9597
SJ-A-9	473	942.1	11.82	0.9421
SJ-A-10	483	966.9	12.13	0.9669
SJ-A-11	518	1042	13.06	1.042
SJ-A-12	537	1072	13.44	1.072
SJ-A-13	554	1114	13.98	1.114
SJ-A-14	558	1115	13.98	1.115

Test Number	Leachant Volume, mL	Mass Cs in Leachant, µg	Mass Cs on soil, µg	µg Cs/g soil	K_d (Cs) mL/g
SJ1-Cs-A-1	12.59	315.9	303.1	606.2	596.7
SJ1-Cs-A-2	12.57	315.4	302.2	604.5	578.8
SJ1-Cs-A-3	12.58	315.6	301.5	603.0	536.2
SJ1-Cs-A-4	12.57	315.4	302.2	604.4	576.6
SJ1-Cs-A-5	12.57	315.4	302.4	604.8	586.9
SJ1-Cs-A-6	12.57	315.4	302.1	604.3	573.8
SJ1-Cs-A-7	12.58	315.6	301.8	603.5	547.1
SJ1-Cs-A-8	12.58	315.6	303.8	607.5	644.8
SJ1-Cs-A-9	12.57	315.4	301.7	603.4	554.3
SJ1-Cs-A-10	12.57	315.4	300.7	601.5	516.3
SJ2-Cs-A-11	12.57	315.4	302.7	605.4	600.2
SJ2-Cs-A-12	12.58	315.6	302.3	604.5	569.0
SJ2-Cs-A-13	12.57	315.4	300.8	601.5	517.1
SJ2-Cs-A-14	12.60	316.1	302.0	604.0	538.6

Table 13. (cont.)

Test Number	Leachant Volume, mL	Mass Sr in Leachant, μg	Mass Sr on soil, μg	μg Sr/g soil	K_d (Sr) mL/g
SJ1-Sr-A-1	12.57	102.2	90.45	180.9	193.7
SJ1-Sr-A-2	12.58	102.3	89.99	180.0	184.4
SJ1-Sr-A-3	12.54	101.9	89.62	179.2	182.4
SJ1-Sr-A-4	12.55	102.0	89.27	178.5	175.7
SJ1-Sr-A-5	12.54	101.9	89.89	179.8	187.0
SJ1-Sr-A-6	12.54	101.9	89.67	179.3	183.3
SJ1-Sr-A-7	12.56	102.1	89.38	178.8	176.5
SJ1-Sr-A-8	12.55	102.0	89.98	180.0	187.5
SJ1-Sr-A-9	12.54	101.9	90.13	180.3	191.3
SJ1-Sr-A-10	12.54	101.9	89.82	179.6	185.8
SJ2-Sr-A-11	12.54	101.9	88.88	177.8	170.7
SJ2-Sr-A-12	12.54	101.9	88.50	177.0	165.2
SJ2-Sr-A-13	12.54	101.9	87.97	175.9	157.9
SJ2-Sr-A-14	12.54	101.9	87.97	175.9	157.9

Note that the tests run for the four shortest durations were conducted with soil from a different bottle of SRM 2709a than the other tests. The dashed lines show the average values of tests run for longer than 10 hours, all of which were conducted with soil from Jar 1. The 6.6-hour tests were also conducted with soil from Jar 1, but not included in the average values indicated by the dashed lines. Soil from Jar 2 was used in tests run for 2 hours or less. The Cs results for the short-term tests with soil from Jar 2 are consistent with the other tests, both with soil from Jar 1 and after longer reaction times. The Sr results for the short-term tests are consistently higher, although the uncertainty ranges of the data points overlap. The Sr result of the 6.6-hour test is consistent with the results of longer duration tests, all run with soil from Jar 1. The results suggest that tests with soil from Jar 2 are approaching an equilibrium Sr concentration that is higher than the equilibrium concentration attained by tests with soil from Jar 1. The result for the 6.6-hour test indicates that this is not an effect of reaction time, but a real difference in the properties of the San Joaquin soil in Jars 1 and 2. The difference could result from more Sr being dissolved from the soil into solution or less Sr being taken up by the soil from solution. Regardless, this indicates that inter-laboratory studies should be conducted using well-mixed soil from a single source, which could be a mixture of soil taken from several jars.
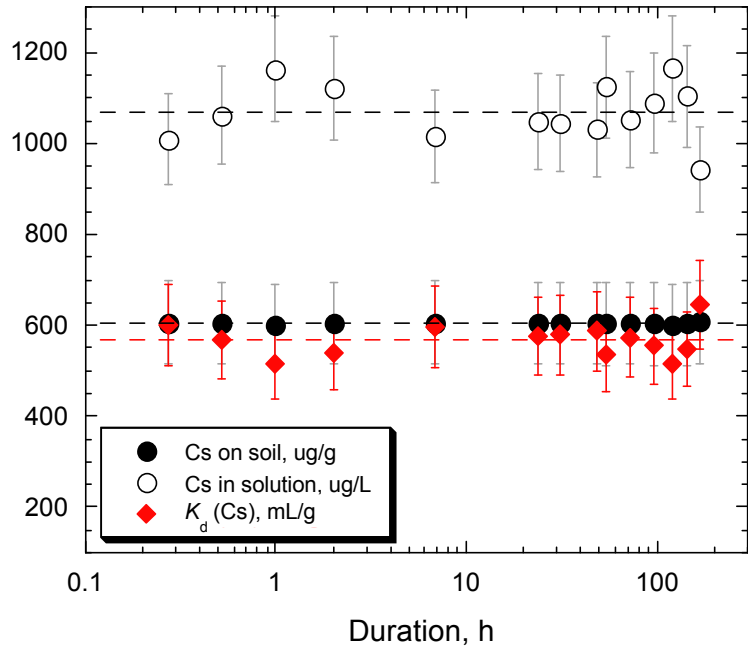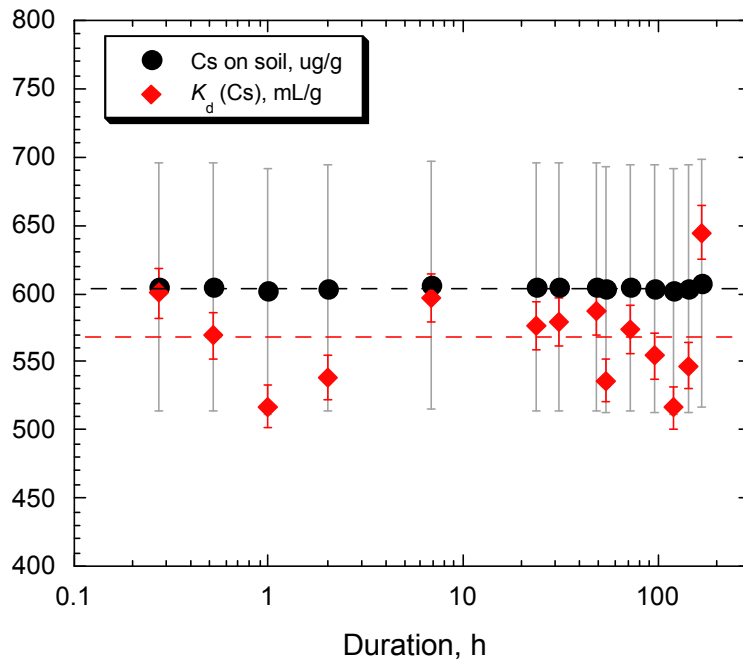
Figure 7. Results of tests conducted for different duration: (a) concentrations of Cs on soil and in solution, and K_d values vs. test duration and (b) expanded view of Cs on soil and K_d values. Dashed lines show average values for tests conducted for longer than 10 hours.
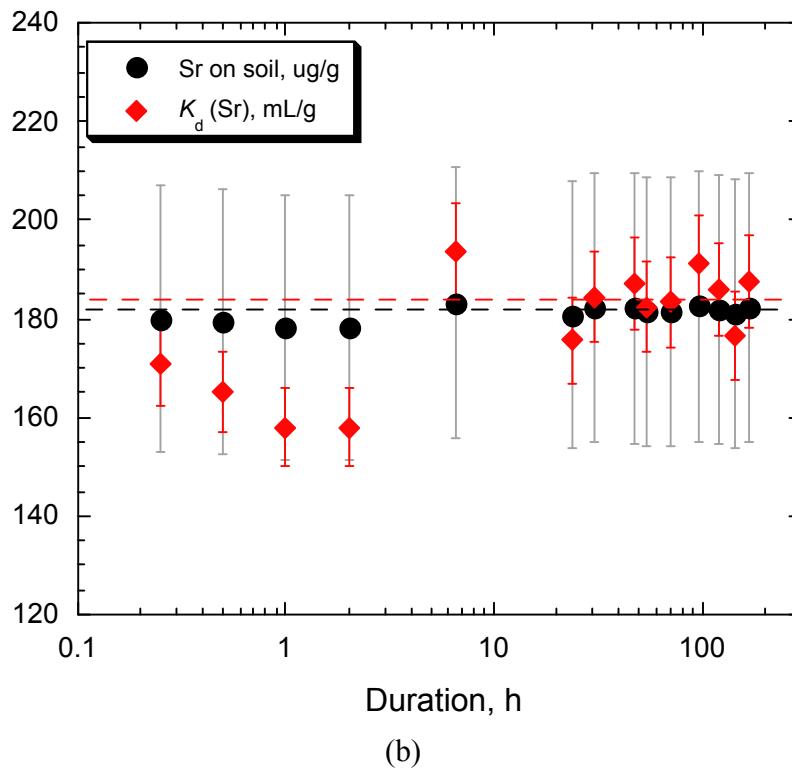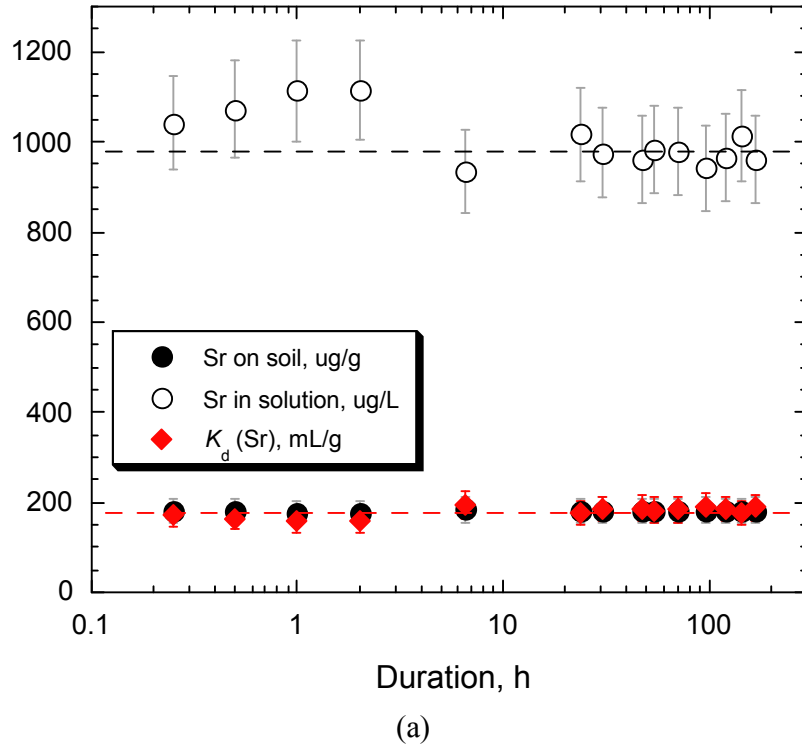
Figure 8. Results of tests conducted for different duration: (a) concentrations of Sr on soil and in solution, and K_d values vs. test duration and (b) expanded view of Sr on soil and K_d values. Dashed lines show average values for tests conducted for longer than 10 hours.

29

3.6 Effects of pH Tests

Portions of Cs solution A and Sr solution A were used to make new solutions with imposed pH values that were either lower or higher than the nominal pH 8.5 values of the base solutions to assess the effects of pH on uptake by the soil. Small amounts of either a dilute HNO_3 solution or a dilute NaOH solution were added to adjust the solutions to targeted pH values of 7.5 (solution A3), 8.0 (solution A4), 8.5 (solution A2), 9.0 (solution A5), or 9.5 (solution A6). These leachants had no buffer capacity, and the solution pH values drifted during the test. Tests were conducted for 1 and 2 days using soil from Jar 2. The test data are summarized in Table 14. Tests with A2 solutions were conducted as part of the effects of duration study discussed in Section 3.7, but are included here for completeness. Note that the intended adjustment of the Cs A6 solution to pH 9.5 was not achieved. Note also that all solutions drifted to final pH values near the leachant blank pH values of about 8.5. The data for the combination of solutions for analysis are given in Table 15 and the results are given in Table 16. Calculations are discussed in Appendix A.

Table 14. Data for tests conducted with leachants adjusted to different pH values

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Initial pH[a]	Final pH
SJ2-Cs-A3-1	24.00	0.50	12.51	7.54	8.44
SJ2-Cs-A3-2	47.88	0.50	12.50	7.54	8.32
SJ2-Cs-A4-1	24.00	0.50	12.50	8.05	8.42
SJ2-Cs-A4-2	47.88	0.50	12.50	8.05	8.38
SJ2-Cs-A2-1	24.00	0.50	12.50	8.56	8.64
SJ2-Cs-A2-2	48.03	0.50	12.52	8.56	8.45
SJ2-Cs-A5-1	22.08	0.50	12.50	9.12	8.43
SJ2-Cs-A5-2	47.88	0.50	12.49	9.12	8.44
SJ2-Cs-A6-1	24.00	0.50	12.50	9.13	8.49
SJ2-Cs-A6-2	47.88	0.50	12.50	9.13	8.46

[a]Values from Table 3.

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Initial pH[a]	Final pH
SJ2-Sr-A3-1	23.57	0.50	12.50	7.42	8.33
SJ2-Sr-A3-2	47.45	0.50	12.51	7.42	8.28
SJ2-Sr-A4-1	23.57	0.50	12.50	8.07	8.39
SJ2-Sr-A4-2	47.45	0.50	12.54	8.07	8.19
SJ2-Sr-A2-1	24.00	0.50	12.50	8.56	8.53
SJ2-Sr-A2-2	48.03	0.50	12.50	8.56	8.42
SJ2-Sr-A5-1	23.57	0.50	12.50	9.13	8.45
SJ2-Sr-A5-2	47.45	0.50	12.53	9.13	8.30
SJ2-Sr-A6-1	23.57	0.50	12.52	9.65	8.72
SJ2-Sr-A6-2	47.45	0.50	12.51	9.65	8.29

[a]Values from Table 3.

Table 14. (cont.)

Test Number	Solution Bottle, g	Bottle + Cs test solution, g	Bottle + Cs test solution + DIW + HNO$_3$, g	Cs Dilution Factor
SJ2-Cs-A3-1	11.15	22.44	22.53	1.008
SJ2-Cs-A3-2	11.19	22.46	22.54	1.007
SJ2-Cs-A4-1	11.17	22.41	22.50	1.008
SJ2-Cs-A4-2	11.25	22.37	22.44	1.006
SJ1-Cs-A2-1	11.10	22.45	22.50	1.004
SJ1-Cs-A2-2	11.15	22.52	22.56	1.004
SJ2-Cs-A5-1	11.20	22.43	22.51	1.007
SJ2-Cs-A5-2	11.15	22.28	22.36	1.007
SJ2-Cs-A6-1	11.14	22.22	22.33	1.010
SJ2-Cs-A6-2	11.22	22.35	22.50	1.013

Test Number	Solution Bottle, g	Bottle + Sr test solution, g	Bottle + Sr solution + DIW + HNO$_3$, g	Sr Dilution Factor
SJ2-Sr-A3-1	11.18	22.35	22.41	1.005
SJ2-Sr-A3-2	11.14	22.23	22.28	1.005
SJ2-Sr-A4-1	11.17	22.33	22.42	1.008
SJ2-Sr-A4-2	11.21	22.41	22.55	1.013
SJ2-Sr-A2-1	11.10	22.45	22.50	1.004
SJ2-Sr-A2-2	11.16	22.38	22.44	1.005
SJ2-Sr-A5-1	11.18	22.27	22.34	1.006
SJ2-Sr-A5-2	11.22	22.41	22.53	1.011
SJ2-Sr-A6-1	11.19	22.45	22.56	1.010
SJ2-Sr-A6-2	11.15	22.27	22.37	1.009

Table 15. Combination of solutions for tests conducted with leachants adjusted to different pH values

Solution Number	Solution 1	Solution 2
SJ-A-1	SJ2-Cs-A3-1	SJ2-Sr-A3-1
SJ-A-2	SJ2-Cs-A3-2	SJ2-Sr-A3-2
SJ-A-3	SJ2-Cs-A4-1	SJ2-Sr-A4-1
SJ-A-4	SJ2-Cs-A4-2	SJ2-Sr-A4-2
SJ-A-5	SJ2-Cs-A5-1	SJ2-Sr-A5-1
SJ-A-6	SJ2-Cs-A5-2	SJ2-Sr-A5-2
SJ-A-7	SJ2-Cs-A6-1	SJ2-Sr-A6-1
SJ-A-8	SJ2-Cs-A6-2	SJ2-Sr-A6-2

Test Number	Mixed Solution Bottle, g	Bottle + Cs test solution, g	Bottle + Cs + Sr test solution, g	Cs Dilution Factor	Sr Dilution Factor
SJ-A-1	11.19	22.43	33.48	1.9831	2.017
SJ-A-2	11.16	22.38	33.37	1.9795	2.021
SJ-A-3	11.06	22.23	33.29	1.9902	2.010
SJ-A-4	11.20	22.20	33.42	2.0200	1.980
SJ-A-5	11.17	22.35	33.35	1.9839	2.016
SJ-A-6	11.16	22.17	33.33	2.0136	1.987
SJ-A-7	11.13	22.15	33.32	2.0136	1.987
SJ-A-8	11.21	22.34	33.44	1.9973	2.003

Table 16. Solution results for tests conducted with leachants adjusted to different pH values

Test Number	Measured Cs, µg/L	Dil.-corrected Cs, µg/L	Cs in test solution, µg	Cs in test solution, µg/mL
SJ2-Cs-A3-1	593	1176	14.79	1.176
SJ2-Cs-A3-2	641	1269	15.95	1.269
SJ2-Cs-A4-1	596	1186	14.91	1.186
SJ2-Cs-A4-2	573	1157	14.55	1.157
SJ-A2-1	552	1101	13.84	1.101
SJ-A2-2	573	1139	14.34	1.139
SJ2-Cs-A5-1	515	1022	12.84	1.022
SJ2-Cs-A5-2	527	1061	13.33	1.061
SJ2-Cs-A6-1	574	1156	14.53	1.156
SJ2-Cs-A6-2	569	1136	14.28	1.136

Test Number	Measured Sr, µg/L	Dil.-corrected Sr, µg/L	Sr in test solution, µg	Sr in test solution, µg/mL
SJ2-Sr-A3-1	715	1442	18.09	1.44
SJ2-Sr-A3-2	680	1374	17.25	1.37
SJ2-Sr-A4-1	637	1280	16.06	1.28
SJ2-Sr-A4-2	616	1220	15.35	1.22
SJ-A2-1	528	1059	13.28	1.059
SJ-A2-2	528	1062	13.32	1.062
SJ2-Sr-A5-1	258	520	6.53	0.520
SJ2-Sr-A5-2	290	576	7.24	0.576
SJ2-Sr-A6-1	82.4	164	2.06	0.164
SJ2-Sr-A6-2	86.5	173	2.17	0.173

Test Number	Leachant Volume, mL	Mass Cs in Leachant, µg	Mass Cs on soil, µg	µg Cs/g soil	K_d (Cs) mL/g
SJ2-Cs-A3-1	12.58	315.62	301	602	511.6
SJ2-Cs-A3-2	12.57	315.37	299	599	472.0
SJ2-Cs-A4-1	12.57	315.37	300	601	506.6
SJ2-Cs-A4-2	12.57	315.37	301	602	519.8
SJ1-Cs-A2-1	12.57	308.63	302	603	547.7
SJ1-Cs-A2-2	12.59	309.13	301	602	528.4
SJ2-Cs-A5-1	12.57	315.37	302	605	592.2
SJ2-Cs-A5-2	12.56	315.12	302	604	568.8
SJ2-Cs-A6-1	12.57	315.37	301	602	520.6
SJ2-Cs-A6-2	12.57	315.37	301	602	529.9

Test Number	Leachant Volume, mL	Mass Sr in Leachant[a], µg	Mass Sr on soil, µg	µg Sr/g soil	K_d (Sr) mL/g
SJ2-Sr-A3-1	12.54	101.9	83.9	168	116.3
SJ2-Sr-A3-2	12.55	102.0	84.8	170	123.4
SJ2-Sr-A4-1	12.54	101.9	85.9	172	134.2
SJ2-Sr-A4-2	12.58	102.3	86.9	174	142.5
SJ2-Sr-A2-1	12.54	102.1	88.8	178	167.7
SJ2-Sr-A2-2	12.54	102.0	88.7	177	167.0
SJ2-Sr-A5-1	12.54	101.9	95.4	191	366.8
SJ2-Sr-A5-2	12.57	102.2	94.9	190	329.6
SJ2-Sr-A6-1	12.56	102.1	100	200	1222
SJ2-Sr-A6-2	12.55	102.0	99.9	200	1153

[a]Calculated using concentration in Sr A solution to be consistent with other tests.

32

Figures 9a and 9b show the masses of Cs and Sr calculated to be in the leachant solution and in the test solutions and on the soil after reacting for 1 or 2 days. Uncertainty bars are drawn at 10% for the masses in the leachants and test solutions, and at 15% for the masses on the soil. The results for the 1- and 2-day tests overlap at all pH values for both Cs and Sr. As determined from the Cs concentrations in the test solutions, almost all of the Cs present in the leachant was taken up by the soil within 1 day at all pH values. The uptake of Sr showed a dependence on the pH, where more Sr was taken up from the solutions with higher imposed pH values.
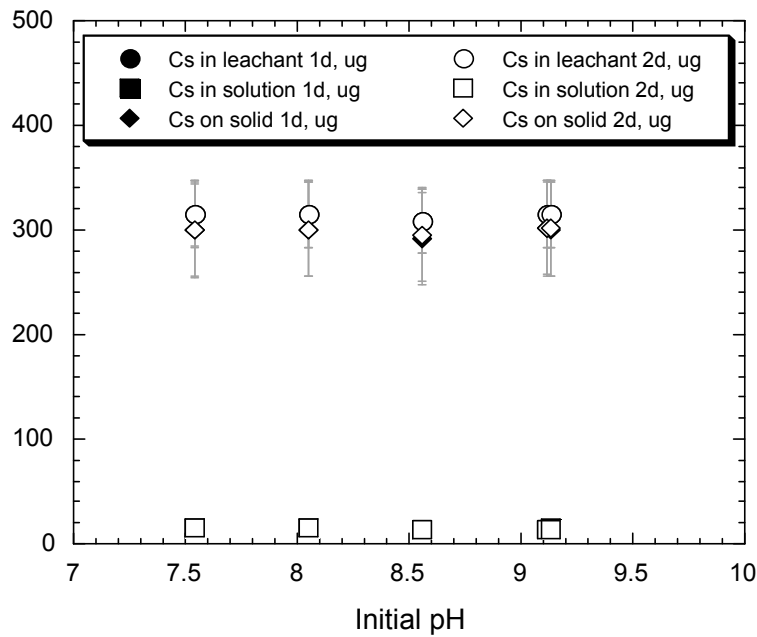
(a)
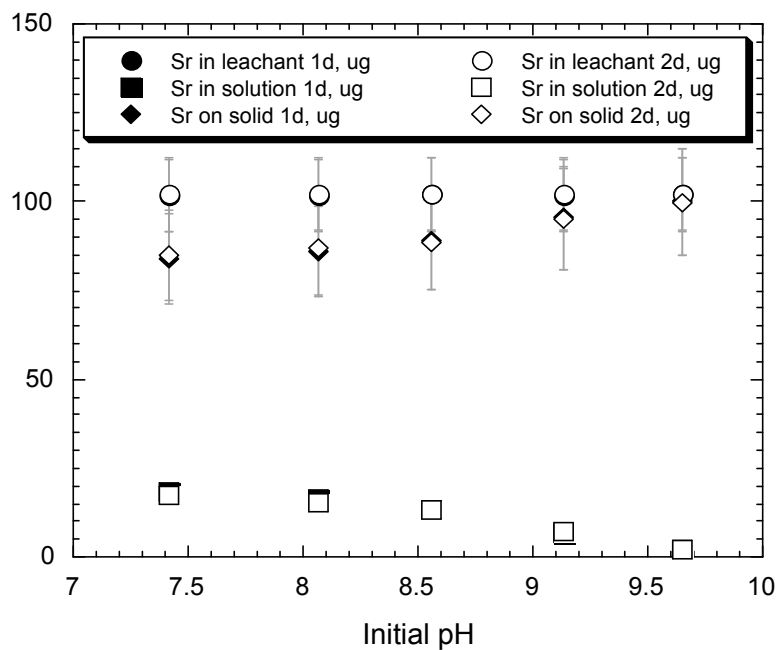
(b)

Figure 9. Results of tests conducted with different imposed pH values: (a) Cs masses in leachant, in test solution, and on soil vs. initial pH and (b) Sr masses in leachant, in test solution, and on soil vs. initial pH.

33

Figures 10a and 10b show the distributions on per mass and per volume bases, and the calculated K_d values. These ratios emphasize the small differences in the distributions. The result of tests with Cs solution A2 and Sr solution A2 conducted for 1 and 2 days as part of the series of tests to study the effects of reaction time are included in the plots at about pH 8.3. Note that these tests were conducted with soil from Jar 1 whereas the tests conducted to study the effects of pH were conducted with soil from Jar 2. The distribution coefficient of Cs is insensitive to the imposed pH, but the distribution coefficient of Sr increases significantly as the imposed pH is increased, despite the fact the pH values drifted to about pH 8.5. This suggests the uptake of Sr is not rapidly reversible, but other tests are needed to verify that.
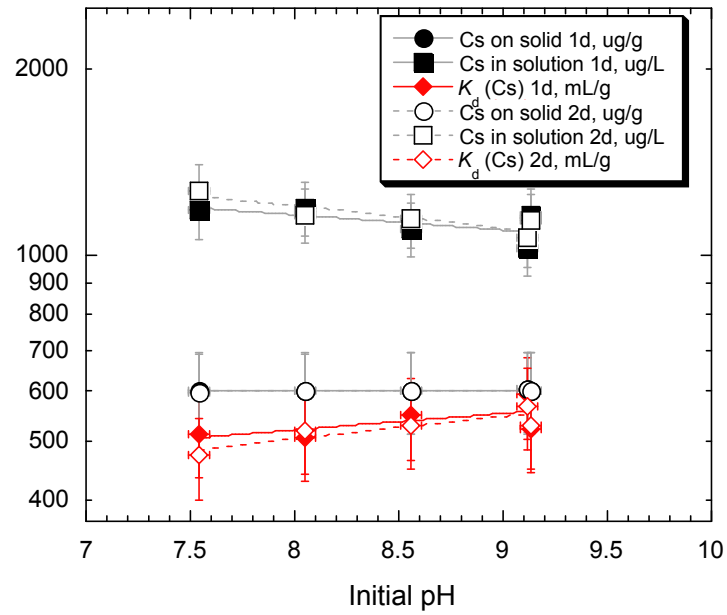
(a)
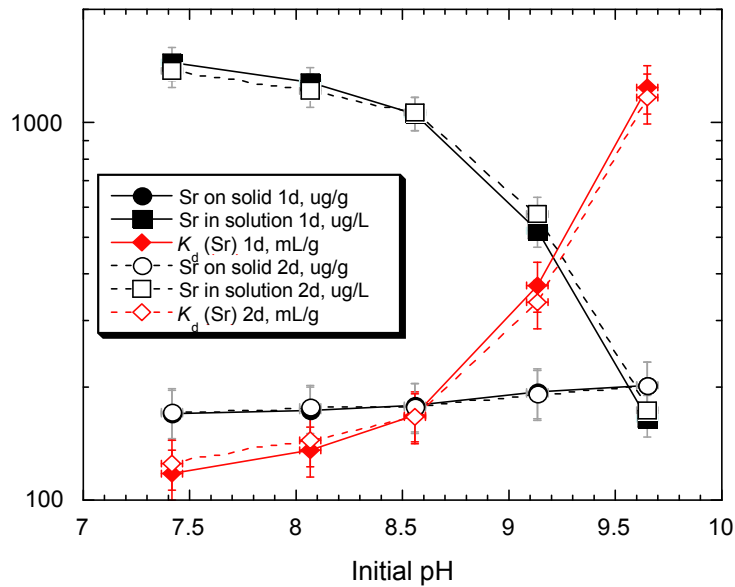
(b)

Figure 10. Results of tests conducted with different imposed pH values: (a) concentrations of Cs on the soil and in solution, and K_d values, and (b) concentrations of Sr on the soil and in solution, and K_d values.

3.7 Effects of Concentration Tests

Tests were conducted using six Cs concentrations and six Sr concentrations to evaluate the effects of contaminant concentration on the values of K_d and the evaluation of analytical sorption isotherms. Tests were run for 1, 2, and 3 days with each concentration to detect possible kinetic effects. The test data are provided in Table 17. Data for the combination of solutions for analysis are summarized in Table 18 and the results are given in Table 19. Calculations are discussed in Appendix B. Though it was not the intent of this work to quantify the sorption behaviors of Cs and Sr, preliminary applications of linear, Freundlich, and Langmuir isotherms are discussed to show the usefulness of the test results.

Table 17. Data for tests run with various Cs solution and Sr solution concentrations

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Soil/Leachant mass ratio	Final pH
SJ1-Cs-A2-0	74.87	—	12.51	—	8.56
SJ1-Cs-A2-1	24.00	0.50	12.50	0.0400	8.64
SJ1-Cs-A2-2	48.03	0.50	12.52	0.0399	8.45
SJ1-Cs-A2-3	74.87	0.50	12.50	0.0400	8.43
SJ1-Cs-B-0	71.63	—	12.50	—	8.53
SJ1-Cs-B-3	24.00	0.50	12.50	0.0400	8.44
SJ1-Cs-B-4	48.03	0.50	12.51	0.0400	8.41
SJ1-Cs-B-5	78.60	0.50	12.50	0.0400	8.37
SJ1-Cs-C-0	71.63	—	12.51	—	8.40
SJ1-Cs-C-3	24.00	0.50	12.50	0.0400	8.45
SJ1-Cs-C-4	48.03	0.50	12.50	0.0400	8.46
SJ1-Cs-C-5	78.60	0.50	12.50	0.0400	8.40
SJ1-Cs-D-0	71.63	—	12.50	—	8.47
SJ1-Cs-D-3	24.00	0.50	12.50	0.0400	8.44
SJ1-Cs-D-4	48.03	0.50	12.50	0.0400	8.44
SJ1-Cs-D-5	78.60	0.50	12.53	0.0399	8.39
SJ1-Cs-E-0	71.63	—	12.50	—	8.44
SJ1-Cs-E-3	24.00	0.50	12.51	0.0400	8.45
SJ1-Cs-E-4	48.03	0.50	12.50	0.0400	8.37
SJ1-Cs-E-5	78.60	0.50	12.51	0.0400	8.41
SJ1-Cs-F-0	71.63	—	12.50	—	8.48
SJ1-Cs-F-4	24.00	0.50	12.53	0.0399	8.44
SJ1-Cs-F-5	78.60	0.50	12.50	0.0400	8.48
SJ1-Cs-F-6	48.03	0.50	12.50	0.0400	8.44

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Soil/Leachant mass ratio	Final pH
SJ2-Sr-A2-0	74.87	—	12.51	—	8.56
SJ2-Sr-A2-1	24.00	0.50	12.50	0.0400	8.53
SJ2-Sr-A2-2	48.03	0.50	12.50	0.0400	8.42
SJ2-Sr-A2-3	74.87	0.50	12.51	0.0400	8.43
SJ1-Sr-B-0	78.93	—	12.51	—	8.10
SJ1-Sr-B-3	24.00	0.50	12.50	0.0400	8.24
SJ1-Sr-B-4	48.03	0.50	12.50	0.0400	8.28
SJ1-Sr-B-5	71.63	0.50	12.50	0.0400	8.23
SJ1-Sr-C-0	78.93	—	12.50	—	8.52

Table 17. (cont.)

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Soil/Leachant mass ratio	Final pH
SJ1-Sr-C-3	24.00	0.50	12.50	0.0400	8.40
SJ1-Sr-C-4	47.97	0.50	12.51	0.0400	8.42
SJ1-Sr-C-5	71.63	0.50	12.51	0.0400	8.43
SJ1-Sr-D-0	78.93	—	12.50	—	8.52
SJ1-Sr-D-3	24.00	0.50	12.51	0.0400	8.45
SJ1-Sr-D-4	47.97	0.40	10.01	0.0400	8.47
SJ1-Sr-D-5	71.63	0.50	12.50	0.0400	8.47
SJ1-Sr-E-0	78.93	—	12.51	—	8.50
SJ1-Sr-E-3	24.00	0.50	12.50	0.0400	8.46
SJ1-Sr-E-4	47.97	0.50	12.53	0.0399	8.42
SJ1-Sr-E-5	71.63	0.50	12.52	0.0399	8.47
SJ1-Sr-F-0	78.93	—	12.50	—	8.50
SJ1-Sr-F-3	24.00	0.50	12.50	0.0400	8.43
SJ1-Sr-F-4	47.97	0.50	12.51	0.0400	8.46
SJ1-Sr-F-5	71.63	0.50	12.51	0.0400	8.47
SJ1-Sr-F-0[a]	47.88	—	12.50	0.0000	8.62

[a]Repeat of blank test SJ1-Sr-F0.

Test Number	Solution Bottle, g	Bottle + Cs test solution, g	Bottle + Cs test solution + DIW + HNO_3, g	Dilution Factor
SJ1-Cs-A2-0	11.16	22.97	23.03	1.005
SJ1-Cs-A2-1	11.10	22.45	22.50	1.004
SJ1-Cs-A2-2	11.15	22.52	22.56	1.004
SJ1-Cs-A2-3	11.18	22.30	22.36	1.005
SJ1-Cs-B-0	11.17	22.82	22.88	1.005
SJ1-Cs-B-3	11.14	22.56	22.61	1.004
SJ1-Cs-B-4	11.16	21.96	22.03	1.006
SJ1-Cs-B-5	11.19	22.24	22.28	1.004
SJ1-Cs-C-0	11.14	23.03	23.11	1.007
SJ1-Cs-C-3	11.14	22.53	22.57	1.004
SJ1-Cs-C-4	11.21	22.46	22.53	1.006
SJ1-Cs-C-5	11.11	22.25	22.28	1.003
SJ1-Cs-D-0	11.16	22.90	22.93	1.003
SJ1-Cs-D-3	11.18	22.48	22.51	1.003
SJ1-Cs-D-4	11.14	22.36	22.42	1.005
SJ1-Cs-D-5	11.13	22.29	22.32	1.003
SJ1-Cs-E-0	11.15	22.89	22.93	1.003
SJ1-Cs-E-3	11.21	22.62	22.66	1.004
SJ1-Cs-E-4	11.17	22.36	22.42	1.005
SJ1-Cs-E-5	11.13	22.33	22.39	1.005
SJ1-Cs-F-0	11.19	22.94	23.00	1.005
SJ1-Cs-F-4	11.12	22.45	22.50	1.004
SJ1-Cs-F-5	11.11	22.37	22.42	1.004
SJ1-Cs-F-6	11.12	22.41	22.49	1.007

Table 17. (cont.)

Test Number	Solution Bottle, g	Bottle + Sr test solution, g	Bottle + Sr test solution + DIW + HNO$_3$, g	Dilution Factor
SJ2-Sr-A2-0	11.16	22.97	23.03	1.005
SJ2-Sr-A2-1	11.10	22.45	22.50	1.004
SJ2-Sr-A2-2	11.16	22.38	22.44	1.005
SJ2-Sr-A2-3	11.18	22.30	22.36	1.005
SJ1-Sr-B-0	11.10	22.73	22.78	1.004
SJ1-Sr-B-3	11.17	22.46	22.52	1.005
SJ1-Sr-B-4	11.18	22.28	22.34	1.005
SJ1-Sr-B-5	11.18	22.43	22.47	1.004
SJ1-Sr-C-0	11.21	22.87	22.94	1.006
SJ1-Sr-C-3	11.14	22.46	22.49	1.003
SJ1-Sr-C-4	11.10	22.25	22.32	1.006
SJ1-Sr-C-5	11.15	22.27	22.32	1.004
SJ1-Sr-D-0	11.19	22.87	22.94	1.006
SJ1-Sr-D-3	11.13	22.37	22.41	1.004
SJ1-Sr-D-4	11.15	20.08	20.17	1.010
SJ1-Sr-D-5	11.15	22.27	22.31	1.004
SJ1-Sr-E-0	11.17	22.91	22.94	1.003
SJ1-Sr-E-3	11.13	22.47	22.50	1.003
SJ1-Sr-E-4	11.19	22.41	22.49	1.007
SJ1-Sr-E-5	11.04	22.32	22.36	1.004
SJ1-Sr-F-0	11.24	22.98	23.02	1.003
SJ1-Sr-F-3	11.11	22.47	22.53	1.005
SJ1-Sr-F-4	11.16	22.41	22.48	1.006
SJ1-Sr-F-5	11.12	21.94	21.99	1.005
SJ1-Sr-F-0[a]	11.11	22.82	22.92	1.009

[a]Repeat of blank test SJ1-Sr-F0.

Table 18. Combination of test solutions for analysis

Solution Number	Solution 1	Solution 2	Solution Number	Solution 1	Solution 2
SJ-A2-0	SJ1-Cs-A2-0	SJ2-Sr-A2-0	SJ-D-0	SJ1-Cs-D-0	SJ1-Sr-D-0
SJ-A2-1	SJ1-Cs-A2-1	SJ2-Sr-A2-1	SJ-D-3	SJ1-Cs-D-3	SJ1-Sr-D-3
SJ-A2-2	SJ1-Cs-A2-2	SJ2-Sr-A2-2	SJ-D-4	SJ1-Cs-D-4	SJ1-Sr-D-4
SJ-A2-3	SJ1-Cs-A2-3	SJ2-Sr-A2-3	SJ-D-5	SJ1-Cs-D-5	SJ1-Sr-D-5
SJ-B-0	SJ1-Cs-B-0	SJ1-Sr-B-0	SJ-E-0	SJ1-Cs-E-0	SJ1-Sr-E-0
SJ-B-3	SJ1-Cs-B-3	SJ1-Sr-B-3	SJ-E-3	SJ1-Cs-E-3	SJ1-Sr-E-3
SJ-B-4	SJ1-Cs-B-4	SJ1-Sr-B-4	SJ-E-4	SJ1-Cs-E-4	SJ1-Sr-E-4
SJ-B-5	SJ1-Cs-B-5	SJ1-Sr-B-5	SJ-E-5	SJ1-Cs-E-5	SJ1-Sr-E-5
SJ-C-0	SJ1-Cs-C-0	SJ1-Sr-C-0	SJ-F-0	SJ1-Cs-F-0	SJ1-Sr-F-0
SJ-C-3	SJ1-Cs-C-3	SJ1-Sr-C-3	SJ-F-3	SJ1-Cs-F-4	SJ1-Sr-F-3
SJ-C-4	SJ1-Cs-C-4	SJ1-Sr-C-4	SJ-F-4	SJ1-Cs-F-5	SJ1-Sr-F-4
SJ-C-5	SJ1-Cs-C-5	SJ1-Sr-C-5	SJ-F-5	SJ1-Cs-F-6	SJ1-Sr-F-5
SJ-A2-0	11.12	22.98	34.80	1.997	2.003
SJ-A2-1	11.12	22.41	33.64	1.995	2.005

Table 18. (cont.)

Solution Number	Mixed Solution Bottle, g	Bottle + Cs test solution, g	Bottle + Cs + Sr test solution, g	Cs Dilution Factor	Sr Dilution Factor
SJ-A2-2	11.16	22.41	33.53	1.988	2.012
SJ-A2-3	11.18	22.20	33.26	2.004	1.996
SJ-B-0	11.16	22.79	34.34	1.993	2.007
SJ-B-3	11.21	22.57	33.76	1.985	2.015
SJ-B-4	11.24	21.89	32.80	2.024	1.976
SJ-B-5	11.18	22.11	33.20	2.015	1.986
SJ-C-0	11.18	23.11	34.77	1.977	2.023
SJ-C-3	11.26	22.56	33.75	1.990	2.010
SJ-C-4	11.21	22.31	33.27	1.987	2.013
SJ-C-5	11.11	22.15	33.11	1.993	2.007
SJ-D-0	11.19	22.86	34.57	2.003	1.997
SJ-D-3	11.19	22.26	33.43	2.009	1.991
SJ-D-4	11.11	22.18	31.05	1.801	2.248
SJ-D-5	11.20	22.20	33.14	1.995	2.005
SJ-E-0	11.19	22.88	34.60	2.003	1.997
SJ-E-3	11.17	22.42	33.62	1.996	2.004
SJ-E-4	11.16	22.20	33.24	2.000	2.000
SJ-E-5	11.20	22.22	33.35	2.010	1.990
SJ-F-0	11.23	22.98	34.66	1.994	2.006
SJ-F-3	11.23	22.39	33.55	2.000	2.000
SJ-F-4	11.17	22.35	33.46	1.994	2.006
SJ-F-5	11.10	22.18	32.82	1.960	2.041
SJ-F-0[a]	11.11	—	22.92	—	1.003

[a]Repeat of blank test SJ1-Sr-F0.

Table 19. Solution results for tests using leachants with different Cs or Sr concentrations

Solution Number	Measured Cs, µg/L	Dil.-corrected Cs, µg/L	Mass Cs in test solution, µg	Cs in test solution, µg/mL
SJ-A2-0	12300	24559	309	24.6
SJ-A2-1	552	1101	13.8	1.10
SJ-A2-2	573	1139	14.3	1.14
SJ-A2-3	531	1064	13.4	1.06
SJ-B-0	63200	125965	1583	126
SJ-B-3	6140	12188	153	12.2
SJ-B-4	5660	11458	144	11.5
SJ-B-5	5190	10456	131	10.5
SJ-C-0	6140	12141	153	12.1
SJ-C-3	182	362	4.55	0.362
SJ-C-4	164	326	4.10	0.326
SJ-C-5	150	299	3.76	0.299
SJ-D-0	1260	2524	31.7	2.52
SJ-D-3	11.3	22.7	0.285	0.0227
SJ-D-4	11.9	21.4	0.269	0.0214
SJ-D-5	7.47	14.9	0.188	0.0149
SJ-E-0	135	270	3.40	0.270
SJ-E-3	0.273	0.545	0.0069	0.0005

Table 19. (cont.)

Solution Number	Measured Cs, µg/L	Dil.-corrected Cs, µg/L	Mass Cs in test solution, µg	Cs in test solution, µg/mL
SJ-E-4	0.203	0.406	0.0051	0.0004
SJ-E-5	0.147	0.296	0.0037	0.0003
SJ-F-0	3140	6261	78.69	6.26
SJ-F-3	52.5	105	1.32	0.105
SJ-F-4	53.5	107	1.34	0.107
SJ-F-5	47.6	93.3	1.17	0.0933

Solution Number	Measured Sr, µg/L	Dil.-corrected Sr, µg/L	Mass Sr in test solution, µg	Sr in test solution, µg/mL
SJ-A2-0	7360	14745	185	14.7
SJ-A2-1	528	1059	13.3	1.06
SJ-A2-2	528	1062	13.3	1.06
SJ-A2-3	484	966	12.1	0.966
SJ-B-0	18000	36125	453	36.1
SJ-B-3	1890	3809	47.8	3.81
SJ-B-4	1730	3419	42.9	3.42
SJ-B-5	2580	5123	64.3	5.12
SJ-C-0	3860	7809	98.0	7.81
SJ-C-3	276	555	6.96	0.555
SJ-C-4	289	582	7.30	0.582
SJ-C-5	252	506	6.35	0.506
SJ-D-0	772	1541	19.3	1.54
SJ-D-3	114	227	2.85	0.227
SJ-D-4	113	254	2.55	0.254
SJ-D-5	102	205	2.57	0.205
SJ-E-0	82.8	165	2.08	0.165
SJ-E-3	76.4	153	1.92	0.153
SJ-E-4	81.7	163	2.05	0.163
SJ-E-5	69.9	139	1.75	0.139
SJ-F-0[b]	790[b]	1585[b]	19.9[b]	1.58[b]
SJ-F-3	164	328	4.11	0.328
SJ-F-4	170	341	4.28	0.341
SJ-F-5	98.1	200	2.51	0.200
SJ-F-0[c]	3830	3840	48.0	3.84

[b]Results of blank test SJ1-Sr-F0 rejected.

[c]Repeat of blank test SJ1-Sr-F0 solution was not mixed with Cs solution.

Test Number	Volume Cs Leachant, mL	Mass Cs in Leachant, µg	Mass Cs on soil, µg	µg Cs/g soil	K_d (Cs) mL/g
SJ1-Cs-A2-0	12.58	309	—	—	—
SJ1-Cs-A2-1	12.57	309	295	590	535.5
SJ1-Cs-A2-2	12.59	309	295	590	517.4
SJ1-Cs-A2-3	12.57	309	295	591	555.0
SJ1-Cs-B-0	12.57	1583	—	—	—
SJ1-Cs-B-3	12.57	1583	1430	2860	234.6
SJ1-Cs-B-4	12.58	1584	1440	2880	251.4
SJ1-Cs-B-5	12.57	1583	1452	2903	277.7

Table 19 (cont.)

Test Number	Volume Cs Leachant, mL	Mass Cs in Leachant, µg	Mass Cs on soil, µg	µg Cs/g soil	K_d (Cs) mL/g
SJ1-Cs-C-0	12.58	153	—	—	—
SJ1-Cs-C-3	12.57	153	148	296	817.3
SJ1-Cs-C-4	12.57	153	148	297	911.1
SJ1-Cs-C-5	12.57	153	148	298	995.8
SJ1-Cs-D-0	12.57	31.7	—	—	—
SJ1-Cs-D-3	12.57	31.7	31.4	62.9	2770
SJ1-Cs-D-4	12.57	31.7	31.5	62.9	2935
SJ1-Cs-D-5	12.60	31.8	31.6	63.2	4243
SJ1-Cs-E-0	12.57	3.40	—	—	—
SJ1-Cs-E-3	12.58	3.40	3.39	6.79	12460
SJ1-Cs-E-4	12.57	3.40	3.39	6.78	16710
SJ1-Cs-E-5	12.58	3.40	3.40	6.79	22990
SJ1-Cs-F-0	12.57	78.7	—	—	—
SJ1-Cs-F-3	12.60	78.9	77.6	155	1477
SJ1-Cs-F-4	12.57	78.7	77.3	155	1450
SJ1-Cs-F-5	12.57	78.7	77.5	155	1661

Test Number	Volume Sr Leachant, mL	Mass Sr in Leachant, µg	Mass Sr on soil, µg	µg Sr/g soil	K_d (Sr) mL/g
SJ2-Sr-A2-0	12.55	185	—	—	—
SJ2-Sr-A2-1	12.54	185	172	343	324.3
SJ2-Sr-A2-2	12.54	185	172	343	323.2
SJ2-Sr-A2-3	12.55	185	173	346	358.0
SJ1-Sr-B-0	12.55	453	—	—	—
SJ1-Sr-B-3	12.54	453	405	811	212.9
SJ1-Sr-B-4	12.54	453	410	821	240.0
SJ1-Sr-B-5	12.54	453	389	778	151.8
SJ1-Sr-C-0	12.54	98.0	—	—	—
SJ1-Sr-C-3	12.54	98.0	91.0	182	328.1
SJ1-Sr-C-4	12.55	98.0	90.7	181	312.0
SJ1-Sr-C-5	12.55	98.0	91.7	183	362.5
SJ1-Sr-D-0	12.54	19.3	—	—	—
SJ1-Sr-D-3	12.55	19.3	16.5	33.0	145.4
SJ1-Sr-D-4	10.04	15.5	12.9	32.3	127.3
SJ1-Sr-D-5	12.54	19.3	16.8	33.5	163.9
SJ1-Sr-E-0	12.55	2.08	—	—	—
SJ1-Sr-E-3	12.54	2.07	0.154	0.307	2.006
SJ1-Sr-E-4	12.57	2.08	0.0250	0.050	0.306
SJ1-Sr-E-5	12.56	2.08	0.330	0.660	4.747
SJ1-Sr-F-0[b]	12.54	—[b]	—[b]	—[b]	—
SJ1-Sr-F-3	12.54	48.0	43.9	87.9	267.8
SJ1-Sr-F-4	12.55	48.1	43.8	87.6	256.8
SJ1-Sr-F-5	12.55	48.1	45.6	91.1	455.1
SJ1-Sr-F-0[a]	12.54	48.0	—	—	—

[a]Repeat of blank test SJ1-Sr-F0.

[b]Results of blank test SJ1-Sr-F0 rejected.

[c]Calculated using concentration in Sr A2 solution to be consistent with other tests in this series.

Linear Isotherm Model

In the linear isotherm model, the amount taken up by the soil is proportional to the amount in the test solution as in the definition of K_d:

$$\text{(concentration on solid)} = K_d \times \text{(concentration in solution)}. \tag{7}$$

Linear plots of the Cs and Sr test results are given in Figures 11 and 12, respectively. Figure 11a shows the mass of Cs taken up by the soil to be linear with the mass of Cs in the initial solution, but Figure 11b shows that the mass of Cs taken up by the soil is not linearly related to the mass of Cs remaining in the solution. Rather, the amounts taken up by the soil at low solution concentrations is related to the solution equilibrium concentration by an empirical power law relationship. Figure 11c shows that the amounts taken up by the soil at higher concentrations are greater than what is predicted based on the relationship followed at low solution concentrations. This may reflect a change in mechanism, such as the added contribution of precipitation to the observed uptake at high CsCl concentrations.

The mass of Sr taken up by the soil is likewise linear with the mass of Sr in the initial solution, as shown in Figure 12a. Figure 12b shows the mass of Sr taken up by the soil is linearly related to the mass of Sr remaining in the solution at low concentrations, but Figure 12c shows the correlation does not extend to higher concentrations, where the amount of Sr on the soil is significantly lower than predicted based on the lower solution concentrations. This may indicate that the surface capacity for Sr has been reached, but addition tests are needed to confirm that hypothesis. Note that the fitted lines in Figure 12b show no Sr is taken up by the soil at Sr solution concentrations below about 0.1 µg/mL. Since the amount taken up by the soil is calculated by subtracting the amount remaining in the test solution from the amount provided in the leachant, this means that Sr has been added from another source. This is consistent with the findings in the soil blank tests of Sr solution concentrations of 0.090, 0.080, and 0.125 µg/mL after reacting 1, 3, and 7 days in demineralized water, and indicates that Sr is being released from the soil during the tests, presumably due to the dissolution of a Sr-bearing phase, even as Sr is being taken up by the soil. Similar amounts of Sr were measured in tests conducted with the Cs solutions in the repeatability test series that were not mixed with Sr test solutions for analysis. While further analyses are needed to determine the processes that are occurring, this is an example of the convoluted processes that are empirically quantified using the distribution coefficient and the sensitivity of the K_d value to both the conditions of the soil and leachant used in the test and the testing parameters.

The Sr results are shown with a single line having a slope of 1 with the value at $x = 0$ equal to the average value of log K_F for all results (regardless of test duration) except the tests with the lowest Sr concentration, which are shown as open symbols. Exclusion of the results for the lowest concentration is justified by the fact that these results are dominated by the effect of Sr release from the soil. The parameter values for the linear isotherms for tests conducted for different durations are summarized in Table 20. The values of the x-intercept values give the solution concentrations when no Sr is calculated to be sorbed to the soil, based on the leachant and test solution concentrations. This represents the additional Sr that is released from the soil during the test.

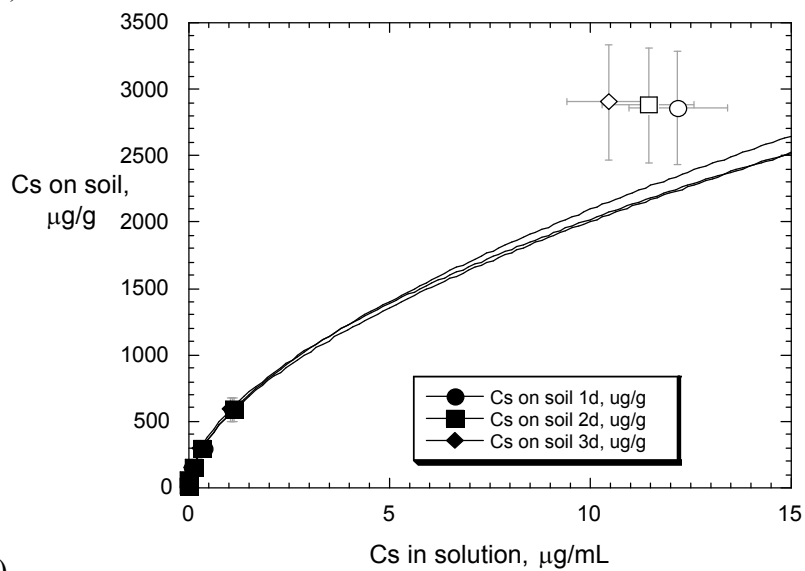Table 20. Values of K_d(Sr) calculated from the linear isotherm model

	Duration, days	K_d(Sr), mL/g	x-intercept Sr, µg/mL
Sr	1	376	0.120
	2	382	0.143
	3	394	0.0759
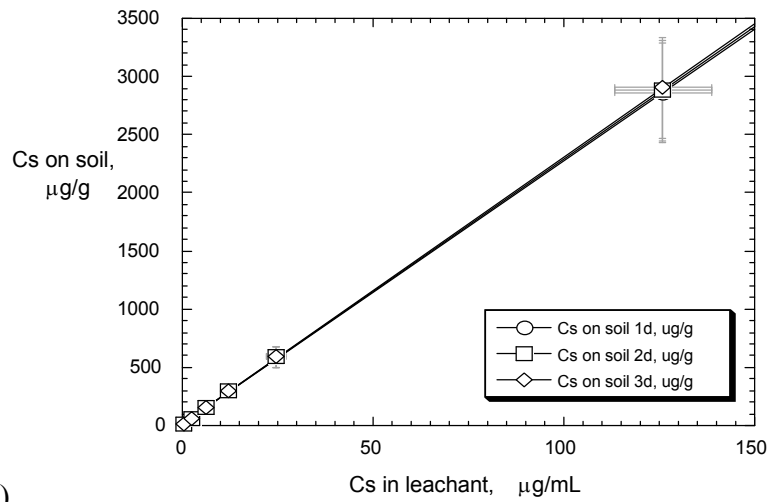
(a)
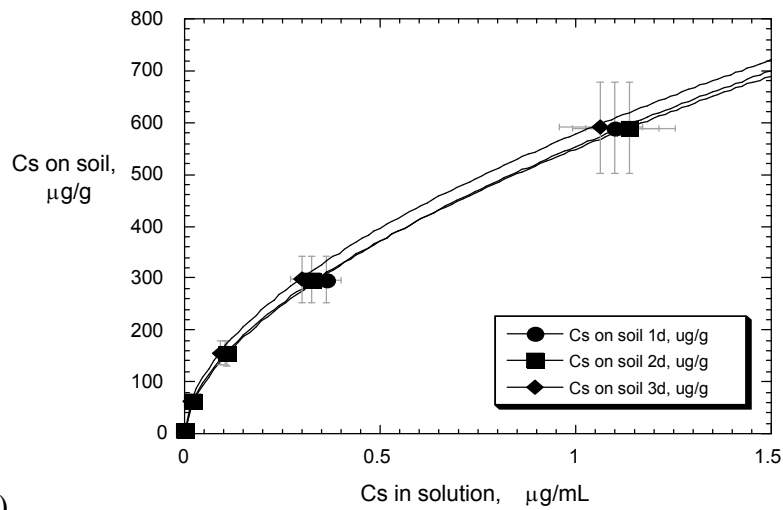
(b)

(c)

Figure 11. Correlation between Cs uptake onto soil and (a) Cs concentration in leachant prior to equilibration, (b) and (c) Cs concentration in solution after equilibration at low and high concentrations.
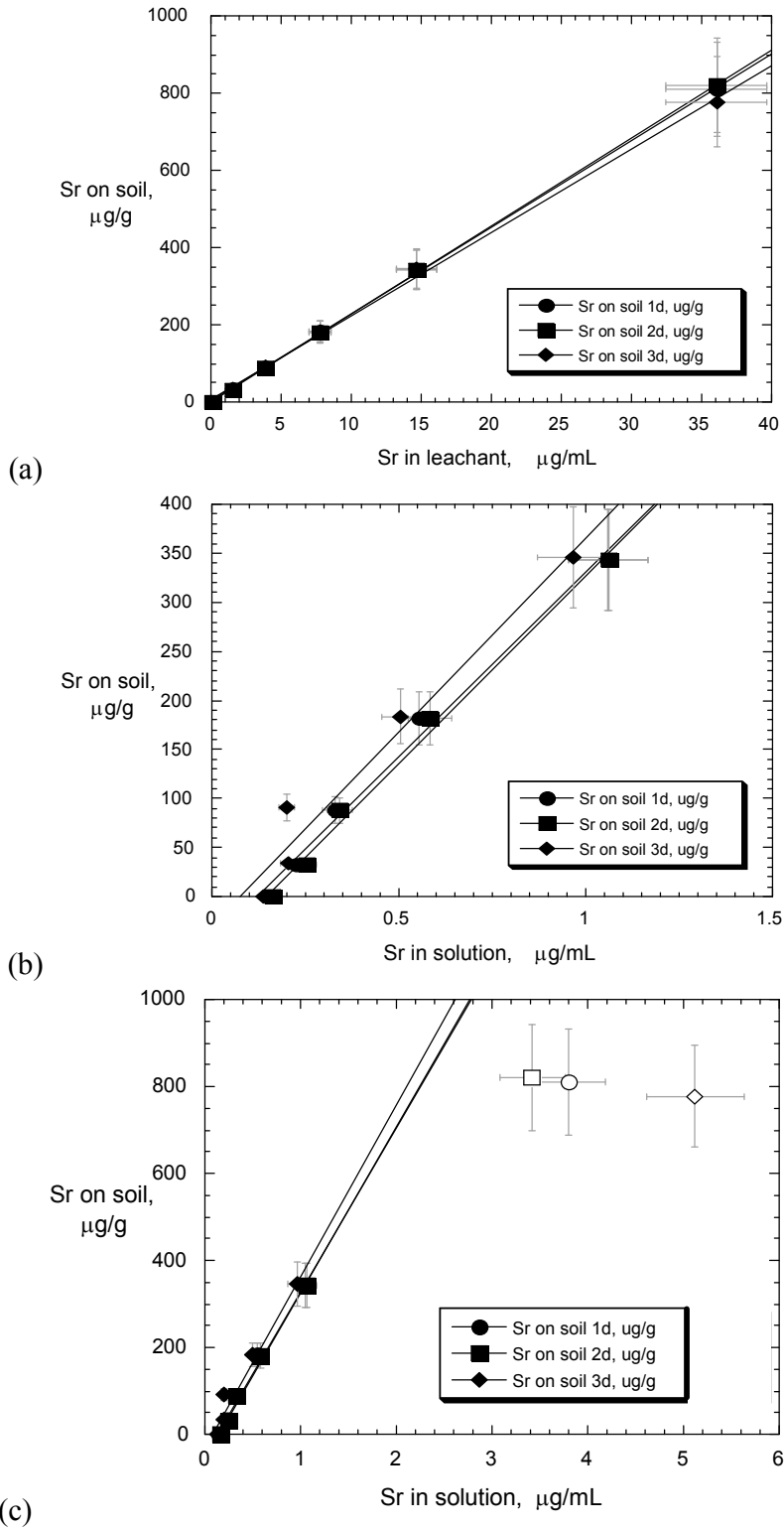
Figure 12. Correlation between Sr uptake onto soil and (a) Sr concentration in leachant prior to equilibration, (b) and (c) Sr concentration in solution after equilibration. The Sr B series solutions are shown as open symbols and were excluded from the regression.

Freundlich Isotherm Model

The Freundlich isotherm is a simple extension of the linear model that has the general form

concentration on solid = K_F × (concentration in solution)^n (8)

where K_F and n are constants, and is applied to systems at equilibrium. The Freundlich model allows for multilayer formation, whereas the Langumuir model discussed below is restricted to a monolayer. The linear isotherm is a special conditions of the Freundlich isotherm with $n = 1$, in which case Equation 7 is equivalent to Equation 1 and K_F equals K_d. In this case, a plot of the concentration on the solid versus the concentration in solution is expected to give a straight line with slope K_d that passes through the origin. The Freundlich isotherm expression can be linearized to evaluate systems for which $n \neq 1$ by taking the logarithm of Equation 8

$\log [Cs]_{soil} = \log K_F + n \log [Cs]_{solution}$ (9)

Plotting the logarithms of the concentrations should result in a linear fit with the slope providing the value of n and a y-intercept (i.e., the y value when $\log x = 0$) providing the value $\log K_F$. Figures 13a and 13b show the Cs and Sr uptakes in a log-log plot fit with Freundlich isotherms. The Cs results for the series tests run at each duration are regressed separately. The equations of the regressed lines are

for 1-day tests: $y = 2.77 + 0.593\ x$ (10)
for 2-day tests: $y = 2.78 + 0.583\ x$ (11)
for 1-day tests: $y = 2.81 + 0.568\ x$ (12)

where y is $\log [Cs]_{soil}$ and x is $\log [Cs]_{solution}$. The slope of each equation gives n and the y-intercept gives $\log K_F$. Note that Equation 8 has the same form as the power law used to empirically fit the Cs results as shown in Figure 11b and extrapolated in Figure 11c, and the parameter values are the same. Values of n significantly less than 1 indicate the the surface is heterogeneous with regard to the uptake of Cs. From the definition of K_d, the values of K_d and K_d are related as

$$K_d = \frac{[Cs]_{soil}}{[Cs]_{solution}} = K_F \times [Cs]_{solution}^{\ n-1}$$ (13)

Values of K_d determined for the experimental concentrations are given in Table 21.

Table 21. Values of K_d(Cs) calculated from the Freundlich isotherm

1 day $n = 0.593$ $K_F = 589$		2 days $n = 0.583$ $K_F = 603$		3 days $n = 0.568$ $K_F = 646$	
Cs in solution, µg/mL	K_d(Cs), mL/g	Cs in solution, µg/mL	K_d(Cs), mL/g	Cs in solution, µg/mL	K_d(Cs), mL/g
0.000545	12500	0.000406	15700	0.000296	21600
0.0227	2750	0.0214	3000	0.0149	3980
0.105	1470	0.107	1530	0.0933	1800
0.362	891	0.326	962	0.299	1090
1.10	566	1.14	571	1.06	629
12.2	213	11.5	218	10.5	234

The pooled Sr test results in Figure 13 b show linear behavior at high concentrations, but the regressed line does not pass through the origin. This is a result of Sr being released from the soil into solution during the test. Since the amount of Sr released from the soil may depend on the Sr solution concentration, how this should be taken into account when calculating K_d is not obvious. The equation of the regressed line in Figure 13b for tests with the higher solution concentrations is

$$\text{Sr:} \quad y = 2.398 + 0.9856\, x \quad (R^2 = 0.885) \tag{14}$$

from which $n = 0.986$ and $K_F = 250.0$. The observation that the value of n is very near 1 is consistent with the fit to the linear isotherm and the effective uniformity of the soil with respect to Sr sorption.
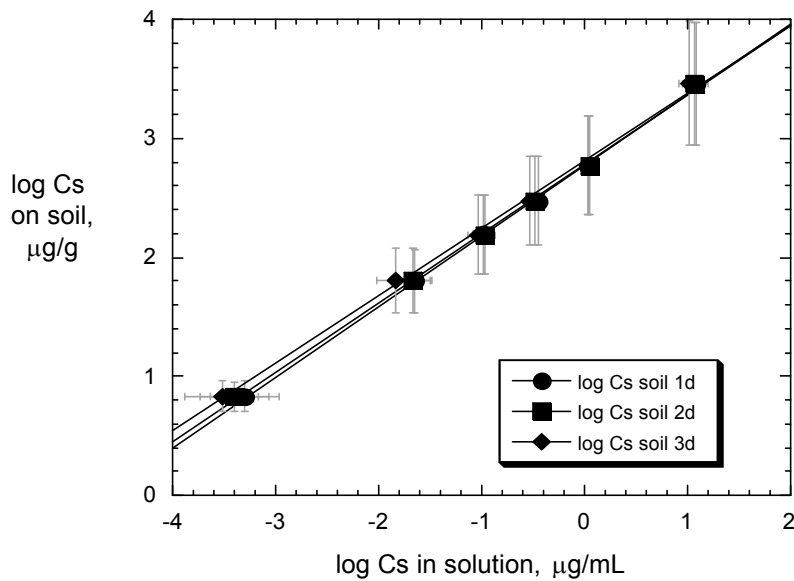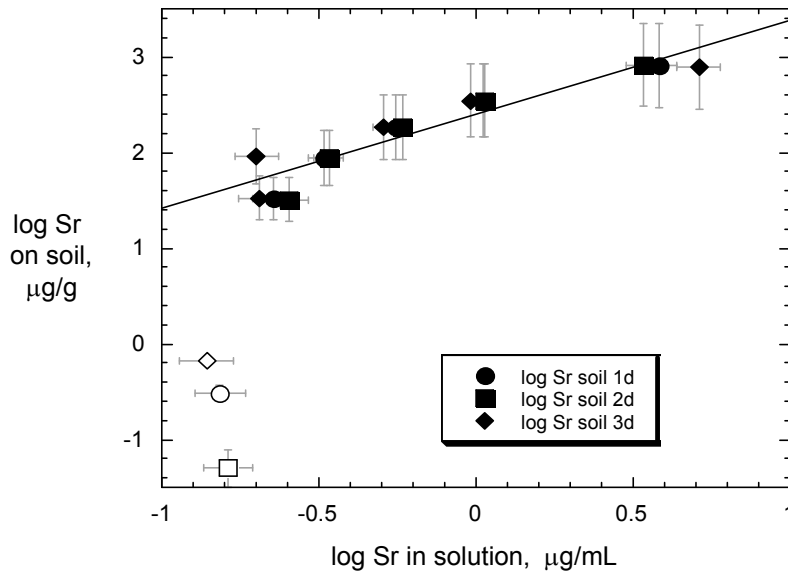
Figure 13. Evaluation of concentration effects using Freundlich isotherm model for tests with (a) Cs and (b) Sr. Results for the Sr E solution are shown as open symbols and were excluded from the regression.

45

Langmuir Isotherm Model

The Langmuir model addresses a sorption of a single species on a uniform surface with a maximum coverage of one monolayer. The isotherm equation has the form

$$C_{soil} = b \frac{K_L C_{solution}}{(1 + K_L C_{solution})},$$ (15)

where b is the sorption capacity of the soil (i.e., the maximum value of C_{soil}) and K_L is the Langmuir constant. This can be rearranged to a linear form that can be used to determine the model parameter values from experimental data as

$$\frac{C_{solution}}{C_{soil}} = \frac{1}{b\,K_L} + \frac{C_{solution}}{b}.$$ (16)

A plot of $C_{solution}/C_{soil}$ against $C_{solution}$ will yield a straight line with slope b^{-1} and intercept $(bK_L)^{-1}$. Data used for the Langmuir plots are provided in Table 22. Results for tests conducted a low concentrations of Cs and Sr in Figures 14a and 14b do not show Langmuir behavior. The results for tests with the D, E, and F solutions are shown as open symbols and were excluded from the linear fits, and the Sr results for the E solutions were excluded from the plot. The pooled results from 1-, 2-, and 3-day tests using higher concentrations were fit with the following equations:

Cs: $y = 0.00130 + 0.000235\,x$ ($R^2 = 0.958$) (17)
Sr: $y = 0.00241 + 0.000688\,x$ ($R^2 = 0.905$) (18)

Table 22. Data used in Langmuir isotherm plots

Test Solution	$Cs_{solution}$, µg/mL	$Cs_{solution}/Cs_{soil}$, mL/g	$Sr_{solution}$, µg/mL	$Sr_{solution}/Sr_{soi}$, mL/g$_1$
SJ-A2-1	1.10	0.001864	1.06	0.003090
SJ-B-3	12.2	0.004266	3.81	0.004698
SJ-C-3	0.362	0.001223	0.555	0.003050
SJ-D-3	0.0227	0.000361	0.227	0.006879
SJ-E-3	0.000500	0.000074	0.153	0.498370
SJ-F-3	0.105	0.000677	0.328	0.003732
SJ-A2-2	1.14	0.001932	1.06	0.003090
SJ-B-4	11.5	0.003993	3.42	0.004166
SJ-C-4	0.326	0.001098	0.582	0.003216
SJ-D-4	0.0214	0.000340	0.254	0.007864
SJ-E-4	0.000406	0.000060	0.163	3.2600
SJ-F-4	0.107	0.000690	0.341	0.003893
SJ-A2-3	1.06	0.001793	0.966	0.002792
SJ-B-5	10.5	0.003617	5.12	0.006581
SJ-C-5	0.299	0.001003	0.506	0.002765
SJ-D-5	0.0149	0.000236	0.205	0.006119
SJ-E-5	0.000296	0.000044	0.139	0.21061
SJ-F-5	0.0933	0.000602	0.200	0.002195
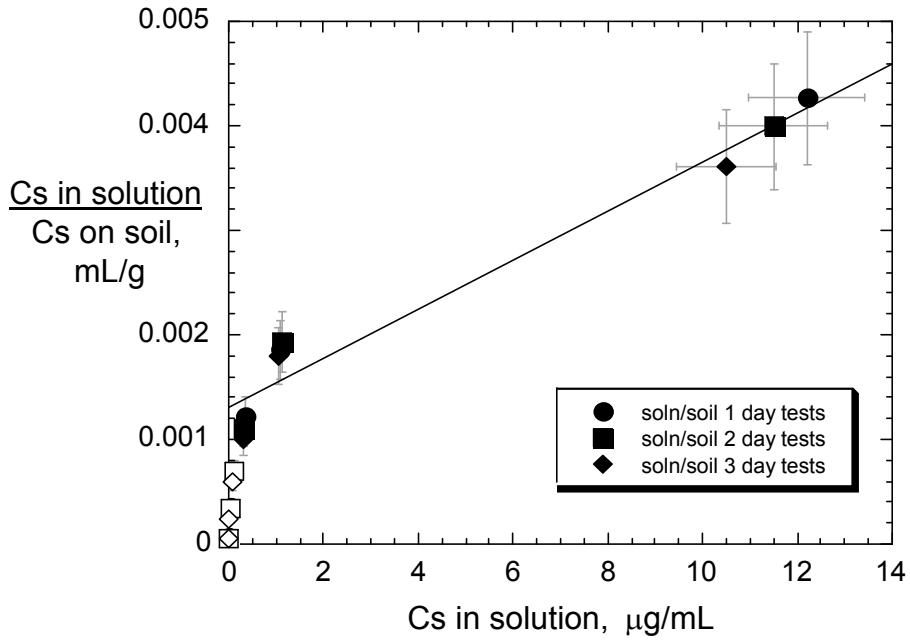
(a)
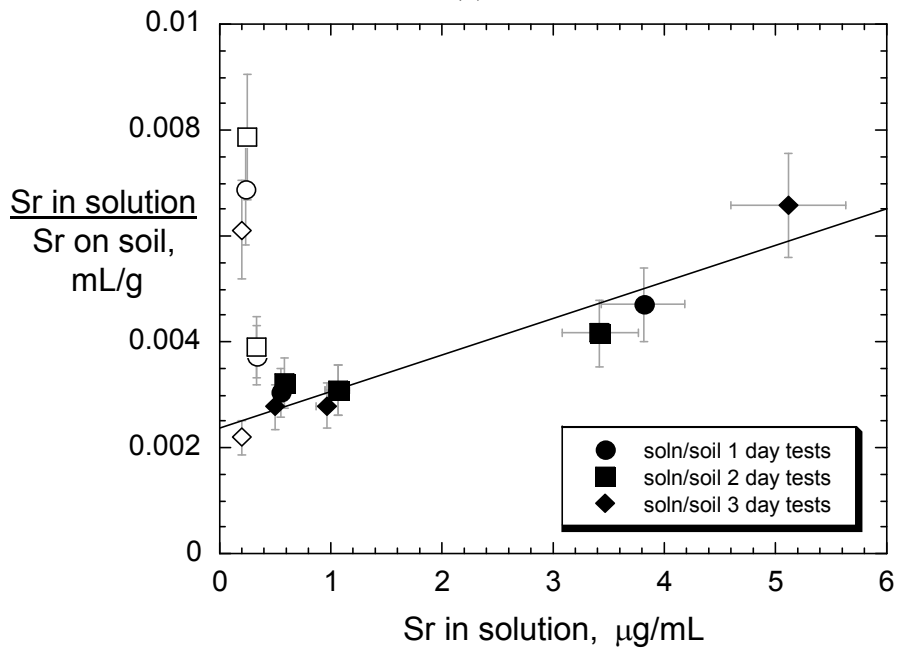
(b)

Figure 14. Evaluation of concentration effects using Langmuir isotherm model for tests with (a) Cs and (b) Sr. Results for the Cs and Sr D, E, and F solution tests are shown as open symbols; Results for the Sr E solution tests were excluded from the plot.

Based on the plotted values, the units of b are the same as C_{soil} (µg/g) and the units of bK_L are mL/g, so the corresponding units of K_F are mL/µg. From the Cs plot, $b = 4255$ µg Cs/g soil, $bK_L = 769.2$ mL/g, and $K_L = 0.181$ mL/µg Cs $= 1.81 \times 10^5$ mL/g Cs. From the Sr plot, $b = 1453$ µg Sr/g soil $bK_L = 414.9$ mL/g, and $K_L = 0.285$ mL/µg Sr $= 2.85 \times 10^5$ mL/g Sr.

3.8 Confirmation Tests

Two series of tests were conducted to confirm the conclusions drawn from the series of tests discussed above. Five replicate tests were conducted to provide a measure of the within-laboratory precision under the conditions recommended for the interlaboratory study (see Section 4.2). These were done due to the added calculations that were required due to our using the wrong solutions in the original series (see Section 3.4). Six tests were conducted to measure the effect of scale over the range of soil mass from 1 to 5 g, as identified in C1733. The original tests were conducted with a maximum of 1.5 g soil. These were also conducted to further evaluate if the effect of scale seen in Figure 5 was real.

Replicate Tests

Five replicate tests were conducted to demonstrate the approach for the interlaboratory study that is proposed above (see Section 4.2). Soil from Jar 1 and Jar 2 were combined (11.54 g from Jar 1 and 21.76 g from Jar 2) and mechanically mixed. A solution of CsCl referred to as Solution G was prepared in the silicate solution (0.0139 g CsCl + 0.0892 g NaHCO$_3$ + 0.0511 g SiO$_2$H$_2$O + 3 drops HNO$_3$) diluted to 500 g with demineralized water and then adjusted to 8.455 by adding a small amount of dilute NaOH. Tests were conducted in Teflon vessels at 30.0 °C for 48.00 hours. The vessels were rocked to suspend the soil several times over the test interval. At the end of the test duration, the vessels were opened and about 10 mL of solution was decanted into syringe filters and passed through 450 nm-pore-size cellulose filters into solution bottles. The test data are summarized in Table 23. The filtrates were acidified with 4 drops of concentrated HNO$_3$ and analyzed with ICP-MS. The analytical results are summarized in Table 24. The Cs concentration in the blank test with Leachant G was measured to be 17200 g/L, which is 9.25×10^{-5} m CsCl. As before, the mass sorbed to the soil was calculated from difference between the mass of Cs in the leachant and the mass remaining in solution. The sorbed mass was normalized to the mass of soil used in the test and used to calculate K_d(Cs) for each test.

Table 23. Data for replicate tests with Leachant G

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Final pH
SJM-Cs-G-1	48.00	1.00	25.01	not measured
SJM-Cs-G-2	48.00	1.00	25.01	not measured
SJM-Cs-G-3	48.00	1.00	25.00	not measured
SJM-Cs-G-4	48.00	1.00	25.00	not measured
SJM-Cs-G-5	48.00	1.00	25.04	not measured
SJM-Cs-G-B1	48.00	—	25.00	not measured

Test Number	Solution Bottle, g	Bottle + test solution, g	Bottle + test solution + DIW + HNO$_3$, g	Dilution Factor
SJM-Cs-G-1	11.16	33.65	33.71	1.0027
SJM-Cs-G-2	11.16	33.92	33.97	1.0022
SJM-Cs-G-3	11.13	34.04	34.08	1.0017
SJM-Cs-G-4	11.17	23.05	23.09	1.0034
SJM-Cs-G-5	11.15	30.61	30.65	1.0021
SJM-Cs-G-B1	11.21	27.94	27.98	1.0024

The mean value of K_d(Cs) is 373 mL/g with a standard deviation of 21 mL/g, giving a relative standard deviation of 5.6 %. This is significantly lower than the value K_d(Cs) = 522 mL/g measured for Leachant A2 having the slightly higher concentration of 1.35×10^{-4} molal CsCl. The mean values of the amounts measured in solution and calculated to be on the soil in the confirmation tests are shown plotted with values measured previously with various leachant concentrations in 2-day tests. The confirmation test results show lower amounts of Cs on the soil than would be predicted based on the concentration in solution. The equation of the regression line is $y = 2.78 + 0.853x$. For the measured solution concentration of 0.726 µg/mL Cs, a sorbed concentration of 459 µg/g Cs is predicted. This is 1.70-times the value of 270 µg/g Cs that was determined from the tests. The concentration dependence of K_d(Cs) must be taken into account to determine the precision of the test method. The pooled results of tests conducted with slightly different CsCl leachant concentrations can be compared to the composition dependence measured in this study (i.e., the results discussed in Section 3.7) to determine the intra-laboratory (between-laboratory) precision of the test method by using this approach.

Table 24. Results for replicate tests with Leachant G

Solution Number	Measured Cs, µg/L	Dil.-corrected Cs in test solution, µg/L	Leachant Volume, mL	Mass Cs in Leachant, µg
SJM-Cs-G-1	727	728.94	25.01	18.23
SJM-Cs-G-2	683	684.50	25.01	17.12
SJM-Cs-G-3	706	707.23	25.00	17.68
SJM-Cs-G-4	715	717.41	25.00	17.94
SJM-Cs-G-5	788	789.62	25.04	19.77
SJM-Cs-G-B1	17200	17241.12	25.00	18.23

Test Numbers	Mass Cs on soil, µg	µg Cs/g soil	Cs in test solution, µg/mL	K_d (Cs), mL/g
SJM-Cs-G-1	270.21	270	0.729	370.69
SJM-Cs-G-2	271.32	271	0.685	396.38
SJM-Cs-G-3	270.76	271	0.707	382.85
SJM-Cs-G-4	270.51	271	0.717	377.06
SJM-Cs-G-5	268.67	269	0.790	340.25
Mean		*270*	0.726	*373.45*
Standard Deviation		—	0.0394	*20.84*
Relative Standard Deviation		—	*5.43%*	*5.58%*

Supplemental Effect of Scale Tests
A series of six tests were conducted to measure the effect of the test scale (i.e., the amounts of soil and solution) over a wider range than was addressed in previous tests (see Section 3.4). The approximately 330 g of Leachant G that remained after conducting the supplemental replicate tests was diluted with about 80 g of demineralized water to provide enough leachant to conduct the series of effect of scale tests. The resulting solution is referred to as Leachant H. The series of tests is summarized in Table 25. The analytical results and calculated values are provided in Table 26. The Cs concentration in the blank test with Leachant H was measured to be 14635 g/L, which is 7.85×10^{-5} molal CsCl. This value was used to calculate the sorbed mass and K_d(Cs) value for each test. The results are summarized in Table 26 and plotted in Figure 16. The values of K_d(Cs) show a slight increase with the amount of soil, but this is well within the uncertainty of each value (which is about 15%). The mean value of K_d(Cs) is 695 mL/g and
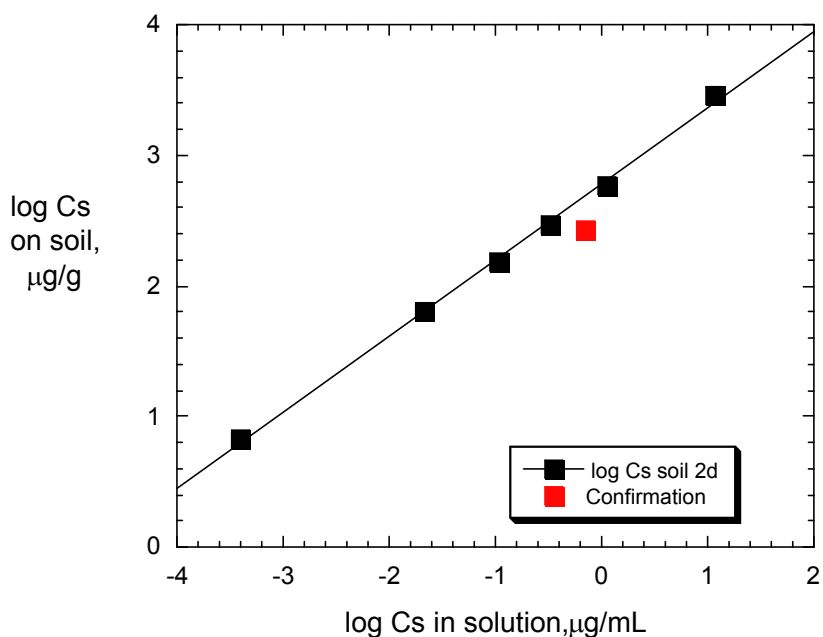
Figure 15. Comparison of confirmation tests result with measured composition dependence of $K_d(Cs)$.

the standard deviation is 17 mL/g. The higher $K_d(Cs)$ value measured with Leachant H compared to that measured with Leachant G is attributed to the lower Na concentration in Leachant H, which competes with Cs for sorption sites. The Na concentration in Leachant H is lower because Leachant G was diluted with demineralized water rather than the silicate solution that was used to make the other leachants.

Table 25. Data for tests conducted with Leachant H and different amounts of soil and solution

Test Number	Duration, h	Mass Soil, g	Mass Leachant, g	Soil/Leachant mass ratio	Final pH
SJM-Cs-H-1	47.22	0.50	12.52	25.04	not measured
SJM-Cs-H-2	47.22	1.00	25.00	25.00	not measured
SJM-Cs-H-3	47.22	2.00	51.99	26.00	not measured
SJM-Cs-H-4	47.22	3.00	75.00	25.00	not measured
SJM-Cs-H-5	47.22	4.01	100.23	25.00	not measured
SJM-Cs-H-6	47.22	5.00	125.00	25.00	not measured

Test Number	Solution Bottle, g	Bottle + Cs solution, g	Bottle + Cs solution + DIW + HNO$_3$, g	Cs Dilution Factor
SJM-Cs-H-1	11.2	22.09	22.14	1.0046
SJM-Cs-H-2	11.17	30.04	30.09	1.0026
SJM-Cs-H-3	11.17	23.79	23.84	1.0040
SJM-Cs-H-4	11.16	25.36	25.41	1.0035
SJM-Cs-H-5	11.18	27.06	27.11	1.0031
SJM-Cs-H-6	11.17	25.94	25.97	1.0020
Leachant H	11.15	32.27	32.32	1.0024

Table 26. Results for tests conducted with Leachant H and different amounts of soil and solution

Test Number	Measured Cs, µg/L	Dil.-corrected Cs in test solution, µg/L	Mass Cs in test solution, µg
SJM-Cs-H-1	518	520.4	6.52
SJM-Cs-H-2	522	523.4	13.08
SJM-Cs-H-3	525	527.1	27.40
SJM-Cs-H-4	505	506.8	38.01
SJM-Cs-H-5	489	490.5	49.17
SJM-Cs-H-6	501	502.0	62.75
Leachant H	14600	14635	—

Test Number	Leachant Volume, mL	Mass Cs in Leachant, µg	Mass Cs on soil, µg	µg Cs/g soil	K_d (Cs) mL/g
SJM-Cs-H-1	12.52	183.22	176.71	353.42	679.16
SJM-Cs-H-2	25.00	365.86	352.78	352.78	674.04
SJM-Cs-H-3	51.99	760.85	733.45	366.72	695.77
SJM-Cs-H-4	75.00	1097.59	1059.58	353.19	696.94
SJM-Cs-H-5	100.23	1466.82	1417.66	353.53	720.70
SJM-Cs-H-6	125.00	1829.32	1766.57	353.31	703.79

The horizontal dashed lines in Figure 16 are drawn at the overall average values. The K_d(Cs) values show a slight upward trend (as opposed to the slight downward trend seen in Figure 5) that is well within the estimated 15% experimental uncertainty in determining the value of K_d(Cs).

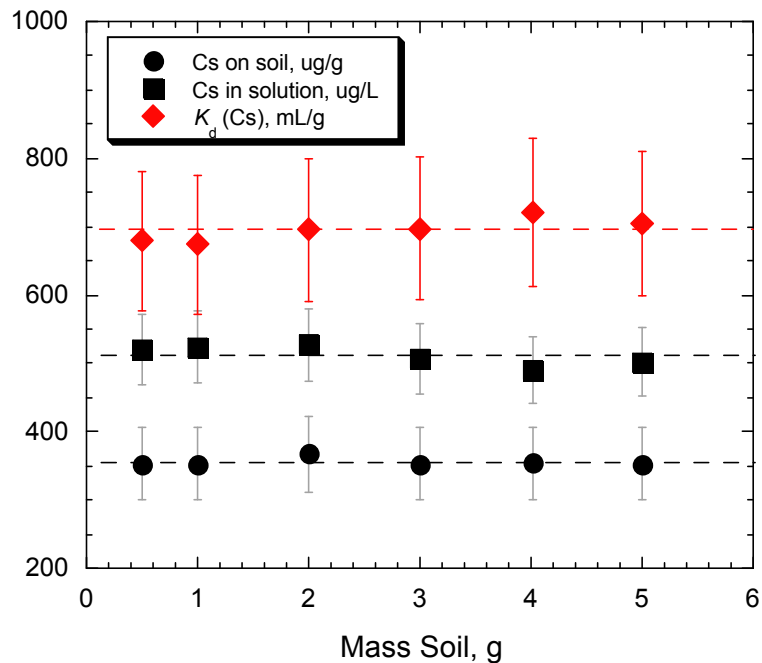Figure 16. Results of effect of scale tests conducted with Leachant H.

51

# 4.	SUMMARY AND RECOMMENDATIONS

Tests were conducted to measure the uptake of Cs and Sr dissolved as CsCl and $Sr(NO_3)_2$ in a dilute $NaHCO_3/SiO_2$ solution (representing contaminants in a silicate groundwater) by a NIST standard reference material of San Joaquin soil (SRM 2709a). Tests were run to measure the repeatability of the method and the sensitivity of the test response to the reaction time, the mass of soil used (at a constant soil-to-solution ratio), the solution pH, and the contaminant concentration. All tests were conducted in screw-top Teflon vessels at 30 °C in an oven. All solutions were passed through a 0.45-m pore size cellulose acetate membrane filter and stabilized with nitric acid prior to analysis with inductively-coupled plasma mass spectrometry (ICP-MS). Scoping tests with soil in demineralized water resulted in a solution pH of about 8.0 and the release of small amounts of Sr from the soil. Solutions were made with targeted concentrations of 1×10^{-6} $\underline{m}$, 1×10^{-5} $\underline{m}$, 2.5×10^{-5} $\underline{m}$, 5×10^{-5} $\underline{m}$, 1×10^{-4} $\underline{m}$, and 5×10^{-4} $\underline{m}$. (The actual Cs concentrations were about 50% higher and the actual Sr concentrations about 30% lower than these targeted values.) The pH values of all solutions were adjusted to about pH 8.5 so that the effects of pH and concentration could be measured separately. The nominal 1×10^{-4} $\underline{m}$ solutions were used to measure the repeatability and the effects of duration, scale, and imposed pH.

## 4.1	Testing Results

Repeatability

Replicate tests run to measure the intra-laboratory precision of the method using the 1.5×10^{-4} $\underline{m}$ Cs solution resulted in a mean value of $K_d(Cs) = 522$ mL/g, standard deviation of 16 mL/g, and relative standard deviation of about 3.1% in $K_d(Cs)$ for the pooled results of 1 and 2-day tests. Replicate tests using the 0.7×10^{-4} $\underline{m}$ Sr solution resulted in a mean value of $K_d(Sr) = 162$ mL/g, standard deviation of 8 mL/g, and relative standard deviation of 5% in $K_d(Sr)$ for 1-day tests. The uncertainty in the calculated value of K_d was determined by propagation of errors method to be about 15% due primarily to uncertainty in the measured concentration of the test solution. The repeatability of the test under the conditions used in this study (the intra-laboratory precision) is well within the uncertainty of the individual $K_d(Cs)$ values. Confirmation tests conducted for 2 days using 9.25×10^{-5} $\underline{m}$ CsCl solution resulted in a mean value of $K_d(Cs) = 373$ mL/g, standard deviation of 21 mL/g, and relative standard deviation of 5.6%.

Effect of Scale

Values of $K_d(Cs)$ in 1-day tests conducted using the 1.5×10^{-4} $\underline{m}$ CsCl solution with 0.2, 0.5, 1.0, and 1.5 g soil with solution masses of 25-times the soil mass showed a small negative correlation outside the uncertainty due to the repeatability of the tests, but within the uncertainties of the individual tests. Confirmation tests using 9.25×10^{-5} $\underline{m}$ CsCl solution and up to 5 g soil showed no effect of scale.

Effect of Duration

Steady-state concentrations were attained within about 1 hour (the shortest test duration was about 15 minutes) in tests conducted with 0.5 g soil in either 12.5 g of the 1.5×10^{-4} $\underline{m}$ Cs solution or 12.5 g of the 0.7×10^{-4} $\underline{m}$ Sr solution and persisted through the longest test duration, which was about 7 days. The rapid uptakes of Cs and Sr measured in these tests are consistent with the uptakes measured for Cs and Sr on similar soilds; for example Cs on humic acid (Celebi et al. 2009), Cs on various clays (Bayulken et al. 2010), Cs on soil (Setiawan 2007), Cs on clay (Ugur and Turhan, 2001), Cs and Sr on Zeolite A (el-Rahman, et al. 2006), Sr on uranium antimontate (Kumar and Sudarsan, 2007), and Sr on synthetic muscovite (Sharma and Shirvastava 2010).

Effect of Imposed pH

The sorption of cesium in 1- and 2-day tests with the 1.5×10^{-4} $\underline{m}$ Cs solution was independent of the imposed pH within the range pH 7.5–pH 9.5, whereas the uptake of strontium from the 1.5×10^{-4} $\underline{m}$ Sr solution increased exponentially over this range. The solution pH drifted to near the "equilibrium" pH of the soil/solution system under these test conditions (i.e., temperature and relative masses of soil and water) in all tests. The use of a pH buffer was avoided to eliminate possible complexation and competitive sorption. Previous tests have shown the value of K_d(Cs) to increase with pH over the range pH 3 to pH 8 (Bayülken et al. 2010) but to decrease between pH 8 and pH 10 (Giannakopoulou et al. 2007). Values of K_d(Ba) on humic acid was seen to increase between pH 2 and pH 4, remain nearly constant between pH 4 and pH 8, and then decrease between pH 8 and pH 10 (Celebi et al. 2009). el-Rahman et al. (2008) found the sorption of Cs and Sr onto Zeolite A increased with pH from pH 2 to pH 8. Westrich et al. (2000) showed the uptake of Sr by quartz and goethite increased from near 0% to 80% between pH 4 and pH 10 and was well-described using a 1-site model, but the uptake on smectite remained between 60% and 80% over this range and was better fit with a 2-site model.

Effect of Concentration

Tests were conducted for about 1, 2, and 3 days with 0.5 g soil and 12.5 g of each solution concentration. The Cs results were well-fitted with a Freundlich isotherm model with K_F(Cs) = 646 mL/g and $n = 0.568$ (for the 3-day tests). The Sr uptake followed a linear isotherm with K_d(Sr) = 394 (for the 3-day tests), except at the highest Sr concentration, which was significantly lower than that predicted and may indicate that the soil was saturated with Sr. The values of both K_F(Cs) and K_d(Sr) increased slightly with the test duration, suggesting that steady state was not attained within one day at the high solution concentrations. Langmuir isotherms adequately described the results of tests at the highest concentration, but the results at low concentrations were not fit well.

The data of Celebi et al. (2009) and Bayulken et al. (2010) for Cs sorption on humic acid and clay, respectively, were fitted well with Freundlich isotherms. Tests conducted by Uğur and Turhan (2011) to measure the sorption of Cs on different size fractions of clay were well fit with Langmuir isotherms.

4.2   Recommended Conditions for an ASTM C1733 Reference Test

The focus of the study described in the report was to (1) evaluate the inherent uncertainty associated with performing the ASTM C1733 standard batch test to generate data used to calculate K_d values and (2) recommend a reference test material, test solution, and test conditions that can be used to establish a test response and precision that can be used to gauge the success of tests conducted by new users and separate the uncertainty from test performance from real effects of test conditions (i.e., other solids, solutions, and testing parameters). The precision of conducting the ASTM C1733 test and the dependencies on test variables that have been evaluated are believed to represent the least-biased conditions. Confounding factors that were not considered in this analysis include contributions to the uncertainty from the day-to-day variance in the analytical measurements (since solutions from groups of tests were analyzed as a single set), test performance by different operators, size and specific surface area the use of a finely divided and compositionally homogeneous NIST standard soil, ionic strength and competitive sorption. Many of these factors will be taken into account in measures of the inter-laboratory precision, while other issues are topics for research studies for effects beyond test execution.

The following provide recommendations for test materials and test conditions that can be used as a reference test to directly compare results of tests conducted at different laboratories to verify tests and

analyses are conducted properly. They are also recommended for use in inter-laboratory studies to measure the reproducibility of the test method and establish test values suitable for direct comparisons.

Reference Soil

The NIST SRM 2709a San Joaquin soil is a suitable standard material for direct use as a sorbent for determining the bias of procedure ASTM C1733. The as-received dried soil can be used in testing without further preparation (washing or sizing). A large amount of soil is available from NIST and accessible to researchers world-wide in 50-g allotments. The composition of the soil is well-characterized and its physical characteristics acceptably uniform. However, it is recommended that a single source be used by participants in inter-laboratory studies to mitigate the effects of any differences between allotments, which could be a mixture of soil from more than one 50-g jar of SRM 2709a.

Reference Solution

The use of CsCl dissolved in a dilute $NaHCO_3/SiO_2$ matrix solution is appropriate for measuring experimental bias and testing precision. The matrix solution is prepared following the recipe provided in ASTM C1220 step 7.4, which includes adjustment to pH 8.0. It is recommended that the pH be adjusted to pH 8.5 (see Reference pH below) after the addition of CsCl and that each solution concentration be adjusted separately. The range of concentrations used in the present study is adequate for studying the concentration dependences and evaluating isotherms. The solutions appear to be stable over time and Cs is not released from the San Joaquin soil to a significant extent. The uptake of Cs occurs rapidly and appears to follow a relatively simple mechanism that displays Freundlich behavior over a fairly wide compositional range. A solution composition of 1.5×10^{-4} $\underline{m}$ CsCl (0.0253 g CsCl/kg solution) is recommended for use in tests to measure the repeatability and reproducibility of the C1733 method.

Reference Imposed pH

It is recommended that the solutions be adjusted to pH 8.5 when they are prepared. The matrix solution that is prepared following the recipe provided in ASTM C1220 step 7.4 includes an adjustment to pH 8.0. The present tests were conducted with solutions that had been adjusted to pH 8.5 to be slightly higher than the pH that was attained in soil blank tests. It is recommended that both the matrix solution and the Cs solutions be adjusted to pH 8.5 to mitigate pH drift during the test. The uptake of Cs was measured to be fairly insensitive to the pH over the range pH 8.0–9.1, so the effect of a small pH drift on the test results will be minor.

Reference Soil and Solution Mass

A sample size of 1 g dry soil and 25 g solution is recommended for reference tests. The use of between 1 and 5 g of soil is recommended in the ASTM C1733 method. The logical reason for this is need for small samples to provide a uniform and reproducible phase assemblage (and relative surface areas) that represents the environment of interest. The present tests show SRM 2709a soil is sufficiently uniform that less than 1 g can be used in scoping tests and when limited amounts of soil are available. For the present tests, 0.5-g sample sizes were used in order to conduct as many tests as possible using the 100-g of soil that was available. The use of 12.5 mL solution in these provides ample solution for analysis without excessive waste, which could be an issue when using hazardous soils or groundwaters. Nevertheless, a sample size of 1 g is recommended for inter-laboratory tests conducted to measure the test precision and reproducibility to be consistent with the range of soil mass called for in the procedure.

Reference Test Duration

A 2-day test duration is recommended for reference tests. Although 1-day tests were adequate to quantify the intra-laboratory precision and effects of test variables, a 2-day during will reduce the impact of uncontrolled testing variables, such as the exact time the system is at temperature. The present tests suggest that kinetic effects may have contributed small differences to the measured effects of pH and contaminant concentration, albeit within the experimental uncertainty.

Reference Temperature

A temperature of 30 °C is recommended for reference tests. Although all tests in this study were conducted at 30 °C and the sensitivity of the test response to temperature was not evaluated, this temperature is easily attainable in the laboratory using an incubator or low-temperature oven and avoids the variance and fluctuations of ambient temperatures common to most laboratories. While temperatures below ambient laboratory temperatures are more representative of subaerial conditions (e.g., 20 °C), refrigeration units are less-common in laboratories and the value of is not expected to be significantly different over a ~20 °C temperature range near ambient.

Replicate Tests

It is recommended that at least five replicate tests be conducted to measure the intra-laboratory precision (repeatability) at each laboratory and to provide data for determining the inter-laboratory precision (reproducibility). The tests should be conducted by a single operator and analyzed as a set using the same instrument calibration. Although it was not done in the present study, a sample of the leachant solution should be analyzed together with the test solutions to eliminate any contribution of the day-to-day analytical variance of solution analysis. Note that the $K_d(Cs)$ values from tests conducted in this study are not suitable for use in inter-laboratory comparisons because the test solutions from test conducted for 1 and 2 days were combined (inadvertently) for analysis and added uncertainty to the results, despite the finding that test duration beyond 1 day did not significantly contribute to the variance. It is expected that small differences in the leachant Cs concentration will provide the most variance in an inter-laboratory study. This is supported by the results of confirmation replicate tests conducted with a less concentrated CsCl solution. It is expected that the effect of CsCl concentration on the value of $K_d(Cs)$ measured in this study can be used to distinguish between the effects of concentration and test precision in an inter-laboratory study.

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Appendix A: Chemical Composition of SRM 2709a

The compositions provided in the NIST Certificate of Analysis for SRM 2709a (NIST 2011) are provided below. The certified values in Table 1 represent the means from analyses using one, two, or three methods conducted at NIST and/or the US Geological Society (Denver, CO) and the expanded uncertainties. The reference values in Table 2 (with uncertainties) and informational values Table 3 (without uncertainties) represent the results of single method analyses performed at NIST.

Table 1. Certified Values[a] (Dry-Mass Basis) for Selected Elements in SRM 2709a

Element	Mass Fraction (%)			Element	Mass Fraction (mg/kg)		
Aluminum	7.37	±	0.16	Antimony	1.55	±	0.06
Calcium	1.91	±	0.09	Barium	979	±	28
Iron	3.36	±	0.07	Cadmium	0.371	±	0.002
Magnesium	1.46	±	0.02	Chromium	130	±	9
Phosphorus	0.0688	±	0.0013	Cobalt	12.8	±	0.2
Potassium	2.11	±	0.06	Lead	17.3	±	0.1
Silicon	30.3	±	0.4	Manganese	529	±	18
Sodium	1.22	±	0.03	Strontium	239	±	6
Titanium	0.336	±	0.007	Vanadium	110	±	11
				Zirconium	195	±	46

Table 2. Reference Values[a] (Dry-Mass Basis) for Selected Elements in SRM 2709a

Element	Mass Fraction (mg/kg)		
Arsenic	10.5	±	0.3
Cerium	42	±	1
Cesium	5.0	±	0.1
Copper	33.9	±	0.5
Europium	0.83	±	0.02
Gadolinium	3.0	±	0.1
Lanthanum	21.7	±	0.4
Mercury[b]	0.9	±	0.2
Nickel	85	±	2
Rubidium	99	±	3
Scandium	11.1	±	0.1
Thallium	0.58	±	0.01
Thorium	10.9	±	0.2
Uranium	3.15	±	0.05
Zinc	103	±	4

Table 3. Information Values[a] (Dry Mass Basis) for Selected Elements in SRM 2709a

Element	Mass Fraction (mg/kg)
Boron	74
Dysprosium	3
Hafnium	4
Lutetium	0.3
Neodymium	17
Samarium	4
Selenium	1.5
Tantalum	0.7
Terbium	0.5
Ytterbium	2

Appendix B: Calculations

B.1   Preparation of synthetic silicate groundwater (SSG)

Bottle 1 tare = 184.40 g
Bottle 1 + 0.1971 g $SiO_2 \cdot 2H_2O$ + 0.3577 g $NaHCO_3$ + DIW = 1184.42 g
1000.02 g solution
Initial pH 9.21. Adjusted to pH 8.99 by adding dilute HNO_3
Bottle 2 tare = 182.17 g
Bottle 2 + 0.1920 g $SiO_2 \cdot 2H_2O$ + 0.3585 g $NaHCO_3$ + DIW = 1182.16 g
999.99 g solution
Initial pH 9.28. Adjusted to pH 8.98 by adding dilute HNO_3.

B.2   Preparation of Cs and Sr solutions

Formula weight: CsCl = 168.36 g/mole

$$\text{CsCl stock bottle 1:} \quad \frac{0.3367 \text{ g CsCl}}{100.00 \text{ g solution}} \frac{1 \text{ mole}}{168.36 \text{ g CsCl}} \frac{1000 \, g}{1 \, kg} = 0.0200 \; molal \tag{B.1}$$

$$\text{CsCl stock bottle 2:} \quad \frac{0.3351 \text{ g CsCl}}{100.00 \text{ g solution}} \frac{1 \text{ mole}}{168.36 \text{ g CsCl}} \frac{1000 \, g}{1 \, kg} = 0.0199 \; molal \tag{B.2}$$

Formula weight: $Sr(NO_3)_2$ = 211.63 g/mole

$$Sr(NO_3)_2 \text{ stock bottle 1:} \quad \frac{0.4233 \text{ g } Sr(NO_3)_2}{100.00 \text{ g solution}} \frac{1 \text{ mole}}{211.63 \text{ g } Sr(NO_3)_2} \frac{1000 \, g}{1 \, kg} = 0.0200 \; molal \tag{B.3}$$

$$Sr(NO_3)_2 \text{ stock bottle 2:} \quad \frac{0.4279 \text{ g } Sr(NO_3)_2}{100.00 \text{ g solution}} \frac{1 \text{ mole}}{211.63 \text{ g } Sr(NO_3)_2} \frac{1000 \, g}{1 \, kg} = 0.0202 \; molal \tag{B.4}$$

B.3   Densities of Cs and Sr solutions

Volume of volumetric flask:
Mass empty flask: 56.17 g;
Mass flask + DIW: 155.19 g;
Mass DIW = 155.37 - 56.17 = 99.20 g
Density of water at 30 °C = 0.99565 g/mL;

$$\text{Volume volumetric flask} = \frac{99.20 \text{ g}}{0.99565 \, g/mL} = 9.963 \text{ mL} . \tag{B.5}$$

Mass empty flask: 56.18 g;
Mass flask + 1.5×10^{-4} $\underline{m}$ CsCl solution = 155.28 g;
Mass Cs solution = 155.28 – 56.17 = 99.09 g.

$$\text{Density CsCl solution} = \frac{99.09 \text{ g}}{9.963 \text{ mL}} = 0.9946 \text{ g/mL} . \tag{B.6}$$

Mass empty flask: 55.99 g;
Mass flask + 0.7 $\times$ 10^{-4} $\underline{m}$ $Sr(NO_3)_2$ solution = 155.28 g;
Mass Sr solution = 155.28 – 55.99 = 99.29 g.

$$\text{Density Sr}(NO_3)_2 \text{ solution} = \frac{99.29 \text{ g}}{9.963 \text{ mL}} = 0.9966 \text{ g/mL} . \tag{B.7}$$

B.4   Bragg's law calculation of d-spacing

Bragg's law (Equation 6) was rearranged to solve for *d* as

$$d = \frac{\lambda}{2\sin\left(2\theta/2\right)} . \tag{B.8}$$

The 2θ values of the peaks were determined by inspection of the spectrum and the digitized file provided by the instrument. The value of 2θ was converted from degrees to radians by multiplying the value in degrees by pi/180 (pi = 3.14159). The *d*-spacing was then calculated for λ= 1.5406 Å. For example, the most intense peak in Table 1 occurs at 2θ = 26.630 degrees, which is

$$26.630 \text{ degrees} \frac{3.14159}{180 \text{ degree}} = 0.46478 \text{ radians}. \tag{B.9}$$

Solving for the *d*-spacing gives

$$d = \frac{1.5406 \text{ A}}{2\sin\left(0.46478/2\right)} = 3.3447 \text{ A}. \tag{B.10}$$

Background intensities were estimated by graphically connecting background signals on both sides of the peak and reading the background intensity at the 2θ value of the peak. The background intensity was subtracted from the peak intensity.

B.5   Test solution dilution factor and dilution-corrected solution concentration

The test solution recovered from the test vessel was passed through a 0.45-m pore size filter directly into a pre-weighed polyethylene solution bottle. An aliquot of the solution was removed for pH analysis and the remaining solution weighed. Demineralized water was added to some test solutions so that about 10 mL was available for analysis. About 0.05 g concentrated nitric acid was added to the solution and the total mass weighed. A dilution factor was calculated from the masses and used to correct the measured concentrations. In the following expressions, parentheses are used to denote measured quantities.

$$\text{dilution factor} = \frac{(\text{mass bottle} + \text{test solution} + \text{DIW} + \text{HNO}_3) - (\text{mass bottle})}{(\text{mass bottle} + \text{test solution}) - (\text{mass bottle})} \tag{B.11}$$

$$\text{dilution-corrected concentration} = \text{measured solution concentration} \times \text{dilution factor} \tag{B.12}$$

For example, in Table 3, the measured masses for Cs-Blank-1 test solution are
mass bottle = 11.09 g
mass bottle + test solution = 23.32 g
mass bottle + test solution + DIW + HNO$_3$ = 23.36 g.

$$\text{The dilution factor for the Cs solution is } \frac{23.36 - 11.09}{23.32 - 11.09} = 1.0033. \tag{B.13}$$

Also from Table 3, the measured masses for Sr-Blank-1 test solution are
mass bottle = 11.10 g
mass bottle + Sr test solution = 23.35 g
mass bottle + Sr test solution + DIW + HNO$_3$ = 23.40 g.

$$\text{The dilution factor for the Sr solution is } \frac{23.40 - 11.10}{23.35 - 11.10} = 1.0041. \tag{B.14}$$

B.6   Dilution factors for mixed test solutions

After they were acidified, test solutions from corresponding tests with Cs and Sr were combined for analysis. The solutions were combined in a new solution bottle. The masses are measured for the empty bottle, the bottle + the Cs solution, and bottle + Cs solution + Sr solution. The dilution factors for the Cs and Sr test solutions due to this mixing were calculated as

$$\text{Cs dilution factor} = \frac{(\text{mass bottle} + \text{Cs test solution} + \text{Sr test solution}) - (\text{mass bottle})}{(\text{mass bottle} + \text{Cs test solution}) - (\text{mass bottle})} \tag{B.15}$$

$$\text{Sr dilution factor} = \frac{(\text{mass bottle} + \text{Cs test solution} + \text{Sr test solution}) - (\text{mass bottle})}{(\text{mass bottle} + \text{Cs test solution} + \text{Sr test solution}) - (\text{mass bottle} + \text{Cs test solution})} \tag{B.16}$$

The test solutions Cs-Blank-1 and Sr-Blank-1 were mixed for analysis. From Table 4, the measured masses were
Mass bottle = 11.09 g
Mass bottle + acidified Cs-Blank-1 test solution (solution 1) = 23.24 g
Mass bottle + Cs solution + acidified Sr-Blank-1 test solution (solution 2) = 35.46 g

$$\text{Cs dilution factor} = \frac{35.46 \text{ g} - 11.09 \text{ g}}{23.24 \text{ g} - 11.09 \text{ g}} = 2.0058 \tag{B.17}$$

$$\text{Sr dilution factor} = \frac{34.46 \text{ g} - 11.09 \text{ g}}{34.46 \text{ g} - 23.24 \text{ g}} = 1.9943. \tag{B.18}$$

The total dilution factor for the Cs solution is 1.0033 × 2.0058 = 2.0124. The dilution due to acidification contributes 0.3% to the total and was ignored in the calculations.

From Table 5, the Cs and Sr concentrations measured in the combined solution Cs/Sr-Blank-1 were 12500 μg/L Cs and 5450 μg/L Sr. The dilution-corrected Cs and Sr concentrations are

$$2.0124 \frac{12500\,\mu g}{L} = 25155\,\mu g/L\ Cs \quad and \quad 1.9943 \frac{5450\,\mu g}{L} = 10869\,\mu g/L\ Sr. \tag{B.19}$$

B.7   Test solution pH

The pH values of commercial pH buffer standards and test solutions were measured using micro-combination electrode and multivoltmeter. The meter settings were not adjusted based on the buffer readings, rather the as-measured values for the buffers and solutions were recorded and later used to calculate the correlation between the certified buffer values and the measured values, and that correlation was used to calculate the true pH values of the test solutions. As and example, buffers for pH 3, 7, and 10 were measured on June 30, 2011:

Buffer	Before Measuring Test Solutions	After Measuring Test Solutions
3	2.137	2.184
7	6.117	6.150
10	8.776	8.790

Linear regression of the certified values against the measured values (using all measurements weighted equally) gave the relationship

actual pH = measured pH x 1.053 + 0.6726. (B.20)

That equation was used to calculate the pH values of the test solutions that were measured.
The pH of the test solution from SJ1-Cs-B-2 was measured to be 7.037. Inserting this value gives a pH of

7.037 x 1.053 + 0.6726 = 8.083 (B.21)

B.8   Calculation of K_d (mass in leachant, mass in test solution, mass on soil)

The value of $K_d(Cs)$ is calculated from the mass Cs in the leachant, the mass of Cs in the test solution, and the concentration of Cs in the test solution. The mass of Cs in the leachant is calculated using the average Cs concentration in the leachant blank tests and the mass of leachant used in the test of interest.
In the data sheet in ASTM C1733 Table 1, these are referred to (using Cs as the contaminant) as:
Total volume of liquid: V
Starting concentration of Cs in leachant (contact solution): Cs
Final concentration of Cs in test solution (contact solution): Cf
Dry mass of solid: M
The quantity of Cs sorbed on the solid (Q_s) is calculated as:

$$Q_s = (V \times C_s) - (V \times C_f) \tag{B.22}$$

62

The concentration of Cs sorbed on the solid (S) is calculated as:

$$S = \frac{(V \times C_s) - (V \times C_f)}{M} = {Q_s}/{M}$$ (B.23)

The distribution coefficient (K_d) is calculated as:

$$K_d = \frac{\dfrac{(V \times C_s) - (V \times C_f)}{M}}{C_f} = {S}/{C_f}$$ (B.24)

The Cs concentrations measured in the three blank tests with the Cs A solution are 25.07 mg/L, 25.25 mg/L, and 24.096 mg/L, having an average of 25.10 mg/L.

The effect of scale test SJ2-Cs-A-Y-13 is used as an example. From Table 9, the mass of leachant used is 25.02 g. From the density of the Cs solution, this corresponds to a volume of

$$25.10 \text{ g} \frac{\text{mL}}{0.9946 \text{ g}} = 25.24 \text{ mL}$$ (B.25)

and provides a mass of Cs equal to

$$25.24 \text{ mL} \frac{25.1 \text{ mg}}{\text{L}} \frac{\text{L}}{1000 \text{ mL}} \frac{1000 \text{ µg}}{\text{mg}} = 633.52 \text{ µg Cs.}$$ (B.26)

From Table 10, the dilution-corrected concentration of Cs in the SJ2-Cs-A-Y-13 test solution is 1072 µg/L. Assuming that the volume of test solution is the same as the volume of leachant used in the test, the mass of Cs remaining in the test solution can be calculated as

$$25.24 \text{ mL} \frac{1072 \text{ µg}}{\text{L}} \frac{\text{L}}{1000 \text{ mL}} = 27.06 \text{ µg Cs.}$$ (B.27)

By difference, the mass of Cs taken up by the soil is

633.52 µg Cs in leachant – 27.06 µg Cs = 606.46 µg Cs on soil. (B.28)

From Table 9, 1.00 g soil was used in test SJ2-Cs-A-Y-13, so the Cs concentration on the soil is 606.46 µg Cs/g soil. The Cs concentration in the test solution is 1.072 µg/mL, so K_d is

$$K_d = \frac{606.46 \text{ µg/g}}{1.072 \text{ µg/mL}} = 565.7 \text{ mL/g.}$$ (B.29)

B.9   Linearization of the Langmuir equation

The Langmuir equation given in Equation 15 is rearranged to the linearized Equation 16 by the following steps

$$C_{soil} = b \frac{K_{\mathrm{L}} C_{solution}}{\left(1 + K_L \, C_{solution}\right)} \qquad (15)$$

$$C_{soil} + K_L \, C_{solution} \, C_{soil} = b \, K_{\mathrm{L}} C_{solution} \qquad (\mathrm{B.30})$$

$$\frac{C_{soil}}{b \, K_{\mathrm{L}}} + \frac{K_L \, C_{solution} C_{soil}}{b \, K_{\mathrm{L}}} = C_{solution} \qquad (\mathrm{B.31})$$

$$\frac{1}{b \, K_{\mathrm{L}}} + \frac{C_{solution}}{b} = \frac{C_{solution}}{C_{soil}} \, . \qquad (16)$$

Appendix C: Propagation of Errors

The uncertainties in the rates calculated for individual tests within each test series were estimated from the measured test values using the propagation of errors method. For a property P that is a function of measured values x_1, x_2, x_3, etc., the probable error associated with P can be expressed in terms of the probable error in the means of the measured values as:

$$Q_p{}^2 = \left(\frac{\partial P}{\partial x_1}\right)^2 \bullet Q_1{}^2 + \left(\frac{\partial P}{\partial x_2}\right)^2 \bullet Q_2{}^2 + \left(\frac{\partial P}{\partial x_3}\right)^2 \bullet Q_3{}^2 + \ldots \tag{C.1}$$

The estimated uncertainties for measured and calculated values are listed below. The partition function K_d is calculated using Eq. C.2:

$$K_d = \frac{C_{sorbed}(g_{species}/g_{soil})}{C_{solution}(g_{species}/mL_{solution})} . \tag{C.2}$$

The mass of a contaminant sorbed per gram solid is calculated as

$$C_{sorbed}(g_{species}/g_{sorbent}) =$$
$$\frac{C_{leachant}(g_{species}/mL_{leachant}) \bullet V_{leachant}(mL) - C_{solution}(g_{species}/mL_{solution}) \bullet V_{solution}(mL)}{M_{sorbent}(g_{soil})} . \tag{C.3}$$

Substituting this into Equation C.2 gives

$$K_d = \frac{C_{leachant}(g_{species}/mL_{leachant}) \bullet V_{leachant}(mL) - C_{solution}(g_{species}/mL_{solution}) \bullet V_{solution}(mL)}{C_{solution}(g_{species}/mL_{solution}) \bullet M_{sorbent}(g_{soil})}, \tag{C.4}$$

which can be rearranged as

$$K_d = \frac{C_{leachant}(g_{species}/mL_{leachant}) \bullet V_{leachant}(mL)}{C_{solution}(g_{species}/mL_{solution}) \bullet M_{sorbent}(g_{soil})} - \frac{C_{solution}(g_{species}/mL_{solution}) \bullet V_{solution}(mL)}{C_{solution}(g_{species}/mL_{solution}) \bullet M_{sorbent}(g_{soil})}, \tag{C.5}$$

and simplified to

$$K_d = \frac{C_{leachant} \bullet V_{leachant}}{C_{solution} \bullet M_{soil}} - \frac{V_{solution}}{M_{soil}} . \tag{C.6}$$

The probable error in the value of K_d that is calculated with Eq. C.6 is given by:

$$Q_{K_d}{}^2 = \left(\frac{\partial K_d}{\partial C_{leachant}}\right)^2 \bullet Q_{C_{leachant}}{}^2 + \left(\frac{\partial K_d}{\partial C_{solution}}\right)^2 \bullet Q_{C_{solution}}{}^2 + \left(\frac{\partial K_d}{\partial M_{soil}}\right)^2 \bullet Q_{M_{soil}}{}^2$$
$$+ \left(\frac{\partial K_d}{\partial V_{leachant}}\right)^2 \bullet Q_{V_{leachant}}{}^2 + \left(\frac{\partial K_d}{\partial V_{solution}}\right)^2 \bullet Q_{V_{solution}}{}^2 . \tag{C.7}$$

The standard deviation is the square root of $Q_{K_d}{}^2$. The partial differential terms in Eq. C.7 are:

$$\frac{\partial K_d}{\partial C_{leachant}} = \frac{V_{leachant}}{C_{solution} \bullet M_{soil}},$$

(C.8)

$$\frac{\partial K_d}{\partial C_{solution}} = -\frac{C_{leachant} \bullet V_{leachant}}{\left(C_{solution}\right)^2 \bullet M_{soil}},$$

(C.9)

$$\frac{\partial K_d}{\partial M_{soil}} = -\left\{\frac{C_{leachant} \bullet V_{leachant}}{C_{solution}} - V_{solution}\right\}\frac{1}{[M_{soil}]^2},$$

(C.10)

$$\frac{\partial K_d}{\partial V_{leachant}} = \frac{C_{leachant}}{C_{solution} \bullet M_{soil}},$$

(C.11)

$$\frac{\partial K_d}{\partial V_{solution}} = \frac{-1}{M_{soil}},$$

(C.12)

The individual uncertainty terms are discussed below.

$QC_{leachant}$
The uncertainty in the concentration of the element of interest in the leachant solution is due to analytical uncertainty in the measured concentration, which is reported by the analyst to be within 10%.

$QC_{solution}$
The uncertainty in the concentration of the element of interest in the test solution is due to analytical uncertainty in the measured concentration, which is reported by the analyst to be within 10%.

QM_{soil}
The uncertainty in the mass of soil used in the test is computed by applying Eq. C.1 to the difference in two measurements:

$$M = mass_1 - mass_2$$

(C.13)

$$Q_M{}^2 = (\frac{\partial M}{\partial M_{mass\ 1}})^2 \bullet Q_1{}^2 + (\frac{\partial M}{\partial M_{mass\ 2}})^2 \bullet Q_2{}^2.$$

(C.14)

If the uncertainty in each measured mass is 0.01 g, then $\frac{\partial M}{\partial mass_1} = 1$, $\frac{\partial M}{\partial mass_2} = -1$, and

$Q_1 = Q_2 = 0.01$. Inserting these values into Eq. C.14 gives:

$$Q_M{}^2 = (1)^2 \bullet (0.01)^2 + (-1)^2 \bullet (0.01)^2 = 0.0002 \text{ g.}$$

(C.15)

The uncertainty in the difference of any two masses to the nearest 0.01 g is $Q_M = (0.0002)^{0.5} = 0.014$ g.

$QV_{leachant}$

The uncertainty in the volume of the leachant solution is due to analytical uncertainty in the measured mass, which is taken to be 0.014 g, and the uncertainty in the density. The densities of the 1 x 10^{-4} molal solutions were measured by weighing solution in a volumetric flask. The volume of the flask was measured by filling with demineralized water. The uncertainty in the leachant volume is due to two weight measurements to determine the density and one weight measurement to determine the mass, with an uncertainty of 0.014 g for each weight measurement. The uncertainty in the leachant volume is is $Q_V =$ [3 x (0.014)^2]$^{1/2}$ = 0.024 mL.

QV$_{solution}$

The mass of the test solution differs from the mass of the leachant solution due to evaporative loss during the test. The average mass loss in tests conducted with 12.5 g leachant was 0.017 g, which is 0.13%. Tests conducted with 25 g leachant lost an average of 0.06 g, which is 0.24%. An uncertainty of 0.03 mL is assigned to the solution volumes of tests conducted with 12.5 g leachant and an uncertainty of 0.06 mL is assigned to the solution volumes of all tests with 25 g leachant.

Table C.1. Experimental values used to calculate K_d for replicate tests

	$V_{leachant}$, mL	$V_{solution}$, mL	$C_{leachant}$, µg/L	$C_{solution}$, µg/L	M_{soil}, g
SJ2-A-Y-1	50.31	50.31	25095	1200	2.00
SJ2-A-Y-2	50.27	50.27	25095	1140	2.00
SJ2-A-Y-3	50.27	50.27	25095	1170	2.00
SJ2-A-Y-4	50.30	50.30	25095	1120	2.00
SJ2-A-Y-5	50.27	50.27	25095	1120	2.00
SJ2-A-Y-6	24.91	24.91	8128	1013	1.00
SJ2-A-Y-7	24.91	24.91	8128	1095	1.00
SJ2-A-Y-8	24.93	24.93	8128	1073	1.00
SJ2-A-Y-9	24.91	24.91	8128	1093	1.00
SJ2-A-Y-10	24.95	24.95	8128	1142	1.00

Table C.2. Differentials in Equation B.8 calculated using experimental values

	$\dfrac{\partial K_d}{\partial C_{leachant}}$	$\dfrac{\partial K_d}{\partial C_{solution}}$	$\dfrac{\partial K_d}{\partial M_{soil}}$	$\dfrac{\partial K_d}{\partial V_{leachant}}$	$\dfrac{\partial K_d}{\partial V_{solution}}$
SJ2-A-Y-1	0.0210	-0.4384	-250.4461	10.4563	-0.5
SJ2-A-Y-2	0.0220	-0.4853	-264.0782	11.0066	-0.5
SJ2-A-Y-3	0.0215	-0.4608	-256.9847	10.7244	-0.5
SJ2-A-Y-4	0.0225	-0.5031	-269.1797	11.2031	-0.5
SJ2-A-Y-5	0.0224	-0.5028	-269.0183	11.2031	-0.5
SJ2-A-Y-6	0.0246	-0.1973	-174.9725	8.0231	-1
SJ2-A-Y-7	0.0228	-0.1690	-160.0628	7.4247	-1
SJ2-A-Y-8	0.0232	-0.1759	-163.8942	7.5732	-1
SJ2-A-Y-9	0.0228	-0.1694	-160.2990	7.4341	-1
SJ2-A-Y-10	0.0218	-0.1554	-152.5777	7.1144	-1

Table C.3. Uncertainties in experimental measurements

	$QC_{leachant}$	$QC_{solution}$	QM_{soil}	$QV_{leachant}$	$QV_{solution}$
	µg/L	µg/L	g	mL	mL
SJ2-A-Y-1	2509.5	120.00	0.014	0.024	0.03
SJ2-A-Y-2	2509.5	114.00	0.014	0.024	0.03
SJ2-A-Y-3	2509.5	117.00	0.014	0.024	0.03
SJ2-A-Y-4	2509.5	112.00	0.014	0.024	0.03
SJ2-A-Y-5	2509.5	112.00	0.014	0.024	0.03
SJ2-A-Y-6	812.8	101.31	0.014	0.024	0.03
SJ2-A-Y-7	812.8	109.47	0.014	0.024	0.03
SJ2-A-Y-8	812.8	107.33	0.014	0.024	0.03
SJ2-A-Y-9	812.8	109.33	0.014	0.024	0.03
SJ2-A-Y-10	812.8	114.25	0.014	0.024	0.03

Table C.4. Calculated terms in Equation C.7

	C leachant term	C solution term	M soil term	V leachant term	V solution term	Sum of terms
SJ2-A-Y-1	2767.25	2767.25	12.294	0.06298	0.000225	5546.9
SJ2-A-Y-2	3061.31	3061.31	13.669	0.06978	0.000225	6136.4
SJ2-A-Y-3	2906.33	2906.33	12.944	0.06625	0.000225	5825.7
SJ2-A-Y-4	3175.42	3175.42	14.202	0.07229	0.000225	6365.1
SJ2-A-Y-5	3171.62	3171.62	14.185	0.07229	0.000225	6357.5
SJ2-A-Y-6	399.55	399.55	6.001	0.03708	0.000900	805.13
SJ2-A-Y-7	342.16	342.16	5.022	0.03175	0.000900	689.38
SJ2-A-Y-8	356.56	356.56	5.265	0.03304	0.000900	718.42
SJ2-A-Y-9	343.04	343.04	5.036	0.03183	0.000900	691.15
SJ2-A-Y-10	315.17	315.17	4.563	0.02915	0.000900	634.94

Table C.5. Propagated uncertainties as standard deviations and ranges of K_d values for replicate tests

	Standard Deviation	K_d (Cs), mL/g		Standard Deviation	K_d (Sr), mL/g
SJ2-A-Y-1	74.48	500.9	SJ2-A-Y-6	28.37	175.0
SJ2-A-Y-2	78.33	528.1	SJ2-A-Y-7	26.26	160.1
SJ2-A-Y-3	76.33	514.0	SJ2-A-Y-8	26.80	163.9
SJ2-A-Y-4	79.78	538.4	SJ2-A-Y-9	26.29	160.3
SJ2-A-Y-5	79.73	538.0	SJ2-A-Y-10	25.20	152.6
	Mean	*523.88*		*Mean*	*162.38*
	Standard Deviation	*16.23*		*Standard Deviation*	*8.17*

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