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Iodine, Neptunium, Radium, and Strontium Sorption to Savannah River Site Sediments

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EXECUTIVE SUMMARY

The Savannah River National Laboratory (SRNL) was requested by Solid Waste Management to determine distribution coefficients (K_d) (contaminant concentration ratios of the solids over the liquids) for neptunium, strontium, iodine, and radium for use in Savannah River Site (SRS) Performance Assessments (PAs). New values for radium and iodine have been determined. Neptunium and strontium values did not change. Baseline Np K_d values were determined to be 9.05 ± 0.61 mL/g and 4.26 ± 0.24 mL/g for the clayey and sandy sediments, respectively. The addition of natural organic matter (NOM) to the clayey sediment resulted in an increase in the K_d value most likely due to the formation of ternary soil-NOM-Np complexes. None of the reductants nor the anaerobic atmosphere resulted in large increases in K_d values for either sediment, indicating that little to no reduction of Np(V) to Np(IV) occurred. Long term equilibration experiments (71 days) indicated that even prolonged equilibration under anoxic conditions do not facilitate reduction of Np(V) to Np(IV). Desorption K_d values were calculated under the baseline and anaerobic conditions and found to approach the sorption K_d values given a long enough equilibration period which indicated fully reversible sorption. This was further confirmed with a flowcell experiment that desorbed >99.9% of sorbed Np from the clayey sediment.

Radium and strontium sorption to the sediments was found to be highly dependent upon ionic strength due to competition for ion exchange sites. Radium K_d values for the clayey sediment were determined to be 185.1 ± 25.63 mL/g and 30.35 ± 0.66 mL/g for ionic strengths of 0.02M (the approximate ionic strength of SRS groundwater) and 0.1M as NaCl which is the approximate ionic strength of groundwater. Radium K_d values for the sandy sediment were determined to be 24.95 ± 2.97 mL/g and 9.05 ± 0.36 mL/g for ionic strengths of 0.02M and 0.1M as NaCl. These values were greater than the strontium sorption K_d values which were consistent with values presently used in SRS PAs.

Iodine can exist as iodate, IO_3^- , or iodide, I^- . The focus of the iodine sorption studies was to measure iodide and iodate sorption under oxidizing and reducing conditions. Only recently was it determined that both iodine species can exist under SRS groundwater conditions. Prior it was assumed that all iodine existed as the weaker sorbing iodide species. Sorption tests demonstrated that iodate K_d values were in the order of four times greater than iodide K_d values for all three sediments tested. However, iodate is reportedly easily reducible to iodide. We observed no noticeable change in the iodate clayey K_d values under either oxidizing or reducing conditions indicating that it remained as iodate. However, under reducing conditions, the wetland soil reduced the iodate to iodide, which resulted in an eight fold decrease in sorption. The final iodate equilibrium K_d value under reducing conditions was equal to that of iodide suggesting complete reduction of the iodate. Below are recommended K_d values based on these tests and a comparison with previously used K_d values.

Rad	Recommended Values Based on this Study		Existing Geochemical Data Package SRNL-STI-2009-00473		Comment
	Sand K_d (mL/g)	Clay K_d (mL/g)	Sand K_d (mL/g)	Clay K_d (mL/g)	
Sr	5	17	5	17	No change recommended
Ra	25	185	5	17	Ra K_d (ionic strength, ~0.02 M, which approximates that of SRS groundwater)
Np	3	9	3	9	Results from this study are included in SRNL-STI-2009-00473
I	0.3	0.9	0.3	0.9	<ul style="list-style-type: none"> - K_d: iodate >> iodide - SRS has both iodate and iodide; it was previously assumed that only iodide was present. - No change in K_ds is recommended at this time because research is on-going.

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LIST OF ABBREVIATIONS

CSTR	Continuous Stirred-tank Reactor
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
K _d	Distribution Coefficient
LSC	Liquid Scintillation Counting
NOM	Natural Organic Matter
NIST	National institute of Standards and Technology
ORWBG	Old Radioactive Waste Burial Ground
PA	Performance Assessment
ppq	parts per quadrillion
RSD	Relative Standard Deviation
SRNL	Savannah River National Laboratory
SRS	Savannah River Site

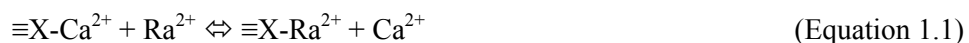
1.0 Introduction

1.1 Radium and Strontium Geochemistry

Radium is present in the environment as a decay product from uranium bearing ores as ^{226}Ra which has a 1602 year half life. Stable ^{88}Sr is found in most rocks while ^{90}Sr is present in the environment due to releases from legacy nuclear weapons wastes, nuclear reactors, or from atmospheric testing of nuclear weapons. ^{90}Sr is a high yield product from the fission of ^{235}U , ^{233}U , and ^{239}Pu . Radium and strontium are both divalent cations existing only in the +2 oxidation state. Sposito (1989) indicates that sorption affinity of the alkaline earth metals follow the trend $\text{Ra}^{2+} > \text{Ba}^{2+} > \text{Sr}^{2+} > \text{Ca}^{2+} > \text{Mg}^{2+}$, where increasing sorption occurs with increasing ionic radii. Because ionic potential (the ratio of the electric charge of the ion to the radius of the ion) decreases with increasing ionic radius, this implies that the larger ions will create a smaller electric field and be more prone to sorption.

It has been estimated that the inventory of $^{226}\text{Ra}/^{228}\text{Ra}$ and ^{90}Sr in the Old Radioactive Waste Burial Ground) ORWBG is 0.18 Ci and 54,000 Ci, respectively (Hiergesell et al., 2008). ^{226}Ra waste is primarily present as a daughter product of uranium disposal. Approximately 17 Ci of ^{238}U is buried in the ORWBG indicating that radium contamination will still be an issue far beyond the 10,000 year assessment period of the PA (Hiergesell et al., 2008).

Sorption of radium and strontium is also highly dependent upon ionic strength and the concentration of competing ions. This effect is shown in Figure 1-1. Divalent cations form outer-sphere complexes which are relatively weak and can easily be displaced by other cations in solution (Chen and Hayes, 1999). This can be shown by the following reaction:



which indicates that higher concentrations of competing cations can prevent radium and strontium from sorbing to the sediment.

Currently, K_d values of 17 mL/g and 5 mL/g for the clayey and sandy sediments, respectively, have been recommended for use in the SRS PA for strontium (Kaplan, 2009). These values were determined using actual SRS groundwater which had an ionic strength ranging from 0.01 to 0.1 M, but they may not be applicable to all groundwater applications. Because no data is available for radium sorption to these same sediments, the strontium K_d values are used. This assumption results in radium and strontium having the same mobility resulting in higher than expected potential risk for strontium. By generating two separate K_d values for these two elements, it may be possible to separate their risks and lower the peak dose.

Looney et al. (1987) recommended a K_d value for radium sorption on SRS soils of 100 mL/g with a range of 10 to 1,000,000 mL/g. These values were based on the sorption of other metals, namely strontium. Thibault et al. (1990) gave radium K_d values for a clay soil of 9,100 mL/g and for a sand soil of 500 mL/g. Nathwani and Phillips (1979) were also able to show that increasing the concentrations of Ca^{2+} resulted in decreasing K_d due to increased competition for surface sites. An objective of this work is to directly measure ^{226}Ra K_d values for SRS sediments and compare those values to ^{90}Sr . Therefore, the work proposed here may be valuable to ^{90}Sr geochemistry as well as $^{226}/^{228}\text{Ra}$ geochemistry.

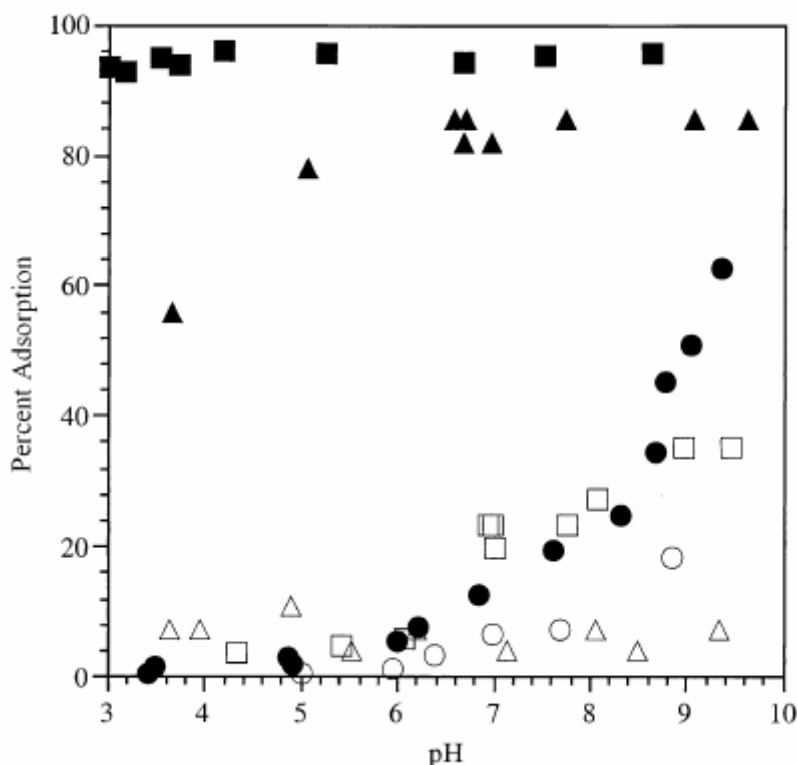


Figure 1-1 Sr (initial concentration = 10^{-6} M) sorption on various solids at two Na ion concentrations. Sr sorption on quartz is both pH and Na ion concentration dependent, but sorption on illite and montmorillonite is pH independent at lower NaCl concentrations. (■) montmorillonite, 0.01 M NaCl; (□) montmorillonite, 0.1 M NaCl; (▲) illite, 0.01 M NaCl; (Δ) illite, 0.1 M NaCl; (●) silica, 0.01 M NaCl; (○) silica, 0.1 M NaCl (Chen and Hayes, 1999).

1.2 Iodine Geochemistry

Iodine is commonly found as an anion in various oxidation states as seen in Figure 1-2. The most common being the reduced iodide (I^-) and the oxidized iodate (IO_3^-). Iodide is the dominate oxidation state under all but the most oxidizing conditions. According to this diagram, at a neutral pH, the redox potential would need to be at least +0.75V for IO_3^- to become the dominate species.

Iodide has been observed to have a lower K_d than IO_3^- and has been used as a groundwater tracer due to its relatively low affinity for solid phases (Kaplan et al., 2000). Iodate showed stronger sorption to several Chinese soils than the reduced iodide (Dai et al., 2009). When Hu et al. (2005) examined IO_3^- and I^- interactions with soils, they found IO_3^- was easily reduced to I^- , especially at low concentrations. Reduction was speculated to be promoted by Fe(II) found in the clays. Sheppard et al. (1995) noted I^- exposed to natural bog water was not readily oxidized to IO_3^- , but in fact remained as the reduced I^- . Kaplan et al. (2000) examined the sorption of I^- to certain sediments and illitic minerals. They noted K_d values less than 1 mL/g for minerals such as calcite, goethite, montmorillonite, and vermiculite. However, illite had a K_d of 15 mL/g, which increased to 27 mL/g when iron oxides, carbonate, and organic matter were removed.

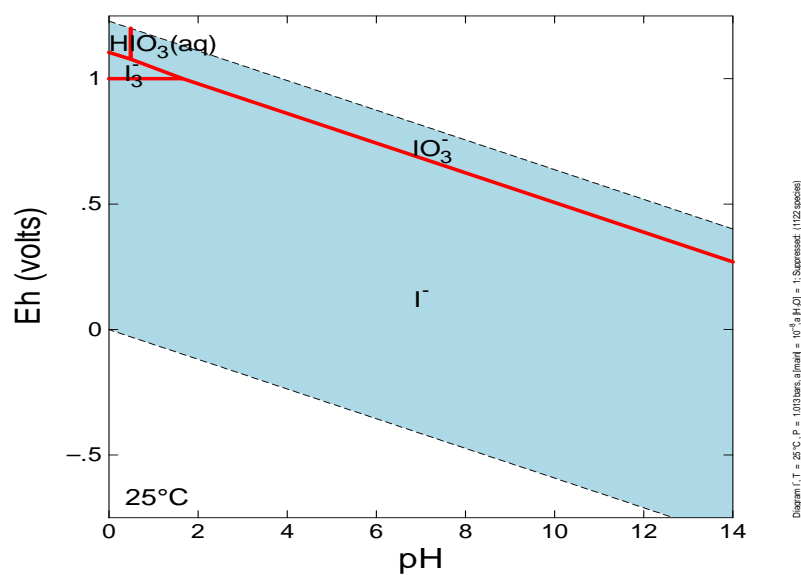


Figure 1-2: Iodine E_H-pH Diagram. Modeled with Geochemist Workbench, LLNL thermochemical database with precipitation of solids suppressed. Total {I} = 1 x 10⁻⁸ M.

1.3 Research Objectives

This research project is designed to validate data and assumptions regarding iodine, radium, and strontium used in SRS Performance Assessments to ensure sound decision making concerning radionuclide transport in the subsurface.

- Radium and Strontium
 - Calculate and compare K_d values for Ra and Sr sorption on SRS end member sediments at varying ionic strengths.
 - Test the current assumption in the SRS PA that Sr sorption behavior can be used to approximate Ra sorption.
- Iodine
 - Determine distribution coefficients (K_d) for iodide and iodate on end member sediments and a representative wetland sediment under oxidizing and reducing conditions.

2.0 Materials and Methods

2.1 Description of Sediments

Three end member sediments from the Savannah River Site were used in this work. The first is was a subsurface yellow sandy sediment, referred to as sandy. This sediment has very little organic material (Table 2.1). The second is a subsurface red clayey sediment, referred to as clayey, which has little organic material but a significantly higher clay fraction than the sandy sediment. The third soil, referred to as wetland, is a wetland soil from Four Mile Branch. This soil is primarily sand with a high organic matter content. Some additional analyses of these three natural materials are given in Table 2.1.

Table 2-1: Characteristics of SRS Sediments used in the current work.

PARAMETER	Subsurface Red Clayey	Subsurface Yellow Sandy	Four Mile Branch Wetland
% sand ($>53\ \mu\text{m}$)	57.9	97	85.5
% silt ($53 - 2\ \mu\text{m}$)	40.6	2.9	11.7
% clay ($<2\ \mu\text{m}$)	1.6	0.2	2.8
Textural classification	Silty clay	Sand	
pH	4.55	5.10	4.10
% OM	NA	0	7.9
CEC (cmol/kg)	1.09 ± 0.31	<0.05	
AEC (cmol/kg)	1.58 ± 0.61	0.06 ± 0.19	
BET surface area (m^2/g)	15.3	1.27	3.76
CDB extractable Fe (mg/g)	15.3	7.06	
Al (ppm)	63.6	16.6	
Na (ppm)	42.9	34.7	
Mg (ppm)	144	98.8	
Ca (ppm)	64.4	24.6	
K (ppm)	183	93.0	
Mineralogy	Kaolinite, goethite, hematite	Kaolinite, goethite, muscovite/14A	

2.2 Experimental Methods for Radium and Strontium Sorption Experiments

2.2.1 Sorption Experimental Protocol

Radium and strontium sorption was examined on the same SRS clayey and sandy sediments as the neptunium sorption experiments (Table 2-1). The experiments were performed with sediment concentrations of 25 g/L and pH 5.5. Two ionic strengths, 0.01 M and 0.1 M, were used to explore possible influences of ion exchange processes. Stable ^{88}Sr was used to spike the sediment suspensions and initial concentration ranged from 50 ppb to 1000 ppb. ^{226}Ra was also spiked into the soil suspensions with initial activity concentrations ranging from 25 to 250 cpm/mL (counts per minute per mL). A summary of the experimental conditions is shown in Table 2-2 where each condition was performed in duplicate for each of the two soils. After spiking, the suspensions were mixed end over end for a week to ensure equilibrium (Kaplan, 2009). The sample vials were then centrifuged to remove particles greater than 100 nm. A 1 mL aliquot of the supernatant was analyzed on the ICP-MS to determine the strontium concentration. An ICP-MS calibration curve and calibration data for ^{88}Sr are shown in Figure 2-1 and Table 2-3.

Table 2-2: Summary of radium-strontium sorption experiments. Each condition was performed in duplicate for each of the two soils.

Ionic Strength (M)	[²²⁶ Ra] (cpm mL ⁻¹)	[²²⁶ Ra] (mol L ⁻¹)	[⁸⁸ Sr] (ppb)	[⁸⁸ Sr] (mol L ⁻¹)
0.01	250	5.0E-10	1000	1.1E-05
0.01	185	3.7E-10	500	5.7E-06
0.01	125	2.5E-10	200	2.3E-06
0.01	60	1.2E-10	100	1.1E-06
0.01	25	5.0E-11	50	5.7E-07
0.1	250	5.0E-10	1000	1.1E-05
0.1	185	3.7E-10	500	5.7E-06
0.1	125	2.5E-10	200	2.3E-06
0.1	60	1.2E-10	100	1.1E-06
0.1	25	5.0E-11	50	5.7E-07
0.01	250	5.0E-10	0	0
0.01	185	3.7E-10	0	0
0.01	125	2.5E-10	0	0
0.01	60	1.2E-10	0	0
0.01	25	5.0E-11	0	0

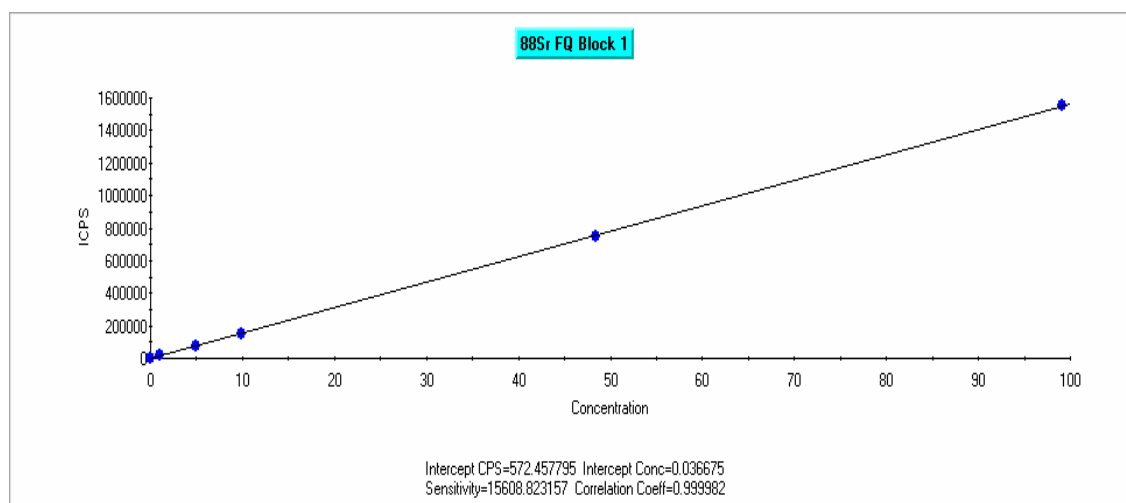


Figure 2-1: Screen capture of a typical strontium calibration curve using Thermo PlasmaLab software to control the data collection and analysis. $R^2=0.999982$, Intercept Conc. (Detection Limit) = 0.037 ppb.

Table 2-3: Example ICP-MS Calibration Curve Data

Sample	Sr standard actual concentration (ppb)	Measured Sr Concentration (ppb)	Mean Sr Ion Counts Per Second (ICPS)	Error	% Error
Wash	0	0.148	2890	0.148	0
0.05ppb Sr	0.049	0.139	2748	0.09	182.12
1 ppb Sr	0.995	1.076	17370	0.081	8.15
5 ppb Sr	4.927	4.913	77251	-0.015	-0.3
10 ppb Sr	9.866	9.788	153355	-0.077	-0.79
50 ppb Sr	48.474	48.015	750029	-0.459	-0.95
100 ppb Sr	99.159	99.391	1551948	0.232	0.23

To quantify the activity of aqueous ^{226}Ra remaining in solution, two different detection methods were employed. The first method involved pipetting approximately 4 mL of the equilibrated supernatant into a liquid scintillation vial along with 15 mL of High Safe 3 cocktail. This counting method assumes that no diffusion of ^{222}Rn out of the cocktail will occur allowing detection of ^{226}Ra and 5 of its daughters (^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po). Therefore, the activity of ^{226}Ra will be 1/6 that of the total activity measured after 30 days as the sample is permitted to reach secular equilibrium. The second detection method was performed by pipetting another 4 mL aliquot of the equilibrated solution into another liquid scintillation vial along with 10 mL of mineral oil scintillating cocktail. The mineral oil scintillating cocktail method is an ASTM standard method for radon measurements (AWWA, 1998) and is useful because ^{222}Rn is the daughter product of ^{226}Ra . After the the 30 days required to reach secular equilibrium passed, each vial was shaken to mix the immiscible fluids. Radon selectively partitions into the mineral oil phase which scintillates when radon and its daughter products decay and can be quantified. The samples were analyzed on the Quantalus Ultra Low Level Liquid Scintillation Counter (LSC) along with a set of standards prepared from a NIST traceable ^{226}Ra source to determine the ^{226}Ra concentration. All data shown in the Results section was generated using the modified AWWA standard method. The calibration curve generated using the ^{226}Ra standards is shown in Figure 2-2.

Initial experiments indicated that native strontium existed on the SRS soils and can desorb into the aqueous phase when dried sediment is suspended in 0.01 M NaCl. An experiment was performed using native strontium to determine long term K_d values for each sediment. Suspensions were made with 25 g L^{-1} of sediment in 10 mL of water. The ionic strength was varied from 0 to 1.0 M (as NaCl) in increments (0.001, 0.005, 0.010, 0.050, 0.1, 0.5, and 1.0 M). These suspensions mixed for 95 days. This was assumed to be sufficient time to allow equilibrium to be reached. The vials were centrifuged to remove particles greater than 100 nm and the resulting supernatant was analyzed on the ICP-MS to determine strontium concentrations. Sediment samples then underwent microwave soil digestion using the same procedure described above. Strontium was separated from the digested sample using a Bio-Rad poly-prep column packed with Eichrom Sr Resin. The column was first washed with distilled-deionized (DDI) H_2O then glass wool was added to the top of the resin to keep it in place. The column was washed with 5 column volumes of 8 M BDH Aristar Ultra HNO_3 . The sample was spiked with ^{90}Sr to a concentration of 2000 cpm mL^{-1} for use in yield calculations and acidified using BDH Aristar Ultra HNO_3 before being loaded onto the column. The column was washed with 5 column volumes of 8M HNO_3 . The $^{88/90}\text{Sr}$ was eluted from the column with 15 column volumes of DDI H_2O into a preweighed vial. A 5 mL aliquot of the resulting sample was analyzed on the Quantalus Ultra Low Level LSC for ^{90}Sr analysis while the resulting solution was analyzed on the ICP-MS to determine the ^{88}Sr concentration in the sediment. Using

the aqueous and sediment strontium concentrations, a K_d was obtained for each ionic strength using the equations described in Section 3.2.2.

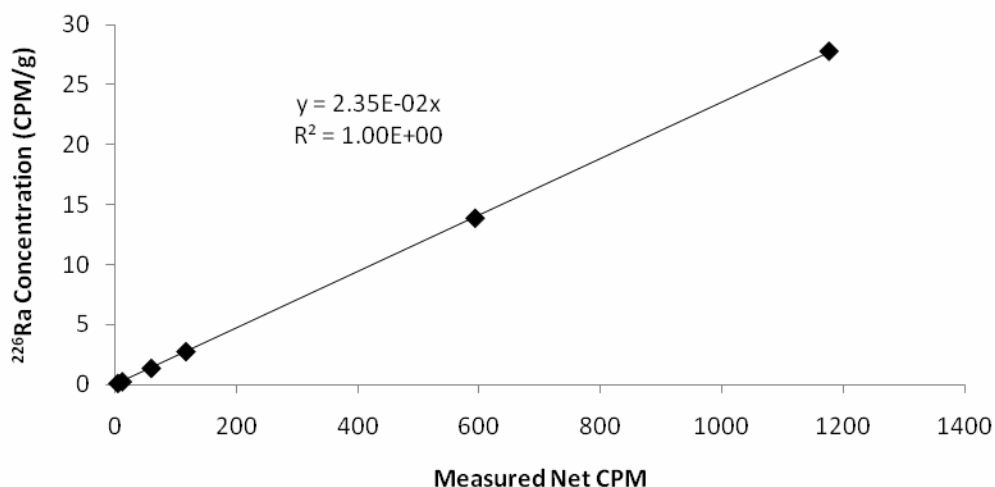


Figure 2-2: ²²⁶Ra calibration curve for radon in mineral oil cocktail standards. Count time of 60 min.

2.2.2 Data Analysis

The sediment concentration of Ra or Sr was calculated using the following equation (written for Ra):

$$[Ra]_{sed} = \frac{([Ra]_{aqu,o} - [Ra]_{aqu})V_L}{m_{sed}} \quad (\text{Equation 2.1})$$

where:

- $[Ra]_{aqu,o}$: Initial aqueous Ra concentration, ppb
- $[Ra]_{aqu}$: Equilibrated (ICP-MS measured) aqueous Ra concentration, ppb
- $[Ra]_{sed}$: Equilibrated sediment Ra concentration, ppb
- V_L : Sample liquid volume, mL
- m_{sed} : Sample sediment mass, g

The sediment water partitioning constant, K_d , was calculated via the following equation:

$$K_d = [Ra]_{sed}/[Ra]_{aqu} \quad (\text{Equation 2.2})$$

The percent of Ra sorbed was calculated via the following equation:

$$f_s = 1 - \frac{[Ra]_{aqu}}{[Ra]_{aqu,o}} \quad (\text{Equation 2.3})$$

2.3 Experimental Methods for Iodine and Iodate Sorption Experiments

2.3.1 *Iodine Analysis via ICP-MS*

Analysis of iodine using ICP-MS required the use of a reducing, basic solution that was capable of reducing iodate to iodine, holding the iodine in solution, and preventing off-gassing of $I_2(g)$. This minimized the loss of I during sample analysis. A 1 L trap solution was prepared by weighing out 0.0500 g $NaHSO_3$ (Fisher Scientific, ACS Grade) on a calibrated Sartorius LA 230S scale and adding it to a 1L volumetric flask. Then 40 mL of 25% w/w tetramethylammonium hydroxide (Alfa Aesar, electronic grade) and 10 mL CFA-C solution (Spectrasol, Inc.) were added to the volumetric flask via a calibrated 1000-5000 μ L Eppendorf Research pipette. The solution was then diluted to volume with DDI water.

For ^{127}I analysis, the ICP-MS must be reconfigured from the standard glass nebulizer setup to accommodate the basic, reducing trap solution. The reconfigured instrument uses an Elemental Science Microflow PFA-100 teflon nebulizer with a flow rate of 100 μ L/min, along with a sapphire torch, and a Teflon spray chamber. This configuration must be run with a low pump speed to prevent back pressure on the system. Two 30-minute stability tests were performed using a 50 ppb iodide solution. Each experiment consisted of 40 separate measurements. After each experiment was completed, the uncorrected mass counts were examined and found to stay steady over the sampling period. The % relative standard deviation (% RSD) over all samples for each experiment was 1.866% and 1.460%, respectively. This shows that there was no significant “memory” or loss of the iodine signal over time and that the reconfigured instrument has a stable iodine signal over time. However, as will be discussed below, some difficulty had been encountered in finding an adequate internal standard for iodine analysis.

A 100 μ g/mL iodide (I^-) stock solution from High Purity Standards (Charleston, SC) was used to make 1, 5, 10, 50, and 100 ppb standards by dilution using a “trap” solution (discussed in Section 3.2 below). These standards were used to calibrate the Thermo Scientific X Series 2 ICP-MS for quantification of ^{127}I . A screen shot of a representative calibration curve is shown in Figure 2-3. The data used to generate this curve are shown in Table 2-4. Although the background counts are higher for iodine, this data illustrates the ICP-MS is still accurate over many orders of magnitude. The use of a reducing, basic trap solution for iodine analysis limits the number of available internal standards that can be used to monitor ICP-MS instrument performance during iodine analysis. Initially there were not any reliable internal standards, so none were used for iodine analysis. This resulted in up to 20% error for QA/QC samples. With such large errors, it was necessary to find suitable internal standards. In house experiments have shown ^{95}Mo , ^{115}In , and ^{187}Re are acceptable internal standards, which were used with iodine analysis in later experiments. Spiked QA/QC samples were frequently analyzed throughout the analysis as a check on instrument performance. The 100 μ g/mL (ppm) stock iodide solution from High Purity Standards was used as the working solution for iodide experiments.

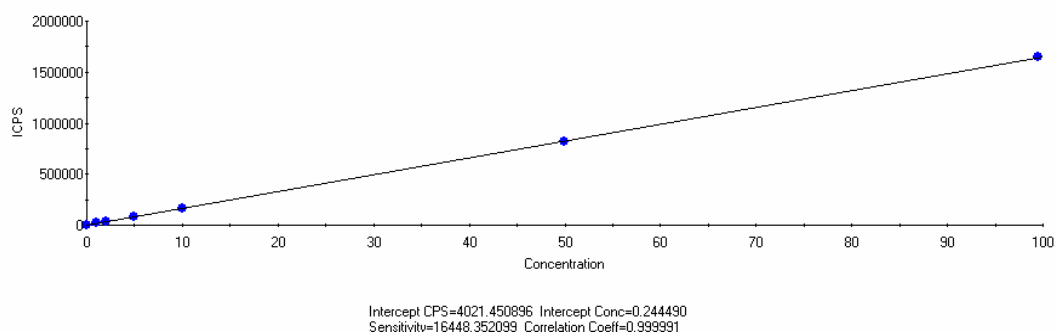


Figure 2-3: Screen Capture of a Typical ^{127}I Calibration Curve using Thermo PlasmaLab Software to Control the Data Collection and Analysis. $R^2=0.999991$, Intercept Conc. (Detection Limit) = 0.24 ppb. y-axis represents ion counts per second (ICPS) measured by the ICP-MS and x-axis represents concentration of ^{127}I in parts per billion.

Table 2-4: Sample iodine calibration data.

Sample	Defined Conc (ppb)	Measured Conc (ppb)	Counts	Error
Blank	0.000	0.000	1.44×10^3	0.000
1 ppb	0.971	1.24	6.61×10^3	0.269
5 ppb	4.90	5.00	2.23×10^4	0.102
10 ppb	9.84	9.88	4.26×10^4	0.083
50 ppb	49.0	50.1	2.10×10^5	1.14
500 ppb	494	494	2.06×10^6	-0.116

2.3.2 Determining Water Content of Wetland Soil

The Four Mile Branch wetland soil is unlike the sandy and clayey soils in that it is saturated with water. Because dehydrating the soil could lead to changes in soil chemistry, it was necessary to determine the water content. This was done by weighing three 15mL Falcon BlueMax 15mL polypropylene vials on a calibrated Sartorius LA 230S scale, and recording the masses. The scale was then zeroed, and $6.0 \pm 0.01\text{g}$ of wetland soil was added. These samples were then placed uncapped in an oven at 100°C overnight. After 24 hours, the vials were reweighed on the Sartorius LA 230S scale, and the dry weight was recorded to within 0.001g. A water/dry soil ratio was then calculated using the initial mass of the “wet” soil and the final dry weight. The resulting water content was $1.044 \pm 0.044\text{g H}_2\text{O/g dry soil}$ or $2.044 \pm 0.044\text{g wetland soil/g dry soil}$.

2.3.3 Preparation of Iodate Stock

Batch sorption experiments were also performed with iodate for comparison with the iodide experiments discussed above. An iodate stock solution was prepared by weighing 0.0122g potassium iodate (Alfa Aesar) on a calibrated Sartorius LA 230S scale, and diluting with 100mL DDI in an amber bottle. The stock concentration was then checked using the ICP-MS and the iodide standards. The iodine concentration of the stock was determined to be 74,280 ppb. This stock concentration was re-checked every time samples were run on the ICP-MS.

2.3.4 Experimental Methods in Aerobic Conditions

2.3.4.1 Experimental Protocol for Iodide Sorption

For each of the three soils, three sets of triplicate samples (n=9) were prepared in Falcon BlueMax 15mL polypropylene vials as describe above, but with 0.30 +/- 0.01g of either sandy or clayey soil added, and the mass recorded to within 0.001g. In the case of the saturated wetland soil, 0.60 +/- 0.01g of soil was added to each tube and the mass recorded to within 0.001g.

The three sets allow for experiments to be run with varying concentrations of iodide. Target initial ^{127}I concentrations were 1000ppb, 500ppb, and 100ppb. A set of controls containing no solids at 1000ppb and 100ppb ^{127}I were also prepared. The solids were equilibrated with the 0.01M NaCl solution before spiking with iodide. This experimental matrix is shown in Table 2-5. This was accomplished by adding 12mL 0.01M NaCl to each tube and soil, and recording the mass. The samples were then placed on a Labquake end-over-end shaker at 8 rpm overnight. After 24 hours, the suspensions were spiked with the iodide stock. For the 1000ppb iodide suspensions, a calibrated pipette was used to add a 120 μL aliquot of the iodide stock solution to the first three tubes for each soil. The 500ppb suspensions were prepared by adding 60 μL of the working solution to the next three tubes for each soil. The final three tubes were used for the initial concentrations of 100ppb. They were prepared by adding 12 μL aliquots of the iodide stock to each tube. A set of solid-free controls (no-solids controls) with ^{127}I concentrations of 100ppb and 1000 ppb were also prepared using this technique.

Table 2-5: Experimental matrix of soil sorption experiments for iodide and iodate under aerobic and reducing conditions. All samples prepared in triplicate.

Experiment	Target Initial Concentration $^{127}\text{I}^-$ or $^{127}\text{IO}_3^-$
Solids-Present	1000 ppb
Solids-Present	500 ppb
Solids-Present	100 ppb
Solids-Free	1000 ppb
Solids-Free	100 ppb

After spiking the samples with the iodide stock solution, the pH values of each sample were recorded. The samples were then placed on an end-over-end shaker at approximately 8 rpm. After 24 hours, the samples were removed from the shaker, and the sediment suspensions settled for an hour. The pH was then recorded using an Orion Ross semi-micro glass electrode, which was calibrated against pH 4, 7, and 10 buffers (Thermo). Each sample was then hand shaken to ensure a homogenous mixture. A transfer pipette was then used to pipette approximately 3 mL of each suspension to a 5 mL syringe. The solution was then passed through a 200 nm nylon syringe filter. The first 0.25-0.50 mL of filtrate was discarded, and the remaining filtrate was collected in a clean polyethylene vial. Then, 1.0 mL of the filtrate was removed and diluted in 5 mL trap solution. Each of these steps involved the use of a calibrated pipette. The iodine concentration in the diluted sample was determined using ICP-MS. The samples were then placed back on the shaker to mix until sampling events at 4 and 8 days using the same procedure.

2.3.4.2 Experimental Protocol for Iodate Sorption

The same sample preparation and sampling procedure described above was used to test iodate sorption to these three soil types. The only differing factor was in the amounts of the iodate stock solution added to each sample versus the amount of iodide stock solutions used in the above experiments.

2.3.5 Experimental Procedure in Reducing Conditions

2.3.5.1 Preparation of 0.01M NaCl

When preparing the samples in the anaerobic glove box, the 0.01M NaCl needed to be prepared in a manner that ensured that it was oxygen free. This was accomplished by bringing 2.5L DDI water to a rolling boil for 30 minutes. This was then cooled using an argon gas purge. While cooling, 1.168g NaCl was weighed on a calibrated Sartorius LA 230S scale, and added to a 2L volumetric flask. The cooled DDI water and volumetric flask containing the NaCl were then placed in the glove box. The DDI water was added to the flask, and the remaining water was saved to use as an electrode wash.

2.3.5.2 Preparation of Iodide Samples

The soil samples used in the glove box were prepared in much the same manner as those under aerobic conditions. The soil was added to the labeled vials under aerobic conditions, and the masses recorded. The masses were the same used for aerobic conditions. The samples were then transferred to the glove box, where they were left uncapped overnight. The 0.01M NaCl described above was then added to each sample in three 4.0mL aliquots using a calibrated pipette. The samples equilibrated overnight. After 24 hours, the predetermined mass of iodide stock was pipetted into the vials to achieve the desired initial concentrations found in the matrix in Table 2-5. This was done using calibrated pipettes. These samples mixed for approximately 1 hour and then the pH was recorded.

2.3.5.3 Preparation of Iodate Samples

The soil samples used for iodate sorption under reducing conditions were prepared using the above method with the only difference being the masses of iodate stock used. The iodide and iodate stocks had different iodine concentrations, so it was important to use the correct masses to ensure initial concentrations found in the matrix in Table 2-5.

2.3.5.4 Sampling of Iodide and Iodate Samples

Both the iodide and iodate samples were collected in the same manner as the previous samples. The sampling events occurred at 1, 4, and 8 day intervals. These samples were then analyzed using the Teflon setup on the ICP-MS.

2.3.6 Data Analysis

The K_d calculation for the sediment experiments was slightly modified from a traditional K_d equation. These sediments had native ^{127}I , which could desorb during the experiments and influence the measurement. This was accounted for by measuring the aqueous iodine for three sediment suspensions without any spiked iodine. These samples were then averaged, and this average was then subtracted from the ICP-MS measurements for each sample. These average values of aqueous iodine in the ICP-MS samples were 16.5, 68.7, and 7.60 ppb for the sandy, clayey, and wetland sediments, respectively. However, there was some variation with time so the unamended iodine samples were analyzed on the same dates as the samples amended with iodine.

The concentration on the solid was then calculated using:

$$[I]_{\text{solid}} = \frac{[I]_{\text{initial}} - [I](t)_{\text{measured}} - [I](t)_{\text{native}}}{m_{\text{solid}}} V_{\text{solution}} \quad (\text{Equation 2.4})$$

$[I]_{\text{solid}}$ = calculated solid phase concentration of the iodine/iodate associated with the sediment (ppb)

$[I]_{\text{initial}}$ = initial aqueous concentration of iodine/iodate following amendment (ppb)

$[I](t)_{\text{measured}}$ = measured iodine/iodate concentration from ICP-MS at sampling interval t .
 $[I](t)_{\text{native}}$ = measured aqueous iodine from unamended sediment suspensions at sampling interval t .
 m_{solid} = mass of the saltstone used in the suspension (g)
 V_{solution} = volume of solution

The distribution coefficient (K_d) can be calculated using the equation:

$$K_d = \frac{[I]_{\text{solid}}}{[I](t)_{\text{measured}}} \quad (\text{Equation 2.5})$$

This K_d equation (2.5) is numerically equivalent to the traditional K_d equation proposed in ASTM D-4646 which has been used in previous experiments (Kaplan et al., 2000; Powell et al., 2002).

2.4 Materials and Methods for the Neptunium Experiments

2.4.1 *Materials: Stock Solution Preparation and Soils*

A compiled ^{237}Np stock solution from the Environmental Engineering and Earth Science, Clemson University inventory (purchased from Isotope Products, Valencia, CA) was evaporated to dryness then the residue was brought up in approximately 5 mL 8.0 M HNO_3 . Then 1.0 M hydroxylamine hydrochloride ($\text{NH}_2\text{OH}\cdot\text{HCl}$, EMD Chemicals, ACS grade) and water were added to achieve a 3 M HNO_3 /0.3M $\text{NH}_2\text{OH}\cdot\text{HCl}$ solution. This solution was purified by extraction chromatography using Eichrom TEVA resin packed in a Bio-Rad poly-prep column. The 3 M HNO_3 /0.3 M $\text{NH}_2\text{OH}\cdot\text{HCl}$ neptunium solution was loaded on a 2 mL column and washed with three column volumes of 3 M HNO_3 . The Np(IV) was eluted with 0.02 M HCl + 0.2M HF . The effluent was evaporated to dryness then redissolved in 1.0 M HNO_3 . The sample was brought up in 10 mL of 1.0 M HNO_3 then evaporated to incipient dryness and redissolved in 5.0 mL of 1.0 M HNO_3 . An aliquot of the stock solution was evaporated to dryness on a stainless steel planchet and counted on the EG&G Ortec Alpha Spectrometer (Octete PC Detectors). Alpha energies besides ^{237}Np were not observed. The approximate ^{237}Np concentration was determined using liquid scintillation counting and little ^{233}Pa was observed. The fuming in HNO_3 as performed at the end of the purification procedure will drive neptunium to the soluble pentavalent state. This is the stable oxidation state of neptunium under the experimental conditions. Therefore, experiments performed here can be assumed to be initially Np(V). The exact neptunium concentration in this solution was determined using ICP-MS calibrated with a NIST standard as discussed below.

Working Solution #1 was created by pipetting an aliquot of the neptunium stock solution into a 100 mL Nalgene Teflon bottle and diluting with 2% BDH Aristar Ultra HNO_3 to give a working solution concentration of approximately 800 ppb. Working Solution #2 was created by pipetting an aliquot of Working Solution #1 with 2% BDH Aristar Ultra HNO_3 in a 250 mL polypropylene bottle to create a target concentration of approximately 50 ppb. Analysis on the ICP-MS calibrated against a National Institute of Standards and Technology (NIST) standard as described below gave concentrations of Working Solution #1 and Working Solution #2 of 820 ppb and 49.6 ppb, respectively, as described below. Calibration of the ICP-MS using the NIST standard is described below.

The sediments used for these experiments were obtained from the Savannah River Site. The subsurface sandy sediment will be referred to as the sandy sediment and the subsurface clayey sediment will be referred to as the clayey sediment. The clayey sediment was baked in an oven at 85°C overnight to

remove excess moisture. The sandy sediment did not receive any treatment. Specific characteristics of each sediment are shown in Table 2-6. As the table indicates, both soils are very low in organic matter.

2.4.2 ICP-MS Calibration Curves – Detection Limits

A NIST, Standard Reference Material (NIST SRM 4341) was used to prepare a stock ^{237}Np solution by dilution in 2% Aristar Optima HNO_3 . All volume additions were monitored gravimetrically. This working solution was then used to make a set of 0.01, 0.05, 1, 2, 5, 10 ppb standards by dilution using 2% HNO_3 . Again all volume additions were monitored gravimetrically. These standards were used to calibrate the Thermo Scientific X Series 2 ICP-MS for quantification of ^{237}Np . A representative calibration curve for ^{237}Np is shown in Figure 2-1. The calibration data from Figure 2-1 is shown in Table 2-7. The instrument performance was monitored using ^{232}Th and ^{238}U as internal standards. The recovery of each sample during analysis was corrected based on the internal standard recovery. The internal standard recoveries remained within standard QA/QC protocols for the instrument (between 80% and 120%).

The calibration curves were used to calculate the measured concentrations of neptunium in the samples being analyzed. The typical calibration curve shown in Figure 2-4 gave a minimum detectable limit of 1.8 ppq (parts per quadrillion). This is consistent with an average minimum detectable quantity of 2 ppq under the configuration of the instrument used for these measurements. Table 2-7 shows the goodness of fit of the calibration curve.

2.4.3 Preliminary Kinetic Sorption Tests

Preliminary experiments were performed to determine the time needed to reach steady state sorption between the aqueous neptunium and the sorbed neptunium. This experiment was performed in 50 mL BD Falcon polypropylene centrifuge tubes. Replicate samples were prepared with sediment concentrations of 5 g/L sediment and 25 g/L sediment. A fifth tube was used as a control blank. The tubes were first filled with the appropriate mass of sediment then 4.5 mL of 0.1M NaCl was added to produce a constant ionic strength of 0.01 M in the final sample. This ionic strength was chosen to be similar to the ionic strength of the actual groundwater at the SRS. The use of this groundwater surrogate was used instead of actual groundwater to aid in experimental control. However, if actual groundwater were used, no changes in aqueous speciation of neptunium would have been expected. Next, 40 mL of distilled deionized water (DDI H_2O) was added along with 0.55 mL of Np Working Solution #1 to obtain an initial neptunium concentration of 10 ppb. The pH was adjusted to 5.5 using 0.1N and 0.01N NaOH. The pH was measured using a VWR Ag/AgCl glass electrode calibrated with pH 4, 7, and 10 buffers (Thermo). The solutions were mixed using an end-over-end rotating tumbler at approximately eight rpm.

After 1, 3, 8, 24, and 48 hours, a 5 mL aliquot of each suspension was removed. Prior to removing the aliquot, a polyethylene transfer pipette was used to re-suspend any settled sediment particles and remove a homogenous suspension. This sample was then placed in a 15 mL BD Falcon polypropylene centrifuge tube and centrifuged in a Beckman Coulter Allegra X-22R Centrifuge at 8000 rpm for 20 minutes. This was sufficient time to allow all particles >100 nm to settle (Jackson, 1958). A 1 mL sample of the supernatant was then placed into an ELKay polystyrene culture tube and diluted with 2% BDH Aristar Ultra HNO_3 for analysis on the ICP-MS. Then 2 mL of the remaining supernatant was placed into a Microsep 10,000 MWCO centrifugal filter. The samples were then centrifuged in a Beckman GS-6 centrifuge at 3000 rpm for 2-3 minutes in order to wet the filter membrane and equilibrate neptunium with the membrane; the filtrate from this step was discarded. This pre-filtration step equilibrates the solution with the filter and washes the sodium azide preservation coating away. This results in a significant reduction in the loss of neptunium to the filter in the subsequent filtration. The sample was then centrifuged for an additional 20 minutes or until the majority of the sample passed through the filter. The filtrate was then transferred into an ELKay polystyrene culture tube and diluted with 2% BDH

Aristar Ultra HNO₃ to determine the neptunium concentration using the ICP-MS. All volumes in the ICPMS sample were monitored gravimetrically.

2.4.4 Sample Preparation – Baseline Batch Sorption Experiments

Samples were prepared in 15 mL BD Falcon polypropylene centrifuge tubes. Each tube was first filled with the appropriate mass of sediment, filled with approximately 6 mL of DDI-H₂O and 1 mL of 0.1M NaCl and the pH was adjusted to approximately 5.5 with 0.1N and 0.01N NaOH and HCl. All additions were monitored gravimetrically. The sediment suspension was then mixed end-over-end at eight rpm for 24 hours to equilibrate with the solution. The samples were then spiked with Np Working Solution #1 (described above) to reach target initial concentrations ranging from 0.1 ppb to 50 ppb. Finally, water was added to reach a 10 mL sample volume and the pH was again adjusted to a pH of 5.5. The mass of each addition of liquid and sediment to the sample tubes was monitored gravimetrically on Sartorius LA230S analytical balance.

2.4.5 Sample Analysis

After the 48 hour equilibration period the pH of each suspension was measured using a VWR Ag/AgCl glass electrode. Then a homogenous suspension was obtained by using a VWR 7 mL polyethylene transfer pipette to suspend the sediment particles. Approximately 1.5 mL of the suspension was transferred into 2 mL polypropylene centrifuge tubes and approximately 2 mL of solution was transferred into Microsep 10k Centrifugal filters. The 2 mL centrifuge tubes were spun at 5000 rpm for 25 minutes in the VWR Galaxy 5D Centrifuge to settle particles greater than 100 nm. An Eppendorf research grade pipette was used to draw off the supernatant, typically 1 mL, and transfer it into an ELKay polystyrene culture tube. The mass of the transferred liquid was monitored gravimetrically. The sample was then diluted with 4 mL of 2% BDH Aristar Ultra HNO₃ for ICP-MS analysis. The suspension in the Microsep 10k centrifugal filter was centrifuged in a Beckman GS-6 centrifuge at 3000 rpm for 2-3 minutes in order to wet the filter membrane and equilibrate Np with the membrane then the filtrate was discarded. Then the remaining suspension was centrifuged for an additional 20 minutes and the effluent from the 10k centrifugal filters was transferred into an ELKay polystyrene culture tube and diluted with 2% BDH Aristar Ultra HNO₃ for ICP-MS analysis. The neptunium concentration in all samples was determined on the ICP-MS.

The sediment concentration of Np was calculated using the following equation:

$$[Np]_{sed} = \frac{([Np]_{aqu,o} - [Np]_{aqu})V_L}{m_{sed}} \quad (\text{Equation 2.6})$$

where:

$[Np]_{aqu,o}$: Initial aqueous Np concentration, ppb

$[Np]_{aqu}$: Equilibrated (ICP-MS measured) aqueous Np concentration, ppb

$[Np]_{sed}$: Equilibrated sediment Np concentration, ppb

V_L : Sample liquid volume, mL

m_{sed} : Sample sediment mass, g

The sediment water partitioning constant, K_d , was calculated via the following equation:

$$K_d = \frac{[Np]_{soil}}{[Np]_{aqu}} \quad (\text{Equation 2.7})$$

The percent of Np sorbed was calculated via the following equation:

$$f_s = 1 - \frac{[Np]_{aqu}}{[Np]_{aqu,o}} \quad (\text{Equation 2-8})$$

The K_d equation (Equation 2.7) is numerically equivalent to the traditional K_d equation proposed in ASTM D-4646 which has been used in previous sorption tests (Kaplan et al., 2008).

3.0 Results

3.1 Radium and Strontium Sorption to End Member Sediments

3.1.1 *Radium Sorption to End Member Sediments*

The initial radium and strontium sorption experiments were performed similarly to the neptunium experiments with 25 g L⁻¹ of soil, pH 5.50, ionic strength concentrations of 0.01 and 0.1 M (as NaCl), initial strontium concentrations ranging from 50 to 1000 ppb, and initial radium concentrations ranging from 250 to 2500 cpm mL⁻¹. Due to the requirement to adjust the pH of the samples using NaOH and HCl, the 0.01 M NaCl solutions were actually at 0.02 M NaCl. These experiments were performed using two SRS sediments. As discussed in Section 2.0, the samples were allowed to equilibrate for 2 days before sampling.

For radium analysis, sorption studies were performed with and without strontium present (see Table 2-2 for experimental matrix). The sorption of radium to the clayey sediment gave K_d values of 30.35 ± 0.66 mL g⁻¹ for [NaCl] = 0.1 M, 185.1 ± 25.63 mL g⁻¹ for [NaCl] = 0.02 M, and 326.2 ± 33.64 mL g⁻¹ for [NaCl] = 0.02 M and no strontium present (Figure 3-1). For the highest initial radium concentration, more pH adjustment was required since the stock solutions were acidic. Therefore, the resultant ionic strength was higher than the rest of the set and these points were neglected when calculating the K_d values. There was less sorption to the sandy sediment which gave K_d values of 9.05 ± 0.36 mL g⁻¹ for [NaCl] = 0.1 M, 24.95 ± 2.97 mL g⁻¹ for [NaCl] = 0.02 M, and 34.55 ± 4.13 mL g⁻¹ for [NaCl] = 0.02 M and no strontium present (Figure 3-2).

The radium K_d values for the samples with strontium added were lower than the radium only samples due to exchange site competition offered by the high mass loading of strontium compared to radium. The mass of strontium added was 6 to 7 orders of magnitude greater than the mass of radium added (Table 2-2). This discrepancy in masses was required to overcome the concentration of native strontium desorbing from the soils as well as to keep the activity of ²²⁶Ra low enough to safely work with it. Recommended Ra K_d values will be based on the 0.02 N NaCl value when Sr is present because 0.02 N is a realistic normality and the presence of a competing cation is always going to be present.

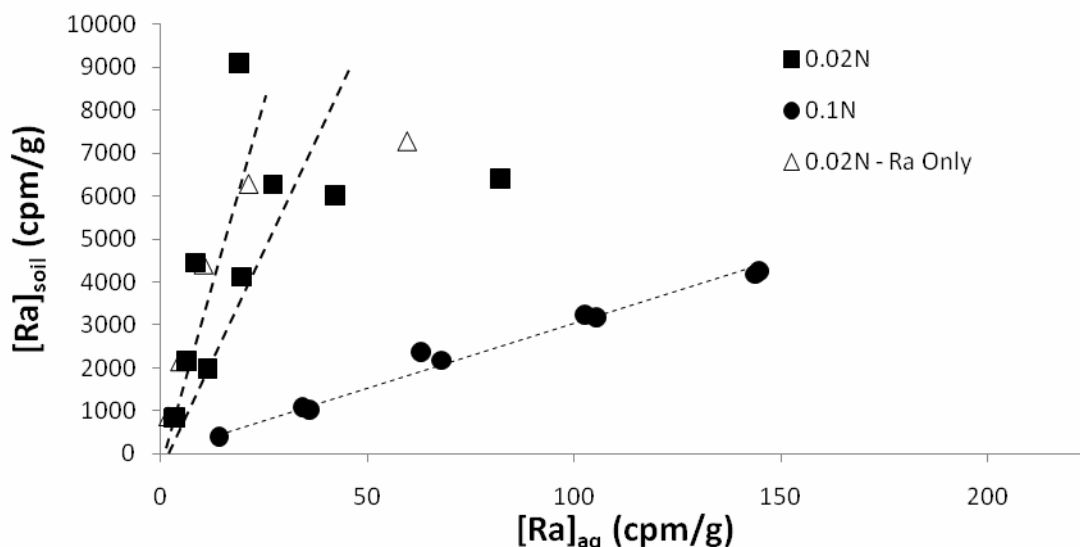


Figure 3-1: Radium sorption to clayey soil. K_d values of 30.35 ± 0.66 mL/g for $[NaCl] = 0.1$ M, 185.1 ± 25.63 mL/g for $[NaCl] = 0.02$ M, and 326.2 ± 33.64 mL/g for $[NaCl] = 0.02$ M and no strontium present were reported.

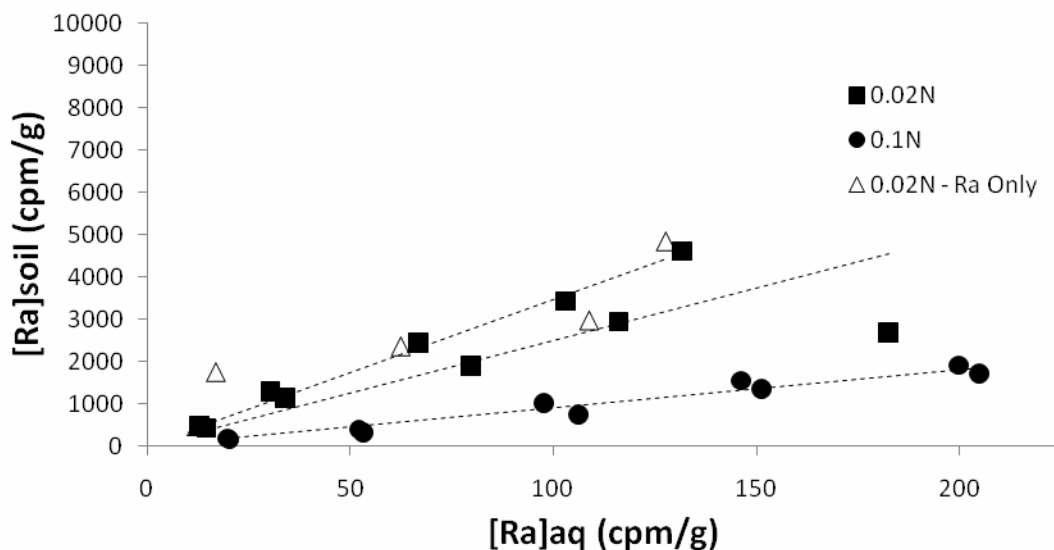


Figure 3-2: Radium sorption to sandy soil gave K_d values of 9.05 ± 0.36 mL/g for $[NaCl] = 0.1$ M, 24.95 ± 2.97 mL/g for $[NaCl] = 0.02$ M, and 34.55 ± 4.13 mL/g for $[NaCl] = 0.02$ M and no strontium present were reported.

These experimentally derived Ra K_d values disagreement with the estimated values currently recommended for use in the SRS PA of 17 mL/g and 5 mL/g for radium and strontium sorption to the clayey and sandy sediments, respectively (Kaplan, 2010). However, the data recommended for use in the SRS PA does not indicate the conditions with which they were determined. The results are consistent with the notion that sorption decreases as competing cation concentration increases. At higher ionic strengths,

there is a higher ratio of competing cations to radium ions which decreases the ability for the radium to sorb to the surface sites which can be shown by the generic ionic exchange reaction:



3.1.2 Strontium Sorption to End Member Sediments

Native strontium was detected in preliminary experiments with the SRS sediments at equilibrium aqueous concentrations up to 5 ppb, so an initial experiment was conducted to determine the total amount of strontium on the sediment. For each sediment, 0.5 g was digested, then the concentration of ^{88}Sr was determined using the ICP-MS. Eichrom strontium resin was used to extract the strontium from the digested solution as discussed in the materials and methods section. The concentration of native strontium on the soils was determined to be $3800 \pm 460 \mu\text{g/g}$ for the clayey soil and $2110 \pm 480 \mu\text{g/g}$ for the sandy soil.

Incorporating the native strontium into the K_d calculations (Equation 3.2), the K_d values for the clayey soil were $8.05 \pm 0.62 \text{ mL g}^{-1}$ for $[\text{NaCl}] = 0.1 \text{ M}$ and $32.06 \pm 3.62 \text{ mL g}^{-1}$ for $[\text{NaCl}] = 0.02 \text{ M}$. For the sandy soil, the K_d values were $6.02 \pm 0.14 \text{ mL g}^{-1}$ for $[\text{NaCl}] = 0.1 \text{ M}$ and $5.86 \pm 0.35 \text{ mL g}^{-1}$ for $[\text{NaCl}] = 0.02 \text{ M}$. The equation to determine the final strontium concentration is shown in Equation 3.2. These experimentally determined K_d values for the clayey sediment were lower than the values used for the SRS PA while the K_d values for the sandy soils were roughly the same (Kaplan, 2010). The sorption isotherms are shown in Figures 3-3 and 3-4.

$$[\text{Sr}]_{\text{soil}} = \frac{([\text{Sr}]_o - [\text{Sr}] - [\text{Sr}]_{\text{Blank}}) * V}{M_{\text{soil}}} \quad (\text{Equation 3.2})$$

where:

- $[\text{Sr}]_{\text{soil}}$ = Final concentration of strontium on soil, ppb
- $[\text{Sr}]_o$ = Initial concentration of strontium in solution, ppb
- $[\text{Sr}]$ = Final aqueous concentration of strontium, ppb
- $[\text{Sr}]_{\text{Blank}}$ = Concentration of strontium desorbed from sediment in blank samples, ppb
- V = Volume of liquid, mL
- M_{soil} = mass of soil

An experiment was also performed to determine the K_d values for the native strontium on the SRS soils at varying ionic strengths. Each soil was suspended in solutions with ionic strength ranging from 0 M to 1.0 M NaCl and was allowed to equilibrate for 95 days. The native strontium concentrations determined by soil digestion were used to calculate the K_d values. Figure 3-5 shows the aqueous strontium concentration after equilibration vs. ionic strength and Figure 3-6 shows K_d values vs ionic strength.

The K_d values reported here for the native strontium on the soils are approximately two orders of magnitude greater than the sorption experiments where strontium was added to the solution. This is likely due to the fact that the concentration of strontium associated with the soil that was determined by soil digestion includes strontium that is within the sediment matrix and possibly not available for dissolution/desorption. This differs from the batch sorption experiments where strontium was added to the solution and the concentration of strontium associated with the soil phase was calculated based on the difference between the initial and final aqueous strontium concentrations. The batch sorption experiments move towards calculating a geologic K_d value that takes into account weathering of soils into smaller particles and possibly allowing more strontium to desorb from the soil. These K_d values may also be more

representative of how a ^{90}Sr release would behave after equilibrating with the subsurface sediment for hundreds of years and may be more valuable than a K_d calculated after an equilibration period of 24 hours.

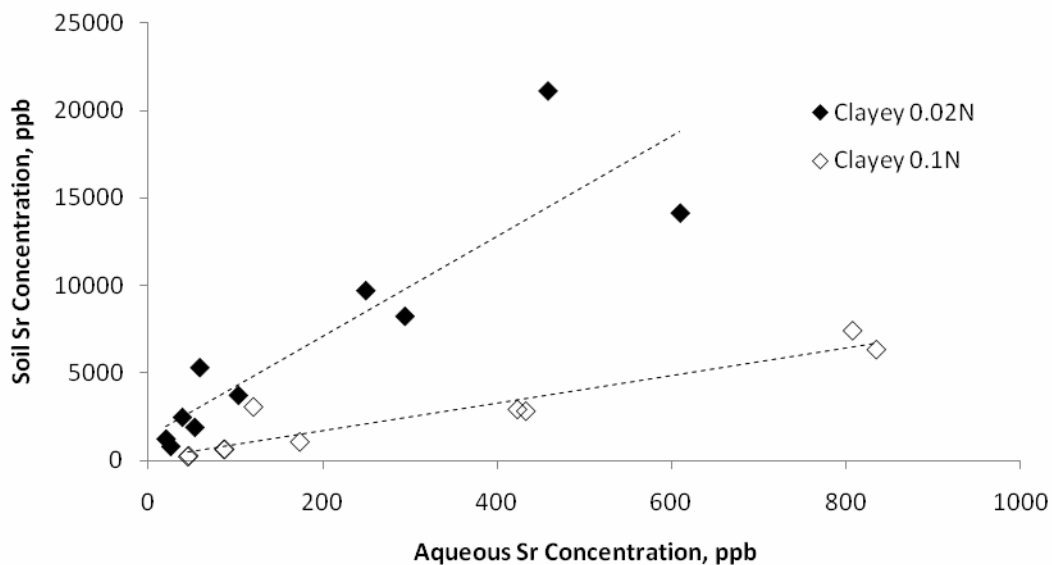


Figure 3-3: Sorption isotherm for strontium sorption on to SRS clayey soil. The K_d values were 8.05 ± 0.62 mL/g for $[\text{NaCl}]$ 0.1 M and 32.06 ± 3.62 mL g^{-1} for $[\text{NaCl}]$ 0.02 M.

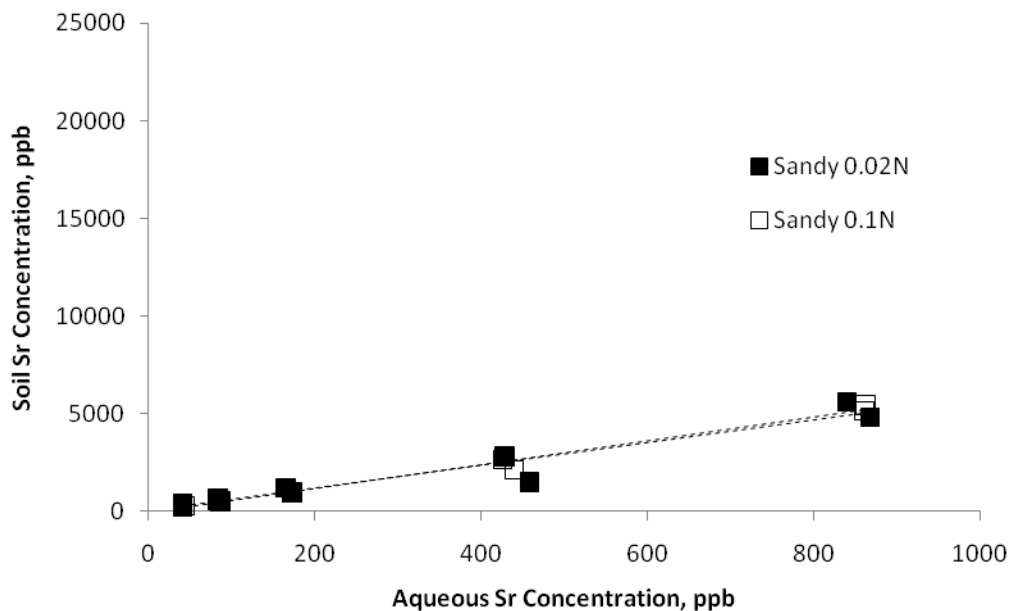


Figure 3-4: Sorption isotherm for strontium sorption on to SRS sandy soil. The K_d values were 6.02 ± 0.14 mL/g for $[\text{NaCl}]$ 0.1 M and 5.86 ± 0.35 mL/g for $[\text{NaCl}]$ 0.02 M

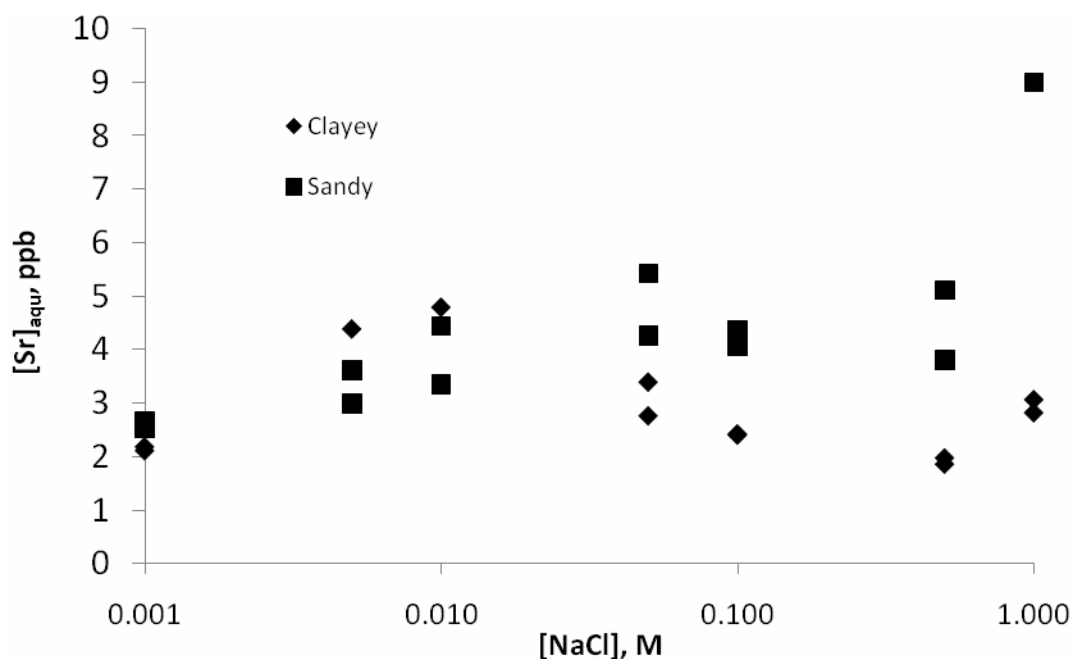


Figure 3-5: Native strontium dissolution concentration vs. ionic strength. Equilibration time of 95 days. Initial soil concentration 25 g/L.

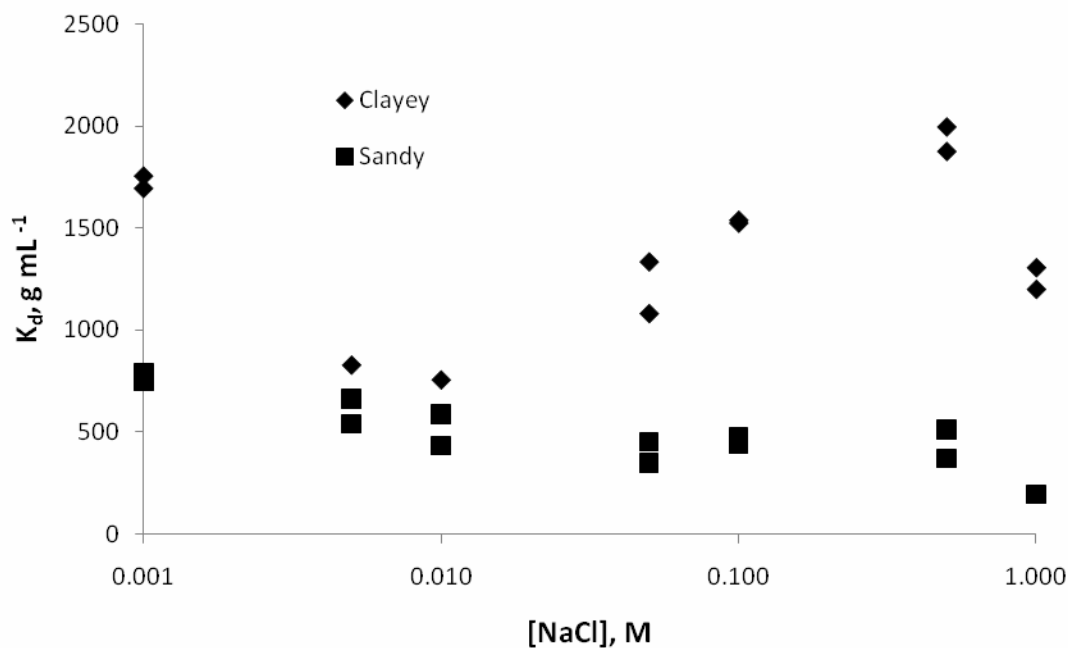


Figure 3-6: Native strontium K_d vs. ionic strength. Equilibration time of 95 day. Initial soil concentration 25 g/L.

3.1.3 Development of Ion Exchange Conceptual and Quantitative Model

This work has shown radium and strontium sorption is highly dependent upon ionic strength which can be highly variable in subsurface environments. The K_d values reported above are only valid for the pH values and ionic strengths for which they were originally determined. Therefore, a cation exchange model would be a better predictor of sorption for these two cations than single K_d values. A generic cation exchange reaction for radium is shown in Equation 3.3 and a mass balance equation for radium in the system is shown in Equation 3.4. An equilibrium constant, K , can then be calculated via Equation 3.5. Using Equation 3.6, a value of fraction sorbed can be calculated based on Equation 4.4.



$$[Ra]_T = [(2\equiv X)-Ra] + [Ra^{2+}] \quad (\text{Equation 3.4})$$

$$K = \frac{[Na^+]^2}{[\equiv X - Na]^2} \frac{[(2\equiv X) - Ra]}{[Ra^{2+}]} \quad (\text{Equation 3.5})$$

$$f_s = \frac{[(2\equiv X) - Ra]}{[Ra]_T} \quad (\text{Equation 3.6})$$

Determination of the ion exchange constants was not performed here and is suggested below as future work to be based on a larger dataset with more ionic strengths and pH values tested.

3.2 Iodide and Iodine Sorption to Natural Sediments

3.2.1 Redox Conditions for the Natural Sediments

The soil experiments below focus on iodide and iodate sorption to natural sediments. As mentioned above, iodide is expected to experience less sorption than iodate. Therefore, it is important to know what the redox conditions will be like for each soil. Table 3-1 shows the redox conditions for the sediments under oxidizing conditions for single suspensions prepared under similar conditions as the samples used for sorption studies. Under oxidizing conditions, the sandy soil is only slightly more oxidizing than the control. The wetland soil is more oxidizing than the sandy, but less than the clayey.

Table 3-1: E_h measurements for soil sediments under oxidizing conditions.

Sample	1 Day E_h (mV)	4 Day E_h (mV)	8 Day E_h (mV)
Control	219	235	250
Sandy	185	269	276
Clayey	251	294	355
Wetland	267	297	320

However, under anaerobic conditions, this order varies slightly (Table 3-2). The wetland soils become the most reducing. The control and the sandy sediment are a little more oxidizing, but still have a negative potential. The clayey sediment remains the most oxidizing, and is the only sediment to have a positive potential.

Table 3-2: E_h measurements for soil sediments under reducing conditions.

Sample	1 Day E_h (mV)	4 Day E_h (mV)	8 Day E_h (mV)
Control	78.1	44.2	-39.0
Sandy	102	114	-34.7
Clayey	101	113	44.2
Wetland	105	82.4	-47.6

3.2.2 Sorption of Iodide to Natural Sediments under Oxidizing Conditions

A plot of the K_d values for iodide in natural soils is present in Figure 3-7 (note y-axis is on log scale). The data is then replotted in Figure 4.8 with a standard y-axis configuration. As expected, the sandy soil experienced the least amount of sorption, followed by the clayey, and the wetland had the most sorption. However, what is surprising is at steady state, there is not a significant difference in the K_d values between the sandy and clayey soils, especially when taking into account the standard deviations. What is apparent is the sandy soil takes longer to reach equilibrium than either the clayey or wetland, which appear to reach it around the 1st day, while the sandy soil takes 4 days.

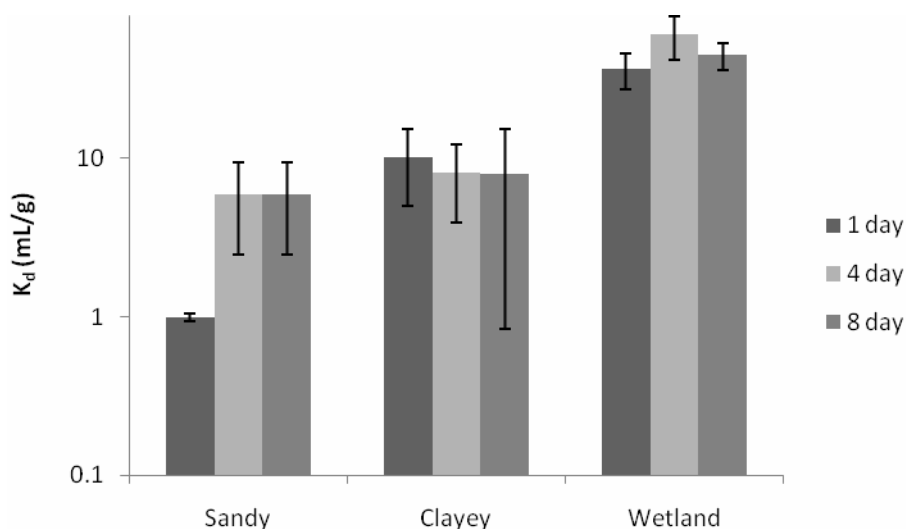


Figure 3-7: Iodide K_d Values for Natural Soils under Oxidizing Conditions. Iodide K_d values measured after 1, 4, and 8 day equilibration times. Represents average K_d values of 6 samples with varying concentrations, except for the 1, and 4 day wetland where $n=5$. The error bars represent the standard deviations. Note the y-axis is on a log scale.

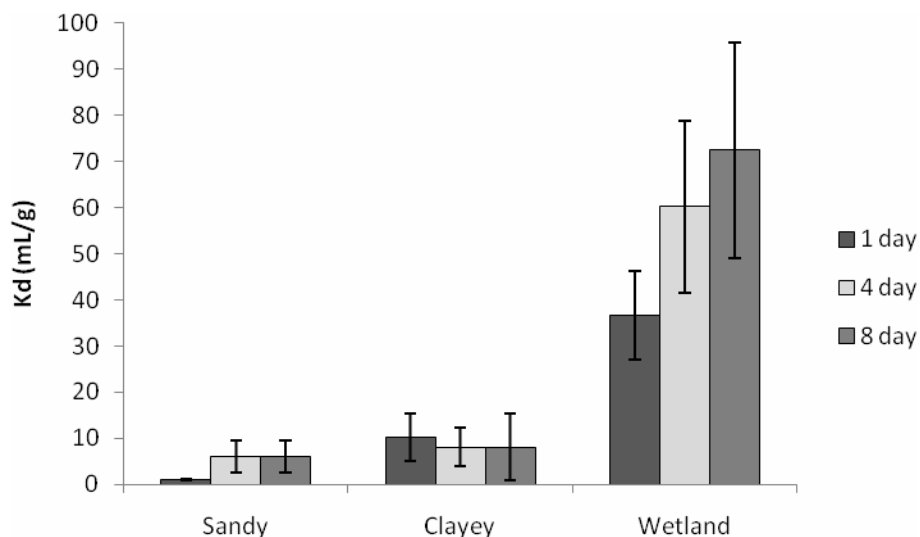


Figure 3-8: Iodide K_d Values for Natural Soils under Oxidizing Conditions. Iodide K_d values measured after 1, 4, and 8 day equilibration times. Represents average K_d values of 6 samples with varying concentrations, except for the 1, and 4 day wetland where $n=5$. The error bars represent the standard deviations.

3.2.2.1 Iodide Sorption to Vial Walls under Oxidizing Conditions

A set of control samples containing no solids was used to monitor iodide sorption to the vial walls. This data is plotted in Figure 3-9. After 24 hours the average iodide aqueous fraction for 1000ppb triplicate samples is 0.97, suggesting minimal sorption to the vial wall. However, by day 4, the fraction of iodide in the aqueous phase drops to approximately 0.90 with an 8% standard deviation. If this drop is attributable to iodide sorption to the vial wall, it is minimal sorption. It is also reversible as seen by the slightly greater than 100% recovery by day 8. When considering the standard deviations in each set of samples, there is overlap, suggesting the observed changes are likely from natural fluctuations in the data.

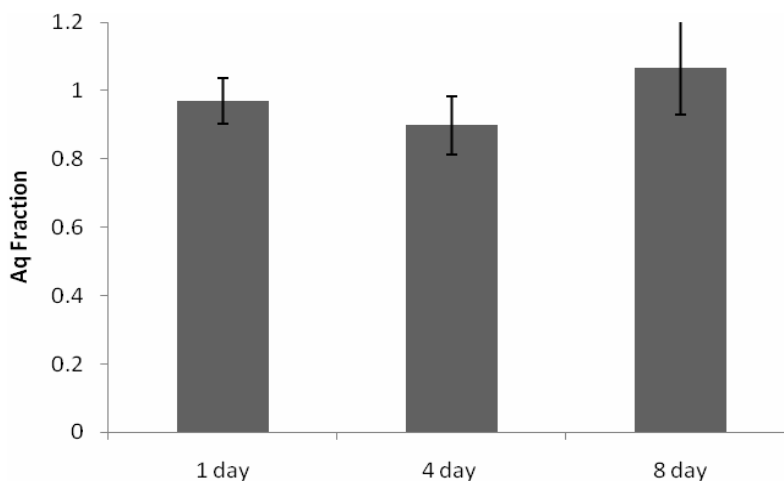


Figure 3-9: Aqueous Fraction of Iodine. Bars represent averages of triplicate 1000ppb samples with the error bars representing the standard deviations.

3.2.3 Iodide Sorption to Natural Sediments under Reducing Conditions

The average iodide K_d values for three natural soils under reducing conditions are plotted below in Figure 3-10. As was the case under oxidizing conditions, there was no sorption to the sandy soil after the 1st day, but equilibrium was reached by day 4 with a $K_d < 10$ mL/g. There is some initial sorption to the clayey soil, which increases from day 1 to day 4, but drops by the 8 day sampling. As for the wetland soil, there is strong sorption at day 1 with an average K_d value of approximately 20 mL/g. It appears to have reached equilibrium by day 4, but like the clayey, the K_d values drop on day 8. The seemingly elevated 4 day K_d values correspond with loss of iodine in solid free control sample (presumably due to sorption to vial walls, or an experimental artifact as discussed below and therefore may represent an artificial K_d increase). The drop in the 8 day K_d values could be due to native iodine being released from the soils adding to the aqueous concentration. In an attempt to correct for this, triplicate unamended samples were used to monitor the release of native iodide from the sediments. The resulting average iodide concentrations were 12 ± 1.8 , 24 ± 0.9 , and 34 ± 3.3 ppb at 1, 4, and 8 days respectively. These concentrations were then subtracted out of the measured aqueous concentrations to minimize any effect the native iodine might have. This would have the most significant effect on the 100ppb samples, in which it accounts for up to a third of the spike iodide concentration. It appears the average equilibrium K_d values for each soil coincide with each other when taking into account the standard deviations. Additionally, Table 3-3 shows the equilibrium K_d values for the sandy and clayey soil samples under oxidizing conditions are comparable, especially when taking their respective standard deviations into account. However, there is a difference between the oxidizing and reducing wetland sediment equilibrium K_d values. Those under oxidizing conditions are approximately 4-5 times greater than the equilibrium K_d s under reducing conditions. These data do not provide any indication as to what mechanism may be responsible for the difference in iodide sorption to the wetland sediment under oxidizing and reducing conditions. Oxidation of iodide to iodate in the SRS subsurface is actively being studied, and it has been found that under ambient wetland conditions, natural microbes can promote this speciation change (Li et al., 2010). It should be added that these oxidizing microbes are not especially common.

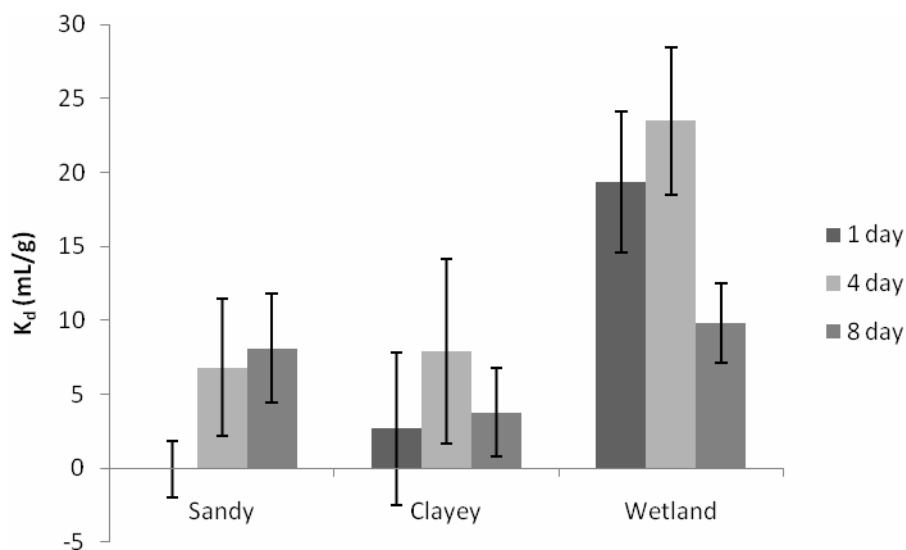


Figure 3-10: Iodide K_d Values for Natural Soils under Reducing Conditions. Iodide K_d values measured after 1, 4, and 8 day equilibration times. Represents average K_d values of 9 samples with varying concentrations, except for the 1 and 4 day clayey, and 1 day wetland where $n=8$, 1 day clayey where $n=7$, and 4 and 8 day sandy, and 4 day wetland where $n=6$. The error bars represent the standard deviations.

Table 3-3: Iodide steady state K_d values determined after 8 days of equilibration.

Soil	Oxidizing	Reducing
Sandy	5.93 ± 3.44	8.09 ± 3.68
Clayey	8.04 ± 7.21	3.78 ± 3.00
Wetland	72.5 ± 23.4	9.79 ± 2.69

As for a comparison with other sediment data, it is difficult to find studies using analogous soils. Kaplan et al. (2000) used sediments with the closest composition to the sandy in this study, but their sorption experiments were at a pH slightly above 8, where as these pH's were around 5. They observed K_d values of approximately 1-2 mL/g after 7 days, which is lower than observed for the 8 day sandy soil. This is likely due to the differences in pH. At a pH of 5 there should be more positively charged binding sites for the anionic I^- than at a pH of 8. Although K_d values were not calculated, Yamaguchi et al. (2006) monitored concentrations of extracted I^- over time. They noted retardation of the spiked I^- with some fraction I^- being more strongly associated with the soils. This conclusion was based on the findings that NO_3^- was able to leach some I^- off the soils, but a retreatment of SO_4^{2-} was able to recover additional I^- .

3.2.4 Iodide Sorption to Vial Walls under Reducing Conditions

A plot of the aqueous iodide fractions for the no-solids controls under reducing conditions is shown in Figure 3-11. There is approximately 100% recovery for the 1 day samples, and over 95% recovery for the 8 day samples. This shows iodide was not sorbing to the vial walls. There is a dip in the recovery of the 4 day samples with only 85% being recovered. This drop could be due to sorption to the vial walls. However, it appears to be easily reversible as the iodide recovery is 95% for the 8 day samples. It is noteworthy that the drop in aqueous iodide after 4 days is consistent with an increase in the sorption K_d at 4 days. Therefore, there is also a possibility of analytical error but none can be found in these datasets. Based on the good mass balance (95%) in the solid free controls after 8 days, these values are assumed to represent the most realistic K_d values.

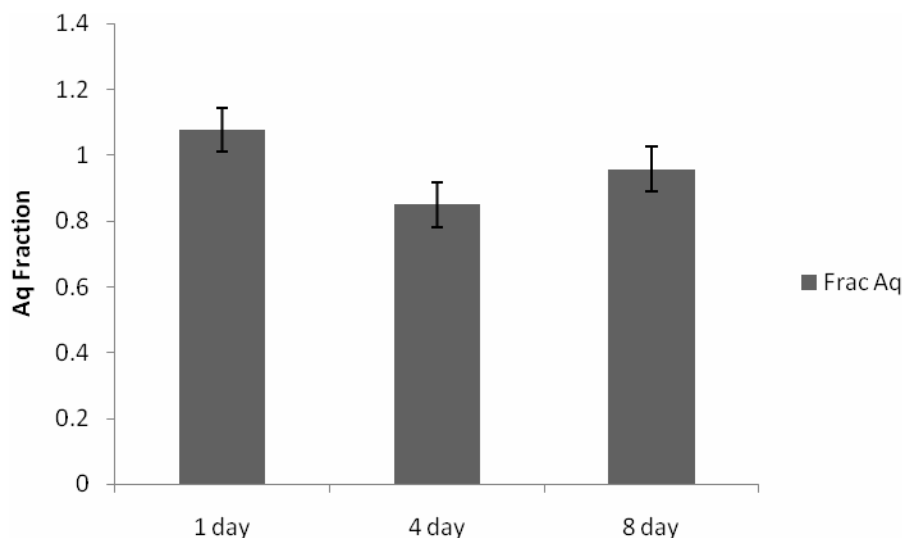


Figure 3-11: Aqueous Fractions of No-Solids Controls under Reducing Conditions. Iodine aqueous fractions above are averages of 6 samples, except for 1 day where n=3. The error bars represent the standard deviation in the samples.

3.2.5 Iodate Sorption to Natural Sediments under Oxidizing Conditions

The previous soil experiments were repeated using the oxidized iodate (IO_3^-) instead of iodide (I^-). The results are plotted in Figure 3-12. Under oxidizing conditions, iodate is expected to have a higher degree of sorption to soil sediments than its reduced form iodide (Fox *et al.*, 2010). There was some sorption to the sandy with an equilibrium K_d value of around 5 mL/g. This was reached by the 1 day sampling event. This value is similar to that of iodide under oxidizing conditions. There was however a noticeable increase for the clayey and wetland soils. There was a dramatic increase in the amount of sorption to the clayey soil as compared to the sandy. The equilibrium K_d value is just over 40 mL/g, and like the sandy, steady-state sorption was quickly reached. As was the case when iodide was used, the wetland soil showed the most iodate sorption. However, it took the longest to reach equilibrium with steady increases between each sampling event. It seemed to still be increasing by the 8 day sampling, so there is not a definite equilibrium K_d . At the 8 day event, the average K_d value is around 80 mL/g with a large standard deviation.

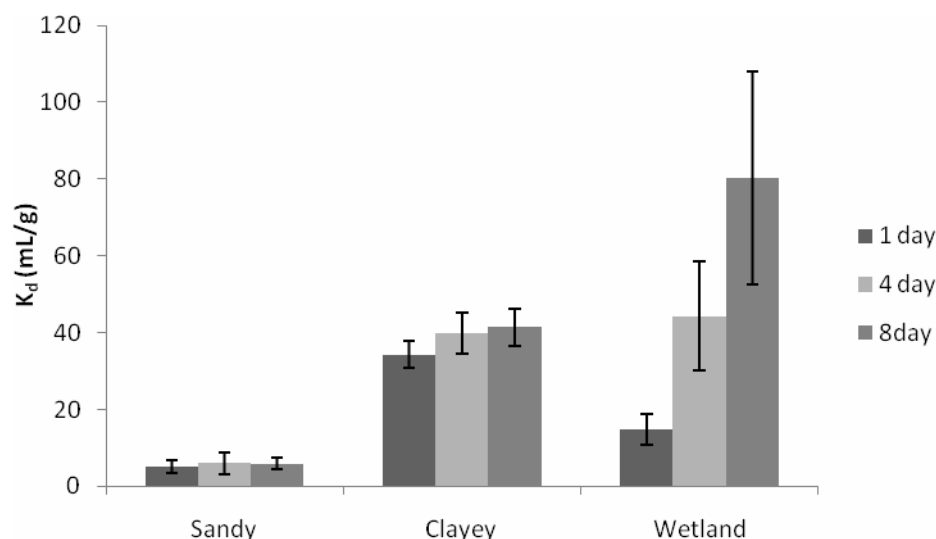


Figure 3-12: Iodate K_d Values for Natural Sediments under Oxidizing Conditions. Iodate K_d values measured after 1, 4, and 8 day equilibration times. The bars represent the average of 9 samples of varying concentrations, except for the following: sandy 4 and 8 day $n=8$, clayey 1,4, and 8 day and the wetland 1 and 4 day $n=6$, and the wetland 4 day $n=5$. The error bars represent the respective standard deviations.

Although the standard deviations seen in Figure 3-12 are small for the sandy and clayey soils, they are large for the wetland soils. This is in large part to the method of calculating K_d values, which can lead to misleadingly large deviations. Another approach is to report the fraction of the initial iodate remaining in the aqueous phase as reported in Table 3-4. This shows that the large standard deviation in the wetland soil K_d value actually represents about an 11% standard deviation in the fraction of the initial iodate remaining in the aqueous phase. Also the relatively small change in the aqueous fractions for the sandy and clayey soils from the 1 and 8 day sampling events is further evidence equilibrium is reached by the 1 day mark. However, there is a significant and steady decrease in the wetland soil from the 1 to 8 day events, which suggests equilibrium has not been reached by the 4 day sampling event.

Table 3-4: Aqueous fraction of iodate for natural soils under oxidizing conditions.

Soil	1 Day	4 Day	8 Day
Sandy	0.891 ± 0.033	$0.872 \pm 0.051^*$	$0.872 \pm 0.032^*$
Clayey	$0.540 \pm 0.025^{**}$	$0.503 \pm 0.034^{**}$	$0.494 \pm 0.029^{**}$
Wetland	$0.707 \pm 0.098^{**}$	$0.496 \pm 0.078^{***}$	$0.326 \pm 0.113^{**}$

Averages and standard deviations of 9 samples

*n=8 for the data set

**n=6 for the data set

***n=5 for the data set

3.2.6 Iodate Sorption to Natural Sediments under Reducing Conditions

Iodate sorption to the clayey and wetland soils was also examined under reducing conditions using an anaerobic glove box (98% N₂(g) and 2% H₂(g) atmosphere). The resulting average K_d values are plotted in Figure 3-13. There are small increases from the 1 to 8 day, but when considering the standard deviations, there does not appear to be a significant difference. Therefore, steady state appears to be reached by the day 1 sampling event. The resulting K_d values agree with those found under oxidizing conditions (Figure 3-12), which suggests iodate is not being reduced to iodide. If iodate was being reduced to iodide, the resulting K_d values would be similar to the equilibrium K_ds for iodide in reducing conditions. As for the wetland soil K_d values, there is significant sorption for the 1 day sorption followed by a significant decrease in sorption for the 4 and 8 day samples. This suggests the iodate in the system is being reduced to iodide by day 4. It appears equilibrium is reached around the 4 day mark, and the resulting average equilibrium K_d value at day 8 is in agreement with the equilibrium K_d value of iodide and the wetland soil under reducing conditions seen in Figure 3-10, further supporting the idea of iodate being reduced.

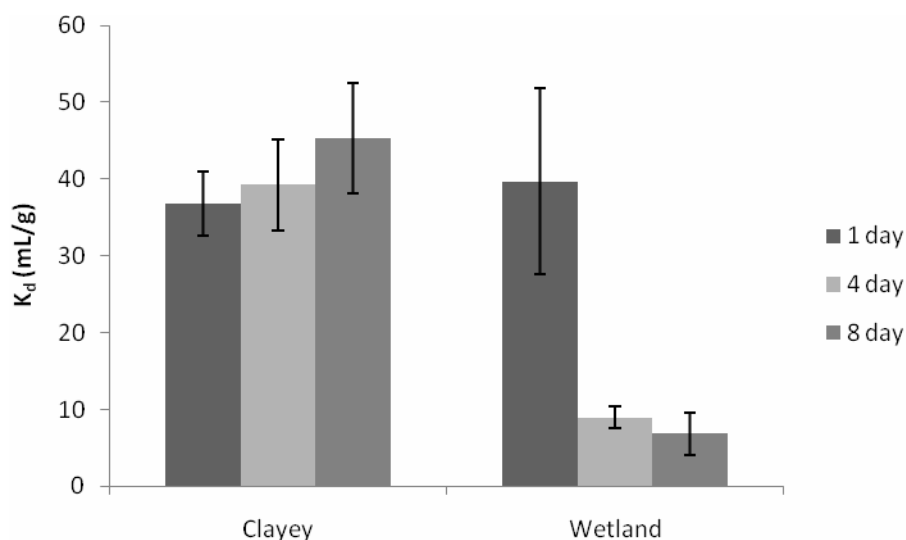


Figure 3-13: Iodate K_d Values for Natural Sediments under Reducing Conditions. Iodate K_d values measured after 1, 4, and 8 day equilibration times. The bars represent the averages of 6 samples except for the wetland 4 and 8 day samples where n=5. The error bars represent the standard deviations.

As was the case with many of the iodine experiments, the standard deviations associated with the K_d values of iodate under reducing conditions are large. The aqueous fractions better represent the agreement of the samples, and are found below in Table 3-5. In the case of the clayey soil, there is a decrease of 5% in the iodine fraction from day 1 to day 8, suggesting equilibrium is reached around day 1. However, for the wetland soil, there is an increase in the fraction of aqueous iodine of 35% from day 1 to day 8. This suggests the iodate is being reduced to iodide causing it to desorb. This increase is most significant from day 1 to day 4. From day 4 to day 8 there is an increase of <5% indicating it may have reached steady state. It is also important to note that the largest standard deviation is the wetland 1 day of 8.4%, while all the others were <5%.

Table 3-5: Aqueous fraction of iodate for natural soils under reducing conditions.

Soil	1 Day	4 Day	8 Day
Clayey	0.526 ± 0.031	0.511 ± 0.0400	0.476 ± 0.0420
Wetland	0.513 ± 0.084	$0.825 \pm 0.0230^*$	$0.862 \pm 0.0480^*$

Averages and standard deviations of 6 samples

*n=5 for the data set

The iodate 8 day K_d values for the clayey and wetland sediments under both oxidizing and reducing conditions are shown below in Table 3-6. There is not a discernible difference between the two sets of clayey data. However this is not the case for the wetland soil. These data indicate that the relatively high K_d values observed for the wetland soil under oxidizing conditions may not persist under reducing conditions. This finding has large implications regarding development of conceptual models of iodine biogeochemical behavior.

Table 3-6: Iodate steady-state K_d values mL/g after 8 day equilibration.

Soil	Oxidizing	Reducing
Clayey	41.4 ± 4.99 (n=6)	45.3 ± 7.19 (n=6)
Wetland	80.3 ± 27.7 (n=5)	6.82 ± 2.72 (n=5)

4.0 Neptunium Baseline Results

The preliminary sorption kinetic experiments were performed to determine the time required for the samples to reach equilibrium. For each of the two sediments, the aqueous neptunium concentration reached a steady state concentration after 24 hours. An equilibration time of 48 hours was used for all subsequent experiments to ensure that a steady state condition was reached.

For the baseline case, the samples were prepared as described above then sampled after 48 hours. The K_d values for the clayey sediment were calculated to be 9.05 ± 0.61 mL/g and 9.99 ± 0.28 mL/g for the centrifuged and filtrate samples, respectively (Figure 4-1). K_d values for the sandy sediment were calculated to be 4.26 ± 0.24 mL/g and 5.32 ± 0.16 mL/g for the centrifuged and filtrate samples, respectively (Figure 4-2). The data shows a stronger interaction of the neptunium with the clayey sediment than the sandy sediment. The stronger interaction of neptunium with the clayey sediment was likely due to the higher surface area of the clayey sediment (Table 3-1) and the relatively poor

interactions of neptunium with silica surfaces which may dominate the sandy sediment (Righetto et al., 1991; Bertetti et al., 1998).

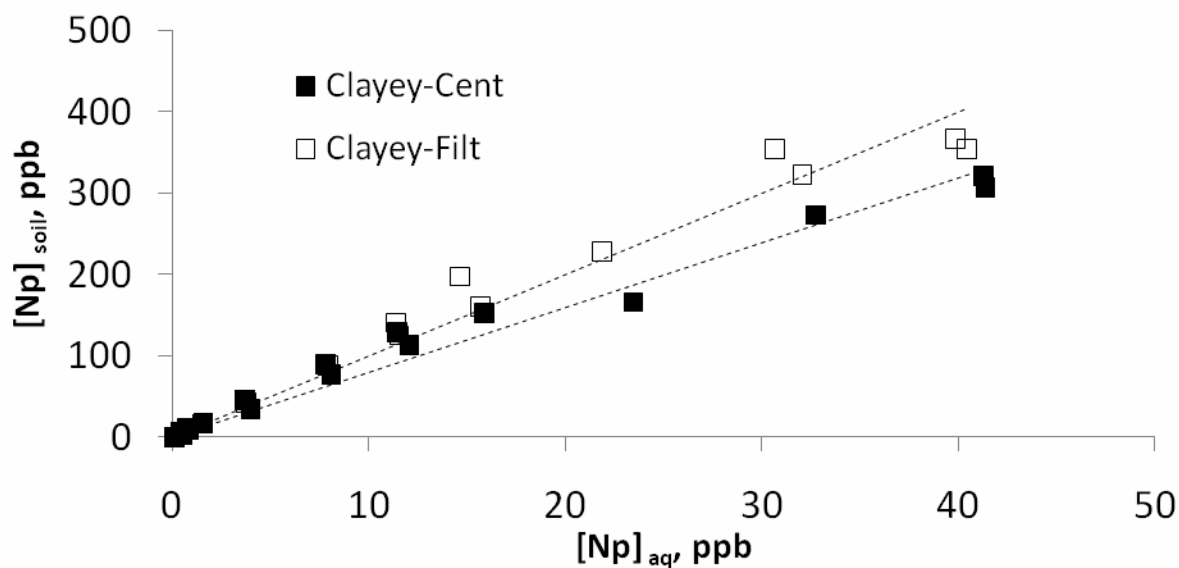


Figure 4-1: Clayey Sediment Baseline Sorption Isotherm Data measured after 48 hr. $[Np]_o$ ranged from 0.1 ppb to 50 ppb. Sediment concentration of 25 g/L. pH = 5.50 ± 0.01 . Measured K_d values of 9.05 ± 0.61 mL/g and 9.99 ± 0.28 mL/g for the centrifuged and filtered samples, respectively. Clayey-Filt samples were both centrifuged and filtered. Error determined using linear regression analysis of data to determine K_d values.

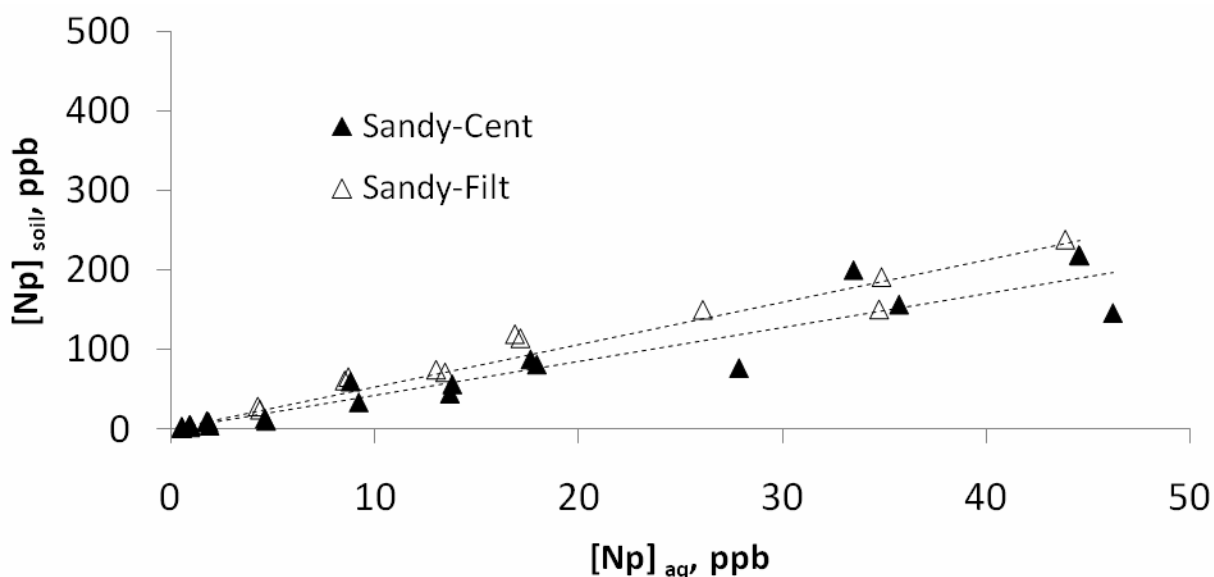


Figure 4-2: Sandy Sediment Baseline Sorption Isotherm. Data measured after 48 hours. [Np]₀ ranged from 0.1ppb to 50ppb. Sediment concentration of 25 g/L. pH = 5.50±0.03. Measured K_d values of 4.26±0.24 mL/g and 5.32±0.16 L/g for the centrifuged and filtered samples, respectively. Sandy-Filt samples were both centrifuged and filtered. Error determined using linear regression analysis of data to determine K_d values.

The data also show that there is an increase in K_d with the filtration step. This indicates that a small fraction of neptunium may be associated with colloidal particles in the 4-100 nm size range. For the clayey sediment, 1.77% of the total neptunium added was removed with the filtration step while 4.17% was removed from the sandy sediment samples. These percentages were calculated using Equation 4-1. However, the small loss of neptunium during this step is most likely attributed to sorption to the filter membrane. The same result was observed when the blank samples without sediment were passed through the filters. The sandy soil samples had a higher loss of neptunium during this step because they had higher aqueous neptunium concentrations. Therefore, a higher percent of the total amount of neptunium added to each vial was retained. If the loss of neptunium was due to association with colloidal particles, it would be expected that a higher percentage of the total neptunium would sorb to them due to their high surface area to mass ratio as compared to the bulk soil.

$$\%M_{Np,f} = \frac{([Np]_{soil,f} - [Np]_{soil,c})M_{soil}}{[Np]_{aqu,o} * V_L} \quad \text{(Equation 4-1)}$$

where:

<p>$\%M_{Np,f}$:</p> <p>$[Np]_{soil,f}$:</p> <p>$[Np]_{soil,c}$:</p>	<p>Percentage of total neptunium removed during filtration (dm)</p> <p>Calculated concentration of neptunium associated with the soil after the filtration step (μg/kg)</p> <p>Calculated concentration of neptunium associated with the soil after the centrifugation step (μg/kg)</p>	<p>Percentage of total neptunium removed during filtration</p> <p>Calculated concentration of neptunium associated with the soil after the filtration step (μg/kg)</p> <p>Calculated concentration of neptunium associated with the soil after the centrifugation step (μg/kg)</p>
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4.1 Neptunium NOM Results

Overall, the addition of the NOM increased sorption of neptunium to the sediment as compared to the baseline case. At low initial neptunium concentrations in the presence of 10 mg L⁻¹ NOM, there was little sorption of neptunium to the sediment while, at higher initial neptunium concentrations, there was increased sorption as compared to the baseline case (Figure 4-3). This added concentration of NOM is greater than would be expected in most SRS ground waters. The apparent K_d values measured for these samples were 12.90 ± 1.83 mL/g and 16.02 ± 2.88 mL/g for the clayey and sandy soils, respectively. The K_d values determined here are referred to as ‘apparent’ K_d values because it cannot be determined if the samples have reached equilibrium. However, pH control was an issue with these samples. The clayey sediment samples had pH values of 4.83 ± 0.66 as compared to 5.50 ± 0.01 for the baseline samples. The data in Powell et al. (2002) show that a decrease in pH from a value of 5.50 to 4.65 results in approximately a 50% decrease in the K_d value. This indicates that the addition of NOM increased sorption even though the pH value dropped. This is consistent with sorption of an NOM-Np complex with NOM as the bridging ligand (described as a Type B complex below). The sandy sediment samples had pH values of 5.71 ± 0.18 as compared to the baseline samples with pH values of 5.50 ± 0.03 . The data in Powell et al. (2002) show that increasing the pH from a value of 5.50 to a value of 5.70 results in a slight increase in the K_d value. Increased sorption with increasing pH is expected for metal cations and has been frequently observed for neptunium sorption to various solids (Girvin, et al. 1990; Righetto, et al. 1991). Even though sorption increased over the baseline case with the addition of NOM, the results are inconclusive because the increase in sorption could have been due to the increase in pH and not due to any interaction with NOM.

Although not shown, there was also an increase in apparent K_d with the filtration step. The filtered samples’ apparent K_d values were 15.77 ± 2.25 mL/g and 17.78 ± 2.99 mL/g for the clayey and sandy sediments, respectively. These increases in apparent K_d values are consistent with the notion that the centrifugal filters themselves retain a fraction of the neptunium resulting in artificially high apparent K_d values. For all of the samples, the NOM concentration decreased from the initial concentration of 10 mg/L indicating sorption of NOM to the sediment surface. Aqueous NOM concentrations were determined using UV-Visible spectroscopy to measure absorbance at 254 nm. The clayey sediment sorbed $37.4\% \pm 13.1\%$ and the sandy sediment sorbed $61.4\% \pm 6.2\%$ of the NOM during the equilibration period. Roughly half of the Suwannee River NOM is >10kDalton and may be removed from solution during the filtration step (Haiber et al., 2001). This indicates that neptunium does not strongly complex with this size fraction. If this were the case, the filtration step would remove a greater fraction of the total neptunium from the system. This does not provide evidence, however, of neptunium association with the smaller <10kDa fraction of the NOM.

The sorption isotherms in Figure 4-3 are non-linear over this neptunium concentration range while the curves for the baseline case were linear (Figures 5-1 and 5-2). This indicates that the K_d construct is not appropriate over this range of neptunium concentrations in the presence of NOM. It also indicates that more complicated interfacial reactions may be occurring. It is noteworthy that sorption increases, not decreases, at higher neptunium concentrations. When competitive sorption occurs, a decrease in sorption is expected and the K_d curve plateaus at elevated aqueous solute concentrations. Neptunium precipitation could explain the increase in K_d with increasing concentration, but this would not be expected due to the low neptunium concentrations used in these experiments.

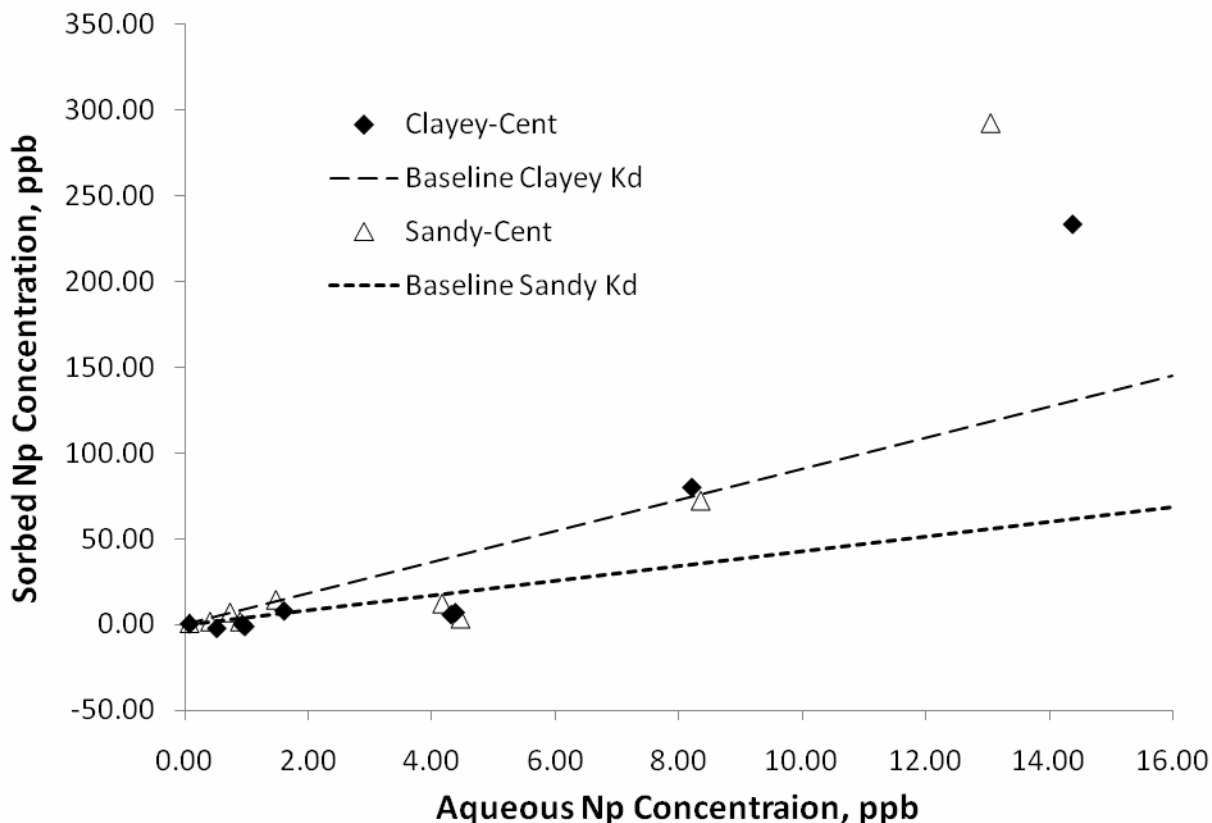


Figure 4-3: Effects of NOM on neptunium sorption measured after 48 hours. Apparent K_d values were calculated to be 12.90 ± 1.83 mL/g and 16.02 ± 2.88 mL/g for the clayey and sandy soils, respectively. $[Np]_0$ ranged from 0.1 ppb to 20 ppb. Sediment concentration of 25 g/L. pH = 4.83 ± 0.66 for clayey sediment and pH = 5.71 ± 0.18 for Sandy Sediment. International Humic Society Suwannee River NOM was added to the samples at a concentration of 10 mg/L and were sampled after an equilibration period of 48 hours.

To further explore the effects of NOM on neptunium sorption, an additional experiment was performed in which the concentration of NOM was varied from 0 to 20 mg L⁻¹ while the initial neptunium concentration was held at 10 ppb. This neptunium concentration was where sorption was elevated, but had not increased sharply in the initial NOM experiments. The results are shown in Figure 4-4. The results showed that varying NOM concentration had no effect on the magnitude of the K_d values. The average apparent K_d values for the clayey sediment were 21.67 ± 1.15 mL/g and 22.86 ± 1.38 mL/g for the centrifuged and filtered samples, respectively. For the sandy sediments, the average apparent K_d values were 5.94 ± 0.88 mL/g and 7.07 ± 0.91 mL/g for the centrifuged and filtered samples, respectively. As a point of comparison, the apparent K_d values from the initial NOM experiments for the clayey and sandy sediments at $[Np]_0 = 10$ ppb are 9.75 mL/g and 8.62 mL/g, respectively.

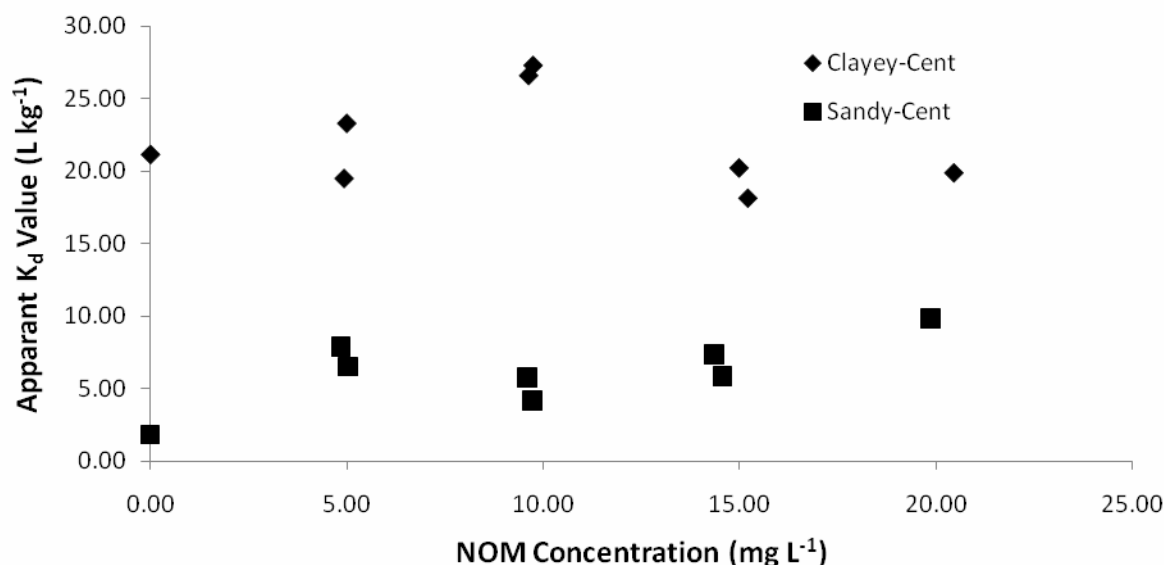


Figure 4-4: Effects of Varying NOM concentrations on Np sorption measured after 48 hours. [Np]₀ = 10ppb. [NOM]₀ ranged from 0 – 20 mg/L. Sediment concentration of 25 g/L. pH = 5.55±0.10 for Clayey sediment and pH = 5.51±0.06 for Sandy sediment.

The increase for the clayey sediment apparent K_d from approximately 13 mL/g in the initial NOM experiment is attributed to the increase in pH between the two experiments. The initial NOM experiment described in Figure 4-3 was performed at pH 4.83 and the experiment described in Figure 4-4 where the NOM concentration was varied was performed at pH 5.5. This increase in the pH, again in the presence of NOM, resulted in a marked increase in the apparent K_d value. This demonstrates a profound effect of pH on these systems that warrants further study.

Schindler et al. (1990) characterized two types of ternary metal (M), ligand (L), surface (SOH) complexes as Type A and Type B ternary surface complexes. Type A complexation involves a metal ligand complex (ML, or in this case M-NOM) forming a further complex with the surface which can be shown as $\equiv\text{SO-Np-NOM}$. Type B complexation is when the ligand acts as a bridging ligand in the ternary system which is shown as $\equiv\text{SO-NOM-Np}$. Because it was previously theorized that the organic matter coated the surface sites which, in turn, increased sorption, Type B complexation is the likely scenario in these studies. The magnitude of the decrease in aqueous NOM concentration was much greater than the decrease in neptunium concentration indicating more NOM sorption to the sediments than neptunium sorption.

Another way to view the data from these two NOM experiments is to plot the apparent K_d values as a function of the ratio of NOM to neptunium (Figure 4-5). A trend emerges that, as the ratio of NOM to neptunium increases, the apparent K_d value decreases. This could indicate that neptunium forms weak aqueous complexes with the NOM and, when the concentration of NOM is high relative to the neptunium concentration, is less prone to sorption to the sediment. In Figure 4-5, some of the data points show negative apparent K_d values. These were the result of minimal neptunium sorption coupled with the measurement error which led to a greater than 100% mass balance.

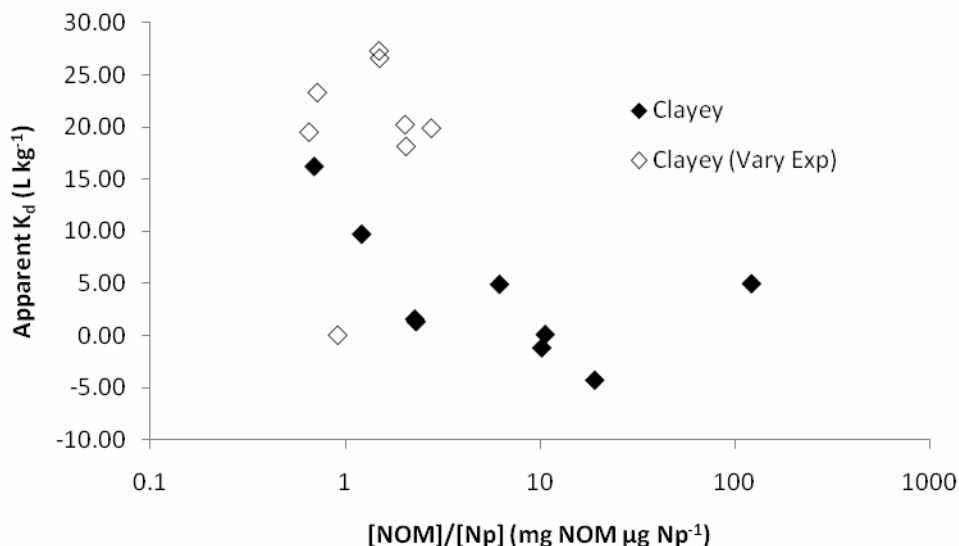


Figure 4-5: Neptunium apparent K_d values as a function of the ratio of NOM concentration to the neptunium concentration. Data obtained from the initial and varying NOM experiments. The trend indicates that sorption decreases as the concentration of NOM increases relative to the neptunium concentration.

4.2 Reducing Conditions

Microbial respiration in subsurface environments may generate a reducing, anaerobic environment. Therefore, experiments were performed in an anaerobic glovebox with an atmosphere of 98% $N_2(g)$ and 2% $H_2(g)$. The $[Np]_0$ ranged from 0.1 to 20 ppb with a fixed sediment concentration of 25 g/L. The results are shown in Figure 4-6. Because it cannot be determined if equilibrium had been reached, the partitioning coefficients will be referred to as apparent K_d values. The apparent K_d values for the clayey sediment were 12.78 ± 0.10 mL/g and 12.51 ± 0.26 mL/g for the centrifuged and filtered samples, respectively. For the sandy sediments, the apparent K_d values were 4.55 ± 0.35 mL/g and 4.84 ± 0.37 mL/g for the centrifuged and filtered samples, respectively. These apparent K_d values were 41% greater for the clayey sediment and 7% greater for the sandy sediment as compared to the baseline case (Table 4-1). The E_H^1 was measured with a VWR combination redox electrode with a Ag/AgCl reference. The measured E_H value of -200 mV indicates that reducing conditions were present and at this pH and E_H , neptunium could have been in the reduced Np(IV) oxidation state under thermodynamic equilibrium (Lilley 2009). This also indicated that an excess of reducing capacity existed and was not used up by the reduction demand of the sediment.

If Np(V) were reduced to Np(IV), the apparent K_d values would increase greatly. The inability for these conditions to reduce Np(V) may be due to kinetic limitations. However, as experiments discussed below show, even after >70 days, little evidence of reduction was seen. It is possible that the type of reductant, in this case dissolved H_2 gas, is unable to interact with the neptunium. It is fathomable that the H_2 gas could reduce the surficial ferric iron which, in turn could serve as a secondary reductant and promote

¹ E_H is a measurement made with a platinum electrode that provides an estimate of the oxidation-reduction potential. The lower the value, the more reduced the system is.

reduction of Np(V) to Np(IV). However, since increased reduction of neptunium was not seen after >70 days, this reduction mechanism may not be occurring.

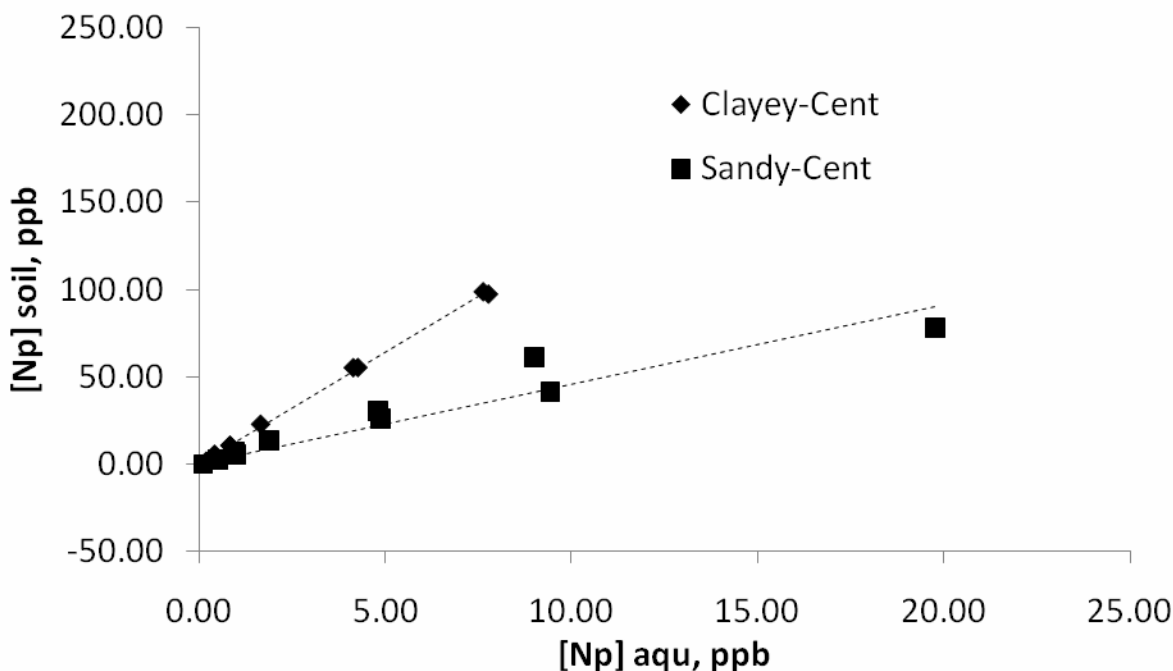


Figure 4-6: Anaerobic conditions data measured after 48 hours. [Np]₀ ranged from 0.1ppb to 10ppb for Clayey sediment and 0.1 ppb to 10 ppb for Sandy sediment. Sediment concentration of 25 g/L. pH = 5.51 ± 0.06 for Clayey sediment and pH = 5.50 ± 0.07 for Sandy sediment. Measured apparent K_d values of 12.78 ± 0.10 L/kg and 12.51 ± 0.26 L kg⁻¹ for the Clayey sediment centrifuged and filtered samples, respectively. Measured apparent K_d values of 4.55 ± 0.35 L/kg and 4.84 ± 0.38 L/kg for the Sandy sediment centrifuged and filtered samples, respectively. Measured E_H = -200 mV Ag/AgCl.

Reductants were added to the experiment to investigate the tendency of Np(V) to reduce to Np(IV). If Np(V) is reduced to Np(IV), neptunium sorption will increase, thus decreasing its subsurface mobility. Ascorbic acid, dithionite, zero valent iron, and hydrogen peroxide were selected as “environmentally relevant” reductants to examine in the sediment-aqueous-Np(V) system.

For each sample, the initial Np(V) concentration was fixed at 10 ppb and the reductant concentration was 10 ppm. The molar ratios of the reductant concentration relative to the neptunium concentration ranged from 1500-7000 indicating that there was an excess of reductant. Compared to the baseline oxic clayey sediment K_d value of 9.05 mL/g and the sandy sediment K_d value of 4.26 mL/g (Figures 4-1 and 4-2), the amount of reduction induced by the various treatments was relatively small (Table 4-1). The increases in sorption are consistent with the three reductants, ascorbic acid, dithionite, and zero valent iron reducing a small fraction of Np(V) to Np(IV). Higher concentrations of the reductants could have been used and may have resulted in more reduction of Np(V) to Np(IV); however, the concentrations used more accurately represented potential environmental conditions.

Table 4-1: K_d Values for Np Sorption under Reducing Conditions

Treatment	Apparent K_d Values ($L\ kg^{-1}$)	
	Clayey	Sandy
Control (No treatment)	9.05 ± 0.61 ^(a)	4.26 ± 0.24 ^(b)
Ascorbic Acid	21.06 ± 0.74	12.76 ± 0.46
Dithionide	22.96 ± 0.56	11.14 ± 0.80
Zero-valent Iron	23.65 ± 0.58	11.99 ± 0.46
Hydrogen Peroxide	12.47 ± 0.46	9.23 ± 0.42
Anaerobic Chamber ^(c)	12.78 ± 0.10	4.55 ± 0.35
^(a) Data from Figure 4-1 for the centrifuged samples.		
^(b) Data from Figure 4-2 for the centrifuged samples.		
^(c) Data from Figure 4-5 for the centrifuged samples		

The overall trend seen with the reductant experiments was determined to be:

Control < Anaerobic \approx Hydrogen Peroxide < Ascorbic Acid \approx Dithionite \approx Zero Valent Iron.

Based on the K_d values in Table 4-1, hydrogen peroxide did not cause an increase in sorption and therefore does not appear to have reduced Np(V) to Np(IV). The ascorbic acid, dithionite, and zero valent iron only promoted a small increase in sorption, and potentially only a small reduction of Np(V) to Np(IV).

4.3 Neptunium Desorption Experiments

To examine the behavior of neptunium desorbing from the soils, further experiments were performed. After the baseline experiments were performed, half of the samples had their supernatants decanted and were refilled with water. After an equilibration period, the supernatant was analyzed as stated previously. The average apparent desorption K_d value for desorbing neptunium from the clayey sediment after two days was 8.08 ± 1.35 mL/g. This was comparable to the sorption K_d of 9.05 ± 0.61 mL/g. Because it was theorized that desorption kinetics were slower than sorption kinetics, the samples were allowed to equilibrate for an additional 67 days. The average apparent desorption K_d value after this longer time was 10.14 ± 1.05 mL/g. When taking into consideration the error associated with these calculated values, it appears neptunium sorption is completely reversible within 67 days of equilibration and possibly within 2 days (Figure 4-7).

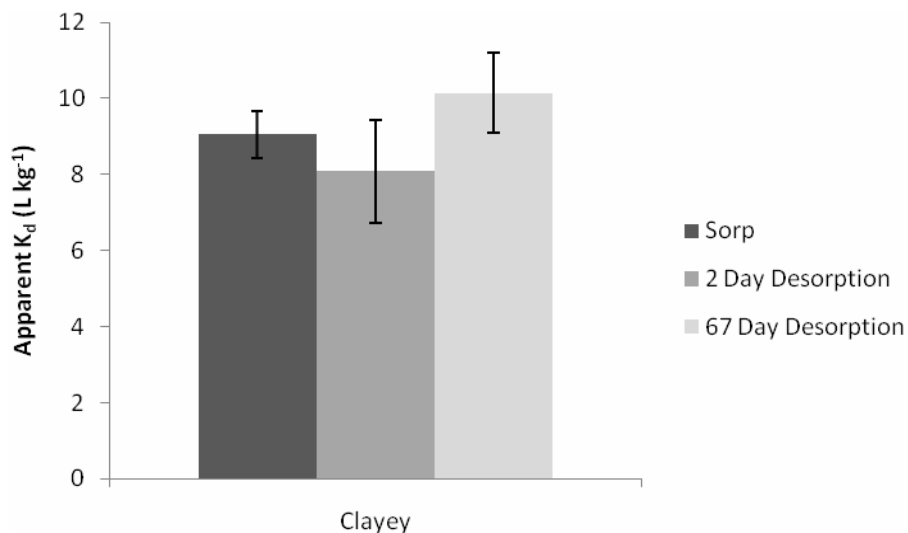


Figure 4-7: Comparison of sorption and desorption K_d values for the clayey sediment under aerobic conditions. Little difference is seen between the K_d values for neptunium sorption and the apparent desorption K_d values for short term (2 day) and long term (67 day) desorption.

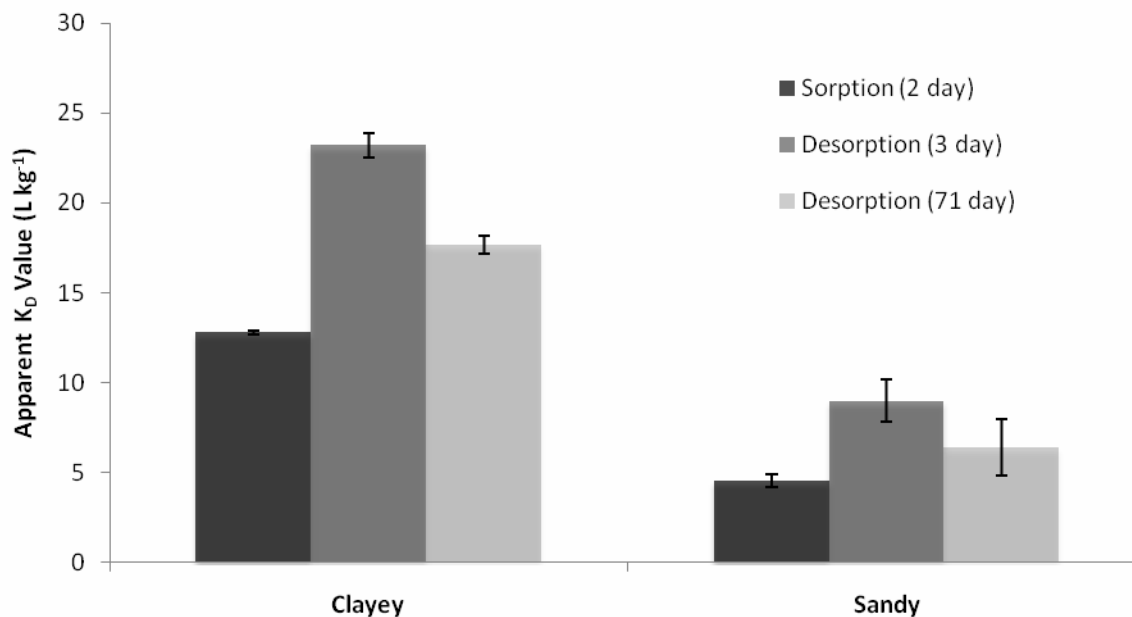


Figure 4-8: Comparison of sorption, short, and long term desorption K_d values under anaerobic conditions.

Due to the small mass of neptunium sorbed to the sandy sediment, the concentration of neptunium desorbed from the soil was very small and led to large uncertainty in the results. There was also a loss of pH control on the long term desorption samples so accurate comparison was not possible and these data were not included in this thesis.

Desorption tests were also performed on the anaerobic samples to examine sorption hysteresis under reducing conditions. It was postulated that the reduction of Np(V) to Np(IV) may be a slow process and long term equilibration periods might show some evidence of this reduction. After 3 days, the apparent desorption K_d for the clayey soil was 23.19 ± 0.69 mL/g. The long term apparent desorption K_d after 71 days for the clayey soil was 17.67 ± 0.52 mL/g. For the sandy soil, the apparent desorption K_d after 3 days was 8.99 ± 1.18 mL/g and after 71 days was 6.37 ± 1.57 mL/g. These results are shown in Figure 4-8 along with the sorption K_d values.

These data show that short term apparent desorption K_d values are greater than the sorption K_d values, but over time they decrease. This shows that the desorption kinetics are slower than the sorption kinetics. If enough time were allowed for equilibration, it may be possible to show that the reaction is fully reversible. Although not performed here, the experiment could have been modified such that the supernatant was decanted after each sampling period and refilled with clean water. This would increase the concentration differential between the neptunium associated with the soil and the liquid phases and lead to an overall faster mass desorption rate.

It is also important to note that this data further shows how neptunium is not reduced to a significant degree under the anaerobic conditions. If reduction of Np(V) to Np(IV) was occurring, it would have resulted in increased apparent desorption K_d values over time. Because the apparent desorption K_d values decreased between the 3 day and the 71 day desorption steps, this reaction can be assumed to be negligible.

4.4 Flowcell

The first flowcell experiment was conducted to determine if the vessel performed as an ideal continuously stirred tank reactor (CSTR). A stock solution of tritiated water (HTO) at a concentration of 680 CPM mL⁻¹ was pumped into the flowcell which was initially filled with clean water at a rate of 0.33 mL min⁻¹. No soil was present. The effluent from the cell was collected using a fractional collector and the HTO concentration was determined via liquid scintillation counting. The results were compared against the theoretical curve for a nonreactive tracer shown in Equation 4-2. The results are shown in Figure 4-9.

$$C = C_o \left(1 - \exp^{-V/V_r} \right) \quad (\text{Equation 4-2})$$

where:	C	Concentration of effluent (CPM mL ⁻¹)
	C _o	Initial concentration (CPM mL ⁻¹)
	V	Total volume of solution passed through cell (mL)
	V _r	Volume of flowcell (mL)

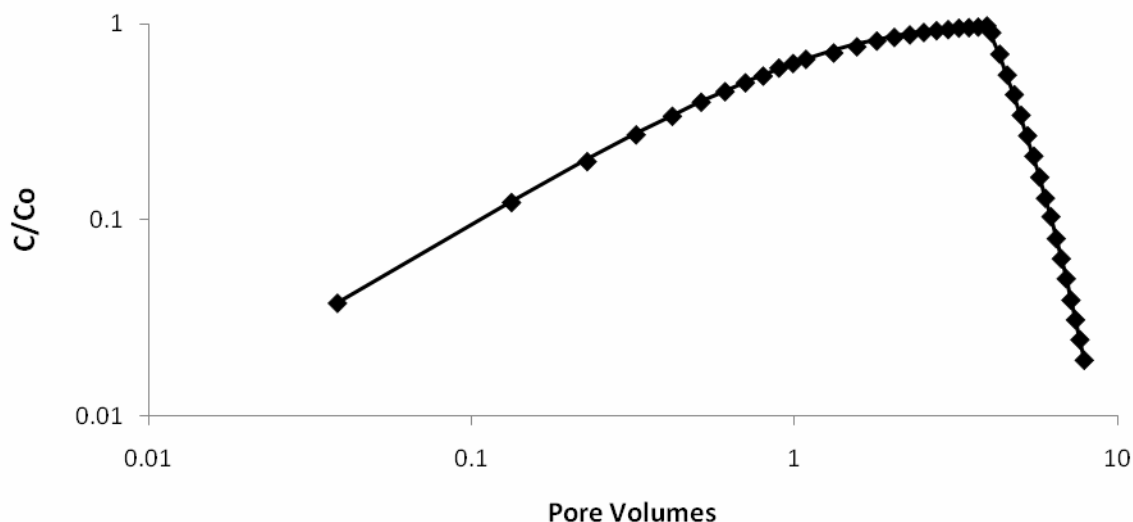


Figure 4-9: Flowcell performance vs. theoretical performance for an ideal CSTR. Black diamonds represent actual data points and solid black line represents theoretical curve. Flowrate 0.33 mL /min. No soil present.

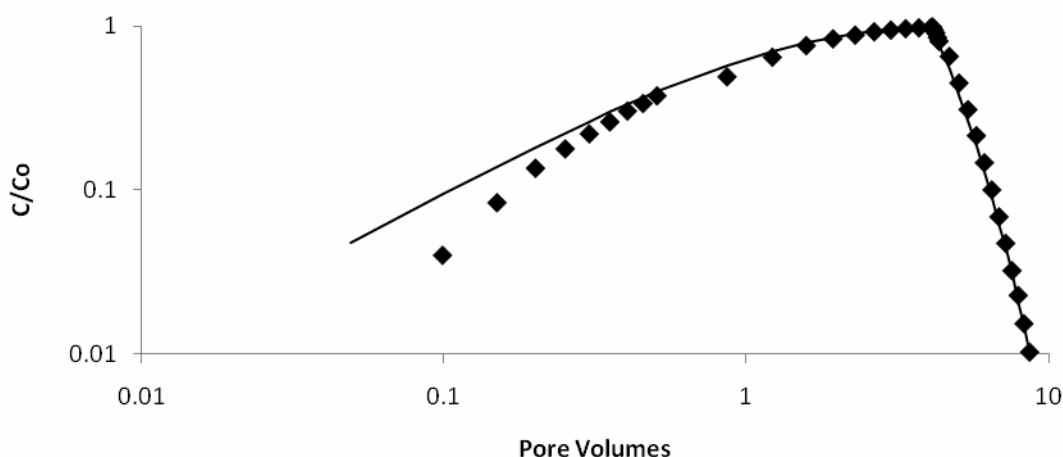


Figure 4-10: Flowcell performance vs. theoretical performance for an ideal CSTR containing 0.5g of the clayey sediment. Black diamonds represent actual data points and solid black line represents theoretical curve. Flowrate 0.33 mL/min.

As Figure 4-9 shows, the flowcell performs as an ideal CSTR at the flowrate of 0.33 mL min⁻¹. The experiment was repeated with a flowrate of 1 mL min⁻¹ and the same performance characteristics were achieved. This proved that the vessel remained ideal even at varying flow rates. To determine if the presence of soil in the flowcell affected the flowcell's performance, 0.5 g of soil was added and the same test was run at a flowrate of 1 mL min⁻¹. As shown in Figure 4-10, there was a lag time associated with the HTO flowing through the flowcell. Tritium is typically used as a non reactive tracer, however it can actually interact with some soils and its migration be retarded. Zuo et al. (2009) determined that tritium was retarded by a Chinese loess media, (which is an unstratified usually buff to yellowish brown loamy deposit found in No. America, Europe, and Asia and believed to be chiefly deposited by the wind), and

found K_d values ranging from 0.05 to 0.25 mL/g. Because this can account for the discrepancy in the flowcell performance, it was determined that the vessel was still ideal while containing soil.

Initial testing of the flowcell with sediment showed that the sediment acted as an abrasive agent between the stir bar and the vessel wall and resulted in the destruction of both in as little as a few days. This resulted in pieces of Teflon from the stir bar coating being suspended in the flowcell. The abrasion also carved into the bottom of the flowcell which increased the volume of the vessel. The sandy sediment showed the greatest abrasive properties due to its large silica particles and was not used in this study. To decrease the abrasion, the clayey sediment was sieved to $<53 \mu\text{m}$ and this eliminated the problem.

Once the final flowcell design was ready and the experimental issues were worked out, the neptunium flowcell experiment was initiated by filling the flowcell with 0.5 g of the sieved clayey soil and a background solution of $[\text{NaCl}] = 0.01 \text{ M}$ at pH 5.50. The sediment had a natural buffering capacity which lowered the pH of the solution passing through it. This was seen previously during some of the equilibrium tests. To mitigate this issue, the background solution was flowed through the flowcell until the effluent pH was equal to the influent pH. The suspension was then left overnight and the pH was checked again the next day. This process was repeated until no change in pH was observed after equilibrating overnight. A total of 1.765 L of background solution was passed through the flowcell over a time period of 6 days to acclimate the soil to the conditions of the experiment.

A spike solution containing 11.3 ± 0.1 ppb neptunium and 1500 CPM mL ^3H with an ionic strength of 0.01 M and a pH of 5.50 was used for the sorption portion of the experiment. The background solution mentioned earlier was used during the desorption portion of the experiment. A constant flowrate of 0.33 mL min^{-1} was used throughout the experiment which resulted in a residence time of 60.6 minutes. Table 4-2 shows the schedule of the sorption and desorption steps as well as the stopped flow periods for the experiment.

Table 4-2: Flowcell Schedule.

	Sample	Start Flow	Stop Flow	Flow time (hrs)	Cell Volumes	Stopped Flow time (hrs)	Stopped Flow time (days)
Sorption	Samples S1-S10	12/2/2009 14:29	12/2/2009 15:19	0.83	1.03		
						2.00	0.08
	Samples S11-S27	12/2/2009 17:19	12/2/2009 18:57	1.63	2.01		
						18.77	0.78
	Samples S28-S44	12/3/2009 13:43	12/3/2009 15:27	1.73	3.14		
						0.00	0.00
	Samples S45-D25	12/3/2009 15:27	12/3/2009 19:28	4.02	7.11		
						26.20	1.09
	Samples D26-D92	12/4/2009 21:40	12/5/2009 10:23	12.72	18.38		
						70.03	2.92
Desorption	Samples D93-D156	12/8/2009 8:25	12/8/2009 20:31	12.10	22.07		
						189.15	7.88
	Samples D157-D187	12/16/2009 17:40	12/17/2009 13:55	20.25	34.64		
						549.45	22.89
	Samples D188-D212	1/9/2010 11:22	1/9/2010 15:32	4.17	27.37		
						1559.65	64.99
	Samples D213-						

The sorption portion of the experiment lasted for 25 hours and a total of 5.15 cell volumes (103 mL) of spiked solution were pumped through the cell. The flow was stopped once for 2 hours after 1.03 cell volumes and again for 18.8 hours after 3.05 cell volumes. Figure 4-11 details the sorption portion of the experiment. The figure shows that there is sorption of both neptunium and tritium to the soil because their concentrations are below the theoretical tracer curve. The aqueous neptunium concentration did not decrease during the stopped flow periods for the sorption phase of the experiment meaning that the kinetic rate constant for sorption was the same or greater than the inverse of the flowcell residence time, or 0.0165 min^{-1} . There was one data point that did show a sharp drop in aqueous neptunium concentration after the 18.8 hour stopped flow period, but the sample immediately following it was at the same concentration as the sample before the stopped flow period. This data point was found to be erroneous due to sampling issues that occurred while the ICP-MS was processing this sample.

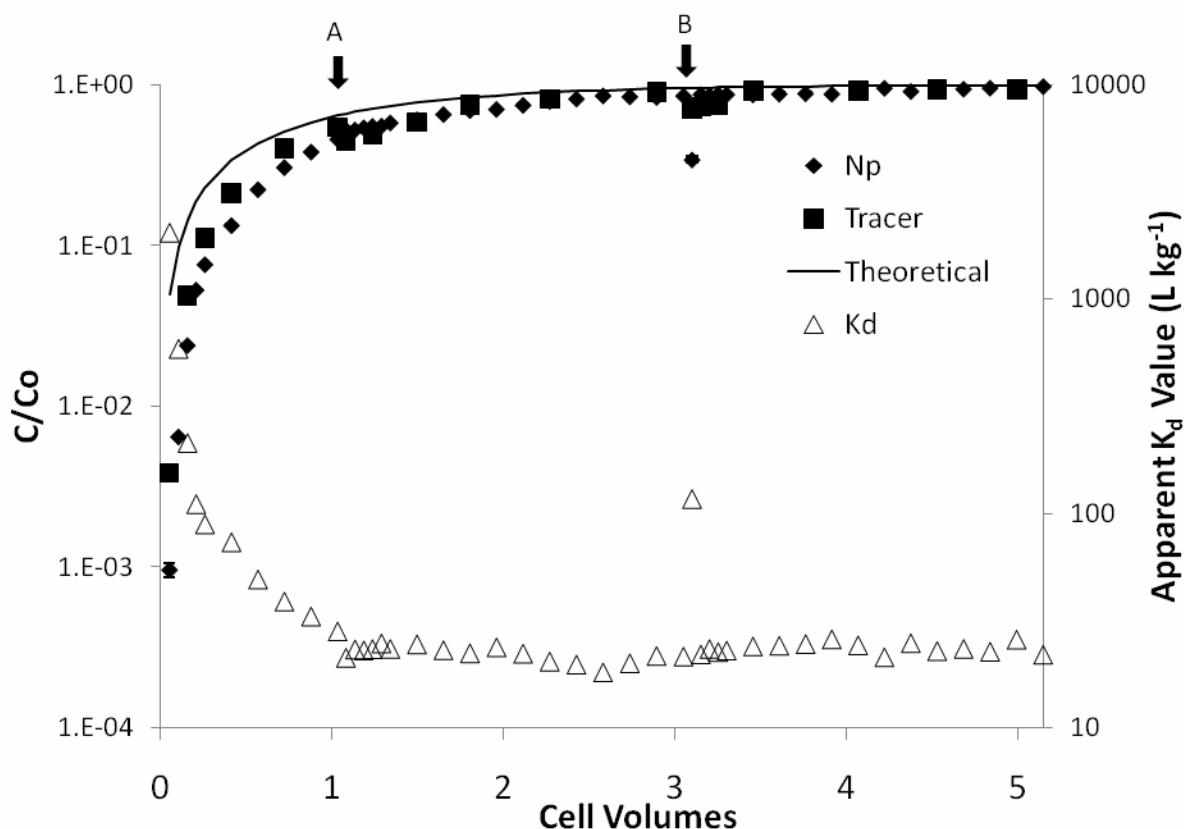


Figure 4-11: Flowcell sorption step results. Point A indicates the 2 hour stopped flow period after 1.03 cell volumes. Point B indicates the 18.8 hour stopped flow period after 3.05 cell volumes. Note: the x-axis is in a linear scale to show detail.

The apparent K_d values during the sorption step agree closely with those from the baseline case. During the last portion of the sorption step (cell volumes 1.0 to 5.1) the average apparent K_d value was $22.85 \pm 1.73 \text{ mL/g}$ (Figure 4-11) as compared to $9.05 \pm 0.61 \text{ mL/g}$ for the baseline case. This increase can be attributed to using the clayey sediment that had been sieved to $<53 \mu\text{m}$ which resulted in a higher surface area per mass of soil and provided more surficial sites for sorption. The apparent K_d values for the samples during the sorption steps remained relatively constant indicating that the rate of neptunium sorption is fast relative to the residence time of the flowcell. During the sorption step, a total of $0.116 \mu\text{g}$ of neptunium sorbed to the sediment reaching a sorbed neptunium concentration of $229.8 \mu\text{g/kg}$.

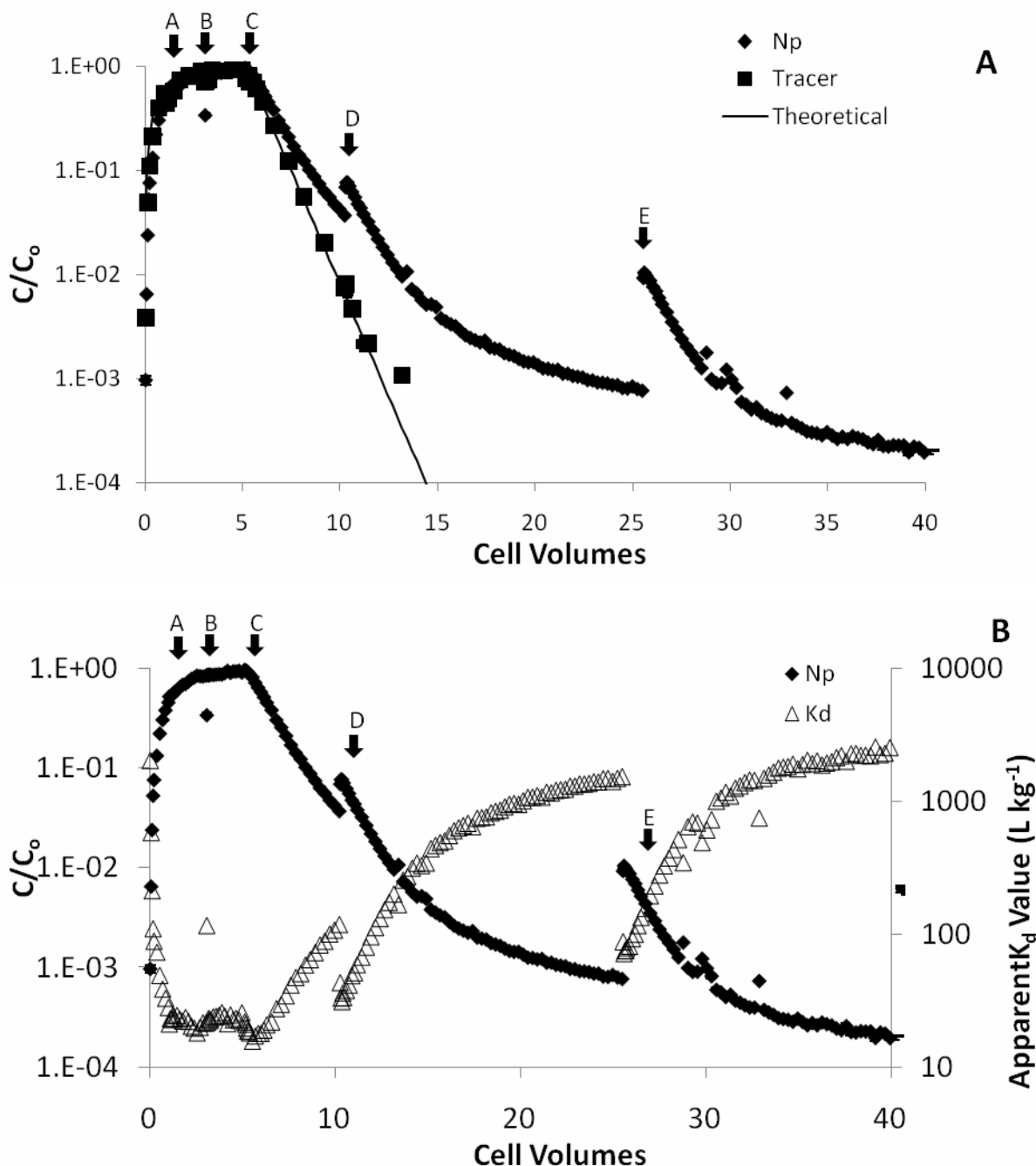


Figure 4-12: Flowcell sorption step and initial desorption step results. Point A indicates the 2 hour stopped flow period after 1.03 cell volumes. Point B indicates the 18.8 hour stopped flow period after 3.05 cell volumes. Point C indicates where the flowcell feed was switched to the background solution after 5.15 cell volumes. Point D indicates a 26.2 hour stopped flow period after 10.24 cell volumes. Point E indicates a 70.0 hour stopped flow period after 25.48 cell volumes. Note: the x-axis is in a linear scale to show detail. (A) Neptunium concentration relative to HTO and theoretical tracer as a function of cell volumes. (B) Neptunium concentration and K_d values as a function of cell volumes.

The results of the first half of the desorption step of the experiment is shown in Figure 4-12. At point C, the feed to the flowcell was changed from the spike solution to the background solution. At this point, the neptunium and tritium in the cell began to wash out of the flowcell. The solid line in the figure represents how a non-reactive theoretical tracer washes out of an ideal CSTR. Any tritium or neptunium data points that lie above this line indicate that it is present due to desorption from the sediment.

The rate of desorption was shown to be slow relative to the residence time of the flowcell due to the large increases in aqueous neptunium concentration after the stopped flow events. This is in agreement with the findings during the sorption equilibrium experiments where it was determined that the rate of sorption was faster than the rate of desorption. Figure 4-12B shows that as each desorption step proceeds, the concentration of sorbed neptunium relative to aqueous neptunium increases, leading to higher K_d values. By comparing the change in concentration of sorbed neptunium with time during the flow periods, a rough approximation of the desorption kinetic rate constant was calculated for every data point using Equation (4-3). Taking an average of the data points, a desorption rate constant of $3.3 \times 10^{-3} \pm 7.9 \times 10^{-3} \text{ min}^{-1}$ was obtained.

$$\frac{-\ln\left(\frac{S_t}{S_{t-1}}\right)}{t} = k \quad (\text{Equation 4-3})$$

where:

S_t :	Solid phase neptunium concentration at time t ($\mu\text{g/kg}$)
S_{t-1} :	Solid phase neptunium concentration at time $t-1$ ($\mu\text{g/kg}$)
t :	Time between samples C_t and C_{t-1} (min)
k :	Kinetic rate constant (min^{-1})

This is a factor of three slower than the sorption rate constant. However, note that this value includes the concentrations measured immediately after stopped flow periods and is therefore artificially high. More rigorous calculations could be performed to obtain more accurate kinetic parameters for this process. A specific example of using a discrete portion of the dataset for this purpose is discussed below. However, as will also be discussed below, neptunium sorption appears to be completely reversible and the overall reactions occur on the order of days. Therefore, for the timeframes used in performance assessment modeling, it can be reasonably assumed that neptunium sorption/desorption achieved a steady state distribution and the specific rate constants will not improve these predictions. Therefore, a rigorous mathematical analysis of these data is considered extraneous and beyond the objective of this work.

Analysis of the aqueous neptunium concentration and sorbed neptunium concentration during each desorption step showed that, as more background solution was passed through the flowcell, the system strayed further from equilibrium. After each stopped flow period, the ratio of sorbed neptunium to aqueous neptunium returned to values approaching the equilibrium K_d value. This data is presented in Figure 4-13. The data is compared against three K_d lines that represent values of 10, 20, and 30 mL/g.

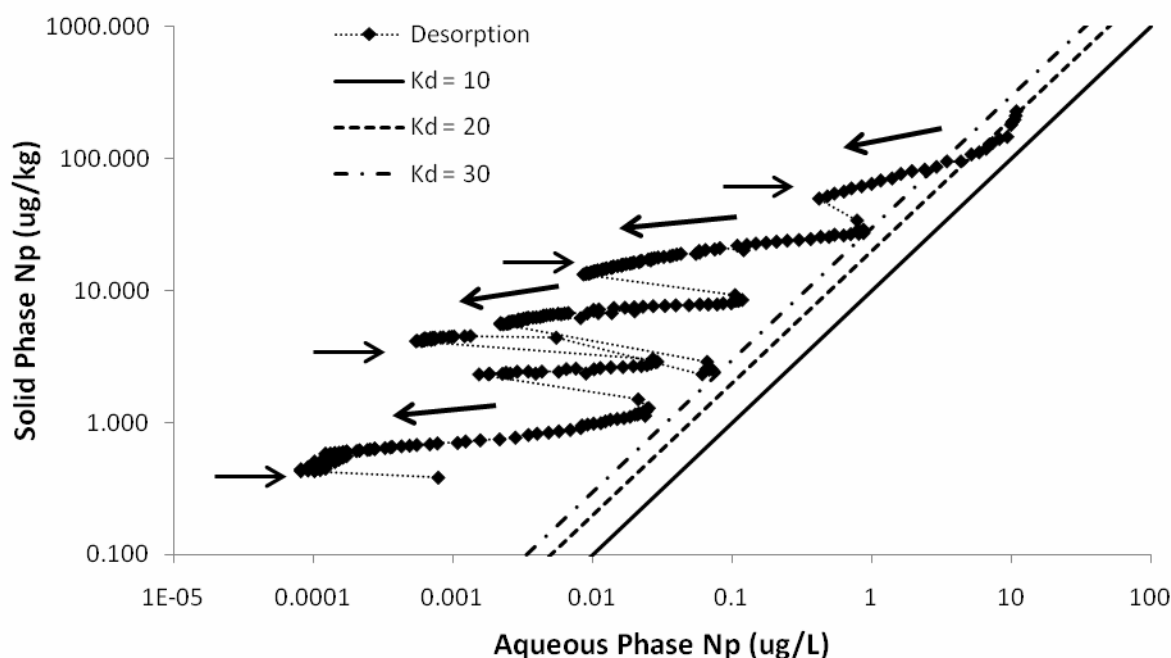


Figure 4-13: Neptunium sorption/desorption isotherm showing departure from equilibrium during the flow events and return to equilibrium during stopped flow events.

The complete results of the experiment are shown in Figure 4-14. The results show that, after 8 cell volumes of background solution were passed through the flow-cell, 90% of the sorbed neptunium desorbed. After 57 cell volumes, 99% of the neptunium desorbed. At the end of the experiment and after 110 total cell volumes of background solution flowed over the sediment, >99.8% of the neptunium was removed from the soil. The final sorbed neptunium concentration was verified by microwave soil digestion and analysis on the ICP-MS. The final concentration was measured to be 0.15 $\mu\text{g/kg}$ which was in good agreement with the calculated value of 0.38 $\mu\text{g/kg}$ when compared to the total sorbed neptunium which was initially at a concentration of 230 $\mu\text{g/kg}$. This represents 0.06% of the total sorbed neptunium concentration after the sorption step.

This experiment also removed more of the sorbed neptunium from the soil because clean water was constantly flowing into the cell and neptunium was always flowing out of the cell. This allowed the aqueous concentration of neptunium to remain low and increase the favorability for neptunium to desorb from the soil. These results are evidence that sorption of neptunium to this soil is a completely reversible process.

The last step in the desorption experiment shown in Figure 4-14 can also be used to calculate a desorption rate constant. During this flow period, a total of 1035 mL of background solution was passed through the flowcell. From Equation 4-2, the aqueous concentration of the flowcell will be <0.001% of its initial concentration after 15 cell volumes. Therefore, any aqueous neptunium concentration after this time can be assumed to have desorbed from the sediment during that time period. This allows for very accurate measurements of desorption kinetic parameters. Figure 4-15 shows the correlation between change in aqueous neptunium concentration in the flowcell and the calculated rate constants. The tail end of the desorption experiment shown by the black box in Figure 4-15 gives an average desorption rate constant of $1.8 \times 10^{-4} \pm 5.9 \times 10^{-5} \text{ min}^{-1}$. This range of data points was selected because, at extended flow periods, the majority of the aqueous neptunium has already been washed out and the remaining neptunium is only

present due to desorption from the soil. This value is smaller than the earlier reported desorption rate constant of $3.3 \times 10^{-3} \text{ min}^{-1}$. However, this earlier value may be larger than the actual rate constant due to its incorporation of data near the stopped flow periods.

One theory suggests that the difference between these two rate constants can be attributed to the ease of removing a mass of neptunium from weak sorption site compared to a strong sorption site. This theory suggests that mineral surfaces contain a distribution of strongly and weakly sorbing sites and the majority of surface sites are weakly sorbing (Dzombak and Morel, 1990). During desorption, neptunium would first be removed from the weak sorption sites. As more neptunium is removed, the ratio of neptunium sorbed to the strong sites relative to the weak sites increases. Desorption from these strong sorption sites is expected to be relatively slower which results in lower desorption kinetics.

The sorption and desorption rate constants can also be used to calculate a distribution ratio similar to a K_d value by taking the ratio of the desorption rate constant to the sorption rate constant. Using the aforementioned sorption rate constant and the smaller desorption rate constant, a value of 0.011 is obtained. This dimensionless number can be compared to the K_d value previously obtained by dividing by the sediment concentration of 25 g/L. This calculation results in a value of 0.44 mL/g which is within an order of magnitude of the calculated equilibrium K_d value for the clayey sediment of 9.05 mL/g. The comparison of these numbers shows that the crude method of rate constant determination performed here resulted in fairly comparable values.

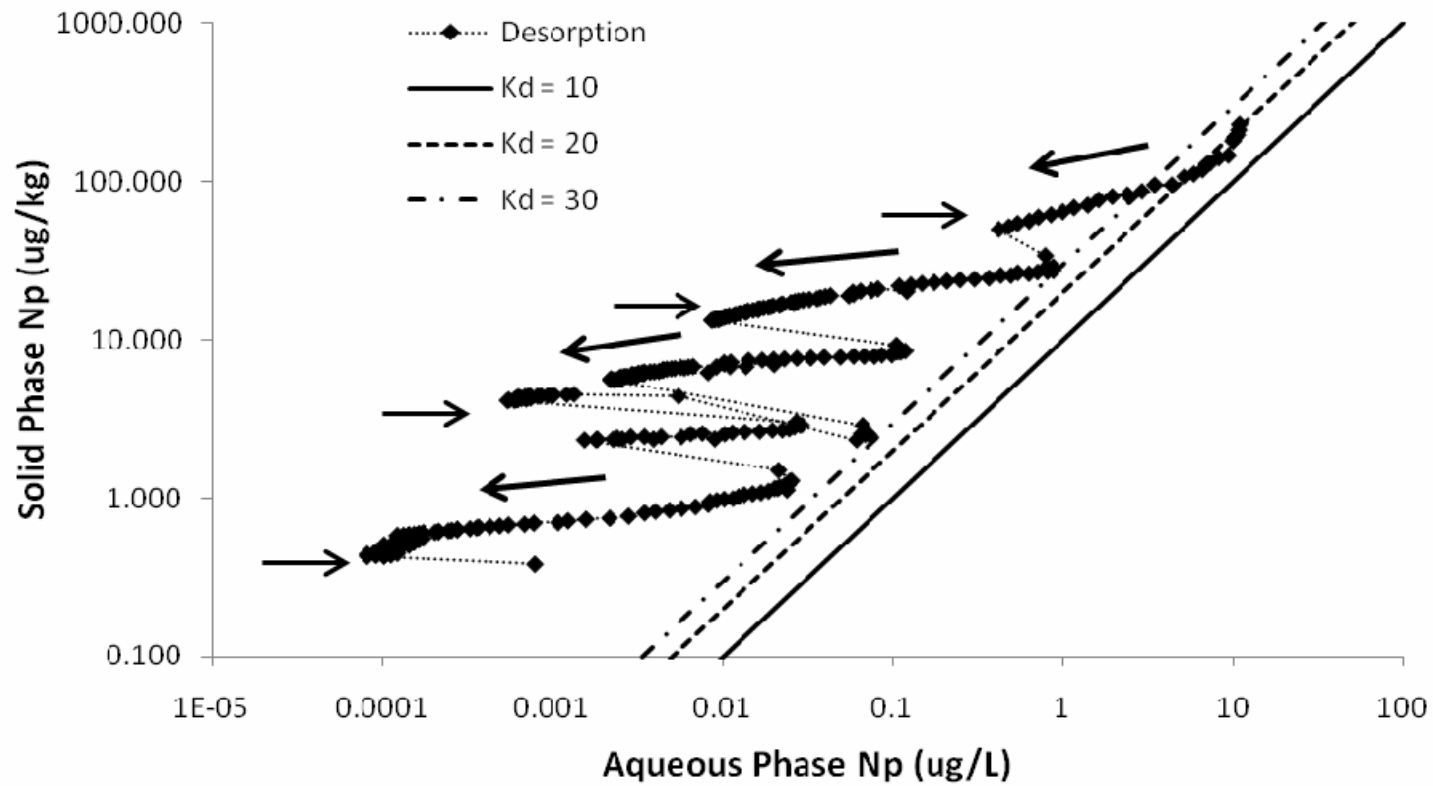


Figure 4-14: Neptunium sorption/desorption isotherm showing departure from equilibrium during the flow events and return to equilibrium during stopped flow events.

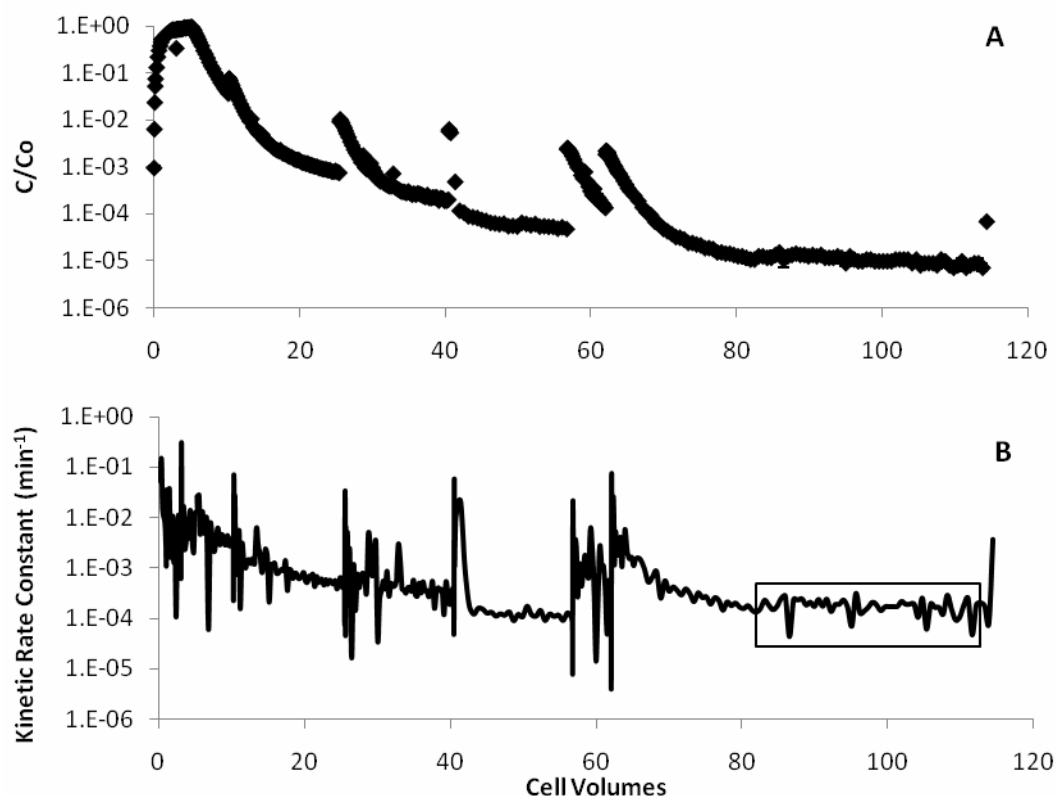


Figure 4-15: (A) Neptunium concentration relative to initial concentration as a function of cell volumes. (B) Absolute value of the kinetic rate constant for each data point as a function of cell volumes. Black box indicates steady state period where the average desorption rate kinetic was calculated to be $2.5E-4 \text{ min}^{-1}$.

5.0 Summary and Results

5.1 Summary of Strontium and Radium Experiments

Radium and strontium sorption to the sediments was found to be highly dependent upon ionic strength due to competition for ion exchange sites (Table 5-1). Radium K_d values for the clayey sediment were determined to be 185.1 ± 25.63 mL/g and 30.35 ± 0.66 mL/g for ionic strengths of 0.02M and 0.1M as NaCl. Radium K_d values for the sandy sediment were determined to be 24.95 ± 2.97 mL/g and 9.05 ± 0.36 mL/g for ionic strengths of 0.02M and 0.1M as NaCl. These values were greater than the strontium sorption K_d values which was consistent with the assumptions in the SRS PA. A more robust model than the K_d model is the cation exchange model, which would permit modeling strontium and radium in the near field and far field of the SRS PA.

Table 5-1: Summary of K_d values (mL/g) for radium and strontium experiments determined as part of this work compared to present values recommended for use in SRS PAs (Kaplan 2010).

Radionuclide	Clayey Sediment		Sandy Sediment	
	0.02N	0.1N	0.02N	0.1N
Sr (measured together with Ra)	32.06 ± 3.62	8.05 ± 0.62	5.86 ± 0.35	6.02 ± 0.14
Ra (measured together with Sr)	185.1 ± 25.63	30.35 ± 0.66	24.95 ± 2.92	9.05 ± 0.36
Ra Only	326.2 ± 33.64		34.55 ± 4.13	
Sr K_d recommended for use in SRS PAs ^a	17	-	5	-
Ra K_d recommended for use in SRS PAs ^a	17	-	5	-

^a SRS performance assessments presently assume the same K_d values for both Sr and Ra, there are measured values for the former, none for the latter. The SRS groundwater has a normality of ~0.02 N. Data from Kaplan (2010).

The effect of competing cations on the sorption of radium and strontium to sediments should be examined further. The influence of ionic strength on strontium and radium sorption shown in this work demonstrate that a single K_d approach may not be valid for the range of water chemistries expected at the SRS which are typically between 0.01 and 0.1 M. Rigorous quantification of Sr and Ra cation exchange constants which can in turn be used to predict K_d values based on groundwater chemistry would result in a more accurate prediction of radium and strontium fate in the subsurface.

5.2 Summary of Iodine Experiments

K_d values for both iodide and iodate were determined for natural end member sediments from the Savannah River Site. Under oxidizing conditions, iodide exhibited minimal sorption to both sandy and clayey soils. Equilibrium was reached by day 1 for the clayey soil, and day 4 for the sandy. In each case, the equilibrium K_d was approximately 10 mL/g. When iodide was spiked in with the wetland soil under oxidizing conditions, it experienced much greater sorption than for

either the sandy or clayey. It had an equilibrium K_d of approximately 50 mL/g, which is 5 times that of either the sandy or clayey.

When these experiments were repeated under reducing conditions, similar results were seen for the sandy and clayey soils. They each reached equilibrium by the 8 day sampling period, and each had a K_d value of around 8 mL/g. This is similar to the K_d values of 10 mL/g each had under oxidizing conditions. However, the wetland soil showed a different behavior. There was a high degree of initial sorption with K_d values of 20 and 25 mL/g at the 1 day and 4 day sampling events, respectively. By the 8th day, the K_d values dropped to 10 mL/g, which is equal to those of the sandy and clayey soils. It appears the reducing conditions altered the wetland soil in some way that hindered the iodide's ability to sorb to the soil.

Spiking iodate into these soil samples produced significantly different results than iodide. Under oxidizing conditions, there was little difference in the sandy soil. It reached equilibrium by the 1st day, and had a K_d of approximately 10 mL/g. When the clayey soil was used, there was a dramatic jump in K_d values. Steady state was approached at day 1, but not actually reached until day 4 with a K_d of about 50 mL/g. Again, the wetland soil had the highest equilibrium K_d values of any soil under oxidizing conditions. The K_d values steadily increased until day 8, where they reached 80 mL/g. Because there was a noticeable increase between each sampling period, it is not possible to confirm this was the equilibrium K_d value.

Only the clayey and wetland soils were examined for iodate sorption under reducing conditions. The iodate in the clayey systems behaved similarly under the reducing conditions as it did under oxidizing conditions. The measured K_d after 8 days was approximately 45 mL/g, which is close to the 50 mL/g from oxidizing conditions. However, there was a dramatic change with the wetland soil. Like iodide under reducing conditions, there was a high degree of initial sorption with a 1 day K_d of 40 mL/g, but this quickly reduced until an equilibrium K_d of about 8 mL/g was reached at the 8 day sampling event. This suggests that the iodate was reduced to iodide in the wetland soil. Therefore, regardless of the initial oxidation state of iodine in reducing wetland soil, an equilibrium K_d of 8-10 mL/g can be expected.

Table 5-2 shows a comparison of the equilibrium K_d values for iodide and iodate under oxidizing and reducing conditions. For the sandy soil, the oxidation state of iodine nor the redox conditions had any effect on sorption. The oxidation state seems to control the extent of sorption for the clayey soils, with significantly more sorption occurring when iodate is used as opposed to iodide. The wetland sediments appear to be affected by both the redox conditions and oxidation state. Under oxidizing conditions there is twice as much sorption for iodate compared to iodide. However, under reducing conditions, there is a dramatic decrease in sorption. For the iodate samples, it appears that under reducing conditions, the iodate is being reduced to iodide with a steady state K_d value of 7 to 10 mL/g being reached.

Table 5-2: Iodide and iodate K_d values mL/g determined after 8 days.

Soil	Iodide		Iodate	
	Oxidizing	Reducing	Oxidizing	Reducing
Sandy	5.93 ± 3.44 (n=5)	8.09 ± 3.68 (n=6)	5.95 ± 1.60 (n=9)	NA
Clayey	8.04 ± 7.21 (n=5)	3.78 ± 3.00 (n=8)	41.4 ± 4.99 (n=6)	45.3 ± 7.19 (n=6)
Wetland	72.4 ± 23.4 (n=6)	9.79 ± 2.69 (n=9)	80.3 ± 27.7 (n=5)	6.82 ± 2.72 (n=5)

K_d values represent averages and standard deviations of 9 samples after 8 days.

It would be beneficial to develop a better understanding of the specific mechanisms involved between iodine and iodate and the sediments, especially the wetland soil. More work should be done to examine the change in the wetland soil chemistry between oxidizing and reducing conditions. Because the wetland soil is high in both organic and inorganic matter, spectroscopy should be used to understand iodine binding with either inorganic or organic components of the wetland soil. It would also be beneficial to determine if the reoxidation of I^- to IO_3^- is possible by transferring the wetland soil from reducing conditions to highly oxidizing ones. If reoxidation does occur, a reaction mechanism could be determined.

5.3 Summary of Neptunium Experiments

Baseline neptunium K_d values were determined to be 9.05 ± 0.61 mL/g and 4.26 ± 0.24 mL/g for the clayey and sandy sediments, respectively. The addition of natural organic matter (NOM) to the clayey sediment resulted in an increase in the K_d value most likely due to the formation of ternary soil-NOM-Np complexes. None of the reductants nor the anaerobic atmosphere resulted in large increases in K_d values for either sediment, indicating that little to no reduction of Np(V) to Np(IV) occurred. Long term equilibration experiments (71 days) indicated that even prolonged equilibration under anoxic conditions do not facilitate reduction of Np(V) to Np(IV). Desorption K_d values were calculated under the baseline and anaerobic conditions and found to approach the sorption K_d values given a long enough equilibration period which indicated fully reversible sorption. This was further confirmed with a flowcell experiment that was able to desorb >99.9% of sorbed neptunium from the clayey sediment. Sorption and desorption rate constants were calculated to be 0.0165 min^{-1} and $2.5 \times 10^{-4} \text{ min}^{-1}$, respectively.

5.4 Summary of Recommended K_d Values

Below are recommended K_d values based on these tests and a comparison with previously recommended K_d values for SRS performance assessments (Kaplan 2010; SRNL-STI-2009-00473).

Table 5-3: Recommended K_d values based on these experiment results compared with previously recommended K_d values used in SRS performance assessments (Kaplan 2010).

Rad ^a	Recommended Values Based on this Study		Existing Geochemical Data Package SRNL-STI-2009-00473		Comment
	Sand K_d (mL/g)	Clay K_d (mL/g)	Sand K_d (mL/g)	Clay K_d (mL/g)	
Sr	5	17	5	17	No change recommended; K_d s are highly dependent on ionic strength
Ra	25	185	5	17	Ra K_d (ionic strength, ~0.02 M, which approximates that of SRS groundwater)
Np	3	9	3	9	Results from this study are included in SRNL-STI-2009-00473
I	0.3	0.9	0.3	0.9	<ul style="list-style-type: none"> - K_d: iodate >> iodide - SRS has both iodate and iodide; it was previously assumed that only iodide was present. - No change in K_ds is recommended at this time because research is on-going.
^a Barium exists between Sr and Ra in the periodic table, and based on known periodicity and first principles of chemical behavior we can assume that sorption behavior of Ba will be between that of Sr and Ra. Assuming Ba has K_d values that are midway between Sr and Ra, we may estimate Ba K_d values in SRS sediment will be 15 mL/g in sandy sediment and 101 mL/g in clayey sediment.					

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7.0 Appendix A: Radium and Strontium Sorption Data

Table 7-1: Data from radium and strontium sorption experiments.

Sample #	[Sr]o (ppb)	[Ra]o (cpm/g)	pH	[Sr] Final (ppb)	[Sr] soil (ppb)	K _d mL/g	[Ra] Final (cpm/mL)	[Ra] soil (cpm/g)	K _d mL/g
1	967.79	244.6	5.42	614.1677	14108.42	23.15179	82.04712	6400.571	78.01092
2	991.10	248.9	5.58	462.5749	21087.28	46.06272	18.91497	9093.735	480.7691
3	490.17	186.3	5.58	298.4474	8203.878	27.93595	42.11063	6021.206	142.9854
4	496.22	187.6	5.3	253.6152	9676.551	38.88738	27.14486	6274.374	231.1441
5	194.72	123.6	5.46	63.2373	5269.668	90.14559	8.411231	4453.428	529.4622
6	197.39	125.3	5.26	107.5105	3688.557	35.90517	19.50884	4122.532	211.3161
7	99.72	60.7	5.49	43.31916	2432.471	63.11687	6.248802	2163.776	346.2705
8	99.88	61.8	5.17	57.58371	1855.473	35.13907	11.34635	1987.337	175.1521
9	50.92	24.7	5.17	24.67949	1191.588	59.88036	3.113201	831.0961	266.9587
10	44.45	25.0	5.18	30.05468	760.2864	30.08096	3.521837	851.8755	241.8839
11	987.03	248.4	5.24	809.5125	7386.369	9.151824	144.5723	4261.224	29.47469
12	993.19	249.0	5.18	836.797	6302.894	7.554012	143.798	4177.201	29.04909
13	496.33	185.3	5.27	425.1006	2882.09	6.818601	102.5408	3238.153	31.57916
14	503.31	186.0	5.21	434.8615	2783.185	6.43598	105.306	3170.577	30.10822
15	199.18	123.3	5.28	175.5893	1022.325	5.903618	67.85783	2179.088	32.11255
16	197.19	123.4	5.38	122.4033	3024.031	25.20378	62.9849	2365.098	37.55024
17	101.96	61.3	5.29	89.34568	605.4381	6.965008	34.31751	1085.463	31.63
18	101.16	61.6	5.19	88.69036	590.0953	6.840069	35.91415	1019.29	28.38129
19	49.35	24.3	5.28	47.29619	173.1983	3.85947	14.13228	394.9117	27.94394
20	51.80	24.3	5.14	48.09614	239.056	5.233716	14.11446	398.226	28.21403
21	0.00	249.5	5.34	3.102883	-118.843	NA	59.6974	7268.783	121.7604
22	0.00	186.0	5.26	2.723872	-103.788	NA	21.27728	6276.707	294.9958
23	0.00	123.0	5.35	2.134819	-83.5607	NA	10.36502	4408.936	425.3669
24	0.00	60.6	5.49	1.857849	-71.7404	NA	4.655659	2159.158	463.7707
25	0.00	24.5	5.47	1.980664	-76.657	NA	1.828532	878.0397	480.1884
26	0.00	0.0	5.45	2.212132	-87.8089	NA	0.0013	-0.0516	-39.6942
27	0.00	0.0	5.45	5.058724	-198.965	NA	0.028516	-1.12155	-39.3311
28	980.76	247.5	5.59	843.6955	5601.011	6.669415	131.6287	4605.925	34.9918
29	987.70	249.3	5.52	871.6462	4825.219	5.560571	182.4742	2688.088	14.73134

Sample #	[Sr]o (ppb)	[Ra]o (cpm/g)	pH	[Sr] Final (ppb)	[Sr] soil (ppb)	K _d mL/g	[Ra] Final (cpm/mL)	[Ra] soil (cpm/g)	K _d mL/g
30	494.58	187.4	5.46	461.9696	1506.289	3.28827	116.117	2940.311	25.32196
31	497.22	186.7	5.53	432.1635	2816.021	6.575287	102.979	3421.031	33.22067
32	194.51	125.5	5.28	168.7247	1225.231	7.43309	79.66451	1892.375	23.75431
33	195.62	125.4	5.53	176.5563	955.1881	5.531989	66.78002	2438.158	36.51029
34	97.34	61.9	5.48	87.58837	557.1775	6.65697	33.93225	1143.203	33.69077
35	100.35	62.3	5.6	87.24966	689.2064	8.267864	30.33181	1294.635	42.6824
36	47.46	24.8	5.43	45.33746	250.7222	6.049158	14.61683	423.0826	28.9449
37	50.10	24.5	5.51	44.96901	378.9435	9.224748	12.95685	483.4038	37.30875
38	988.11	246.9	5.33	865.2813	5159.777	5.992276	199.7242	1916.334	9.594899
39	995.92	247.0	5.36	866.1717	5436.39	6.306997	204.842	1710.612	8.350883
40	494.38	186.0	5.37	443.8878	2136.144	4.858431	151.3088	1354.139	8.949508
41	492.18	184.8	5.31	429.9604	2670.322	6.272037	146.1693	1554.769	10.63677
42	195.73	123.9	5.24	176.8751	961.3467	5.567695	106.1075	740.9795	6.983288
43	197.25	121.8	5.37	177.0791	1019.271	5.896202	97.58632	1012.15	10.37184
44	98.65	61.2	5.26	89.39601	543.5705	6.380983	53.13978	327.4635	6.162305
45	98.97	61.6	5.25	90.51391	519.9299	6.024407	52.07426	392.5962	7.53916
46	51.35	24.3	5.41	47.9635	303.0574	6.926472	20.27228	162.3237	8.007173
47	51.37	24.4	5.45	48.06261	295.5077	6.738657	19.76146	180.3928	9.128516
48	0.00	245.5	5.63	2.899499	-118.865	NA	127.7542	4828.585	37.79591
49	0.00	184.5	5.4	2.820967	-110.873	NA	108.8819	2970.202	27.27913
50	0.00	122.3	5.46	2.925361	-115.258	NA	62.44195	2360.137	37.7973
51	0.00	60.8	5.69	3.344592	-133.56	NA	16.86183	1753.755	104.0074
52	0.00	24.3	5.62	3.008051	-117.875	NA	12.10794	476.7793	39.3774
53	0.00	0.0	5.69	2.610305	-108.019	NA	0.110692	-4.58063	-41.3817
54	0.00	0.0	5.65	-0.05619	2.280571	NA	0.443612	-18.0047	-40.5865

8.0 Appendix B: Iodine Sorption Data

8.1 Data Tables for Iodine Sorption to Natural Sediments under Oxidizing Conditions

8.1.1 *Data Tables for Sandy Sediment*

Table 8-1: Iodide 1 day Sandy Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	989.43	882.20	0.89	5.04
B	989.43	744.32	0.75	13.00
C	989.43	725.22	0.73	15.17
D	495.02	448.88	0.91	4.20
E	495.02	369.90	0.75	13.48
F	495.02	539.32	1.09	-3.34
G	94.55	87.27	0.92	3.27
H	94.55	92.64	0.98	0.82
I	94.55	72.13	0.76	12.75
No Spike A	8.57	NA	NA	NA
No Spike B	9.81	NA	NA	NA
No Spike C	10.01	NA	NA	NA

Table 8-2: Iodide 4 day Sandy Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	989.43	733.92	0.74	14.44
B	989.43	879.90	0.89	4.92
C	989.43	898.33	0.91	4.22
D	495.02	399.00	0.81	9.84
E	495.02	419.55	0.85	7.17
F	495.02	532.44	1.08	-2.85
G	94.55	9.85	0.10	336.91
H	94.55	90.87	0.96	1.61
I	94.55	79.68	0.84	7.66
No Spike A	5.38	NA	NA	NA
No Spike B	5.09	NA	NA	NA
No Spike C	7.33	NA	NA	NA

Table 8-3: Iodide 8 day Sandy Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	989.43	803.45	0.81	9.60
B	989.43	952.30	0.96	1.54
C	989.43	891.22	0.90	4.59
D	495.02	333.23	0.67	19.86
E	495.02	443.54	0.90	4.63
F	495.02	616.19	1.24	-7.99
G	94.55	1.30	0.01	2808.71
H	94.55	113.82	1.20	-6.74
I	94.55	90.16	0.95	2.00
No Spike A	6.28	NA	NA	NA
No Spike B	26.47	NA	NA	NA
No Spike C	-1.79	NA	NA	NA

Table 8-4: Iodate 1 day Sandy Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	976.2634	912.7058	0.934897	2.886027
B	978.5878	869.4016	0.888425	5.05035
C	982.0745	882.5475	0.898656	4.672555
D	481.1584	444.7884	0.924412	3.208962
E	481.1584	424.4509	0.882144	5.255035
F	479.4151	438.4474	0.914547	3.870304
G	98.20745	86.13671	0.877089	5.59664
H	95.3019	78.55848	0.824312	8.449937
I	98.78856	86.21404	0.872713	6.012749
No Spike A	-	17.09743	NA	NA
No Spike B	-	10.07596	NA	NA
No Spike C	-	32.47042	NA	NA

Table 8-5: Iodate 4 day Sandy Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	976.2634	881.149	0.902573	4.473637
B	978.5878	869.0302	0.888045	5.069696
C	982.0745	906.9015	0.923455	3.434421
D	481.1584	425.2543	0.883814	5.159045
E	481.1584	412.984	0.858312	6.493074
F	479.4151	441.236	0.920363	3.584064
G	98.20745	56.53933	0.575713	29.43305
H	95.3019	73.35418	0.769703	11.86224
I	98.78856	82.21607	0.832243	8.309807
No Spike A	-	16.72397	NA	NA
No Spike B	-	8.945201	NA	NA
No Spike C	-	43.98373	NA	NA

Table 8-6: Iodate 8 day Sandy Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	976.2634	854.9915	0.87578	5.87844
B	978.5878	846.5002	0.865022	6.274933
C	982.0745	861.939	0.877672	5.774928
D	481.1584	417.6883	0.868089	5.963364
E	481.1584	406.1864	0.844184	7.259988
F	479.4151	442.7763	0.923576	3.427502
G	98.20745	62.13334	0.632674	23.18744
H	95.3019	78.26686	0.821252	8.629139
I	98.78856	89.30558	0.904007	4.377496
No Spike A	-	20.77287	NA	NA
No Spike B	-	12.28154	NA	NA
No Spike C	-	NA	NA	NA

8.1.2 Data Tables for Clayey Sediment

Table 8-7: Iodide 1 day Clayey Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	989.43	882.20	0.89	5.04
B	989.43	744.32	0.75	13.00
C	989.43	725.22	0.73	15.17
D	495.02	448.88	0.91	4.20
E	495.02	369.90	0.75	13.48
F	495.02	539.32	1.09	-3.34
G	94.55	87.27	0.92	3.27
H	94.55	92.64	0.98	0.82
I	94.55	72.13	0.76	12.75
No Spike A	-	8.57	NA	NA
No Spike B	-	9.81	NA	NA
No Spike C	-	10.01	NA	NA

Table 8-8: Iodide 4 day Clayey Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	989.43	733.92	0.74	14.43881
B	989.43	879.90	0.89	4.9154
C	989.43	898.33	0.91	4.221467
D	495.02	399.00	0.81	9.842039
E	495.02	419.55	0.85	7.170036
F	495.02	532.44	1.08	-2.85414
G	94.55	9.85	0.10	336.9098
H	94.55	90.87	0.96	1.609739
I	94.55	79.68	0.84	7.661719
No Spike A	-	5.38	NA	NA
No Spike B	-	5.09	NA	NA
No Spike C	-	7.33	NA	NA

Table 8-9: Iodide 8 day Clayey Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	989.43	803.45	0.81	9.60
B	989.43	952.30	0.96	1.54
C	989.43	891.22	0.90	4.59
D	495.02	333.23	0.67	19.86
E	495.02	443.54	0.90	4.63
F	495.02	616.19	1.24	-7.99
G	94.55	1.30	0.01	2808.71
H	94.55	113.82	1.20	-6.74
I	94.55	90.16	0.95	2.00
No Spike A	-	6.28	NA	NA
No Spike B	-	26.47	NA	NA
No Spike C	-	-1.79	NA	NA

Table 8-10: Iodate 1 day Clayey Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	982.0745	530.1518	0.539828	33.79046
B	983.8178	551.1853	0.560251	31.99989
C	979.1689	553.3505	0.565123	30.8541
D	479.4151	243.5707	0.508058	39.67223
E	481.1584	268.084	0.557164	32.01223
F	480.5773	245.9679	0.511818	37.55343
G	97.62634	15.38073	0.157547	209.1124
H	92.39636	20.70096	0.224045	139.5125
I	96.46412	32.61735	0.338129	7.970924
No Spike A		42.15201	NA	NA
No Spike B		16.20041	NA	NA
No Spike C		45.10597	NA	NA

Table 8-11: Iodate 4 day Clayey Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	982.0745	511.2966	0.520629	36.49835
B	983.8178	525.2333	0.533872	35.59541
C	979.1689	531.6714	0.542982	33.74707
D	479.4151	227.2619	0.47404	45.45943
E	481.1584	232.7153	0.483656	42.99889
F	480.5773	222.8688	0.463752	45.52628
G	97.62634	9.145919	0.093683	378.3239
H	92.39636	64.79401	0.701261	17.16024
I	96.46412	23.71879	0.245882	12.48909
No Spike A	-	50.73997	NA	NA
No Spike B	-	28.72917	NA	NA
No Spike C	-	48.1117	NA	NA

Table 8-12: Iodate 8 day Clayey Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	982.0745	502.0471	0.511211	37.90108
B	983.8178	507.6823	0.516033	38.23538
C	979.1689	516.5596	0.527549	35.9073
D	479.4151	217.5106	0.4537	49.33428
E	481.1584	233.6441	0.485587	42.66788
F	480.5773	226.465	0.471235	44.17811
G	97.62634	17.60932	0.180375	177.6985
H	92.39636	27.76803	0.300532	93.75404
I	96.46412	43.12961	0.447105	5.035599
No Spike A	-	80.5132	NA	NA
No Spike B	-	-	NA	NA
No Spike C	-	56.98451	NA	NA

8.1.3 Data Tables for Wetland Sediment

Table 8-13: Iodide 1 day Wetland Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	952.26	585.97	0.62	26.46
B	952.24	487.27	0.51	40.35
C	952.29	527.41	0.55	34.14
D	488.26	215.28	0.44	53.93
E	488.21	272.46	0.56	33.55
F	488.23	280.93	0.58	31.30
G	97.85	38.31	0.39	64.39
H	97.92	43.54	0.44	53.03
I	97.89	12.66	0.13	282.81
No Spike A	-	6.843498	NA	NA
No Spike B	-	12.11982	NA	NA
No Spike C	-	15.67949	NA	NA

Table 8-14: Iodide 4 day Wetland Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	952.26	463.72	0.49	44.59
B	952.24	421.39	0.44	53.27
C	952.29	487.73	0.51	40.36
D	488.26	208.74	0.43	56.96
E	488.21	166.18	0.34	82.09
F	488.23	163.71	0.34	84.08
G	97.85	17.92	0.18	184.78
H	97.92	27.46	0.28	108.94
I	97.89	28.80	0.29	100.81
No Spike A	-	10.59795	NA	NA
No Spike B	-	10.62104	NA	NA
No Spike C	-	58.31568	NA	NA

Table 8-15: Iodide 8 day Wetland Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	952.26	407.10	0.43	56.68
B	952.24	434.06	0.46	50.48
C	952.29	521.44	0.55	35.01
D	488.26	153.98	0.32	92.34
E	488.21	257.14	0.53	38.07
F	488.23	239.76	0.49	43.96
G	97.85	99.61	1.02	-0.73
H	97.92	100.99	1.03	-1.29
I	97.89	100.68	1.03	-1.16
No Spike A	-	120.19	NA	NA
No Spike B	-	84.71	NA	NA
No Spike C	-	-2.29	NA	NA

Table 8-16: Iodate 1 day Wetland Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1147.632	773.2083	0.673742	20.4967
B	1147.811	525.5291	0.457853	50.4305
C	1147.42	580.5031	0.50592	41.0368
D	543.0128	280.998	0.51748	38.88809
E	543.0981	238.9247	0.439929	53.42176
F	543.0622	262.8321	0.483982	44.62422
No Spike A	-	11.9349	NA	NA
No Spike B	-	10.47097	NA	NA
No Spike C	-	14.12309	NA	NA

Table 8-17: Iodate 4 day Wetland Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1147.632	1667.059	1.452607	-13.1883
B	1147.811	954.6366	0.831702	8.618165
C	1147.42	900.9985	0.785238	11.49247
D	543.0128	447.4822	0.824073	8.903505
E	543.0981	458.3814	0.844012	7.755328
F	543.0622	455.0511	0.837936	8.094903
No Spike A	-	25.40584	NA	NA
No Spike B	-	24.11284	NA	NA
No Spike C	-	102.9779	NA	NA

Table 8-18: Iodate 8 day Wetland Sediment, Oxidizing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1147.632	1786.242	1.556459	-15.1325
B	1147.811	1033.811	0.90068	4.696421
C	1147.42	920.4051	0.802152	10.36417
D	543.0128	451.6583	0.831764	8.435561
E	543.0981	466.2475	0.858496	6.916542
F	543.0622	499.0634	0.91898	3.689942
No Spike A	-	37.53084	NA	NA
No Spike B	-	32.4315	NA	NA
No Spike C	-	31.94767	NA	NA

8.1.4 Data Tables for No-Solids Controls

Table 8-19: Iodate 1 day No-Solids Controls, Oxidizing

Sample ID	Initial Aq. Conc. (ppb)	Equil. Aq. Conc. (ppb)	Fraction Aq
A	981.4456	975.3759	0.993816
B	982.6225	981.5382	0.998897
C	980.3578	1012.35	1.032633
D	97.5239	101.8619	1.044482
E	96.11832	95.4685	0.993239
F	98.4629	99.62809	1.011834
No Spike A	-	7.772271	NA
No Spike B	-	4.83976	NA
No Spike C	-	7.636699	NA

Table 8-20: Iodate 4 day No-Solids Controls, Oxidizing

Sample ID	Initial Aq. Conc. (ppb)	Equil. Aq. Conc. (ppb)	Fraction Aq
A	981.4456	982.9123	1.0014944
B	982.6225	984.4555	1.0018654
C	980.3578	940.4741	0.9593172
D	97.5239	93.1772	0.9554294
E	96.11832	90.09079	0.9372905
F	98.4629	97.18953	0.9870675
No Spike A	-	8.15584	NA
No Spike B	-	4.70369	NA
No Spike C	-	8.302445	NA

Table 8-21: Iodate 8 day No-Solids Controls

Sample ID	Initial Aq. Conc. (ppb)	Equil. Aq. Conc. (ppb)	Fraction Aq
A	981.4456	1005.094	1.024096
B	982.6225	1006.585	1.024386
C	980.3578	1008.81	1.029022
D	97.5239	99.73406	1.022663
E	96.11832	94.43032	0.982438
F	98.4629	98.83888	1.003818
No Spike A	-	10.44137	NA
No Spike B	-	8.120995	NA
No Spike C	-	14.18115	NA

8.2 Data Tables for Iodine Sorption to Natural Sediments under Reducing Conditions

8.2.1 Data Tables for Sandy Sediments

Table 8-22: Iodide 1 day Sandy Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1082.96	1065.68	0.98	0.64
B	1082.96	1110.69	1.03	-1.01
C	1082.96	1042.48	0.96	1.60
D	543.97	562.81	1.03	-1.36
E	543.97	535.39	0.98	0.64
F	543.97	551.35	1.01	-0.53
G	107.12	97.97	0.91	3.64
H	107.12	111.61	1.04	-1.60
I	107.12	114.49	1.07	-2.58
No Spike A	-	124.73	NA	NA
No Spike B	-	125.93	NA	NA
No Spike C	-	123.47	NA	NA

Table 8-23: Iodide 4 day Sandy Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1082.96	992.91	0.92	3.59
B	1082.96	1027.39	0.95	2.19
C	1082.96	1011.91	0.93	2.89
D	543.97	443.88	0.82	9.18
E	543.97	440.64	0.81	9.30
F	543.97	403.69	0.74	13.71
G	107.12	-21.32	-0.20	-234.52
H	107.12	-16.89	-0.16	-292.16
I	107.12	-14.57	-0.14	-334.29
No Spike A	-	-0.57	NA	NA
No Spike B	-	-1.16	NA	NA
No Spike C	-	621.38	NA	NA

Table 8-24: Iodide 8 day Sandy Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1082.96	958.04	0.88	5.17
B	1082.96	972.04	0.90	4.63
C	1082.96	976.24	0.90	4.49
D	543.97	425.70	0.78	11.31
E	543.97	417.15	0.77	12.06
F	543.97	426.05	0.78	10.92
G	107.12	-15.39	-0.14	-310.01
H	107.12	-15.53	-0.14	-314.34
I	107.12	-15.67	-0.15	-313.67
No Spike A	-	0.36	NA	NA
No Spike B	-	-0.07	NA	NA
No Spike C	-	-1.19	NA	NA

8.2.2 Data Tables for Clayey Sediment

Table 8-25: Iodide 1 day Clayey Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1008.01	1105.24	1.10	-3.62
B	1008.01	886.74	0.88	5.49
C	1008.01	793.40	0.79	11.06
D	504.32	871.18	1.73	-16.94
E	504.32	539.05	1.07	-2.62
F	504.32	469.33	0.93	3.05
G	96.32	96.69	1.00	-0.15
H	96.32	97.82	1.02	-0.62
I	96.32	106.80	1.11	-3.87
No Spike A	-	8.20	NA	NA
No Spike B	-	8.90	NA	NA
No Spike C	-	8.29	NA	NA

Table 8-26: Iodide 4 day Clayey Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1008.01	704.68	0.70	17.73215
B	1008.01	772.00	0.77	12.26273
C	1008.01	833.66	0.83	8.554869
D	504.32	886.81	1.76	-17.3521
E	504.32	482.23	0.96	1.864089
F	504.32	388.54	0.77	12.20718
G	96.32	90.26	0.94	2.704267
H	96.32	98.06	1.02	-0.71445
I	96.32	79.49	0.83	8.346071
No Spike A	-	10.24	NA	NA
No Spike B	-	9.37	NA	NA
No Spike C	-	9.37	NA	NA

Table 8-27: Iodide 8 day Clayey Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1008.01	0.94	2332.38	2.45
B	1008.01	0.93	2721.78	2.90
C	1008.01	0.90	4297.72	4.76
D	504.32	2.40	-28437.76	-23.48
E	504.32	0.89	2173.31	4.82
F	504.32	0.96	890.77	1.85
G	96.32	0.94	220.22	2.42
H	96.32	0.80	793.82	10.35
I	96.32	0.98	66.09	0.70
No Spike A	-	34.36	NA	NA
No Spike B	-	21.44	NA	NA
No Spike C	-	30.57	NA	NA

Table 8-28: Iodate 1 day Clayey Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1193.2	664.7293	0.557098	31.97579
B	1193.2	659.2334	0.552492	33.36228
C	1193.2	657.3759	0.550935	33.82536
D	560.6345	282.1416	0.503254	40.35747
E	560.6345	274.6436	0.48988	41.45062
F	560.6345	281.7404	0.502538	40.05198
No Spike A	-	8.202494	NA	NA
No Spike B	-	8.895463	NA	NA
No Spike C	-	8.287347	NA	NA

Table 8-29: Iodate 4 day Clayey Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1193.2	661.8256	0.554664	32.29255
B	1193.2	630.8946	0.528742	36.71101
C	1193.2	663.1401	0.555766	33.17062
D	560.6345	269.8751	0.481374	44.05018
E	560.6345	265.2083	0.47305	44.34149
F	560.6345	265.6607	0.473857	44.92517
No Spike A	-	10.24026	NA	NA
No Spike B	-	9.368601	NA	NA
No Spike C	-	9.368601	NA	NA

Table 8-30: Iodate 8 day Clayey Sediments, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1193.2	637.6626	0.534414	35.04028
B	1193.2	608.4773	0.509954	39.58096
C	1193.2	585.3439	0.490567	43.09468
D	560.6345	249.2877	0.444653	51.06465
E	560.6345	246.7233	0.440079	50.64598
F	560.6345	243.6284	0.434558	52.64698
No Spike A	-	34.35532	NA	NA
No Spike B	-	21.43609	NA	NA
No Spike C	-	30.57083	NA	NA

8.2.3 Data Tables for Wetland Sediment

Table 8-31: Iodide 1 day Wetland Sediment, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	967.00	673.02	0.70	18.50
B	967.22	740.67	0.77	13.08
C	967.11	1022.59	1.06	-2.31
D	495.68	316.75	0.64	23.77
E	495.56	357.66	0.72	16.08
F	495.63	368.37	0.74	14.48
G	99.43	60.97	0.61	26.75
H	99.42	67.65	0.68	19.83
I	99.40	64.81	0.65	22.37
No Spike A	-	11.93	NA	NA
No Spike B	-	10.47	NA	NA
No Spike C	-	14.12	NA	NA

Table 8-32: Iodide 4 day Wetland Sediment, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	967.00	692.55	0.72	16.79
B	967.22	638.90	0.66	21.97
C	967.11	650.99	0.67	20.66
D	495.68	321.27	0.65	22.85
E	495.56	NA	NA	NA
F	495.63	657.68	1.33	-10.33
G	99.43	58.52	0.59	29.64
H	99.42	58.98	0.59	28.95
I	99.40	143.33	1.44	-12.85
No Spike A	-	25.41	NA	NA
No Spike B	-	24.11	NA	NA
No Spike C	-	102.98	NA	NA

Table 8-33: Iodide 8 day Wetland Sediment, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	967.00	777.01	0.80	10.36
B	967.22	770.10	0.80	10.94
C	967.11	790.83	0.82	9.48
D	495.68	400.70	0.81	9.98
E	495.56	422.59	0.85	7.20
F	495.63	359.54	0.73	15.86
G	99.43	84.99	0.85	7.20
H	99.42	84.61	0.85	7.39
I	99.40	80.77	0.81	9.67
No Spike A	-	37.53	NA	NA
No Spike B	-	32.43	NA	NA
No Spike C	-	31.95	NA	NA

Table 8-34: Iodate 1 day Wetland Sediment, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1147.632	773.2083	0.673742	20.4967
B	1147.811	525.5291	0.457853	50.4305
C	1147.42	580.5031	0.50592	41.0368
D	543.0128	280.998	0.51748	38.88809
E	543.0981	238.9247	0.439929	53.42176
F	543.0622	262.8321	0.483982	44.62422
No Spike A	-	11.9349	NA	NA
No Spike B	-	10.47097	NA	NA
No Spike C	-	14.12309	NA	NA

Table 8-35: Iodate 4 day Wetland Sediment, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc. (ppb)	Frac. Aq.	K _d
A	1147.632	1667.059	1.452607	-13.1883
B	1147.811	954.6366	0.831702	8.618165
C	1147.42	900.9985	0.785238	11.49247
D	543.0128	447.4822	0.824073	8.903505
E	543.0981	458.3814	0.844012	7.755328
F	543.0622	455.0511	0.837936	8.094903
No Spike A	-	25.40584	NA	NA
No Spike B	-	24.11284	NA	NA
No Spike C	-	102.9779	NA	NA

Table 8-36: Iodate 8 day Wetland Sediment, Reducing

Sample	Initial Conc. (ppb)	Equil. Aq. Conc (ppb)	Frac. Aq.	K _d
A	1147.632	1786.242	1.556459	-15.1325
B	1147.811	1033.811	0.90068	4.696421
C	1147.42	920.4051	0.802152	10.36417
D	543.0128	451.6583	0.831764	8.435561
E	543.0981	466.2475	0.858496	6.916542
F	543.0622	499.0634	0.91898	3.689942
No Spike A	-	37.53084	NA	NA
No Spike B	-	32.4315	NA	NA
No Spike C	-	31.94767	NA	NA

8.2.4 Data Tables for No-Solids Controls

Table 8-37: Iodate 1 day No-Solids Controls, Reducing

Sample ID	Initial Aq. Conc. (ppb)	Equil. Aq. Conc. (ppb)	Fraction Aq
A	1048.541	1005.102	0.958572
B	1048.541	995.1827	0.949112
C	1048.541	946.7963	0.902965
D	524.4306	499.8426	0.953115
E	524.4306	488.7721	0.932005
F	524.4306	457.8073	0.872961
No Spike A	-	57.29749	NA
No Spike B	-	59.48766	NA
No Spike C	-	NA	NA

Table 8-38: Iodate 4 day No-Solids Controls, Reducing

Sample ID	Initial Aq. Conc. (ppb)	Equil. Aq. Conc. (ppb)	Fraction Aq
A	1048.541	1008.182	0.9615096
B	1048.541	990.4855	0.9446321
C	1048.541	982.8139	0.9373157
D	524.4306	478.2518	0.9119448
E	524.4306	486.7246	0.928101
F	524.4306	447.2751	0.8528775
No Spike A	-	57.58494	NA
No Spike B	-	57.22211	NA
No Spike C	-	NA	NA

Table 8-39: Iodate 8 day No-Solids Controls, Reducing

Sample ID	Initial Aq. Conc. (ppb)	Equil. Aq. Conc. (ppb)	Fraction Aq
A	1048.541	972.7033	0.927673
B	1048.541	961.6051	0.917089
C	1048.541	961.6818	0.917162
D	524.4306	465.8763	0.888347
E	524.4306	471.916	0.899863
F	524.4306	433.7992	0.827181
No Spike A	-	57.10949	NA
No Spike B	-	57.15117	NA
No Spike C	-	NA	NA

9.0 Appendix C: Neptunium Sorption and Flowcell Data

9.1 Neptunium Baseline Sorption Experiments

Table 9-1: Clayey Centrifugal Data from Baseline Sorption

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Mass Liq	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
C-1-C	0.09	0.2550	0.09	9.86	0.17	1.92	5%	5.53
C-2-C	0.56	0.2555	0.49	9.93	2.91	5.98	13%	5.50
C-3-C	0.57	0.2432	0.43	9.92	5.72	13.17	24%	5.48
C-4-C	1.02	0.2519	0.75	9.99	10.72	14.37	27%	5.49
C-5-C	1.01	0.2479	0.79	9.95	8.96	11.38	22%	5.51
C-6-C	1.99	0.2415	1.58	9.98	16.88	10.70	21%	5.51
C-7-C	1.97	0.2828	1.52	9.99	16.17	10.66	23%	5.51
C-8-C	4.90	0.2684	3.98	9.98	34.33	8.62	19%	5.51
C-9-C	4.81	0.2523	3.66	10.11	45.92	12.54	24%	5.53
C-10-C	10.14	0.2567	7.83	10.08	90.58	11.57	23%	5.49
C-11-C	10.02	0.2545	8.09	10.18	77.23	9.55	19%	5.49
C-12-C	14.78	0.2425	12.04	10.03	113.14	9.39	19%	5.49
C-13-C	14.56	0.2459	11.43	10.15	129.07	11.29	21%	5.50
C-14-C	19.74	0.2606	14.89	10.10	187.84	12.62	25%	5.49
C-15-C	19.75	0.2544	15.87	10.02	152.89	9.64	20%	5.49
C-16-C	27.81	0.2613	23.46	10.00	166.23	7.08	16%	5.49
C-17-C	40.30	0.2538	28.94	9.93	444.34	15.35	28%	5.50
C-18-C	39.88	0.2591	32.76	9.94	273.28	8.34	18%	5.50
C-19-C	49.81	0.2640	41.29	9.95	321.21	7.78	17%	5.51
C-20-C	49.10	0.2536	41.36	10.05	306.75	7.42	16%	5.52

Table 9-2: Clayey Filtrate Data from Baseline Sorption

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Mass Liq	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
C-1-F	0.09	0.2550	0.09	9.86	0.13	1.50	4%	5.53
C-2-F	0.56	0.2555	0.47	9.93	3.61	7.69	17%	5.50
C-3-F	0.57	0.2432	0.40	9.92	6.95	17.22	30%	5.48
C-4-F	1.02	0.2519	0.80	9.99	8.59	10.74	21%	5.49
C-5-F	1.01	0.2479	0.77	9.95	9.80	12.79	24%	5.51
C-6-F	1.99	0.2415	1.58	9.98	16.91	10.72	21%	5.51
C-7-F	1.97	0.2828	1.51	9.99	16.23	10.71	23%	5.51
C-8-F	4.90	0.2684	3.76	9.98	42.40	11.27	23%	5.51
C-9-F	4.81	0.2523	3.67	10.11	45.57	12.41	24%	5.53
C-10-F	10.14	0.2567	7.90	10.08	87.85	11.12	22%	5.49
C-11-F	10.02	0.2545	7.77	10.18	89.97	11.58	22%	5.49
C-12-F	14.78	0.2425	11.38	10.03	140.69	12.37	23%	5.49
C-13-F	14.56	0.2459	11.53	10.15	125.01	10.84	21%	5.50
C-14-F	19.74	0.2606	14.64	10.10	197.37	13.48	26%	5.49
C-15-F	19.75	0.2544	15.69	10.02	159.98	10.20	21%	5.49
C-16-F	27.81	0.2613	21.84	10.00	228.30	10.45	21%	5.49
C-17-F	40.30	0.2538	32.05	9.93	322.61	10.07	20%	5.50
C-18-F	39.88	0.2591	30.65	9.94	354.19	11.56	23%	5.50
C-19-F	49.81	0.2640	40.41	9.95	354.33	8.77	19%	5.51
C-20-F	49.10	0.2536	39.84	10.05	367.00	9.21	19%	5.52

Table 9-3: Sandy Centrifugal Data from Baseline Sorption

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Mass Liq	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
S-1-C	0.10	0.2511	0.13	9.30	-1.03	-8.03	-28%	5.48
S-2-C	0.58	0.2511	0.55	9.78	1.26	2.30	6%	5.49
S-3-C	0.58	0.2469	0.54	9.68	1.62	3.01	7%	5.50
S-4-C	1.05	0.2608	0.93	9.73	4.41	4.74	11%	5.54
S-5-C	1.04	0.2595	0.96	9.76	2.81	2.91	7%	5.52
S-6-C	2.04	0.2570	1.91	9.71	4.82	2.53	6%	5.49
S-7-C	2.04	0.2447	1.78	9.67	10.37	5.84	13%	5.49
S-8-C	4.97	0.2563	4.62	9.84	13.57	2.94	7%	5.53
S-9-C	4.94	0.2434	4.68	9.87	10.69	2.28	5%	5.48
S-10-C	10.36	0.2477	8.83	9.67	59.84	6.78	15%	5.51
S-11-C	10.13	0.2616	9.23	9.87	33.86	3.67	9%	5.50
S-12-C	14.78	0.2433	13.70	10.11	44.89	3.28	7%	5.55
S-13-C	15.29	0.2556	13.82	9.80	56.48	4.09	10%	5.45
S-14-C	19.89	0.2531	17.66	9.98	87.88	4.98	11%	5.47
S-15-C	20.04	0.2541	17.97	9.99	81.28	4.52	10%	5.45
S-16-C	29.79	0.2475	27.88	10.01	76.85	2.76	6%	5.54
S-17-C	39.73	0.2544	35.74	9.97	156.42	4.38	10%	5.54
S-18-C	38.48	0.2555	33.51	10.25	199.60	5.96	13%	5.47
S-19-C	49.95	0.2510	46.23	9.86	145.88	3.16	7%	5.52
S-20-C	49.96	0.2433	44.56	9.84	218.47	4.90	11%	5.46

Table 9-4: Sandy Filtrate Data from Baseline Sorption

Sample	[Np] _o	Mass Soil	[Np] _{aq}	Mass Liq	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
S-1-F	0.10	0.2511	0.12	9.30	-0.70	-5.82	-19%	5.48
S-2-F	0.58	0.2511	0.50	9.78	3.14	6.27	14%	5.49
S-3-F	0.58	0.2469	0.52	9.68	2.52	4.88	11%	5.50
S-4-F	1.05	0.2608	0.91	9.73	5.04	5.53	13%	5.54
S-5-F	1.04	0.2595	0.89	9.76	5.46	6.12	14%	5.52
S-6-F	2.04	0.2570	1.82	9.71	8.17	4.49	11%	5.49
S-7-F	2.04	0.2447	1.80	9.67	9.25	5.13	11%	5.49
S-8-F	4.97	0.2563	4.23	9.84	28.39	6.71	15%	5.53
S-9-F	4.94	0.2434	4.34	9.87	24.52	5.65	12%	5.48
S-10-F	10.36	0.2477	8.68	9.67	65.53	7.55	16%	5.51
S-11-F	10.13	0.2616	8.50	9.87	61.27	7.21	16%	5.50
S-12-F	14.78	0.2433	12.98	10.11	74.68	5.75	12%	5.55
S-13-F	15.29	0.2556	13.43	9.80	71.53	5.33	12%	5.45
S-14-F	19.89	0.2531	16.85	9.98	119.57	7.10	15%	5.47
S-15-F	20.04	0.2541	17.13	9.99	114.40	6.68	15%	5.45
S-16-F	29.79	0.2475	26.07	10.01	150.22	5.76	12%	5.54
S-17-F	39.73	0.2544	34.84	9.97	191.45	5.49	12%	5.54
S-18-F	38.48	0.2555	34.73	10.25	150.66	4.34	10%	5.47
S-19-F	49.95	0.2510	43.87	9.86	238.65	5.44	12%	5.52
S-20-F	49.96	0.2433	44.57	9.84	218.20	4.90	11%	5.46

Table 9-5: Blank Sample Data from Baseline Sorption

Sample	[Np] _o	[Np] _{aq}	Mass Liq	% Sorbed	pH
	ppb	ppb	g		
B-10-1-C	9.42	9.13	10.79	3%	5.52
B-10-2-C	10.62	10.48	10.13	1%	5.48
B-50-C	47.93	47.69	10.41	0%	5.50
B-10-1-F	9.42	8.67	10.79	8%	5.52
B-10-2-F	10.62	9.64	10.13	9%	5.48
B-50-F	47.93	45.97	10.41	4%	5.50

Table 9-6: Neptunium-NOM Clayey Soil Centrifuged Data

Sample	[Np] _o	Mass Soil	[Np] _{aq}	Liq Mass	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
C-1-C	0.09	0.24	0.08	9.92	0.41	5.00	11%	5.25
C-2-C	0.46	0.26	0.52	9.84	-2.24	-4.28	-13%	3.05
C-3-C	0.95	0.26	0.98	9.90	-1.14	-1.17	-3%	4.56
C-4-C	0.94	0.25	0.94	9.97	0.10	0.11	0%	4.79
C-5-C	1.82	0.26	1.61	10.12	7.95	4.93	11%	5.15
C-6-C	4.57	0.27	4.38	10.10	6.99	1.60	4%	4.98
C-7-C	4.46	0.26	4.31	10.08	5.77	1.34	3%	5.08
C-8-C	10.29	0.26	8.21	10.17	80.04	9.75	20%	5.01
C-9-C	20.06	0.26	14.37	10.53	233.61	16.26	28%	5.26

Table 9-7: Neptunium-NOM Clayey Soil Filtrate Data

Sample	[Np] _o	Mass Soil	[Np] _{aq}	Liq Mass	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
C-1-F	0.09	0.24	0.07	9.92	0.76	10.37	20%	5.25
C-2-F	0.46	0.26	0.52	9.84	-2.05	-3.96	-12%	3.05
C-3-F	0.95	0.26	0.95	9.90	-0.27	-0.29	-1%	4.56
C-4-F	0.94	0.25	0.92	9.97	1.02	1.11	3%	4.79
C-5-F	1.82	0.26	1.58	10.12	8.99	5.68	13%	5.15
C-6-F	4.57	0.27	4.28	10.10	11.03	2.58	6%	4.98
C-7-F	4.46	0.26	4.10	10.08	14.06	3.43	8%	5.08
C-8-F	10.29	0.26	7.99	10.17	88.55	11.09	22%	5.01
C-9-F	20.06	0.26	13.44	10.53	271.87	20.23	33%	5.26

Table 9-8: Neptunium-NOM Sandy Soil Centrifuged Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Liq Mass	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
S-1-C	0.09	0.26	0.07	9.73	0.56	7.52	17%	5.69
S-2-C	0.45	0.27	0.41	10.09	1.53	3.75	9%	5.70
S-3-C	0.91	0.26	0.73	9.94	6.86	9.37	20%	5.74
S-4-C	0.93	0.26	0.89	9.87	1.57	1.76	5%	5.55
S-5-C	1.84	0.26	1.47	9.94	14.25	9.66	20%	5.68
S-6-C	4.48	0.26	4.17	10.07	12.12	2.91	7%	5.66
S-7-C	4.55	0.26	4.47	9.99	3.18	0.71	2%	5.43
S-8-C	10.18	0.26	8.36	10.27	72.03	8.62	18%	5.68
S-9-C	20.21	0.25	13.06	10.40	292.03	22.36	35%	5.91

Table 9-9: Neptunium-NOM Sandy Soil Filtrate Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Liq Mass	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
S-1-F	0.09	0.26	0.07	9.73	0.68	9.45	20%	5.69
S-2-F	0.45	0.27	0.40	10.09	1.92	4.85	11%	5.70
S-3-F	0.91	0.26	0.70	9.94	8.10	11.58	23%	5.74
S-4-F	0.93	0.26	0.85	9.87	3.12	3.67	9%	5.55
S-5-F	1.84	0.26	1.91	9.94	-2.84	-1.48	-4%	5.68
S-6-F	4.48	0.26	4.02	10.07	17.82	4.43	10%	5.66
S-7-F	4.55	0.26	4.28	9.99	10.30	2.41	6%	5.43
S-8-F	10.18	0.26	8.03	10.27	84.92	10.57	21%	5.68
S-9-F	20.21	0.25	12.67	10.40	308.06	24.32	37%	5.91

Table 9-10: Neptunium-Varying NOM Clayey Soil Centrifuged Data

Sample	[Np] _o	[NOM] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	mg L ⁻¹	g	ppb	ppb	L kg ⁻¹	
C-1-C	10.90	0.00	0.23	6.58	182.47	27.74	5.55
C-2-C	11.35	4.92	0.24	7.09	167.20	23.58	5.45
C-3-C	11.36	4.99	0.26	6.59	177.20	26.90	5.60
C-4-C	10.60	9.62	0.25	6.12	184.05	30.06	5.49
C-5-C	11.08	9.73	0.24	6.24	192.29	30.82	5.44
C-6-C	11.16	14.97	0.24	7.01	165.90	23.67	5.49
C-7-C	11.25	15.20	0.27	6.95	152.02	21.88	5.66
C-8-C	11.24	20.44	0.25	6.94	166.01	23.93	5.69

Table 9-11: Neptunium-Varying NOM Clayey Soil Filtrate Data

Sample	[Np] _o	[NOM] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	mg L ⁻¹	g	ppb	ppb	L kg ⁻¹	
C-1-F	10.90	0.00	0.23	6.38	190.90	29.93	5.55
C-2-F	11.35	4.92	0.24	6.88	175.41	25.49	5.45
C-3-F	11.36	4.99	0.26	6.30	187.97	29.85	5.60
C-4-F	10.60	9.62	0.25	5.81	196.73	33.84	5.49
C-5-F	11.08	9.73	0.24	5.95	203.72	34.23	5.44
C-6-F	11.16	14.97	0.24	6.84	172.65	25.24	5.49
C-7-F	11.25	15.20	0.27	6.79	157.60	23.21	5.66
C-8-F	11.24	20.44	0.25	6.60	179.05	27.13	5.69

Table 9-12: Neptunium-Varying NOM Sandy Soil Centrifuged Data

Sample	[Np] _o	[NOM] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	mg L ⁻¹	g	ppb	ppb	L kg ⁻¹	
S-1-C	11.12	5.03	0.26	8.50	98.80	11.62	5.56
S-2-C	11.10	4.86	0.24	8.36	106.67	12.76	5.53
S-3-C	10.54	9.60	0.25	8.38	88.28	10.54	5.49
S-4-C	10.81	9.73	0.26	8.84	74.96	8.48	5.41
S-5-C	10.74	14.56	0.25	8.47	90.31	10.66	5.48
S-6-C	10.79	14.36	0.25	8.33	98.50	11.83	5.49
S-7-C	11.10	19.86	0.24	8.02	122.79	15.31	5.61
S-8-C	11.15	0.00	0.25	9.56	60.45	6.32	5.49

Table 9-13: Neptunium-Varying NOM Sandy Soil Filtrate Data

Sample	[Np] _o	[NOM] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	mg L ⁻¹	g	ppb	ppb	L kg ⁻¹	
S-1-F	11.12	5.03	0.26	8.03	116.52	14.51	5.56
S-2-F	11.10	4.86	0.24	8.07	117.86	14.60	5.53
S-3-F	10.54	9.60	0.25	8.31	91.06	10.96	5.49
S-4-F	10.81	9.73	0.26	8.68	80.97	9.32	5.41
S-5-F	10.74	14.56	0.25	8.30	97.42	11.74	5.48
S-6-F	10.79	14.36	0.25	8.11	107.35	13.24	5.49
S-7-F	11.10	19.86	0.24	7.79	131.77	16.91	5.61
S-8-F	11.15	0.00	0.25	9.23	72.84	7.89	5.49

Table 9-14: Reductant Addition Clayey Soil Centrifuged Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	g	ppb	ppb	L kg ⁻¹	
Ascorbic Acid	10.56	0.26	6.78	142.87	21.06	5.30
Zero Valent Iron	11.23	0.24	7.02	166.13	23.65	5.29
Dithionide	11.39	0.24	7.22	165.85	22.96	5.39
Hydrogen Peroxide	10.84	0.26	8.13	101.39	12.47	5.28

Table 9-15: Reductant Addition Clayey Soil Filtrate Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	g	ppb	ppb	L kg ⁻¹	
Ascorbic Acid	11.06	0.25	8.36	106.63	12.76	5.30
Zero Valent Iron	10.81	0.25	8.40	100.76	11.99	5.29
Dithionide	11.04	0.24	8.61	95.98	11.14	5.39
Hydrogen Peroxide	10.99	0.25	8.87	81.86	9.23	5.28

Table 9-16: Reductant Addition Sandy Soil Centrifuged Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	g	ppb	ppb	L kg ⁻¹	
Ascorbic Acid	10.56	0.26	6.41	157.14	24.53	5.84
Zero Valent Iron	11.23	0.24	6.80	175.05	25.75	5.70
Dithionide	11.39	0.24	6.94	176.97	25.49	5.73
Hydrogen Peroxide	10.84	0.26	8.01	105.90	13.22	5.63

Table 9-17: Reductant Addition Sandy Soil Filtrate Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	[Np] _{soil}	K _D	pH
	ppb	g	ppb	ppb	L kg ⁻¹	
Ascorbic Acid	11.06	0.25	8.33	107.50	12.90	5.84
Zero Valent Iron	10.81	0.25	8.31	104.50	12.57	5.70
Dithionide	11.04	0.24	8.68	93.46	10.77	5.73
Hydrogen Peroxide	10.99	0.25	8.85	82.44	9.31	5.63

Table 9-18: Anaerobic Glovebox Clayey Soil Centrifuged Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Liq Mass	[Np] _{soil}	K _D	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹	
C-1-C	0.25	0.26	0.21	9.60	1.47	6.99	5.44
C-2-C	0.59	0.26	0.44	9.36	5.47	12.57	5.51
C-3-C	1.14	0.27	0.85	9.59	10.56	12.48	5.42
C-4-C	1.14	0.26	0.85	9.58	10.47	12.27	5.48
C-5-C	2.28	0.26	1.67	9.61	22.65	13.55	5.58
C-6-C	5.65	0.26	4.15	9.67	55.04	13.26	5.59
C-7-C	5.73	0.25	4.28	9.50	55.10	12.88	5.56
C-8-C	10.12	0.25	7.64	9.99	98.45	12.89	5.53
C-9-C	10.41	0.26	7.78	9.74	97.11	12.48	5.50
C-10-C	21.23	0.26	13.75	9.57	280.66	20.42	5.46

Table 9-19: Anaerobic Glovebox Clayey Soil Filtrate Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Liq Mass	[Np] _{soil}	K _D	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹	
C-1-F	0.25	0.26	0.21	9.60	1.36	6.38	5.44
C-2-F	0.59	0.26	0.43	9.36	5.71	13.32	5.51
C-3-F	1.14	0.27	0.84	9.59	10.65	12.61	5.42
C-4-F	1.14	0.26	0.79	9.58	12.77	16.15	5.48
C-5-F	2.28	0.26	1.67	9.61	22.89	13.74	5.58
C-6-F	5.65	0.26	4.14	9.67	55.64	13.45	5.59
C-7-F	5.73	0.25	4.23	9.50	56.97	13.47	5.56
C-8-F	10.12	0.25	7.67	9.99	97.05	12.65	5.53
C-9-F	10.41	0.26	7.90	9.74	92.75	11.75	5.50
C-10-F	21.23	0.26	13.75	9.57	280.70	20.42	5.46

Table 9-20: Anaerobic Glovebox Sandy Soil Centrifuged Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Liq Mass	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
S-1-C	0.12	0.26	0.12	9.55	-0.06	-0.51	-1%	5.46
S-2-C	0.58	0.26	0.51	9.47	2.72	5.37	13%	5.52
S-3-C	1.15	0.26	0.96	9.53	6.88	7.17	16%	5.56
S-4-C	1.13	0.26	0.99	9.54	5.33	5.39	13%	5.40
S-5-C	2.23	0.25	1.88	9.82	13.46	7.16	16%	5.60
S-6-C	5.62	0.26	4.81	9.72	30.20	6.27	14%	5.47
S-7-C	5.54	0.25	4.88	9.84	25.88	5.30	12%	5.51
S-8-C	10.68	0.26	9.02	9.49	61.38	6.81	16%	5.60
S-9-C	10.53	0.26	9.44	9.65	41.24	4.37	10%	5.44
S-10-C	21.90	0.25	19.77	9.24	77.88	3.94	10%	5.48

Table 9-21: Anaerobic Glovebox Sandy Soil Filtrate Data

Sample	[Np] _o	Mass Soil	[Np] _{aqu}	Liq Mass	[Np] _{soil}	K _D	% Sorbed	pH
	ppb	g	ppb	g	ppb	L kg ⁻¹		
S-1-F	0.12	0.26	0.10	9.55	0.63	6.13	14%	5.46
S-2-F	0.58	0.26	0.48	9.47	3.79	7.93	18%	5.52
S-3-F	1.15	0.26	0.95	9.53	7.36	7.77	17%	5.56
S-4-F	1.13	0.26	0.94	9.54	7.06	7.49	17%	5.40
S-5-F	2.23	0.25	1.88	9.82	13.57	7.23	16%	5.60
S-6-F	5.62	0.26	4.80	9.72	30.86	6.43	15%	5.47
S-7-F	5.54	0.25	4.76	9.84	30.62	6.44	14%	5.51
S-8-F	10.68	0.26	8.97	9.49	62.92	7.01	16%	5.60
S-9-F	10.53	0.26	9.23	9.65	48.93	5.30	12%	5.44
S-10-F	21.90	0.25	19.71	9.24	80.06	4.06	10%	5.48

9.2 Flow Cell Experiments

Two versions of the flowcell were constructed. Figure 9-1 was the first design and was made from Teflon. This is the flowcell that was used during the experiments. Figure 9-2 shows the second design that was constructed of polycarbonate. Figure 9-3B shows some of the issues encountered during the initial testing of the flowcell. Figure 9-3 shows the comparison between a typical sandy soil suspension and the sandy soil suspension removed from the flowcell. The difference between the two is due to bits of Teflon from the stir bars being suspended in solution as well as dissolved iron from the center of the stir bar. Figures 9-3B and 9-3C show the aftermath of the effects of the abrasion due to the sediments on the stir bars. The solution to this problem was to sieve the sediments to less than $53\mu\text{m}$.



Figure 9-1: Version 1 of the flowcell.



Figure 9-2: Version 2 of the flowcell.



Figure 9-3: Trials and tribulations during flowcell testing. (A) Teflon and iron suspended in sandy sediment suspensions. (B) and (C) Stir bar destruction due to abrasive properties of unsieved sediments.

Table 9-22: Summary of Flow and Stopped Flow Periods During Flowcell Experiment.

	Start Flow	Stop Flow	Sample Date	Flow time (hrs)	Stopped Flow time (hrs)	Stopped Flow time (days)
Samples S1-S10	12/2/2009 14:29	12/2/2009 15:19	12/12/2009 17:33	0.83	2.00	0.08
Samples S11-S27	12/2/2009 17:19	12/2/2009 18:57	12/12/2009 17:33	1.63	18.77	0.78
Samples S28-S44	12/3/2009 13:43	12/3/2009 15:27	12/12/2009 17:33	1.73	0.00	0.00
Samples S45-D25	12/3/2009 15:27	12/3/2009 19:28	12/12/2009 17:33	4.02	26.20	1.09
Samples D26-D92	12/4/2009 21:40	12/5/2009 10:23	12/12/2009 17:33	12.72	70.03	2.92
Samples D93-D156	12/8/2009 8:25	12/8/2009 20:31	12/12/2009 17:33	12.10	189.15	7.88
Samples D157-D187	12/16/2009 17:40	12/17/2009 13:55	12/17/2009 16:00	20.25	549.45	22.89
Samples D188-D212	1/9/2010 11:22	1/9/2010 15:32	1/9/2010 15:45	4.17	1559.65	64.99
Samples D213-D322	3/15/2010 15:11	3/17/2010 18:45	3/17/2010 20:00	51.56		

Table 9-23: Data from Flowcell Experiment.

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C ₀)	Theoretical Tracer (In Place of 3H data)	Np/Np ₀	Solid Phase Np (μg/kg soil)	K _d L/kg
S1	1.016	0.0108898	0.051	0.004	0.050	0.001	22.309	2048.631
S2	1.045	0.0732292	0.103		0.098	0.006	43.092	588.450
S3	1.043	0.2712567	0.155	0.049	0.144	0.024	58.045	213.985
S4	1.018	0.6003125	0.206		0.186	0.053	66.579	110.907
S5	1.019	0.8642311	0.257	0.111	0.227	0.077	77.181	89.307
S6	3.093	1.5124212	0.412	0.212	0.337	0.134	111.450	73.690
S7	3.097	2.5301162	0.567		0.433	0.224	124.845	49.344
S8	3.074	3.4733032	0.720	0.403	0.513	0.308	135.049	38.882
S9	3.117	4.3370565	0.876		0.584	0.384	143.721	33.138
S10	3.096	5.1996905	1.031	0.544	0.643	0.461	146.846	28.241
S11	0.974	5.9606484	1.080	0.449	0.660	0.528	126.899	21.289
S12	1.056	5.9510095	1.132		0.678	0.528	138.459	23.266
S13	1.029	6.1399277	1.184		0.694	0.544	141.462	23.040
S14	1.014	6.2681535	1.235	0.488	0.709	0.556	146.468	23.367
S15	1.046	6.2862635	1.287		0.724	0.557	156.126	24.836
S16	1.034	6.5847883	1.339		0.738	0.584	153.906	23.373
S17	3.107	6.8820356	1.494	0.584	0.775	0.610	169.240	24.592
S18	3.091	7.4220314	1.648		0.808	0.658	171.473	23.103
S19	3.095	7.8544659	1.803	0.747	0.835	0.696	175.354	22.325
S20	3.093	7.9901523	1.958		0.859	0.708	190.179	23.802
S21	3.099	8.4717228	2.113		0.879	0.751	188.332	22.231

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C ₀)	Theoretical Tracer (In Place of 3H data)	Np/Np ₀	Solid Phase Np (μg/kg soil)	K _d L/kg
S22	3.106	8.9630501	2.268	0.811	0.896	0.795	183.108	20.429
S23	3.126	9.2729616	2.424		0.911	0.822	183.256	19.762
S24	3.101	9.6948737	2.579		0.924	0.859	176.257	18.180
S25	3.125	9.568543	2.736		0.935	0.848	191.904	20.056
S26	3.114	9.4931716	2.891	0.896	0.944	0.842	205.955	21.695
S27	3.147	9.6814509	3.049		0.953	0.858	208.467	21.533
S28	0.999	3.8746231	3.099	0.706	0.955	0.343	453.914	117.151
S29	1.020	9.8840968	3.150	0.728	0.957	0.876	217.941	22.050
S30	1.011	9.7248072	3.200		0.959	0.862	227.394	23.383
S31	1.015	9.8927732	3.251	0.749	0.961	0.877	223.518	22.594
S32	0.999	9.8751268	3.301		0.963	0.875	227.007	22.988
S33	3.064	9.8496379	3.454	0.915	0.968	0.873	236.727	24.034
S34	3.068	9.9457574	3.607		0.973	0.882	241.042	24.236
S35	3.071	10.000248	3.761		0.977	0.887	246.685	24.668
S36	3.052	9.9347762	3.914		0.980	0.881	257.445	25.914
S37	3.065	10.274989	4.067	0.912	0.983	0.911	250.045	24.335
S38	3.075	10.819576	4.221		0.985	0.959	231.218	21.370
S39	3.087	10.320584	4.375		0.987	0.915	256.931	24.895
S40	3.067	10.711664	4.528	0.932	0.989	0.950	244.853	22.859
S41	3.087	10.676376	4.683		0.991	0.946	249.958	23.412
S42	3.089	10.841869	4.837		0.992	0.961	246.071	22.696
S43	3.092	10.400745	4.992	0.932	0.993	0.922	269.002	25.864
S44	3.098	11.074337	5.147		0.994	0.982	243.501	21.988

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C ₀)	Theoretical Tracer (In Place of 3H data)	Np/Np ₀	Solid Phase Np (μg/kg soil)	K _d L/kg
S45	1.016	10.866496	5.197	0.778	0.994	0.963	229.831	21.150
D1	1.007	10.756171	5.248	0.760	0.946	0.954	212.692	19.774
D2	1.036	10.558492	5.300	0.820	0.898	0.936	198.811	18.830
D3	1.034	10.214882	5.351	0.711	0.853	0.906	191.488	18.746
D4	1.032	9.9563416	5.403		0.810	0.883	181.340	18.214
D5	3.104	9.3636151	5.558	0.710	0.693	0.830	147.142	15.714
D6	3.089	8.2252973	5.713	0.611	0.594	0.729	141.894	17.251
D7	3.107	7.3326751	5.868		0.509	0.650	132.104	18.016
D8	3.114	6.618449	6.024	0.456	0.435	0.587	119.531	18.060
D9	3.106	5.8844774	6.179		0.373	0.522	112.386	19.099
D10	3.117	5.179493	6.335		0.319	0.459	108.319	20.913
D11	5.196	4.3582169	6.595	0.271	0.246	0.386	95.958	22.018
D12	5.213	3.4586367	6.855		0.190	0.307	95.884	27.723
D13	5.204	2.9172771	7.115		0.146	0.259	87.233	29.902
D14	5.210	2.3973131	7.376	0.124	0.113	0.213	83.081	34.656
D15	5.234	1.9437019	7.638		0.087	0.172	80.894	41.619
D16	5.203	1.6184515	7.898		0.067	0.143	77.089	47.632
D17	5.207	1.4004097	8.158	0.056	0.052	0.124	71.265	50.889
D18	5.217	1.1616258	8.419		0.040	0.103	68.712	59.152
D19	5.218	0.9995114	8.680		0.031	0.089	64.791	64.823
D20	5.202	0.8476622	8.940		0.024	0.075	62.064	73.218
D21	5.214	0.7164514	9.201	0.020	0.018	0.064	59.856	83.545
D22	5.222	0.6333032	9.462		0.014	0.056	56.590	89.356
D23	5.203	0.5410311	9.722		0.011	0.048	54.663	101.034

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D24	5.202	0.481129	9.982		0.008	0.043	52.071	108.226
D25	5.233	0.4203009	10.244	0.008	0.006	0.037	50.118	119.244
D26	0.898	0.7860724	10.288	0.008	0.006	0.070	34.180	43.483
D27	0.937	0.8731492	10.335		0.006	0.077	29.095	33.322
D28	0.943	0.8742648	10.382	0.007	0.006	0.078	27.413	31.355
D29	0.939	0.7973077	10.429		0.005	0.071	28.984	36.352
D30	0.926	0.8078707	10.476		0.005	0.072	27.078	33.518
D31	2.941	0.707789	10.623	0.005	0.004	0.063	26.919	38.032
D32	2.917	0.6309094	10.769		0.004	0.056	26.317	41.712
D33	2.944	0.5423204	10.916		0.003	0.048	26.665	49.169
D34	2.945	0.4955406	11.063	0.002	0.003	0.044	25.624	51.709
D35	2.931	0.431532	11.210		0.002	0.038	25.654	59.450
D36	4.868	0.3658981	11.453	0.002	0.002	0.032	24.723	67.569
D37	4.890	0.3018944	11.698		0.001	0.027	24.334	80.603
D38	4.894	0.2487322	11.942		0.001	0.022	24.028	96.600
D39	4.925	0.2082217	12.188		0.001	0.018	23.600	113.339
D40	4.950	0.1756555	12.436		0.001	0.016	23.166	131.885
D41	4.941	0.1480874	12.683		0.001	0.013	22.808	154.017
D42	4.972	0.1275321	12.932		0.000	0.011	22.365	175.367
D43	4.979	0.1089946	13.181	0.001	0.000	0.010	22.023	202.060
D44	4.964	0.121028	13.429		0.000	0.011	20.352	168.156
D45	4.977	0.0818861	13.678		0.000	0.007	21.097	257.642
D46	4.995	0.0751207	13.927		0.000	0.007	20.621	274.500
D47	4.972	0.0644409	14.176		0.000	0.006	20.408	316.699

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D48	4.964	0.0581309	14.424		0.000	0.005	20.086	345.526
D49	4.996	0.0586565	14.674		0.000	0.005	19.483	332.148
D50	4.997	0.0551088	14.924		0.000	0.005	19.076	346.161
D51	5.014	0.0430367	15.174		0.000	0.004	19.127	444.445
D52	4.996	0.0401265	15.424		0.000	0.004	18.845	469.636
D53	4.985	0.0375629	15.674		0.000	0.003	18.575	494.494
D54	4.998	0.0359559	15.923		0.000	0.003	18.281	508.442
D55	5.004	0.0324259	16.174		0.000	0.003	18.099	558.176
D56	5.020	0.0295548	16.425		0.000	0.003	17.919	606.288
D57	4.976	0.0277421	16.673		0.000	0.002	17.716	638.612
D58	4.993	0.0264465	16.923		0.000	0.002	17.506	661.924
D59	4.972	0.0251289	17.172		0.000	0.002	17.310	688.834
D60	4.968	0.0261582	17.420		0.000	0.002	17.011	650.295
D61	4.980	0.0224856	17.669		0.000	0.002	16.934	753.103
D62	4.975	0.0219862	17.918		0.000	0.002	16.736	761.227
D63	4.959	0.0215073	18.166		0.000	0.002	16.544	769.210
D64	4.984	0.0198337	18.415		0.000	0.002	16.414	827.569
D65	4.948	0.0191683	18.662		0.000	0.002	16.252	847.844
D66	4.956	0.0185727	18.910		0.000	0.002	16.092	866.458
D67	4.987	0.0174272	19.160		0.000	0.002	15.965	916.116
D68	4.957	0.0165247	19.407		0.000	0.001	15.838	958.468
D69	4.936	0.0161401	19.654		0.000	0.001	15.695	972.450
D70	4.976	0.0161829	19.903		0.000	0.001	15.534	959.887
D71	4.975	0.0151357	20.152		0.000	0.001	15.426	1019.166

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D72	4.990	0.0141434	20.401		0.000	0.001	15.325	1083.540
D73	5.016	0.0140183	20.652		0.000	0.001	15.190	1083.597
D74	5.034	0.0135545	20.904		0.000	0.001	15.073	1112.033
D75	5.050	0.0137134	21.156		0.000	0.001	14.929	1088.650
D76	5.066	0.0125615	21.410		0.000	0.001	14.848	1182.062
D77	5.072	0.0125988	21.663		0.000	0.001	14.720	1168.370
D78	5.075	0.0119667	21.917		0.000	0.001	14.625	1222.098
D79	5.073	0.01175	22.171		0.000	0.001	14.515	1235.297
D80	5.080	0.0115702	22.425		0.000	0.001	14.405	1245.008
D81	5.085	0.0109004	22.679		0.000	0.001	14.322	1313.855
D82	5.089	0.0108247	22.933		0.000	0.001	14.215	1313.210
D83	5.106	0.0103521	23.189		0.000	0.001	14.129	1364.821
D84	5.086	0.0102969	23.443		0.000	0.001	14.027	1362.255
D85	5.089	0.0101175	23.697		0.000	0.001	13.932	1377.003
D86	5.126	0.009757	23.954		0.000	0.001	13.847	1419.169
D87	5.104	0.0097583	24.209		0.000	0.001	13.748	1408.828
D88	5.085	0.0091277	24.463		0.000	0.001	13.681	1498.800
D89	5.119	0.0090503	24.719		0.000	0.001	13.592	1501.785
D90	5.096	0.0094571	24.974		0.000	0.001	13.480	1425.362
D91	5.107	0.0089284	25.229		0.000	0.001	13.410	1501.963
D92	5.104	0.0086175	25.484		0.000	0.001	13.335	1547.449
D93	0.996	0.1050064	25.534		0.000	0.009	9.297	88.539
D94	1.002	0.118411	25.584		0.000	0.010	8.529	72.027
D95	0.994	0.1127829	25.634		0.000	0.010	8.530	75.630

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D96	0.985	0.110824	25.683		0.000	0.010	8.391	75.712
D97	0.990	0.1056881	25.733		0.000	0.009	8.387	79.356
D98	3.046	0.0987565	25.885		0.000	0.009	8.065	81.663
D99	3.041	0.0863432	26.037		0.000	0.008	8.036	93.074
D100	3.032	0.0782349	26.188		0.000	0.007	7.887	100.814
D101	3.038	0.0676114	26.340		0.000	0.006	7.901	116.863
D102	3.041	0.0587092	26.492		0.000	0.005	7.900	134.566
D103	5.047	0.0491627	26.745		0.000	0.004	7.787	158.385
D104	5.043	0.0395476	26.997		0.000	0.004	7.772	196.534
D105	5.031	0.0330638	27.248		0.000	0.003	7.700	232.870
D106	5.040	0.0272055	27.500		0.000	0.002	7.660	281.559
D107	5.067	0.0230283	27.754		0.000	0.002	7.594	329.772
D108	5.064	0.019909	28.007		0.000	0.002	7.518	377.604
D109	5.081	0.0170927	28.261		0.000	0.002	7.457	436.274
D110	5.090	0.0142381	28.516		0.000	0.001	7.427	521.594
D111	5.101	0.020062	28.771		0.000	0.002	6.992	348.508
D112	5.111	0.0111274	29.026		0.000	0.001	7.234	650.091
D113	5.120	0.0101725	29.282		0.000	0.001	7.168	704.672
D114	5.099	0.0101934	29.537		0.000	0.001	7.064	693.012
D115	5.107	0.0137644	29.792		0.000	0.001	6.783	492.762
D116	5.101	0.0110297	30.048		0.000	0.001	6.779	614.658
D117	5.103	0.0091995	30.303		0.000	0.001	6.759	734.709
D118	5.095	0.0067105	30.557		0.000	0.001	6.790	1011.830
D119	5.064	0.0063083	30.811		0.000	0.001	6.742	1068.820

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C ₀)	Theoretical Tracer (In Place of 3H data)	Np/Np ₀	Solid Phase Np (μg/kg soil)	K _d L/kg
D120	5.142	0.0056758	31.068		0.000	0.001	6.710	1182.146
D121	5.146	0.0059806	31.325		0.000	0.001	6.636	1109.648
D122	5.133	0.0052236	31.582		0.000	0.000	6.613	1266.020
D123	5.154	0.004977	31.839		0.000	0.000	6.572	1320.468
D124	5.125	0.0046827	32.096		0.000	0.000	6.536	1395.761
D125	5.126	0.0044565	32.352		0.000	0.000	6.500	1458.465
D126	5.165	0.0044456	32.610		0.000	0.000	6.454	1451.870
D127	5.205	0.0082303	32.870		0.000	0.001	6.219	755.613
D128	5.156	0.0042196	33.128		0.000	0.000	6.335	1501.338
D129	5.178	0.0039633	33.387		0.000	0.000	6.304	1590.691
D130	5.153	0.0037475	33.645		0.000	0.000	6.275	1674.343
D131	5.179	0.0034814	33.904		0.000	0.000	6.249	1795.068
D132	5.165	0.0034129	34.162		0.000	0.000	6.217	1821.670
D133	5.213	0.0033529	34.422		0.000	0.000	6.185	1844.599
D134	5.205	0.0032056	34.683		0.000	0.000	6.157	1920.865
D135	5.212	0.0034414	34.943		0.000	0.000	6.112	1776.146
D136	5.232	0.0031854	35.205		0.000	0.000	6.090	1911.692
D137	5.237	0.0029494	35.467		0.000	0.000	6.068	2057.468
D138	5.226	0.0031069	35.728		0.000	0.000	6.030	1940.770
D139	5.244	0.0029493	35.990		0.000	0.000	6.005	2036.187
D140	5.218	0.0031463	36.251		0.000	0.000	5.965	1895.782
D141	5.220	0.0030283	36.512		0.000	0.000	5.938	1960.851
D142	5.250	0.0029393	36.775		0.000	0.000	5.911	2010.989
D143	5.222	0.0027333	37.036		0.000	0.000	5.891	2155.174

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D144	5.219	0.002635	37.297		0.000	0.000	5.867	2226.659
D145	5.224	0.0028906	37.558		0.000	0.000	5.827	2015.912
D146	5.207	0.0025271	37.818		0.000	0.000	5.815	2301.286
D147	5.232	0.0024972	38.080		0.000	0.000	5.791	2318.876
D148	5.237	0.0025758	38.342		0.000	0.000	5.761	2236.516
D149	5.250	0.0025559	38.604		0.000	0.000	5.735	2243.773
D150	5.265	0.0025655	38.868		0.000	0.000	5.708	2224.759
D151	5.255	0.002202	39.130		0.000	0.000	5.699	2588.208
D152	5.250	0.0024969	39.393		0.000	0.000	5.661	2267.325
D153	5.229	0.0024284	39.654		0.000	0.000	5.639	2322.027
D154	5.257	0.0021921	39.917		0.000	0.000	5.625	2566.189
D155	5.256	0.0021921	40.180		0.000	0.000	5.602	2555.722
D156	5.273	0.0023	40.444		0.000	0.000	5.574	2423.478
D157	0.980	0.0663403	40.493		0.000	0.006	2.900	43.716
D158	0.987	0.0723861	40.542		0.000	0.006	2.518	34.786
D159	0.978	0.0688232	40.591		0.000	0.006	2.526	36.700
D160	0.993	0.0651025	40.640		0.000	0.006	2.545	39.096
D161	2.996	0.0612668	40.790		0.000	0.005	2.333	38.079
D162	12.155	0.0055095	41.398		0.000	0.000	4.416	801.453
D163	12.051	0.0013391	42.000		0.000	0.000	4.549	3397.250
D164	12.088	0.0012154	42.605		0.000	0.000	4.525	3723.107
D165	12.114	0.0010196	43.211		0.000	0.000	4.508	4421.490
D166	12.091	0.0009785	43.815		0.000	0.000	4.486	4585.037
D167	12.106	0.0009372	44.420		0.000	0.000	4.465	4764.461

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C ₀)	Theoretical Tracer (In Place of 3H data)	Np/Np ₀	Solid Phase Np (μg/kg soil)	K _d L/kg
D168	12.141	0.0008548	45.027		0.000	0.000	4.448	5203.841
D169	12.140	0.0008136	45.634		0.000	0.000	4.430	5445.203
D170	12.154	0.0007415	46.242		0.000	0.000	4.415	5954.534
D171	12.162	0.0007209	46.850		0.000	0.000	4.398	6101.751
D172	12.201	0.0006899	47.460		0.000	0.000	4.383	6353.106
D173	12.166	0.0007105	48.069		0.000	0.000	4.365	6143.114
D174	12.237	0.0006281	48.680		0.000	0.000	4.353	6930.868
D175	12.261	0.0006383	49.293		0.000	0.000	4.337	6794.484
D176	12.265	0.0006177	49.907		0.000	0.000	4.323	6997.997
D177	12.233	0.0007413	50.518		0.000	0.000	4.300	5800.272
D178	12.292	0.0007	51.133		0.000	0.000	4.284	6120.268
D179	12.301	0.0006588	51.748		0.000	0.000	4.270	6480.960
D180	12.309	0.0007	52.363		0.000	0.000	4.251	6073.060
D181	12.303	0.0006177	52.979		0.000	0.000	4.239	6863.549
D182	12.325	0.0006279	53.595		0.000	0.000	4.224	6726.250
D183	12.348	0.0006073	54.212		0.000	0.000	4.210	6931.466
D184	12.378	0.0006073	54.831		0.000	0.000	4.195	6907.373
D185	12.450	0.0005557	55.454		0.000	0.000	4.183	7527.179
D186	12.517	0.0005968	56.080		0.000	0.000	4.166	6981.549
D187	12.526	0.0005453	56.706		0.000	0.000	4.155	7619.212
D188	0.9542	0.0271147	56.754		0.000	0.002	3.048	112.399
D189	0.9913	0.0291466	56.803		0.000	0.003	2.910	99.823
D190	0.9789	0.0284476	56.852		0.000	0.003	2.882	101.308
D191	2.9776	0.0262973	57.001		0.000	0.002	2.812	106.924

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D192	2.9862	0.0245844	57.150		0.000	0.002	2.734	111.210
D193	2.9669	0.0223805	57.299		0.000	0.002	2.690	120.180
D194	2.9685	0.0196064	57.447		0.000	0.002	2.684	136.907
D195	4.9634	0.0165389	57.695		0.000	0.001	2.643	159.808
D196	4.963	0.013544	57.943		0.000	0.001	2.629	194.072
D197	4.9864	0.0113709	58.193		0.000	0.001	2.602	228.848
D198	5.0234	0.0102676	58.444		0.000	0.001	2.544	247.728
D199	5.0487	0.0076122	58.696		0.000	0.001	2.573	337.978
D200	5.0502	0.0065011	58.949		0.000	0.001	2.552	392.494
D201	5.0597	0.0089797	59.202		0.000	0.001	2.363	263.136
D202	5.0902	0.0057283	59.456		0.000	0.001	2.434	424.932
D203	5.0829	0.0043504	59.710		0.000	0.000	2.445	562.008
D204	5.0955	0.003476	59.965		0.000	0.000	2.445	703.264
D205	5.0866	0.0028899	60.220		0.000	0.000	2.439	843.828
D206	5.0847	0.0039184	60.474		0.000	0.000	2.358	601.811
D207	5.1085	0.0024063	60.729		0.000	0.000	2.394	994.826
D208	5.1025	0.0025812	60.984		0.000	0.000	2.361	914.585
D209	5.1108	0.0022623	61.240		0.000	0.000	2.350	1038.954
D210	5.122	0.0018303	61.496		0.000	0.000	2.349	1283.383
D211	5.0785	0.0017999	61.750		0.000	0.000	2.332	1295.646
D212	5.1362	0.0015422	62.007		0.000	0.000	2.326	1508.508
D213	0.9314	0.0212653	62.053		0.000	0.002	1.503	70.697
D214	0.9658	0.025312	62.102		0.000	0.002	1.294	51.122
D215	0.9598	0.0246027	62.150		0.000	0.002	1.275	51.835

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C ₀)	Theoretical Tracer (In Place of 3H data)	Np/Np ₀	Solid Phase Np (μg/kg soil)	K _d L/kg
D216	0.9311	0.0243819	62.196		0.000	0.002	1.239	50.814
D217	0.9641	0.0239954	62.244		0.000	0.002	1.208	50.357
D218	0.9364	0.0230725	62.291		0.000	0.002	1.202	52.100
D219	0.8887	0.0238571	62.336		0.000	0.002	1.129	47.314
D220	0.9617	0.0215572	62.384		0.000	0.002	1.179	54.691
D221	0.9274	0.0211032	62.430		0.000	0.002	1.158	54.880
D222	0.9635	0.0202136	62.478		0.000	0.002	1.155	57.130
D223	2.9059	0.0186268	62.623		0.000	0.002	1.110	59.608
D224	2.9012	0.0168208	62.769		0.000	0.001	1.085	64.510
D225	2.9236	0.0150425	62.915		0.000	0.001	1.068	71.025
D226	2.9127	0.0134515	63.060		0.000	0.001	1.054	78.338
D227	2.9243	0.0123789	63.207		0.000	0.001	1.024	82.759
D228	2.9311	0.0114803	63.353		0.000	0.001	0.993	86.524
D229	2.9163	0.0101955	63.499		0.000	0.001	0.985	96.640
D230	2.9283	0.0092456	63.645		0.000	0.001	0.969	104.833
D231	2.929	0.0084088	63.792		0.000	0.001	0.954	113.401
D232	2.9314	0.0082454	63.938		0.000	0.001	0.912	110.612
D233	5.8809	0.0069388	64.232		0.000	0.001	0.883	127.239
D234	5.8919	0.0057549	64.527		0.000	0.001	0.863	149.883
D235	5.9087	0.0048567	64.822		0.000	0.000	0.841	173.212
D236	5.9352	0.0040197	65.119		0.000	0.000	0.827	205.761
D237	5.8426	0.00348	65.411		0.000	0.000	0.808	232.220
D238	11.3137	0.0028083	65.977		0.000	0.000	0.772	274.788
D239	11.3902	0.0021647	66.547		0.000	0.000	0.748	345.682

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C ₀)	Theoretical Tracer (In Place of 3H data)	Np/Np ₀	Solid Phase Np (μg/kg soil)	K _d L/kg
D240	11.3565	0.0015725	67.114		0.000	0.000	0.736	468.244
D241	11.4303	0.0012354	67.686		0.000	0.000	0.722	584.160
D242	11.4214	0.0010823	68.257		0.000	0.000	0.703	649.743
D243	11.38242	0.000776	68.826		0.000	0.000	0.698	899.222
D244	11.38242	0.0006841	69.395		0.000	0.000	0.686	1002.736
D245	11.38242	0.0005514	69.964		0.000	0.000	0.679	1231.086
D246	11.38242	0.0004901	70.533		0.000	0.000	0.670	1367.323
D247	11.38242	0.0004289	71.103		0.000	0.000	0.663	1545.717
D248	11.38242	0.0003676	71.672		0.000	0.000	0.657	1787.343
D249	11.38242	0.0003574	72.241		0.000	0.000	0.649	1816.930
D250	11.38242	0.0003267	72.810		0.000	0.000	0.643	1968.377
D251	11.38242	0.0002757	73.379		0.000	0.000	0.639	2317.635
D252	11.38242	0.0002757	73.948		0.000	0.000	0.633	2295.019
D253	11.38242	0.0002553	74.517		0.000	0.000	0.628	2459.184
D254	11.38242	0.0002451	75.086		0.000	0.000	0.623	2540.690
D255	11.38242	0.0002144	75.656		0.000	0.000	0.619	2886.707
D256	11.38242	0.0002144	76.225		0.000	0.000	0.614	2864.092
D257	11.38242	0.0002042	76.794		0.000	0.000	0.610	2986.668
D258	11.38242	0.0001736	77.363		0.000	0.000	0.607	3498.124
D259	11.38242	0.0001736	77.932		0.000	0.000	0.603	3475.508
D260	11.38242	0.0001634	78.501		0.000	0.000	0.600	3672.595
D261	11.38242	0.0001634	79.070		0.000	0.000	0.596	3649.980
D262	11.38242	0.0001532	79.639		0.000	0.000	0.593	3873.345
D263	11.38242	0.000143	80.208		0.000	0.000	0.590	4130.236

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D264	11.38242	0.000143	80.778		0.000	0.000	0.587	4107.620
D265	11.38242	0.0001327	81.347		0.000	0.000	0.585	4404.032
D266	11.38242	0.0001225	81.916		0.000	0.000	0.582	4751.731
D267	11.38242	0.0001225	82.485		0.000	0.000	0.579	4729.115
D268	11.38242	0.000143	83.054		0.000	0.000	0.575	4025.235
D269	11.38242	0.000143	83.623		0.000	0.000	0.572	4002.619
D270	11.38242	0.0001327	84.192		0.000	0.000	0.570	4290.954
D271	11.38242	0.0001327	84.761		0.000	0.000	0.567	4268.339
D272	11.38242	0.0001532	85.331		0.000	0.000	0.562	3671.313
D273	11.38242	0.0001736	85.900		0.000	0.000	0.558	3212.103
D274	11.38242	0.0001225	86.469		0.000	0.000	0.557	4544.421
D275	11.38242	0.000143	87.038		0.000	0.000	0.553	3866.926
D276	11.38242	0.0001532	87.607		0.000	0.000	0.549	3583.866
D277	11.38242	0.0001634	88.176		0.000	0.000	0.545	3334.775
D278	11.38242	0.0001532	88.745		0.000	0.000	0.542	3537.127
D279	11.38242	0.0001532	89.314		0.000	0.000	0.538	3514.512
D280	11.38242	0.000143	89.884		0.000	0.000	0.535	3745.771
D281	11.38242	0.0001532	90.453		0.000	0.000	0.532	3470.788
D282	11.38242	0.000143	91.022		0.000	0.000	0.529	3698.924
D283	11.38242	0.0001532	91.591		0.000	0.000	0.525	3427.065
D284	11.38242	0.0001327	92.160		0.000	0.000	0.523	3937.803
D285	11.38242	0.0001327	92.729		0.000	0.000	0.520	3915.188
D286	11.38242	0.0001327	93.298		0.000	0.000	0.517	3892.572
D287	11.38242	0.000143	93.867		0.000	0.000	0.513	3589.077

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D288	11.38242	0.0001327	94.437		0.000	0.000	0.510	3845.601
D289	11.38242	0.0001021	95.006		0.000	0.000	0.509	4988.587
D290	11.38242	0.000143	95.575		0.000	0.000	0.505	3529.308
D291	11.38242	0.0001225	96.144		0.000	0.000	0.503	4101.533
D292	11.38242	0.0001123	96.713		0.000	0.000	0.500	4455.396
D293	11.38242	0.0001123	97.282		0.000	0.000	0.498	4432.781
D294	11.38242	0.0001225	97.851		0.000	0.000	0.495	4037.455
D295	11.38242	0.0001225	98.420		0.000	0.000	0.492	4014.840
D296	11.38242	0.0001123	98.989		0.000	0.000	0.490	4360.822
D297	11.38242	0.0001123	99.559		0.000	0.000	0.487	4338.207
D298	11.38242	0.0001123	100.128		0.000	0.000	0.485	4315.591
D299	11.38242	0.0001123	100.697		0.000	0.000	0.482	4292.975
D300	11.38242	0.0001123	101.266		0.000	0.000	0.480	4270.360
D301	11.38242	0.0001225	101.835		0.000	0.000	0.476	3888.570
D302	11.38242	0.0001225	102.404		0.000	0.000	0.474	3865.954
D303	11.38242	0.0001225	102.973		0.000	0.000	0.471	3843.338
D304	11.38242	0.0001225	103.542		0.000	0.000	0.468	3820.723
D305	11.38242	0.0001021	104.112		0.000	0.000	0.467	4570.199
D306	11.38242	0.0001225	104.681		0.000	0.000	0.463	3779.261
D307	11.38242	9.19E-05	105.250		0.000	0.000	0.462	5029.645
D308	11.38242	0.0001021	105.819		0.000	0.000	0.459	4500.091
D309	11.38242	0.0001021	106.388		0.000	0.000	0.457	4477.475
D310	11.38242	0.0001021	106.957		0.000	0.000	0.455	4454.860
D311	11.38242	9.19E-05	107.526		0.000	0.000	0.453	4931.644

Sample	Sample Mass (g)	Actual Np Conc (ppb)	Cumulative Cell Volumes	3H Tracer (C/C _o)	Theoretical Tracer (In Place of 3H data)	Np/Np _o	Solid Phase Np (μg/kg soil)	K _d L/kg
D312	11.38242	0.0001225	108.095		0.000	0.000	0.449	3666.183
D313	11.38242	0.0001123	108.665		0.000	0.000	0.447	3980.469
D314	11.38242	9.19E-05	109.234		0.000	0.000	0.446	4851.233
D315	11.38242	8.169E-05	109.803		0.000	0.000	0.444	5439.989
D316	11.38242	9.19E-05	110.372		0.000	0.000	0.442	4808.515
D317	11.38242	0.0001123	110.941		0.000	0.000	0.439	3904.399
D318	11.38242	8.169E-05	111.510		0.000	0.000	0.438	5360.834
D319	11.38242	9.19E-05	112.079		0.000	0.000	0.435	4738.155
D320	11.38242	0.0001021	112.648		0.000	0.000	0.433	4237.750
D321	11.38242	0.0001021	113.217		0.000	0.000	0.430	4215.135
D322	11.38242	8.169E-05	113.787		0.000	0.000	0.429	5256.237
Last	11.38242	0.0007862	114.356		0.000	0.000	0.384	487.878

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