



**Keywords:** *Neptunium,  
Sediment, pH, Redox*

**Retention:** *Permanent*

## **Neptunium IV and V Sorption to End-Member Subsurface Sediments of the Savannah River Site**

Todd J. Miller,<sup>(a)</sup> Brian A. Powell,<sup>(a)</sup>  
Daniel I. Kaplan

November 13, 2009

<sup>(a)</sup> Department of Environmental Engineering and Earth Sciences  
Clemson University, Clemson, SC

Savannah River National Laboratory  
Savannah River Nuclear Solutions  
Aiken, SC 29808

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Prepared for the U.S. Department of Energy under  
contract number DE-AC09-08SR22470.



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**Prepared for  
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## ABSTRACT

Migration of Np through the subsurface is expected to be primarily controlled by sorption to sediments. Therefore, understanding and quantifying Np sorption to sediments and sediments from the Savannah River Site (SRS) is vital to ensure safe disposal of Np bearing wastes. In this work, Np sorption to two sediments representing the geological extremes with respect to sorption properties expected in the SRS subsurface environment (named “subsurface sandy sediment” and “subsurface clayey sediment”) was examined under a variety of conditions. First a series of baseline sorption tests at pH 5.5 under an oxic atmosphere was performed to understand Np sorption under typical subsurface conditions. These experiments indicated that the baseline  $K_d$  values for the subsurface sandy and subsurface clayey sediments are  $4.26 \pm 0.24 \text{ L kg}^{-1}$  and  $9.05 \pm 0.61 \text{ L kg}^{-1}$ , respectively. These Np  $K_d$  values of SRS sediments are the first to be reported since Sheppard et al. (1979). The previous values were 0.25 and 0.16  $\text{L kg}^{-1}$  for a low pH sandy sediment.

To examine a possible range of  $K_d$  values under various environmental scenarios, the effects of natural organic matter (NOM, also a surrogate for cellulose degradation products), the presence of various chemical reductants, and an anaerobic atmosphere on Np sorption were examined. The presence of NOM resulted in an increase in the Np  $K_d$  values for both sediments. This behavior is hypothesized to be the result of formation of a ternary Np-NOM-sediment complex. Slight increases in the Np sorption ( $K_d$  13 – 24  $\text{L kg}^{-1}$ ) were observed when performing experiments in the presence of chemical reductants (dithionite, ascorbic acid, zero-valent iron) or under anaerobic conditions. Presumably, the increased sorption can be attributed to a slight reduction of Np(V) to Np(IV), the stronger sorbing form of Np. The most significant result of this study is the finding that Np weakly sorbs to both end member sediments and that Np only has a slight tendency to reduce to its stronger sorbing form, even under the most strongly reducing conditions expected under natural SRS conditions. Also, it appears that pH has a profound effect on Np sorption. Based on the these new measurements and the revelations about Np redox chemistry, the following changes to “Best  $K_d$ ” values, as defined in Kaplan (2006), for SRS performance assessment calculations are recommended.

	Previous Recommended “Best $K_d$ ” Values <sup>(a)</sup> ( $\text{L kg}^{-1}$ )	New Recommended “Best $K_d$ ” Values ( $\text{L kg}^{-1}$ )
Sandy Sediment	0.6	3
Clay Sediment	35 <sup>(b)</sup>	9
<sup>(a)</sup> Kaplan (2006; Geochemical Data Package, WSRC-TR-2006-00004)		
<sup>(b)</sup> An estimate that was based without experimental data.		

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## LIST OF ABBREVIATIONS

[Np]	Neptunium concentrations
[Np] <sub>0</sub>	initial Neptunium concentrations
DDI water	Distilled deionized water
K <sub>d</sub>	Distribution Coefficient
NOM	Natural Organic Matter
ppb	parts per billion
ppq	parts per quadrillion
QA/QC	Quality Assurance/Quality Control
SRNL	Savannah River National Laboratory
SRS	Savannah River Site

# Neptunium IV and V Sorption to End-Member Subsurface Sediments of the Savannah River Site

## 1.0 Introduction

In several scenarios at the SRS's E-Area Low-Level Waste Facility, Np has become a key risk driver because of its perceived high mobility, long-half life, and high inventory (WSRC 2008). Neptunium generally exists in surface aquifers as the oxyanion  $\text{NpO}_2^+$ . This cation sorbs quite weakly to sediments. For example, the last measurement of Np  $K_d$  values using Savannah River Site sediments was only 0.25 and 0.16  $\text{L kg}^{-1}$  for a sandy sediment (Sheppard et al. 1979). This value is an especially low  $K_d$  value when compared to other transuranic elements, such as U or Pu, which have values of about 200 and 290, respectively for sandy SRS sediment (Kaplan 2009). There has been very little laboratory or field research conducted with Np to understand how it interacts with SRS sediments. Little is known about its tendency to change from its Np(V) form as  $\text{NpO}_2^+$ , to its less mobile form, Np(IV). By chemical analogy, Pu, which also exists primarily in the +5 oxidation state in the aqueous phase,  $\text{PuO}_2^+$ , and which has only one atomic number more than Np (Pu = 94), readily reduces to  $\text{PuO}_{2(s)}$  in a matter of minutes when it comes in contact with vadose zone SRS sediments (Kaplan et al. 2004). Once the  $\text{PuO}_{2(s)}$  forms, Pu solubility decreases sharply to  $10^{-9}$  M ( $K_d$  values of 10,000  $\text{L kg}^{-1}$ ; Kaplan et al. 2006). It was this observation that motivated us to expect that some  $\text{NpO}_2^+$  might be reduced by SRS sediments.

A second parameter known to have a strong influence on transuranic sorption to sediments is natural organic matter complexation (Artinger et al. 2000). Artinger et al. (2000) reported that when Np complexed with humic substances, it moved faster than the averaged velocity of groundwater in a column experiment. Furthermore, they reported that Np(IV) complexed stronger than Np(V) to the humic substances. The E-Area Low-Level Waste Facility contains cellulose degradation products, materials that can degrade to form organic complexes that may behave very similarly to humic acids.

### 1.1 Objectives

The objectives of this study were:

1. to determine if Np(V) reduces under SRS environmental conditions to Np(IV), and if so, quantify the difference in Np sorption under oxidizing and reducing conditions, and
2. to quantify the influence of the presence of aqueous humic acids on Np(V) sorption to SRS sediments.



## 2.0 Materials and Methods

### 2.1 Materials: Stock Solution Preparation and Sediments

A composite  $^{237}\text{Np}$  stock solution from the Environmental Engineering and Earth Science, Clemson University inventory (primarily purchased from Isotope Products, Valencia, CA) was evaporated to dryness then the residue was brought up in approximately 5 mL 8.0 M  $\text{HNO}_3$ . 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  $\text{HNO}_3$ /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  $\text{HNO}_3$ /0.3 M  $\text{NH}_2\text{OH}\cdot\text{HCl}$  Np solution was loaded on a 2 mL column and washed with 3 column volumes of 3 M  $\text{HNO}_3$ . The Np(IV) was eluted with 0.02 M  $\text{HCl}$  + 0.2M  $\text{HF}$ . The effluent was evaporated to dryness then redissolved in 1.0 M  $\text{HNO}_3$ . Additional 1.0 M  $\text{HNO}_3$  was added to maintain a approximately 10 mL then the solution was evaporated to incipient dryness and redissolved in a 5.0 mL of 1.0 M  $\text{HNO}_3$ . 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). No other alpha energies besides  $^{237}\text{Np}$  were observed. The approximate concentration was determined using liquid scintillation counting and little  $^{233}\text{Pa}$  was observed. The fuming in  $\text{HNO}_3$  as performed at the end of the purification procedure will drive Np to the soluble pentavalent state. This is the stable oxidation state of Np under the experimental conditions. Therefore, experiments performed here can be assumed to be initially Np(V). The exact Np concentration in this solution was determined using ICP-MS calibrated with a NIST standard as discussed in Section 2.2 below.

Working Solution #1 was created by pipetting an aliquot of the Np stock solution into a 100 mL Nalgene Teflon bottle and diluting with 2% BDH Aristar Ultra  $\text{HNO}_3$  to give a working solution Np concentration of approximately 800 ppb. Working Solution #2 was created by pipetting an aliquot of Working Solution #1 with 2% BDH Aristar Ultra  $\text{HNO}_3$  in a 250 mL polypropylene bottle to created a target Np concentration of approximately 50 ppb. Analysis on the ICP-MS calibrated against NIST standards as described below gave concentrations of Working Solution #1 and Working Solution #2 of 820 ppb and 49.6 ppb, respectively. Calibration using the NIST standard is described in Section 3.1.2 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 1.

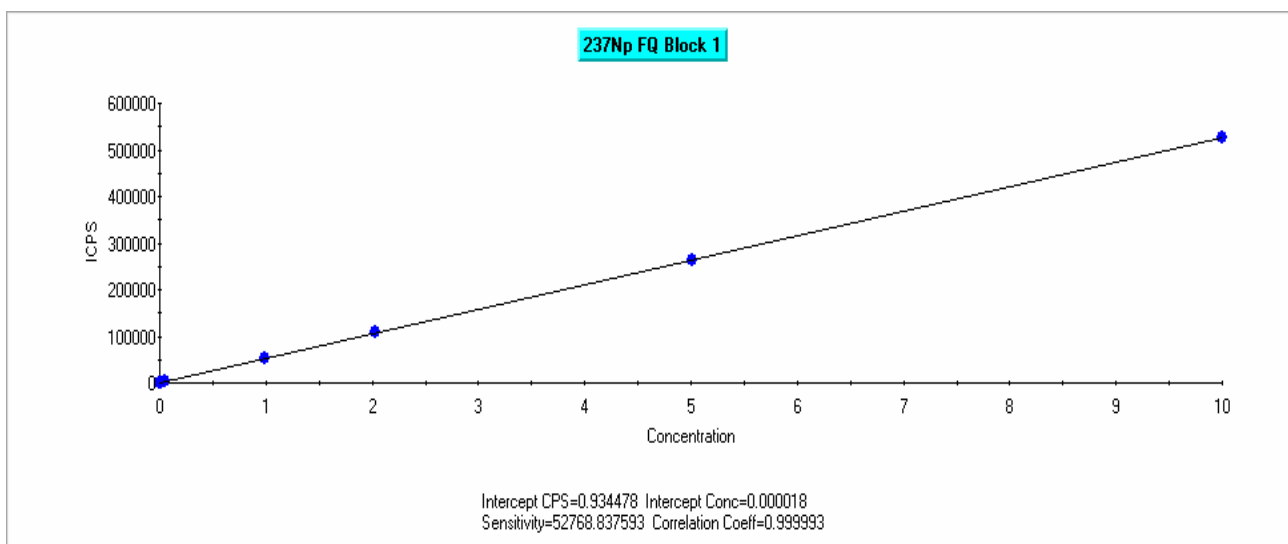
**Table 1: Descriptions of SRS sediments used in this work**

Name	Description	Sand/Silt/Clay (wt%)	Surface Area (m <sup>2</sup> /g)	pH	Organic Matter (wt-%)
Subsurface Sandy	Subsurface Yellow Sandy Sediment Low Organic Matter	97/2/1	1.27	5.10	<0.01
Subsurface Clayey	Subsurface Red Clayey Burial Ground Sediment Low Organic Matter	58/30/12	15.31	4.55	NA

## 2.2 ICP-MS Calibration Curves – Detection Limits

A National Institute of Standards and Technology, Standard Reference Material (NIST SRM 4341) was used to prepare a stock <sup>237</sup>Np solution by dilution in 2% Aristar Optima HNO<sub>3</sub>. 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<sub>3</sub>. Again all volume additions were monitored gravimetrically. These standards were used to calibrate the Thermo Scientific X Series 2 ICP-MS for quantification of <sup>237</sup>Np. A representative calibration curve for <sup>237</sup>Np is shown in Figure 2.1. The calibration data from Figure 2.1 are shown in Table 2.2. The instrument performance was monitored using <sup>232</sup>Th and <sup>238</sup>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 Np in the samples being analyzed. The typical calibration curve shown in Figure 2.1 gave a minimum detectable limit of 1.8 ppq (parts per quadrillion). This result is consistent with an average minimum detectable quantity of 2 ppq under the configuration of the instrument used for these measurements.



**Figure 2.1:** Screen capture of a typical  $^{237}\text{Np}$  calibration curve using Thermo PlasmaLab software to control the data collection and analysis.  $R^2=0.999993$ , Intercept Conc. (Detection Limit) = 0.000018 ppb.

**Table 2.2: Example ICP-MS Calibration Curve Data**

Sample	Actual NIST Np Concentration (ppb)	Measured Np Concentration (ppb)	Mean Np Ion Counts Per Second (ICPS)	Error	% Error
Blank	0	0	1	0	0
0.01ppb Np	0.01	0.01	528	0	0.82
0.05ppb Np	0.05	0.05	2653	0	0.53
1ppb Np	0.991	0.993	52410	0.002	0.2
2ppb Np	2.028	2.06	108686	0.032	1.56
5ppb Np	5.01	4.995	263575	-0.015	-0.3
10ppb Np	9.998	9.999	527649	0.001	0.01

### 2.3 Preliminary Kinetic Tests

Preliminary experiments were performed to determine the time needed to reach steady state sorption between the aqueous Np and the sorbed Np. This experiment was performed in 45 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 final constant ionic strength of 0.01 M. Next, 40 mL of distilled deionized water (DDI H<sub>2</sub>O) was added along with 0.55 mL of Np Working Solution #1 to obtain an initial Np 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 8 rpm.

After 1, 3, 8, 24, and 48 hours, a 5 mL aliquot of each suspension was removed. 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 time was sufficient 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<sub>3</sub> for analysis on the ICP-MS. Then 2 mL of the supernatant was placed into a Microsep 10K Centrifugal filter. The samples were then centrifuged in a Beckman GS-6 centrifuge at 3000 rpm for 2-3 minutes to wet the filter membrane and equilibrate Np with the membrane then the filtrate was discarded. This pre-filtration step equilibrates the solution with the filter and washes the filter preservation coatings away. This treatment results in a significant reduction in the loss of Np to the filter in the subsequent filtration. The sample was then centrifuged for an additional 20 minutes, or until the entire sample passed through the filter. The filtrate was then transferred into an ELKay polystyrene culture tube and diluted with 2% BDH Aristar Ultra HNO<sub>3</sub> to determine the Np concentration using the ICP-MS.

#### 2.4 Sample Preparation for Sorption Experiments

The 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-H<sub>2</sub>O and 1 mL of 0.1M NaCl and the pH was adjusted to an approximate value of 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 approximately 8 rpm for 24 hours to equilibrate with the solution. The samples were then spiked with the Np Working Solution #1 (described in Section 2.1) 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.

#### 2.5 Sample Analysis by ICP-MS

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 from each suspension was transferred into a 2 mL polypropylene centrifuge tube and approximately 2 mL from each suspension was transferred into a Microsep 10K Centrifugal filter. 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 1mL, 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 HNO<sub>3</sub> for ICP-MS analysis. Then the suspension in the Microsep 10K centrifugal filter was centrifuged in a Beckman GS-6 centrifuge at 3000 rpm for 2-3 min to wet the filter membrane and equilibrate Np with the membrane then the filtrate was discarded. 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 HNO<sub>3</sub> for ICP-MS analysis. The Np concentration in all samples was determined on the Thermo Scientific X Series 2 ICP-MS.

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

$$[Np]_{sed} = \frac{([Np]_{aqu,o} - [Np]_{aqu}) V_L}{m_{sed}} \quad (1)$$

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:

$$K_d = \frac{[Np]_{sed}}{[Np]_{aqu}} \quad (2)$$

The percent of Np sorbed can be calculated via the following equation:

$$f_s = 1 - \frac{[Np]_{aqu}}{[Np]_{aqu,o}} \quad (3)$$

This  $K_d$  equation (1 and 2) 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).

## 2.6 NOM Experiment and Redox Experiments.

In addition to the baseline experiments described above, this study also aimed to capture the effects of various perturbations to the system to the sorption behavior of Np on these sediments. The Natural Organic Matter (NOM) used was Suwannee River NOM from the International Humic Substance Society (IHSS). These experiments were performed analogous to the baseline case with the NOM being added before the first equilibration period. The Np spike was then added 24 hours later as in the baseline case. The NOM stock solution was prepared by mixing the dry Suwannee River NOM with DDI water to make a 250 mg L<sup>-1</sup> stock solution. A 30 mg L<sup>-1</sup> working solution was also prepared and used to make NOM standards of concentrations 0.1, 1.0, 2.0, 10, 20 and 30 mg L<sup>-1</sup>. A Cary 50 Bio UV-Visible Spectrometer was used to measure the absorbance at 254 nm to determine NOM concentrations. This calibration curve was used to determine the NOM concentration in the aqueous phase of centrifuged and filtered samples as described for the Np analyses above. This information was used to calculate the fraction of NOM sorbed to each sediment using similar equations as 1 and 3, except instead of solving for Np, we solved for NOM.

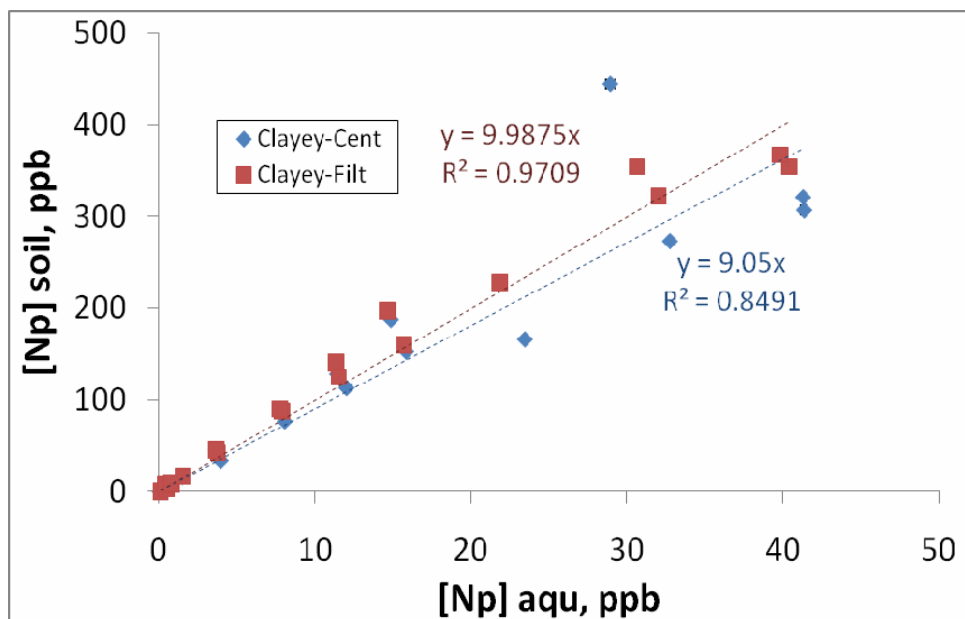
## 2.7 Redox Experiments

A series of experiments was also performed under reducing conditions or in the presence of chemical reductants in an effort to reduce the weakly sorbing Np(V) to the more strongly sorbing Np(IV). The reducing conditions used consisted of the addition of reductants such as ascorbic acid, dithionite, zero-valent iron, and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>). Stock solutions of each

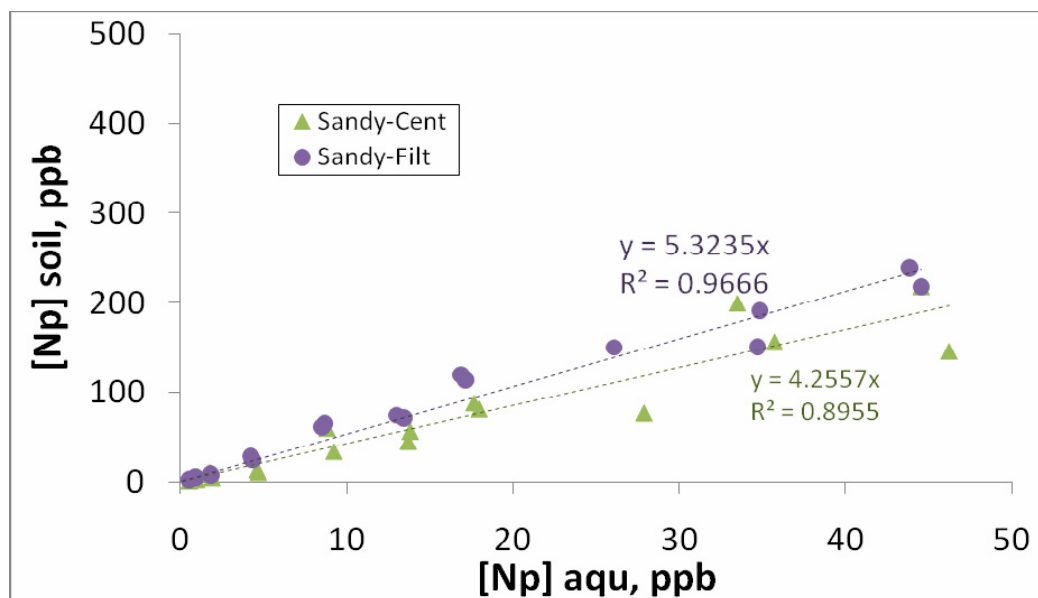
reductant were prepared at 1000 ppm. The stock solutions were added separately to sample vials to obtain a reductant concentration of 10 ppm at the same time the Np spike was added. The Np concentration in each sample was analyzed as stated above. The preliminary experiment was also performed in an anaerobic glovebox with an atmosphere of 98% N<sub>2</sub> and 2% H<sub>2</sub>. All water used for the experiment was deoxygenated by boiling for 30 min then cooling under a nitrogen gas purge. The Np stock solutions were deoxygenated by uncapping the bottles and stirring the solution for 24 h within the glovebox. The samples were analyzed as described in Section 2.5 above.

### 3.0 Results and Discussion

For the baseline case, the samples were prepared then sampled after 48 hours. The  $K_d$  values for the clayey sediment were calculated to be  $9.05 \pm 0.61 \text{ L kg}^{-1}$  and  $9.99 \pm 0.28 \text{ L kg}^{-1}$  for the centrifuged and filtrate samples, respectively (Figure 3.1). The slightly larger filter  $K_d$  value is the result of filtering out  $>100 \text{ nm}$  colloid particles holding Np, that were measured originally in the aqueous phase, thereby producing a lower  $K_d$  (because  $K_d = N_{p\text{sed}}/N_{p\text{aq}}$ ).  $K_d$  values for the sandy sediment were calculated to be  $4.26 \pm 0.24 \text{ L kg}^{-1}$  and  $5.32 \pm 0.16 \text{ L kg}^{-1}$  for the centrifuged and filtrate samples, respectively (Figure 3.3). The data shows a stronger interaction of the Np with the clayey sediment than the sandy sediment. The stronger interaction of Np with the clayey sediment was likely due to the higher surface area of the clayey sediment and the relatively poor interactions of Np with silica surfaces which may dominate the sandy sediment (Righetto et al., 1991; Bertetti et al., 1998). The data also show that there is an increase in  $K_d$  with the filtration step. This behavior indicates that a small fraction of Np may be associated with colloidal particles in the 4-100 nm size range.



**Figure 3.1: Clayey Sediment Baseline Sorption Isotherm Data measured after 48 hr. [Np]<sub>0</sub> ranged from 0.1 ppb to 50 ppb. Sediment concentration of 25 g L<sup>-1</sup>. pH = 5.50±0.01. Measured  $K_d$  values of  $9.05 \pm 0.61 \text{ L kg}^{-1}$  and  $9.99 \pm 0.28 \text{ L kg}^{-1}$  for the centrifuged and filtered samples, respectively. Error determined using linear regression analysis of data to determine  $K_d$  values.**

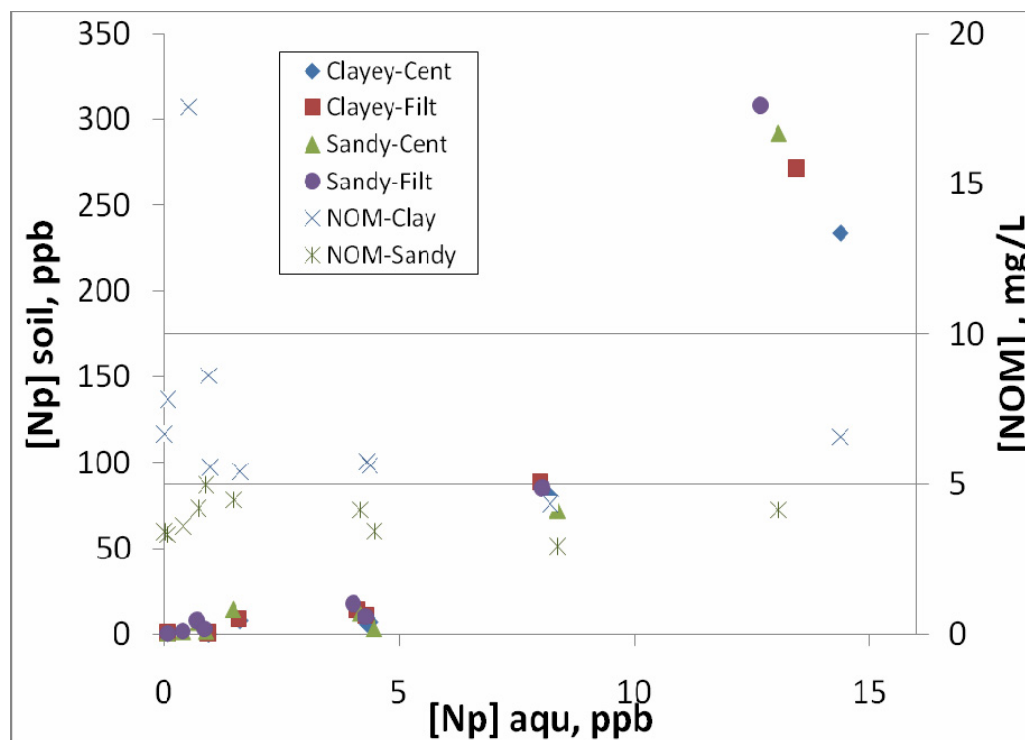


**Figure 3.2: Sandy Sediment Baseline Sorption Isotherm. Data measured after 48 hours.  $[Np]_0$  ranged from 0.1ppb to 50ppb. Sediment concentration of  $25 \text{ g L}^{-1}$ .  $\text{pH} = 5.50 \pm 0.03$ . Measured  $K_d$  values of  $4.26 \pm 0.24 \text{ L kg}^{-1}$  and  $5.32 \pm 0.16 \text{ L kg}^{-1}$  for the centrifuged and filtered samples, respectively. Error determined using linear regression analysis of data to determine  $K_d$  values.**

It has been shown that the sorption of Np onto amorphous silica and  $\gamma$ -alumina at low pH increases with increasing humic acid concentration (Righetto, et al. 1991). This behavior is attributed to the sediments becoming coated with the humic acids causing the surface charge to become more negative thus more strongly attracting the cationic  $\text{NpO}_2^+$ , thus forming a ternary metal-ligand-surface complex. There was evidence of this effect in the final NOM concentration. For most of the samples, the NOM concentration decreased from the initial concentration of  $10 \text{ mg L}^{-1}$  indicating sorption of NOM to the sediment surface. There was also increased sorption of Np to the sediments.

At low initial Np concentrations, " $[Np]_0$ ," there was little sorption of Np to the sediment (Figure 3.3). Overall, the addition of the NOM increased sorption of Np to the sediment. The effect of NOM addition was seen more with the sandy sediment than the clayey sediment, but this was seen only at the highest NOM concentration. However, pH control was an issue with these samples, so the higher pH for the sandy samples (5.7 versus 5.0 for the clayey sediments) could have added to the increase in sorption. Increased sorption with increasing pH is expected for metal cations and has been frequently observed for Np sorption to various solids (Righetto, et al. 1991; Girvin, et al. 1990). There was also an increase in  $K_d$  with the filtration step, again, a result of colloidal Np being removed from what was assumed to be aqueous Np ( $K_d = Np_{\text{sed}}/Np_{\text{aq}}$ ). This result indicates that there is small fraction of Np associated with colloidal particles in the 4-100 nm size range. However, it should be noted that the filtration step used in these experiments will likely remove some fraction of the NOM. The data shown in

Figure 3.3 indicate that the pH of the suspension has an impact on Np sorption as observed in previous studies (Righetto, et al. 1991; Girvin, et al. 1990; Bertetti et al., 1998).



**Figure 3.3: Effects of NOM on Np Sorption Isotherm Data measured after 48 hours. [Np]<sub>0</sub> ranged from 0.1ppb to 20ppb. [NOM]<sub>0</sub> = 10 mg L<sup>-1</sup>. Sediment concentration of 25 g L<sup>-1</sup>. pH = 5.02±0.22 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<sup>-1</sup> and were sampled after an equilibration period of 48 hours.**

The sorption isotherm in

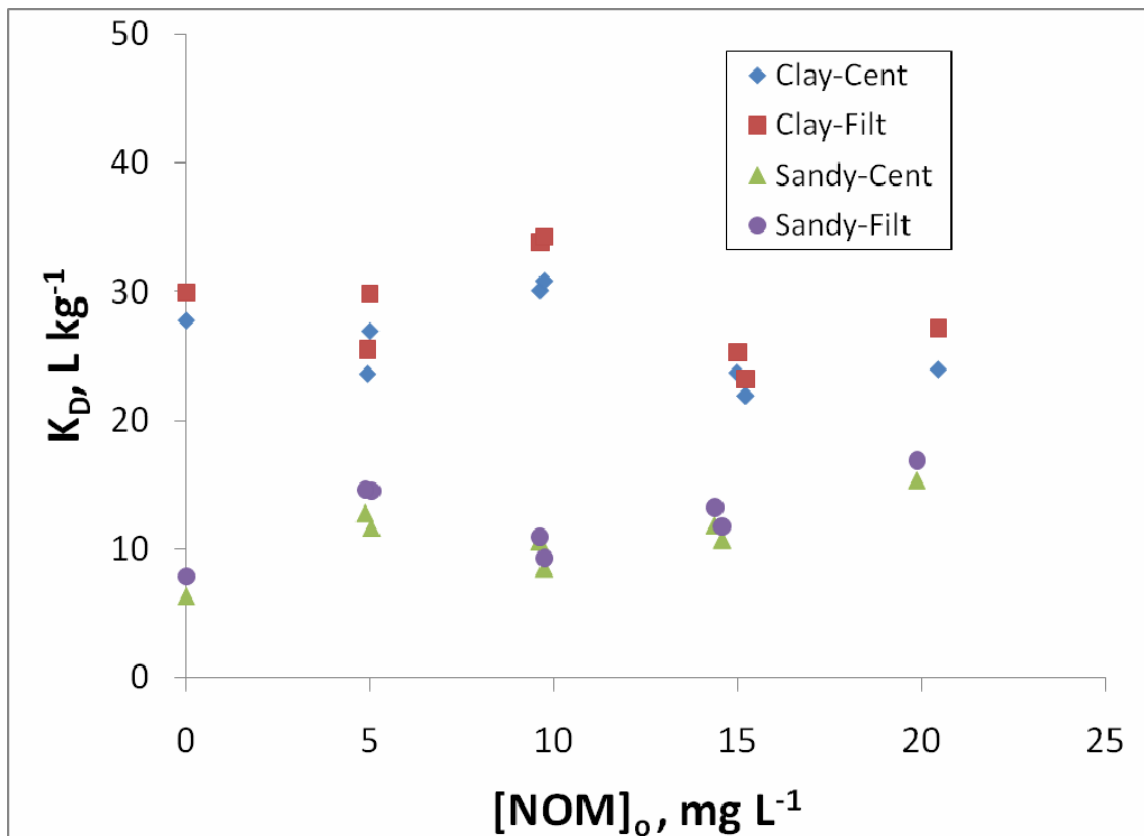
Figure 3.3 is, instead of being linear, exponential over this Np concentration range. This behavior indicates that the  $K_d$  construct is not appropriate over this entire range, 0 to 15 ppb. It also indicates that more complicating surface chemistry may be occurring. It is important to note that sorption sharply increases, not decreases, at higher Np concentrations. When competitive sorption occurs, then decreased sorption occurs and a plateau sorption occurs at elevated aqueous solute concentrations. However, in this study, the increase is not steep enough to suggest precipitation, but perhaps some early stages of precipitation.

### 3.1 Influence of NOM on Np Sorption to Sediments

To explore the effects of NOM on Np sorption further, an additional experiment was performed in which the concentration of NOM was varied and the initial Np concentration was held at 10 ppb, where Np sorption was elevated, but had not increased sharply. The resulting  $K_d$  values obtained from this experiment are reported in Figure 3.4. As a point of comparison, the  $K_d$  values for the clayey sediment at  $[Np]_0 = 10$  ppb are  $9.75 \pm 1.28$  L kg<sup>-1</sup> and  $11.09 \pm 1.35$  L kg<sup>-1</sup> for the centrifuged and filtered samples, respectively. For the sandy sediment,  $K_d$  values of  $8.62 \pm 1.38$  L kg<sup>-1</sup> and  $10.57 \pm 1.31$  L kg<sup>-1</sup> for the centrifuged and filtered samples, respectively (data from

Figure 3.3 above).





**Figure 3.4: Effects of Varying NOM concentrations on Np Sorption Isotherm Data measured after 48 hours. [Np]<sub>0</sub> = 10ppb. [NOM]<sub>0</sub> ranged from 0 – 20 mg L<sup>-1</sup>. Sediment concentration of 25 g L<sup>-1</sup>. pH = 5.55±0.10 for Clayey sediment and pH = 5.51±0.06 for Sandy sediment.**

The NOM initial concentration, [NOM]<sub>0</sub>, was varied from 0 – 20 mg L<sup>-1</sup> and [Np]<sub>0</sub> for all samples was 10 ppb. The data show no effect of [NOM] on K<sub>d</sub> values. The K<sub>d</sub> values for the clayey sediment were 25.78 ± 1.14 L kg<sup>-1</sup> and 28.19 ± 1.40 L kg<sup>-1</sup> for the centrifuged and filtered samples, respectively. For the sandy sediments, the K<sub>d</sub> values were 10.70 ± 0.98 L kg<sup>-1</sup> and 12.13 ± 1.07 L kg<sup>-1</sup> for the centrifuged and filtered samples, respectively. The increase for the clayey sediment K<sub>d</sub> from approximately 10 L kg<sup>-1</sup> is attributed to the increase in pH between the two experiments. The initial NOM experiment described in Figure 3.4 was performed at pH 5.0 and the experiment described in Figure 3.4 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 K<sub>d</sub> value. Again, this demonstrates a profound effect of pH on these systems that warrants further study.

The K<sub>d</sub> values measured in the presence of NOM were greater than those for the baseline study presented in Figure 3.1 and Figure 3.2. Therefore, the presence of NOM appears to increase sorption of Np. This result is likely due to the formation of ternary surface complexes as described by Schindler et al. (1990).

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) forming a further complex with the surface. This can be

shown as  $\equiv\text{S-O-M-L}$ , or, in this case,  $\equiv\text{S-O-Np-NOM}$ . Type B complexation is when the ligand acts as a bridging ligand in the ternary system. This can be shown as  $\equiv\text{S-O-L-M}$ , or, in this case,  $\equiv\text{S-O-NOM-Np}$ . Since 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 which results in increased Np sorption in the presence of NOM. This conclusion is also consistent with observed sorption of NOM to the solid phases at the pH ( $\sim 5.5$ ) of the batch experiments as shown in

Figure 3.3.

### 3.2 Influence of Redox Status on Np Sorption to Sediments

The purpose of the addition of reductants was to investigate the tendency of Np(V) reduction in sediments to Np(IV). Actinides follow the sorption affinity trend of:

$$\text{An(IV)} > \text{An(VI)} > \text{An(III)} < \text{An(V)} \quad (4)$$

with tetravalent actinides sorbing the strongest and pentavalent actinides sorbing the weakest. If Np(V) is reduced to Np(IV), Np sorption will increase, thus decreasing its subsurface mobility. Ascorbic acid, dithionide, 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. Compared to the baseline oxic clayey sediment that had a  $K_d$  value of  $9.05 \text{ L kg}^{-1}$  (Figure 3.1) and the sandy sediment, which had a  $K_d$  of  $4.26 \text{ L kg}^{-1}$  (Figure 3.2), the amount of reduction induced by the various treatments was relatively small (Table 3.1). The increases in sorption are consistent with the three reductants, ascorbic acid, dithionide, 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, this concentration was used to simulate potential environmental conditions.

**Table 3.1:  $K_d$  Values for Np Sorption under Reducing Conditions**

Treatment	$K_d$ Values ( $\text{L kg}^{-1}$ )	
	Clayey	Sandy
Control (No treatment)	$9.05 \pm 0.61^{(a)}$	$4.26 \pm 0.24^{(b)}$
Ascorbic Acid	$21.06 \pm 0.74$	$12.76 \pm 0.46$
Dithionide	$22.96 \pm 0.56$	$11.14 \pm 0.80$
Zero-valent Iron	$23.65 \pm 0.58$	$11.99 \pm 0.46$
Hydrogen Peroxide	$12.47 \pm 0.46$	$9.23 \pm 0.42$
Anaerobic Chamber <sup>(c)</sup>	$12.78 \pm 0.10$	$4.55 \pm 0.35$

<sup>(a)</sup> Data from Figure 3.1 for the centrifuged samples.

<sup>(b)</sup> Data from Figure 3.2 for the centrifuged samples.

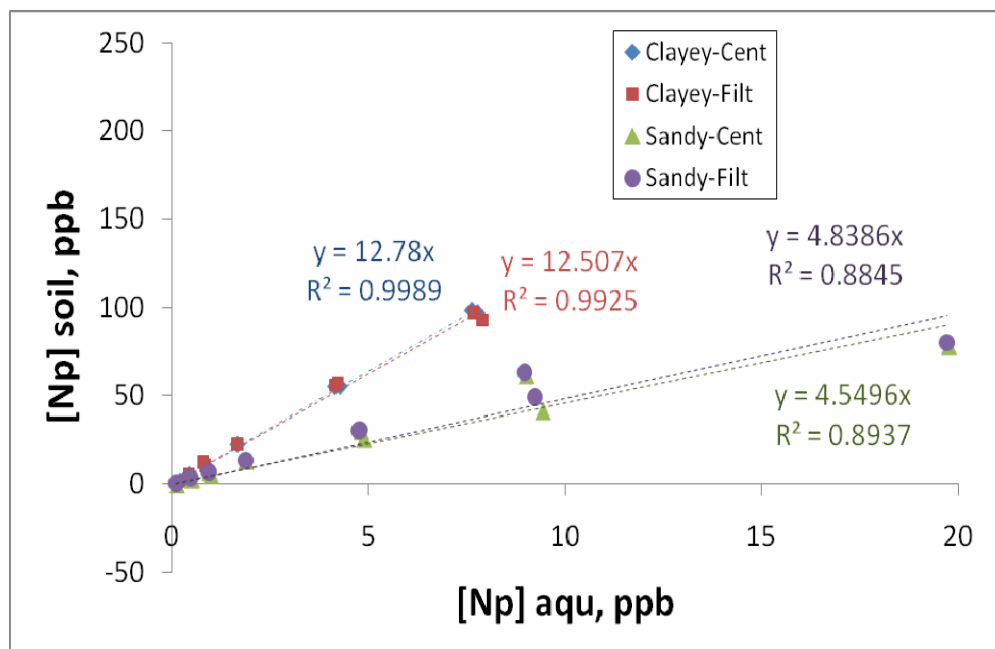
<sup>(c)</sup> Data from Figure 3.5 for the centrifuged samples

A ranking of the treatments by their redox status at the pH of the sediment, pH 5.5, was not measured but is expected to be:

$$\text{hydrogen peroxide} < \text{control} < \text{ascorbic acid} < \text{dithionite} < \text{zero-valent iron.} \quad (5)$$

Based on the  $K_d$  values in Table 3.1, certainly the oxidation did not occur by hydrogen peroxide. The ascorbic acid, dithionide, and zero-valent iron only promoted a small increase in sorption, and potentially only a small reduction in Np(V) to Np(IV).

Microbial respiration in subsurface environments may also 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 – 20 ppb with a fixed sediment concentration of 25 g  $L^{-1}$ . The  $K_d$  values for the clayey sediment were  $12.78 \pm 0.10$  L  $kg^{-1}$  and  $12.51 \pm 0.26$  L  $kg^{-1}$  for the centrifuged and filtered samples, respectively. For the sandy sediments, the  $K_d$  values were  $4.55 \pm 0.35$  L  $kg^{-1}$  and  $4.84 \pm 0.37$  L  $kg^{-1}$  for the centrifuged and filtered samples, respectively. There was little change in the  $K_d$  values from the baseline case. This behavior indicates that little or no reduction of Np(V) to Np(IV) occurred under anaerobic (reducing) conditions.



**Figure 3.5: 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  $L^{-1}$ . pH =  $5.51 \pm 0.06$  for Clayey sediment and pH =  $5.50 \pm 0.07$  for Sandy sediment. Measured  $K_d$  values of  $12.78 \pm 0.10$  L  $kg^{-1}$  and  $12.51 \pm 0.26$  L  $kg^{-1}$  for the Clayey sediment centrifuged and filtered samples, respectively. Measured  $K_d$  values of  $4.55 \pm 0.35$  L  $kg^{-1}$  and  $4.84 \pm 0.38$  L  $kg^{-1}$  for the Sandy sediment centrifuged and filtered samples, respectively.**

## 4.0 Summary and Conclusions

The general conclusions from this work are as follows:

- The baseline  $K_d$  for the clayey sediment was  $9.05 \pm 0.61 \text{ L kg}^{-1}$ 
  - The range of clayey sediment  $K_d$  values was 9.05 (baseline) to 33.9 (10 ppm NOM/100nm-filtered)  $\text{L kg}^{-1}$
- The baseline  $K_d$  for the sandy sediment was  $4.26 \pm 0.24 \text{ L kg}^{-1}$ 
  - The range of sandy sediment  $K_d$  values for all experiments was 4.26 (baseline) to 16.9 (20 ppm NOM/100nm-filtered)  $\text{L kg}^{-1}$
- There is a stronger interaction (sorption) of the Np with the clayey sediment than the sandy sediment.
  - Due to metals poor interaction with silica which comprises the majority of the sandy sediment
  - Greater surface area per mass for clayey sediment ( $15.07 \text{ m}^2/\text{g}$  vs.  $1.24 \text{ m}^2/\text{g}$ )
- There was an increase in the  $K_d$  value with an additional filtration step. This result may indicate that some Np is associated with 4 to 100nm-colloidal material particles
- The additional of Suwannee River NOM resulted in increased Np sorption
  - NOM had a greater effect on the  $K_d$  for the sandy sediment as compared with the clayey sediment.
  - Varying the NOM concentration did not appear to have a significant effect on Np sorption.
  - Changes in the solution pH between similar experiments in the presence of NOM indicated a profound effect of pH on the degree of Np sorption.
- Although minor increases in  $K_d$  values occurred, the addition of reductants or performing the experiments under anaerobic (reducing) conditions did not result in any substantial reduction of Np(V) to Np(IV). It is important to note here that reduction (or lack thereof) is inferred from observed sorption behavior, not from direct measurement of the Np oxidation state using spectroscopic or wet chemistry techniques.
- Based on these new measurements and the revelations about Np redox chemistry, the following changes to “Best  $K_d$ ” values, as defined in Kaplan (2006), for SRS performance assessment calculations are recommended.

	Previous Recommended “Best $K_d$ ” Values <sup>(a)</sup> ( $\text{L kg}^{-1}$ )	New Recommended “Best $K_d$ ” Values ( $\text{L kg}^{-1}$ )
Sandy Sediment	0.6	3
Clay Sediment	35 <sup>(b)</sup>	9
<sup>(a)</sup> Kaplan (2006; Geochemical Data Package, WSRC-TR-2006-00004)		
<sup>(b)</sup> An estimate that was based without experimental data.		

## 5.0 References

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## 6.0 Appendix: Data Tables

Below is the raw data obtained in this work and used to generate the above figures. For the sample descriptions, the first letter denotes the sediment used, C for clayey and S for sandy. The following letter indicates whether the sample was the centrifuged (C) sample or the filtered (F) sample. The initial Np concentration in each sample is noted as “[Np]<sub>o</sub>”. The “Mass Sediment” column indicates the amount of sediment in each sample. “[Np]<sub>aqu</sub>” is the measured aqueous phase concentration of Np after equilibration. “Mass liquid” gives the total mass of liquid in each sample including any liquid used to adjust the pH. “[Np]<sub>sediment</sub>” was calculated based on the [Np]<sub>o</sub>, [Np]<sub>aqu</sub>, Mass Liquid and Mass Sediment via equation 1. The K<sub>d</sub> was calculated using equation 2. The % Sorbed was calculated using equation 3. The pH values listed below indicate the pH reading after the equilibration period.

### 6.1 Baseline Sorption Data

**Table 6.1: Clayey Centrifugal Data from Baseline Sorption**

Sample	[Np] <sub>o</sub>	Mass Sediment (M <sub>sediment</sub> )	[Np] <sub>aqu</sub>	Mass Liq	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb	g	ppb	L kg <sup>-1</sup>		
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 6.2: Clayey Filtrate Data from Baseline Sorption**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Mass Liq	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb	g	ppb	L kg <sup>-1</sup>		
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 6.3: Sandy Centrifugal Data from Baseline Sorption**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Mass Liq	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb	g	ppb	L kg <sup>-1</sup>		
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 6.4: Sandy Filtrate Data from Baseline Sorption**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Mass Liq	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb	g	ppb	L kg <sup>-1</sup>		
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 6.5: Blank Sample Data from Baseline Sorption**

Sample	[Np] <sub>o</sub>	[Np] <sub>agu</sub>	Mass Liq	% Sorbed	pH
Units	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

## 6.2 NOM Experimental Data

**Table 6.6: Neptunium-NOM Clayey Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
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 6.7: Neptunium-NOM Clayey Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
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 6.8: Neptunium-NOM Sandy Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
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 6.9: Neptunium-NOM Sandy Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
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 6.10: Neptunium-Varying NOM Clayey Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	[NOM] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorption	pH
Units	ppb	mg L <sup>-1</sup>	g	ppb	g	ppb	L kg <sup>-1</sup>		
C-1-C	10.90	0.00	0.23	6.58	9.52	182.47	27.74	40%	5.55
C-2-C	11.35	4.92	0.24	7.09	9.48	167.20	23.58	38%	5.45
C-3-C	11.36	4.99	0.26	6.59	9.57	177.20	26.90	42%	5.60
C-4-C	10.60	9.62	0.25	6.12	10.10	184.05	30.06	42%	5.49
C-5-C	11.08	9.73	0.24	6.24	9.65	192.29	30.82	44%	5.44
C-6-C	11.16	14.97	0.24	7.01	9.68	165.90	23.67	37%	5.49
C-7-C	11.25	15.20	0.27	6.95	9.56	152.02	21.88	38%	5.66
C-8-C	11.24	20.44	0.25	6.94	9.54	166.01	23.93	38%	5.69

**Table 6.11: Neptunium-Varying NOM Clayey Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	[NOM] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorption	pH
Units	ppb	mg L <sup>-1</sup>	g	ppb	g	ppb	L kg <sup>-1</sup>		
C-1-F	10.90	0.00	0.23	6.38	9.52	190.90	29.93	41%	5.55
C-2-F	11.35	4.92	0.24	6.88	9.48	175.41	25.49	39%	5.45
C-3-F	11.36	4.99	0.26	6.30	9.57	187.97	29.85	45%	5.60
C-4-F	10.60	9.62	0.25	5.81	10.10	196.73	33.84	45%	5.49
C-5-F	11.08	9.73	0.24	5.95	9.65	203.72	34.23	46%	5.44
C-6-F	11.16	14.97	0.24	6.84	9.68	172.65	25.24	39%	5.49
C-7-F	11.25	15.20	0.27	6.79	9.56	157.60	23.21	40%	5.66
C-8-F	11.24	20.44	0.25	6.60	9.54	179.05	27.13	41%	5.69

**Table 6.12: Neptunium-Varying NOM Sandy Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	[NOM] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorption	pH
Units	ppb	mg L <sup>-1</sup>	g	ppb	g	ppb	L kg <sup>-1</sup>		
S-1-C	11.12	5.03	0.26	8.50	9.70	98.80	11.62	24%	5.56
S-2-C	11.10	4.86	0.24	8.36	9.54	106.67	12.76	25%	5.53
S-3-C	10.54	9.60	0.25	8.38	10.10	88.28	10.54	21%	5.49
S-4-C	10.81	9.73	0.26	8.84	9.86	74.96	8.48	18%	5.41
S-5-C	10.74	14.56	0.25	8.47	9.91	90.31	10.66	21%	5.48
S-6-C	10.79	14.36	0.25	8.33	9.97	98.50	11.83	23%	5.49
S-7-C	11.10	19.86	0.24	8.02	9.63	122.79	15.31	28%	5.61
S-8-C	11.15	0.00	0.25	9.56	9.58	60.45	6.32	14%	5.49

**Table 6.13: Neptunium-Varying NOM Sandy Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	[NOM] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorption	pH
Units	ppb	mg L <sup>-1</sup>	g	ppb	g	ppb	L kg <sup>-1</sup>		
S-1-F	11.12	5.03	0.26	8.03	9.70	116.52	14.51	28%	5.56
S-2-F	11.10	4.86	0.24	8.07	9.54	117.86	14.60	27%	5.53
S-3-F	10.54	9.60	0.25	8.31	10.10	91.06	10.96	21%	5.49
S-4-F	10.81	9.73	0.26	8.68	9.86	80.97	9.32	20%	5.41
S-5-F	10.74	14.56	0.25	8.30	9.91	97.42	11.74	23%	5.48
S-6-F	10.79	14.36	0.25	8.11	9.97	107.35	13.24	25%	5.49
S-7-F	11.10	19.86	0.24	7.79	9.63	131.77	16.91	30%	5.61
S-8-F	11.15	0.00	0.25	9.23	9.58	72.84	7.89	17%	5.49

### 6.3 Reductant Addition Data

**Table 6.14: Reductant Addition Clayey Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] sediment	K <sub>d</sub>	% Sorption	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
Ascorbic Acid	10.56	0.26	6.78	9.83	142.87	21.06	36%	5.30
Zero Valent Iron	11.23	0.24	7.02	9.66	166.13	23.65	37%	5.29
Dithionide	11.39	0.24	7.22	9.59	165.85	22.96	37%	5.39
Hydrogen Peroxide	10.84	0.26	8.13	9.71	101.39	12.47	25%	5.28

**Table 6.15: Reductant Addition Clayey Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] sediment	K <sub>d</sub>	% Sorption	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
Ascorbic Acid	11.06	0.25	8.36	9.68	106.63	12.76	24%	5.30
Zero Valent Iron	10.81	0.25	8.40	10.28	100.76	11.99	22%	5.29
Dithionide	11.04	0.24	8.61	9.68	95.98	11.14	22%	5.39
Hydrogen Peroxide	10.99	0.25	8.87	9.72	81.86	9.23	19%	5.28

**Table 6.16: Reductant Addition Sandy Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aq</sub>	Liq Mass	[Np] sediment	K <sub>d</sub>	% Sorption	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
Ascorbic Acid	10.56	0.26	6.41	9.83	157.14	24.53	39%	5.84
Zero Valent Iron	11.23	0.24	6.80	9.66	175.05	25.75	39%	5.70
Dithionide	11.39	0.24	6.94	9.59	176.97	25.49	39%	5.73
Hydrogen Peroxide	10.84	0.26	8.01	9.71	105.90	13.22	26%	5.63

**Table 6.17: Reductant Addition Sandy Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aq</sub>	Liq Mass	[Np] sediment	K <sub>d</sub>	% Sorption	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
Ascorbic Acid	11.06	0.25	8.33	9.68	107.50	12.90	25%	5.84
Zero Valent Iron	10.81	0.25	8.31	10.28	104.50	12.57	23%	5.70
Dithionide	11.04	0.24	8.68	9.68	93.46	10.77	21%	5.73
Hydrogen Peroxide	10.99	0.25	8.85	9.72	82.44	9.31	19%	5.63



#### 6.4 Anaerobic Data

**Table 6.18: Anaerobic Glovebox Clayey Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
C-1-C	0.25	0.26	0.21	9.60	1.47	6.99	16%	5.44
C-2-C	0.59	0.26	0.44	9.36	5.47	12.57	26%	5.51
C-3-C	1.14	0.27	0.85	9.59	10.56	12.48	26%	5.42
C-4-C	1.14	0.26	0.85	9.58	10.47	12.27	25%	5.48
C-5-C	2.28	0.26	1.67	9.61	22.65	13.55	27%	5.58
C-6-C	5.65	0.26	4.15	9.67	55.04	13.26	26%	5.59
C-7-C	5.73	0.25	4.28	9.50	55.10	12.88	25%	5.56
C-8-C	10.12	0.25	7.64	9.99	98.45	12.89	25%	5.53
C-9-C	10.41	0.26	7.78	9.74	97.11	12.48	25%	5.50
C-10-C	21.23	0.26	13.75	9.57	280.66	20.42	35%	5.46

**Table 6.19: Anaerobic Glovebox Clayey Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
C-1-F	0.25	0.26	0.21	9.60	1.36	6.38	15%	5.44
C-2-F	0.59	0.26	0.43	9.36	5.71	13.32	27%	5.51
C-3-F	1.14	0.27	0.84	9.59	10.65	12.61	26%	5.42
C-4-F	1.14	0.26	0.79	9.58	12.77	16.15	31%	5.48
C-5-F	2.28	0.26	1.67	9.61	22.89	13.74	27%	5.58
C-6-F	5.65	0.26	4.14	9.67	55.64	13.45	27%	5.59
C-7-F	5.73	0.25	4.23	9.50	56.97	13.47	26%	5.56
C-8-F	10.12	0.25	7.67	9.99	97.05	12.65	24%	5.53
C-9-F	10.41	0.26	7.90	9.74	92.75	11.75	24%	5.50
C-10-F	21.23	0.26	13.75	9.57	280.70	20.42	35%	5.46

**Table 6.20: Anaerobic Glovebox Sandy Sediment Centrifuged Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
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 6.21: Anaerobic Glovebox Sandy Sediment Filtrate Data**

Sample	[Np] <sub>o</sub>	Mass Sediment	[Np] <sub>aqu</sub>	Liq Mass	[Np] <sub>sediment</sub>	K <sub>d</sub>	% Sorbed	pH
Units	ppb	g	ppb		ppb	L kg <sup>-1</sup>		
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

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