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Estimated Neptunium Sediment Sorption Values as a Function of pH and Measured Barium and Radium K_d Values

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EXECUTIVE SUMMARY

The objective of this document is to provide traceability and justification for a select few new geochemical data used in the Special Analysis entitled "Special Analysis for the Dose Assessment of the Final Inventories in Center Slit Trenches One through Five" (Collard et al. 2010; SRNL-STI-2010-00760). Most values used in the Special Analysis came from the traditional geochemical data package (Kaplan 2010; SRNL-STI-2009-00473); however, some recent laboratory measurements have made it possible to estimate barium K_d values. Additionally, some recent calculations were made to estimate neptunium K_d values as a function of pH. The assumptions, justifications, and calculations needed to generate these new values are presented in this document, and the values are summarized below.

Rad	Best Sand K_d	Best Clay K_d	High-pH Solution Impact Factor, f(High-pH)[a]
	(mL/g)	(mL/g)	(unitless)
Ba	15	101	NC[b]
Ra	25	185	NC[b]
Np	NC[b]	NC[b]	20[c]
Np pH 5.5	3	9	--
Np pH 6	6	18	--
Np pH 6.5	18	54	--
Np pH 7	30	90	--
Np pH 7.5	42	126	--
Np pH 8	50	151	--
Np pH 8.5	55	166	--
Np pH 9	60	180	--
Np pH 10	60	180	--
Np pH 11	60	180	--
Np pH 12	60	180	--
Np pH 12.5	55	166	--

[a] $f(\text{High-pH}) = K_{d\text{High-pH}}/K_d$

[b] NC = no change in value with respect to the value reported in the original geochemical data package (Kaplan 2010; SRNL-STI-2009-00473)

[c] By chemical analogy to Np, Pa as PaO_2^+ can be assigned the same "High-pH Solution Impact Factor, f(High-pH)" of 20 (unitless).

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LIST OF ABBREVIATIONS

K_d	Distribution Coefficient
SRNL	Savannah River National Laboratory
SRS	Savannah River Site

1.0 INTRODUCTION

1.1 Objectives

The objectives of this report are twofold.

1. Provide estimates of neptunium (Np) distribution coefficient (K_d) values as a function of pH, which will simulate conditions following *in situ* high-pH injection treatment of a SRS slit trench.
2. Explain sources of new barium (Ba) and radium (Ra) K_d values.

1.2 Np K_d Values as a Function of pH in a High-pH Treated Slit Trench Waste Zone

Np K_d values in SRS sediments were recently measured by Powell et al. (2010a) who reported average values in sandy SRS sediments of 3 mL/g and average values in clayey sediments of 9 mL/g.

Powell (2010) measured Np sorption to an aluminum oxide, boehmite, versus pH (Figure 1-1[A]). Three important features about these data and their implications to SRS data are as follows:

- The NpO_2^+ sorption curve follows the typical behavior of a monovalent in that it had the characteristic sigmoidal curve that showed an increase in sorption with increases in pH; >99% Np sorption was observed between pH 10 and 12.
- A drop in Np sorption is beyond pH 12.4.
- Boehmite is not a common soil mineral, but its sorption behavior relative to metals is similar to that of other iron and aluminum oxyhydroxides (Dixon and Weed 1989). It differs from SRS sediments in the degree of sorption; i.e., the line may shift to the right or left depending on the "point-of-zero-charge" of the solid phase and the height of the sorption maximum. As such, this data must be adjusted for SRS conditions.

The upper plot of Figure 1-1 shows sorption of Np; the experiment was conducted by introducing aqueous Np(V) at various pH conditions to the solid phase, boehmite. Powell has since repeated this Np(V) sorption experiment with boehmite between pH 3.7 and pH 11.0 and under slightly different experimental conditions (Powell et al. 2010b). Though the follow-up study was conducted under slightly different conditions, the results are near identical to those from the original study. The lower plot in Figure 1-1 shows the solubility of Np as a function of pH and was conducted with a solid Np phase. The solubility decreases from pH 6 to 11 and then stabilizes at pH 12, after which it starts to increase.

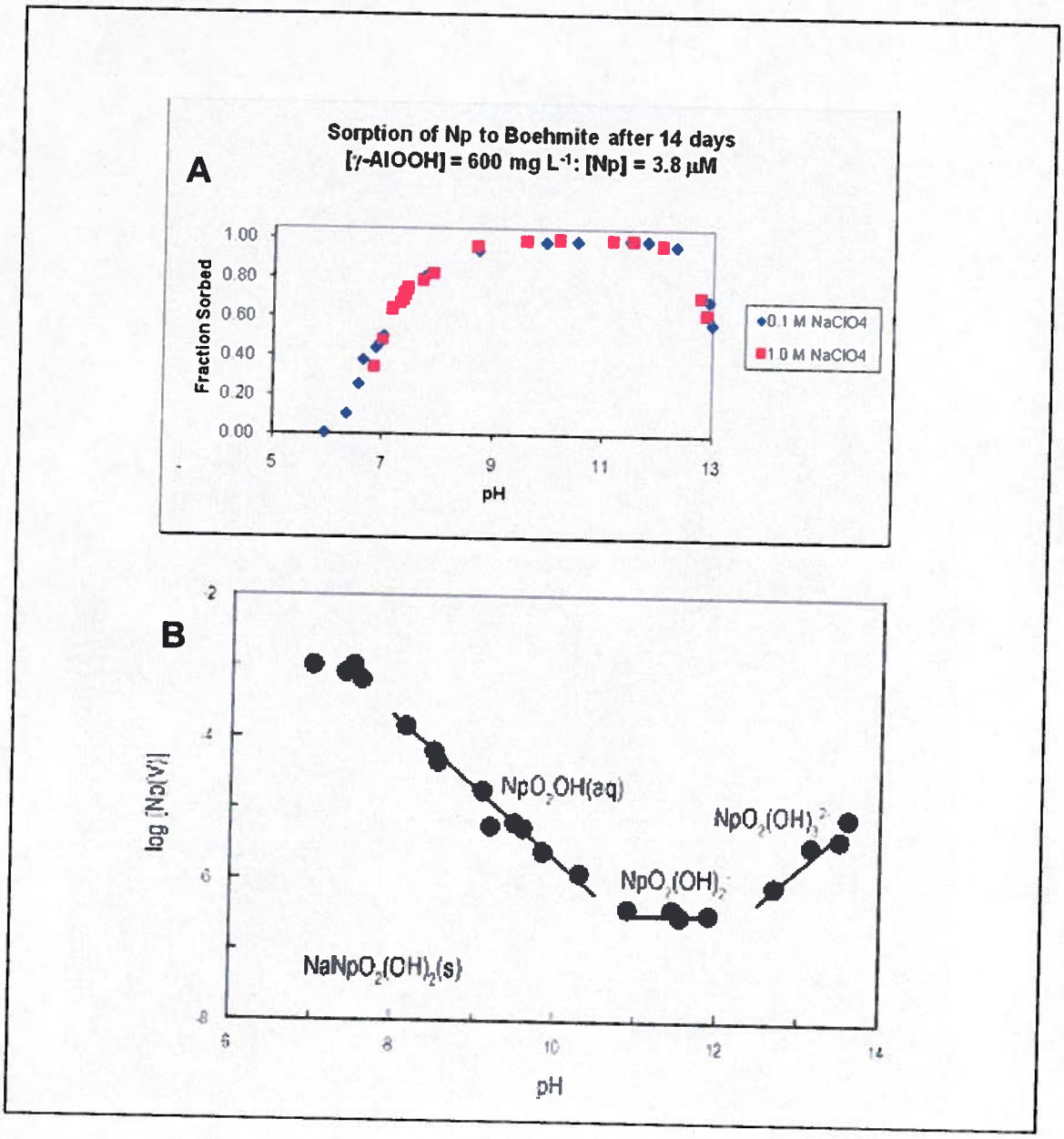

Figure 1-1. (A) Fraction of Aqueous Np(V) Sorbed to Boehmite as a Function of pH, I = 0.1 and 1.0 M $NaClO_4$, (B) Concentration of Aqueous Np(V) as a Function of pH with NpO_2OH(s) in Solution

Note: Figure 1A from Powell 2010
Figure 1B from Rao et al. 2006

Wong et al. (2009) recently conducted a literature review of Np sorption to cementitious materials. A list of data is presented in Table 1-1. All data seem to indicate that uptake of neptunium on cement materials increases with contact time and is relatively independent of the redox conditions of the experiments. Hoglund et al. (1985) and Bayliss et al. (2000) found that cement/concrete composition had little influence on K_d values.

Sylwester et al. (2000) provided spectroscopic evidence that Np(V), the mobile form of neptunium, is first sorbed on cement followed by reduction to Np(IV), the far less mobile form of neptunium. The mechanism responsible for reduction of neptunium was unclear (no reducing agents were added in the experiments and reducing capacity of the cement is not known). But a very important discovery made was only about six months was required for about half of the Np(V) to be reduced (immobilized) to Np(IV). This same research group published very similar results with a little extra data in a second manuscript by Zhao et al. (2000).

When time is not limiting sorption, neptunium K_d values are typically >1000 mL/g (Table 1-1). This behavior is certainly more characteristic of stronger sorbing Np(IV) than that of Np(V) and is consistent with the spectroscopic findings of Sylwester et al. (2000) and Zhao et al. (2000) that demonstrated that Np(V) reduces to Np(IV).

The above studies link Np reduction to sorption on cementitious solids. However, the transition to more stable Np species is a function of pH, irrespective of the solid phase. Np(IV) is thermodynamically more stable at high pH under the equilibrium redox conditions found in the SRS subsurface. Redox potential in the SRS environment typically ranges from 0.2 to 0.4 volts in pH ~5 groundwater. Figure 1-2 is an Eh-pH diagram of Np. The speciation of Np under high pH conditions in SRS soils is approximately represented by the region within the blue rectangle. Note that redox potential decreases as pH is increased, leading to the reduced Np(IV) form becoming the more stable species. Given these conditions, it is recommended that Np K_d values be increased under high pH conditions.

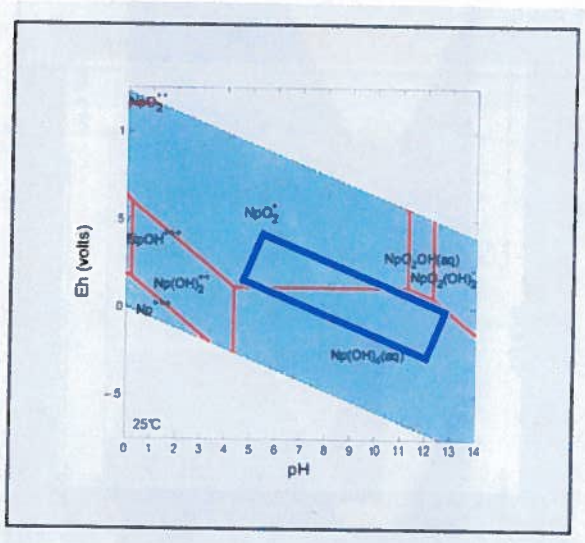

Figure 1-2. Eh-pH Diagram of Neptunium (Np activity 10^{-8} M)

Note: Calculated from Geochemist Workbench, Wong et al. 2009

Table 1-1. Np K_d Values in Cementitious Materials (Wong et al. 2009)[a]

REFERENCE	EXPERIMENTAL CONDITIONS						
	Cementitious Material	**S/L (kg/L)**	**Initial Concentration [M]**	**Contact Solution**	**pH**	**Equilibration Time**	**K_d (mL/g)**
Allard et al. (1984)	Crushed Concretes	0.02	1.9×10^{-7} as Np(V)	Artificial Pore Waters	12 to 13.6	100 d	2,000 to 30,000 Np(V)
Hoglund et al. (1985)	Cements	0.02	1.9×10^{-7} as Np(V)	Artificial Pore Waters	12.5 to 13.4	6 hr to 90 d	300 to 20,000 Np(V)
Bayliss et al. (1996)	BFS/OPC	0.02	10^{-9} as Np(V)	Equilibrated Water (Reducing)	11.8-12.2	1-68 d	5,300 to > 10^6 Np(IV)
Bayliss et al. (1996)	BFS/OPC	0.02	10^{-9} as Np(V)	Equilibrated Water (Aerobic)	11.8-12.2	1-28 d	9,000 to 140,000 Np(IV,V)
Bayliss et al. (2000)	NRVB	0.02	2.5×10^{-10} as Np(IV)	NRVB Equilibrated Water	12.5	23 d	60,000 Np(IV,V)
Bayliss et al. (2000)	NRVB	0.02	2.5×10^{-10} as Np(IV)	NRVB Equilibrated Saline Water	12.5	23 d	200,000 Np(IV,V)

[a] S/L = Solid/Liquid, BFS/OPC = Blast Furnace Slag/Ordinary Portland Cement, NRVB = Nirex Reference Vault Backfill

2.0 RESULTS

2.1 Estimating Neptunium K_d Values as a Function of pH

The following assumptions were made in deriving an estimate of Np K_d values as a function of pH:

1. **The measured K_d for SRS sandy sediment of 3 mL/g and Clay K_d of 9 mL/g (measured in Powell et al. 2010) serves as the lower bound estimate under natural background conditions.**
2. **The upper range K_d's in a high-pH solution environment are assumed to be 20x greater than the lower bound set at 3 mL/g for sandy sediment and 9 mL/g for clayey sediment.** The "20x" value was selected to produce a maximum K_d value of 60 and 180 in sandy and clayey sediments, respectively, to reflect the large increase in sorption observed when Np enters a high pH environment. These values are lower than all values reported in the literature and are therefore conservative for most modeling scenarios (Figure 1-1 and Table 1-1).
3. **Np sorption to SRS soils is assumed to be similar to boehmite.** Experimental sorption data of Np(V) onto boehmite (alpha-AlOOH) by Powell (2010) at 0.1 and 1.0 M $NaClO_4$ (Figure 1-1) were used to provide fraction sorbed as a function of pH. In Figure 1-1, Powell shows essentially complete sorption between pH 10–12.4. This is reinforced by additional experimental data in Powell et al. (2010b) which is provided in Appendix B. Starting with the lower bound K_d measurements (i.e., 3 mL/g for sandy sediment and 9 mL/g for clayey sediment) the maximum sorption at high pH (10 – 12.4) was assumed to be 20x these values (i.e., 60 mL/g for sandy sediment and 180 mL/g for clayey sediment). The K_d's at other pH values were obtained by scaling these maximum values by the fraction sorbed from the Powell et al. (2010a) data. The resulting K_d data are shown in Figure 2-1 and Table 2-1.

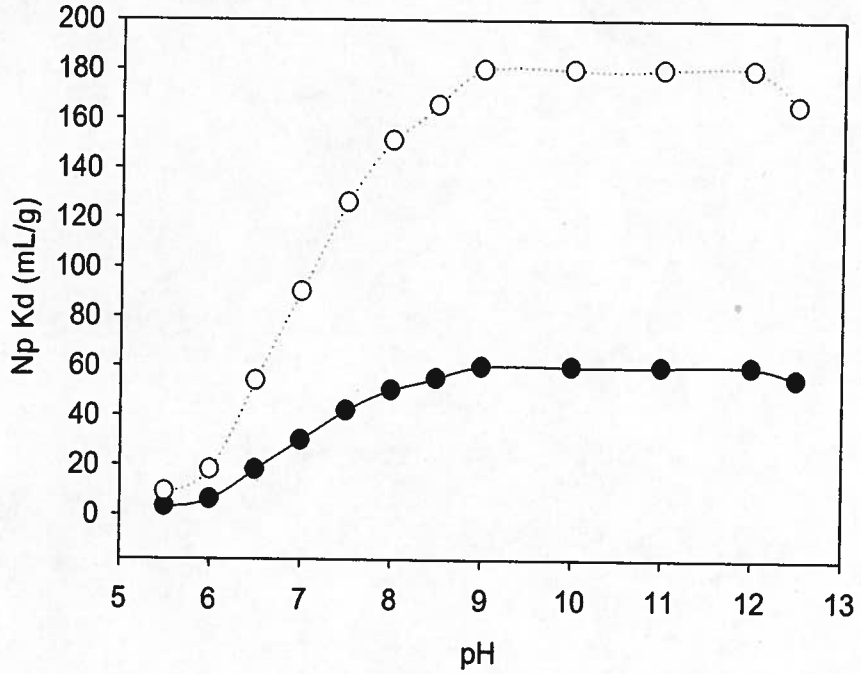

Figure 2-1. Estimated K_d Values for SRS Conditions Based on Measured K_d Value at pH 5.5

Note: Filled circles represent sandy sediment and open circles clayey sediment Np(V) K_d values

Table 2-1. Estimation of Np K_d Values as a Function of pH Using Literature Fraction Sorbed Data and Site Specific Sorption Data (Data used in Figure 2-1)

pH	Fraction Sorbed[a] (Np_{sorb}/Np_{total})	Sand Np K_d (mL/g)	Clay Np K_d (mL/g)
5.5	NA	3[(b)]	9[(b)]
6	0.1	6	18
6.5	0.3	18	54
7	0.5	30	90
7.5	0.7	42	126
8	0.84	50	151
8.5	0.92	55	166
9	0.995	60	180
10	0.995	60	180
11	0.995	60	180
12	0.995	60	180
12.5	0.92	55	166

[a] Powell (2010)
[b] Measured values

2.2 Barium and Radium K_d Values

In the past, the SRS performance assessments have always assumed all IIA elements in the periodic table had the same K_d values as Sr, for which SRS has several good measurements. SRS made this assumption because no actual measurements had been made with the other elements, Be, Mg, Ca, (Sr), Ba, and Ra, listed in increasing atomic weight and, assumed, sorptive strength to soil. As noted in this list, barium exists between Sr and Ra in the periodic table, and based on known first principles of chemical behavior related to its periodicity (where it exists on the periodic chart in relation to other elements), SRS can assume that sorption behavior of Ba will be between that of Sr and Ra (Sr, Ba, and then Ra are directly on top of one another on the periodic chart). Kaplan (2010) reported Sr K_d values of 5 mL/g and 17 mL/g for sand and clay, respectively. Powell et al. (2010a) recently measured similar values. They also measured new Ra K_d values (Table 2-2). From the Sr and Ra K_d values, Ba values were estimated assuming their magnitude were in the middle of the K_d values of Sr and Ra. The result was a Ba sandy sediment K_d value of 15 mL/g and Ba clayey sediment K_d value of 101 mL/g (Table 2-2).

Table 2-2. Recommended K_d Values Based on These Experimental Results Compared with Previously Recommended K_d Values Used in SRS Performance Assessments (Kaplan 2010)

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; K_d's are highly dependent on ionic strength (Kaplan 2010).
Ba	15	101	5	17	Estimated based on Sr and Ra measured K_d values. Ba K_d values were estimated to be midway between Sr and Ra K_d values.
Ra	25	185	5	17	Ra K_d (ionic strength, ~0.02 M, which approximates that of SRS groundwater) (Powell et al. 2010a)

3.0 CONCLUSIONS

Some new geochemical parameters have been identified in this document. The geochemical values are new to this document or were issued since the writing of the last geochemical data package (Kaplan et al. 2010). The database used to build the sediment Kd table in the geochemical data package (Kaplan et al. 2010) has been updated with this new information (Geochem Data Package 09 v2c.xls) and inserted into Appendix A.

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APPENDIX A: Sediment Sorption Constant Table From Database "Geochem Data Package 09 v2c.xls"

Sediment Sorption Constant Table

Rad	(mL/g) Best Sand K_d	(mL/g) Best Clay K_d	(mL/g) Best Sand K_d(CemLech)	(mL/g) Best Clay K_d(CemLech)	(mL/g) Best Sand K_d(CDP)	(mL/g) Best Clay K_d (CDP)	(mL/g) Best Cement Leachate with CDP Sand K_d	(mL/g) Best Cement Leachate with CDP Clay K_d	(unitless) Cement Leachate Impact Factor, f(CemLech)
Ac	1100	8500	1650	12750	605	4675	1650	12750	1.5
Ag	60	150	192	480	60	150	192	480	3.2
Al	1300	1300	1950	1950	715	715	1950	1950	1.5
Am	1100	8500	1650	12750	605	4675	1650	12750	1.5
Ar	0	0	0	0	0	0	0	0	1
As	100	200	140	280	100	200	100	200	1.4
At	0.3	0.9	0.0	0.1	0.3	0.9	0.3	0.9	0.1
Ba	15[a]	101[a]	45	303	15	101	15	101	3
Bi	1100	8500	1650	12750	605	4675	1650	12750	1.5
Bk	1100	8500	1650	12750	605	4675	1650	12750	1.5
C	10	400	50	2000	10	400	10	400	5
Ca	5	17	15	51	5	17	5	17	3
Cd	15	30	45	90	15	30	45	90	3
Ce	1100	8500	1650	12750	605	4675	1650	12750	1.5
Cf	1100	8500	1650	12750	605	4675	1650	12750	1.5
Cl	0	0	0	0	0	0	0	0	0.1
Cm	1100	8500	1650	12750	605	4675	1650	12750	1.5
Co	40	100	128	320	40	100	128	320	3.2
Cr	4	10	6	14	4	10	4	10	1.4
Cs	10	50	10	50	10	50	10	50	1
Cu	50	70	160	224	50	70	160	224	3.2
Eu	1100	8500	1650	12750	605	4675	1650	12750	1.5
F	0	0	0	0	0	0	0	0	0.1
Fe	200	400	300	600	110	220	300	600	1.5
Fr	10	50	10	50	10	50	10	50	1
Gd	1100	8500	1650	12750	605	4675	1650	12750	1.5
H	0	0	0	0	0	0	0	0	1
Hg	800	1000	2560	3200	800	1000	2560	3200	3.2
I	0.3	0.9	0.0	0.1	0.3	0.9	0.3	0.9	0.1
K	5	25	5	25	5	25	5	25	1
Kr	0	0	0	0	0	0	0	0	1
Lu	1100	8500	1650	12750	605	4675	1650	12750	1.5
Mn	15	200	21	280	15	200	15	200	1.4
Mo	1000	1000	1400	1400	1000	1000	1000	1000	1.4

Sediment Sorption Constant Table-continued

Rad	(mL/g) Best Sand K_d	(mL/g) Best Clay K_d	(mL/g) Best Sand K_d(CemLech)	(mL/g) Best Clay K_d(CemLech)	(mL/g) Best Sand K_d(CDP)	(mL/g) Best Clay K_d (CDP)	(mL/g) Best Cement Leachate with CDP Sand K_d	(mL/g) Best Cement Leachate with CDP Clay K_d	(unitless) Cement Leachate Impact Factor, f(CemLech)
N	0	0	0	0	0	0	0	0	0.1
Na	5	25	5	25	5	25	5	25	1
Nb	0	0	0	0	0	0	0	0	1.4
Ni	7	30	22	96	7	30	22.4	96	3.2
Np	3	9	60	180	3	9	60	180	20 [b]
Pa	3	9	60	180	3	9	60	180	20 [b]
Pb	2000	5000	6400	16000	2000	5000	6400	16000	3.2
Pd	7	30	22	96	7	30	22	96	3.2
Po	2000	5000	4000	10000	2000	5000	4000	10000	2
Pt	7	30	22	96	7	30	22.4	96	3.2
Pu(combo)	290	5950	580	11900	290	5950	580	11900	2
Pu(III/IV)	300	6000	600	12000	300	6000	600	12000	2
Pu(V/VI)	16	5000	32	10000	16	5000	16	5000	2
Ra	25 [a]	185 [a]	75	555	25	185	25	185	3
Rb	10	50	10	50	10	50	10	50	1
Re	0.6	1.8	0.1	0.2	0.6	1.8	0.6	1.8	0.1
Rn	0	0	0	0	0	0	0	0	1
Sb	2500	2500	3500	3500	2500	2500	2500	2500	1.4
Sc	1000	1000	1400	1400	1000	1000	1000	1000	1.4
Sm	1100	8500	1650	12750	605	4675	1650	12750	1.5
Sn	2000	5000	6000	15000	2000	5000	6000	15000	3
Sr	5	17	15	51	5	17	5	17	3
Tc	0.6	1.8	0.1	0.2	0.6	1.8	0.6	1.8	0.1
Te	1000	1000	1400	1400	1000	1000	1000	1000	1.4
Th	900	2000	1800	4000	900	2000	1800	4000	2
Tl	10	50	10	50	10	50	10	50	1
U	200	300	600	900	200	300	600	900	3
Y	1100	8500	1650	12750	605	4675	1650	12750	1.5
Zn	15	30	45	90	15	30	45	90	3
Zr	900	2000	1800	4000	900	2000	1800	4000	2

FOOTNOTES

[a] SRNL-STI-2010-00527; Iodine Neptunium, Radium, and Strontium Sorption to Savannah River Site Sediments

[b] New data from this document

APPENDIX B: Figure of Np(V) Sorption to Boehmite as a Function of pH 3.7 to 11.0 (Powell et al. 2010b)

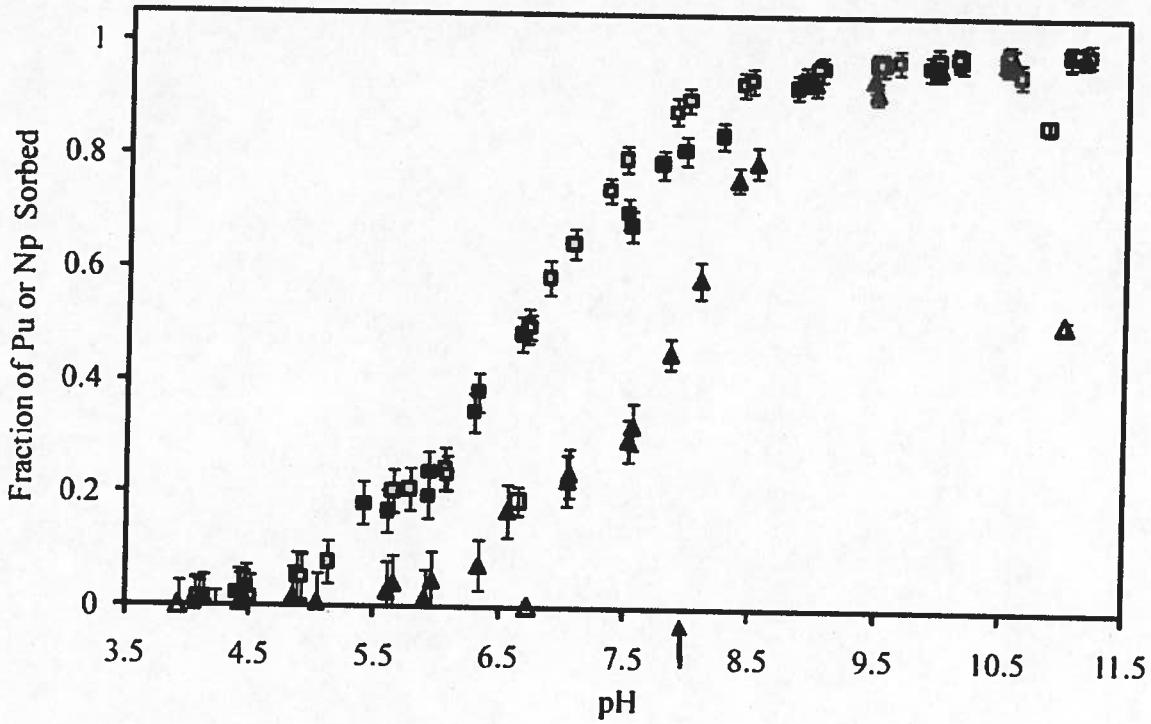

Figure of Sorption of Pu(V) (■,□) and Np(V) (▲) on boehmite after ten days in the absence of a-Hydroxyethane-1,1-diphophonic Acid (HEDPA). Open symbols represent boehmite-free control solutions. Background solution was 1M NaCl (■) or 1M $NaClO_4$ (□). The bold arrow indicates the boehmite point-of-zero-salt-effect. Additional solution conditions: [γ-AlOOH] = 660mgL^{-1}; [Pu(V)] = 2.1 μM; [Np(V)] = 10.6 μM.

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