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Distribution Coefficient Values Describing Iodine, Neptunium, Selenium, Technetium, and Uranium Sorption to Hanford Sediments

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Summary

Burial of vitrified low-level waste (LLW) in the vadose zone of the Hanford Site is being considered as a long-term disposal option. Regulations dealing with LLW disposal require that performance assessment (PA) analyses be conducted. Preliminary modeling efforts for the Hanford Site vitrified LLW PA have identified ^{129}I, ^{237}Np, ^{79}Se, ^{99}Tc, and ^{234,235,238}U as the waste components posing the greatest potential health hazard. The purpose of this document is 1) to compile and describe statistically the available Hanford Site K_d data for I, Np, Se, Tc, and U, and 2) use these data to provide a best estimate of each radionuclide K_d value for future modeling efforts.

K_d values were compiled into four categories of alkaline, low organic material solutions. These categories were based on the ionic strength (a measure of the total concentration of salts in solution) and redox status (the potential of a system to donate or accept electrons) of the system. The redox status was broadly defined as oxic, a system in which ferric (Fe III) minerals exists (Eh > 0.17 V), and anoxic, a system in which ferrous (Fe II) minerals exists (Eh ≤ 0.17). The four categories of alkaline, low organic material solutions were 1) low ionic strength and oxic solutions, 2) low ionic strength and anoxic solutions, 3) high ionic strength and oxic solutions, and 4) high ionic strength and anoxic solutions. Category 1 was selected to approximate the plume in the far field. The best estimate K_d value (in units of mL/g) in Category 1 for I was 3, for Np 15, for Se 0, for Tc 0, and for U 0.6. Category 2 was selected to approximate the plume in a near-field barrier where an anoxic environment exist, (such as a sulphur-polymer cement barrier, a mixture of 95-percent sulfur and 5-percent organic binders). The best estimate K_d value (in units of mL/g) in Category 2 for I was 3, for Np 200, for Se 0, for Tc 200, and for U 100. Category 3 was selected to approximate the plume in a near-field barrier that created a high ionic strength/oxic environment (such as vitrified glass or concrete vault leachate). The best estimate K_d value (in units of mL/g) in Category 3 for I was 0.5, for Np 2, for Se 0, for Tc 0, and for U 0. Category 4 was selected to approximate the plume in a near-field barrier that created a high ionic strength and anoxic environment. The best estimate K_d value (in units of mL/g) in Category 4 for I was 0, for Np 100, for Se 0, for Tc 200, and for U 100. Broad definitions were chosen for these categories because the precise burial site and burial facility have not been identified. Refinement of these categories will likely be required as more information about the burial design becomes available.

The best estimate K_d values were based on laboratory derived values generated in all cases from the <2-mm sediment fraction; hence the diminutive effect of gravel-size material on K_d values is not reflected in these values. It is well known that large portions of the Hanford Site's vadose zone contain large amounts of gravel-sized material. Thus it is important to note that these best estimate K_d values, when used in transport codes, provide a maximum. As a first approximation, it can be assumed that the gravel does not participate in sorption due to its low specific surface area (i.e., low surface area per mass of material). For example, if the sediment to be modelled contains 70-percent gravel and its <2-mm fraction K_d value is 2 mL/g, then the "gravel-corrected K_d value" would be 0.6 mL/g [(1.00 - 0.70) x 2 mL/g = 0.6 mL/g].

An unexpected result from the literature review was that I sorbed to every Hanford sediments tested. Iodine K_d values ranged from 0.7 to 15 mL/g and had a median of 7 mL/g. This finding may have significant implications for the LLW-PA because it suggests that the most conservative estimate for the I-K_d value, 0 mL/g, may not be appropriate. An I-K_d value of only 0.5 mL/g may be sufficient to exclude ^{129}I as a key dose contributor from preliminary LLW-PA simulations.

The compilation of reported K_d values and best-estimate K_d values are meant to provide early guidance to members of the LLW PA team until site-specific values are generated. Additionally, the list of key radionuclides for the PA may change as more information about the location and design of the burial site, waste glass recipe, waste inventory, and engineered barrier(s) become available.

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1.0 Introduction

1.1 Background

Burial of vitrified low-level waste (LLW) in the vadose zone of the Hanford Site is being considered as a long-term disposal option. Regulations dealing with LLW disposal require that performance assessment (PA) analyses be conducted. Preliminary modeling efforts for the Hanford Site LLW PA were conducted to evaluate the potential health risk of a number of radionuclides, including Ac, Am, C, Ce, Cm, Co, Cs, Eu, I, Nb, Ni, Np, Pa, Pb, Pu, Ra, Ru, Se, Sn, Sr, Tc, Th, U, and Zr (Piepho et al. 1994). The radionuclides, ^{129}I, ^{237}Np, ^{79}Se, ^{99}Tc, and ^{234,235,238}U, were identified as posing the greatest potential health hazard. It was also determined that the outcome of these simulations were very sensitive to the parameter describing the extent to which radionuclides sorbed to the subsurface matrix, described as a distribution coefficient (K_d). The distribution coefficient is a ratio of the radionuclide concentration associated with the solid phase to that in the liquid phase. The literature-derived K_d values used in these simulations were conservative, i.e., lowest values within the range of reasonable values used to provide an estimate of the maximum health threat. Thus, these preliminary modeling results reflect a conservative estimate rather than a best estimate of what is likely to occur. The potential problem with providing only a conservative estimate is that it may mislead us into directing resources to resolve nonexisting problems.

1.2 Purpose

The purpose of this report is to: 1) compile and describe statistically the available Hanford Site-specific K_d data and 2) provide best estimates of I, Np, Se, Tc, and U K_d values for future modeling efforts. The list of key radionuclides for the performance assessment (PA) may change as more information about the location and design of the burial site, waste glass recipe, waste inventory, and engineered barrier(s) becomes available.

1.3 Approach

An in depth literature search through the general scientific literature and Hanford Site reports was conducted to identify reports describing the sorption of I, Np, Se, Tc, and U on Hanford Site sediments. Additionally, a number of the authors of these reports were contacted to provide additional details pertaining to their experimental techniques and sediment characterization. K_d values were compiled into four categories of alkaline, low organic material solutions. These categories were based on the ionic strength (a measure of the total concentration of salts in solution) and redox status (the potential of a system to donate or accept electrons) of the system. The redox status was broadly defined as oxic, a system in which ferric (Fe III) minerals exists (Eh > 0.17 V), and anoxic, a system in which ferrous (Fe II) minerals exists (Eh ≤ 0.17). The four categories of alkaline, low organic material solutions were 1) low ionic strength and oxic solutions (Table 1.1), 2) low ionic strength and anoxic solutions (Table 1.2), 3) high ionic strength and oxic solutions (Table 1.3), and 4) high ionic strength and anoxic solutions (Table 1.4). Category 1 was selected to approximate the plume in the far field. Category 2 was selected to approximate the plume in a near-field barrier where an anoxic environment exist (such as a sulphur-polymer cement barrier, a mixture of 95-percent sulfur and 5-percent organic binders). Category 3 was selected to approximate the plume in a near-field barrier that created a high ionic strength/oxic environment (such as vitrified glass or concrete vault leachate). Category 4 was selected to approximate the plume in a near-field barrier that created a high ionic strength and anoxic environment. Broad definitions were chosen for these categories because the precise burial site and burial facility have not been identified. Refinement of these categories will likely be required as more information about the burial design becomes available.

Table 1.1. K_d Values for Hanford Sediments in Neutral-to-High pH, Low Ionic Strength ($\leq$ 0.01M), Low Organic, Oxic Solutions

Radionuclide	Low K_d	High K_d	Number of Samples	Mean ± Std. Err.	Variance	Median	Best Estimate
I	0.7	15.0	9	6.5±1.7	25.02	7	3
Np	2.4	21.7	4	14.9±4.4	76.94	17.8	15
Se	-3.4(a)	0.8	3	-1.7±1.3	4.83	-2.4	0
Tc	-2.8	0.6	15	-0.3±0.2	0.85	0.1	0
U	0.1	79.3	13	7.0±6.0	472.3	0.6	0.6

(a) Negative K_d values are physically impossible and represent analytical or laboratory error.

Table 1.2. Estimated K_d Values for Hanford Sediments in Neutral-to-High pH, Low Ionic Strength ($\leq$ 0.01M), Low Organic, Anoxic Solutions

Radionuclide	Low Estimate	High Estimate	Best Estimate	References (a)
I	0	15	3	1, 2, 3, 4
Np	200	1000	200	2, 5, 6
Se	0	3	0	2, 7, 8, 9, 10, 11
Tc	200	1000	200	2, 5, 12, 13,
U	100	1000	100	2, 14, 15, 16

(a) References: 1 = Serne and Wood 1990; 2 = Strenge and Peterson 1989; 3 = Muramatu et al. 1990; 4 = Tichnor and Cho 1990; 5 = Bondietti and Francis 1979; 6 = Ames and Rai 1978; 7 = Rai and Zachara 1984; 8 = Singh et al. 1981; 9 = Frost and Griffin 1977; 10 = Leckie et al. 1980; 11 = Benjamin and Bloom 1981; 12 = Gu and Schulz 1991; 13 = Hanke et al. 1986; 14 = Rai et al. 1990; 15 = Carnahan 1988; 16 = Bruno et al. 1988.

Attempts were made while compiling these tables to include K_d values generated from 1) Hanford Site sediments, 2) trace concentrations of sorbate (radionuclide), 3) pH levels between 7 and 11, 4) natural Hanford groundwater or Na and/or Ca background electrolyte solution, and 5) solution ionic strengths of $\leq$0.01 M (low ionic strength, Tables 1.1 and 1.2) or 0.01 to 1.0 M (high ionic strength, Tables 1.3 and 1.4). Where no K_d values were available that fit these requirements, estimates of the ranges were made based on experiments in which precipitation and/or adsorption occurred (K_d values are a measure of adsorption and not solubility) or K_d values generated from non-Hanford Site sediments. References used to generate these estimates are provided. Finally, for each radionuclide, a single best estimate of the K_d value in the four general aqueous categories is provided. When the authors' best estimates could not be reduced to a single number, the lower value of the resulting range is provided.

It is important to note that the laboratory derived K_d values reported in this document were in all cases generated from the <2-mm sediment fraction; hence the diminutive effect of gravel-size material on K_d values is not reflected in these values. Portions of the vadose zone of the 200-East

Table 1.3. K_d Values for Hanford Sediments in Neutral-to-High pH, High Ionic Strength (1.0 M > x > 0.01 M), Low Organic, Oxic Solutions

Radionuclide	Low K_d	High K_d	Number of Samples	Mean ± Std. Err.	Variance	Median	Best Estimate
I	0.4	0.7	4	0.5 ± 0.06	0.015	0.5	0.5
Np	0.3	3.8	8	2.2 ± 0.6	2.61	2.3	2
Se(a)	0	2					0
Tc	-3(b)	0.04	3	-0.98 ± 1.0	3.07	0.03	0
U(a)	0	4					0

(a) All selenium and uranium values are estimates based on Table 1.1, Serne and Wood (1990), and Strenge and Peterson (1989).
(b) Negative K_d values are physically impossible and represent analytical or laboratory error.

Table 1.4. Reported Best-Estimate K_d Values for Hanford Sediments in Neutral-to-High pH, High Ionic Strength (1.0 M > x > 0.01 M), Low Organic, Anoxic Solutions

Radionuclide	Low Estimate	High Estimate	Best Estimate	References(a)
I	0	15	0	1, 2, 3, 4
Np	100	1000	100	2, 5, 6
Se	0	2	0	2, 7, 8, 9, 10, 11
Tc	100	1000	200	2, 5, 12, 13
U	100	1000	100	2, 14, 15, 16

(a) References: 1 = Serne and Wood 1990; 2 = Strenge and Peterson 1989; 3 = Muramatu et al. 1990; 4 = Tichnor and Cho 1990; 5 = Bondietti and Francis 1979; 6 = Ames and Rai 1978; 7 = Rai and Zachara 1984; 8 = Singh et al. 1981; 9 = Frost and Griffin 1977; 10 = Leckie et al. 1980; 11 = Benjamin and Bloom 1981; 12 = Gu and Schulz 1991; 13 = Hanke et al. 1986; 14 = Rai et al. 1990; 15 = Carnahan 1988; 16 = Bruno et al. 1988.

Area contain large amounts of gravel-sized material (DOE 1993; Kincaid et al. 1993). Thus it is important to note that these K_d values, when used in transport codes, provide a maximum. As a first approximation, it can be assumed that the gravel does not participate in sorption. For example, if the sediment to be modelled contains 70-percent gravel and its <2-mm fraction K_d value is 2 mL/g, then the "gravel-corrected K_d value" would be 0.6 mL/g [(1.00 - 0.70) x 2 mL/g = 0.6 mL/g].

2.0 Iodine

Iodine will likely exist in waste form leachates and groundwater as an anion, I^- or IO_3 (Muramatu et al. 1990; Tichnor and Cho 1990; Gu and Schulz 1991). Spiking trace concentrations of ^{125}I into Hanford groundwater, Serne et al. (1993) reported that the average K_d value of a sediment collected from the 200 East Area (sample CGS-1; additional characterization of sediments referenced in this report appear in Appendix A) was 0.7 ± 0.5 mL/g (Table 2.1). In the same experiment, a Touchet sand (sample TBS-1) had an average K_d value of 1.4 ± 1.1 mL/g, and a sediment from Trench 8 in 200 West Area had an average K_d value of 1.3 ± 1.4 mL/g. Gee and Campbell (1980) obtained appreciably higher K_d values than Serne et al. (1993) for a surface sediment collected from the 200 Area (Rupert sand). Varying the chemical composition of the aqueous phase, Gee and Campbell (1980) determined K_d values ranging from 4 ± 2 to 15 ± 18 mL/g. It is not apparent which sediment properties or aqueous conditions were responsible for the differences between these two sets of K_d

Table 2.1. I-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

	Experimental Conditions				
I-K_d (mL/g)	^{125}I (nCi/mL)	Aqueous Phase	Solid Phase[(a)]	Expt'l Method	Reference[(b)]
0.7	200	Hanford groundwater	CGS-1 sand	Batch	1
1.4	200	Hanford groundwater	TBS-1 loamy sand	Batch	1
1.3	200	Hanford groundwater	Trench-8 loamy sand	Batch	1
7	200	0.003 M carbonate/sulfate	Rupert sand	Batch	2
8	200	0.0006 M carbonates/sulfates	Rupert sand	Batch	2
11	200	0.014 M carbonate/sulfate	Rupert sand	Batch	2
15	200	0.003 M nitrate/chloride	Rupert sand	Batch	2
10	200	0.0006 M nitrate/chloride,	Rupert sand	Batch	2
4	200	0.014 M nitrate/chloride	Rupert sand	Batch	2

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand; however, in the field, its texture may be classified as a course gravel/sand.

(b) References: 1 = Serne et al. 1993; 2 = Gee and Campbell 1980.

values; the particle distribution and mineralogy were quite similar in the sediments used in both studies. Five column studies conducted by Gee and Campbell (1980) at high ionic strength (1.0 M > x >0.01 M) yielded K_d values between 0.4 to 0.7 mL/g (Table 2.2). Two column studies conducted at >1 M ionic strength yielded K_d values of 0.04 and 0.06 mL/g; due to their high ionic strength, these data were not included in Table 2.2. Together, these data show a general trend common with many sorbates; i.e., the greater the competition of ions in solutions for exchange sites, the lower the K_d value for the tracer ion of interest. I-K_d values of low ionic strength solutions were measured in four different sediments: three sediments had K_d values of <1.4 mL/g and the fourth had an average K_d value of about 9.1 mL/g. The best estimate for an I-K_d value in low ionic strength solutions, 3 mL/g, is approximately equal to the mean of the four sediments, 3.1 mL/g (Table 1.1). Iodine is not expected to precipitate under reducing conditions (Muramatu et al. 1990), and, therefore, the best-estimate for a K_d value under reducing conditions is the same.

A very important and unexpected result from this literature review was that I sorbed to every Hanford sediments tested. Sorption of I^- or IO_3 species has been previously reported in sediments containing organic matter or oxide minerals, especially at low pH levels. However, these sediment constituents exist at extremely low concentrations in the sediments tested. The mechanism by which anions sorbed to these sediments is not known. One mechanism that could be invoked to explain these results deals with the fact that although the mineral surfaces in these studies have a net negative charge, they likely possess a small number of positively charged sites. The positively charged sites may exist on the edges of 2:1 clays, such as smectite and illite, on Al- and Fe-oxide surfaces, and 1:1 clays, such as kaolinite. All the experiments described in Table 2.1 were conducted at extremely low ^{125}I concentrations, approximately 12 parts per trillion. Therefore, only a small number of positively charged sites would be needed to sorb a measurable proportion of dissolved ^{125}I. Consistent with this mechanism, had larger initial concentrations been used in these experiments, say 1 part per million (1,000,000 parts per trillion), only a small proportion of ^{125}I may have been sorbed to the solid phase, resulting in immeasurable ^{125}I sorption. Importantly, should ^{129}I enter the LLW plume, it would more likely exist in the parts-per-trillion concentration range than the parts-per-million range (Piepho et al. 1994). These I sorption results have significant implications for the LLW-PA because it suggests that the most conservative estimate for the I-K_d value, 0 mL/g, may not be appropriate. An I-K_d value of only 0.5 mL/g would be sufficient to exclude ^{129}I as a key dose contributor from preliminary LLW-PA simulations (Piepho et al. 1994). Research has been initiated under this task to provide greater insight as to the mechanism controlling I sorption. Additionally, measurement of I-K_d values will be conducted with site-specific sediments using appropriate I concentration.

Table 2.2. I-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, High-Ionic Strength (1.0 M > x > 0.01 M) Conditions

	Experimental Conditions				
I-K_d (mL/g)	^{125}I (nCi/mL)	Aqueous Phase	Solid Phase(a)	Expt'l Method	Reference(b)
0.5	200	0.16 M nitrate	Rupert sand	Batch	1
0.5	200	0.32 M nitrate	Rupert sand	Batch	1
0.4	200	0.32 M nitrate	Rupert sand	Column	1
0.7	200	0.16 M nitrate	Ringold loam	Batch	1
0.6	200	0.16 M nitrate	Ringold loam	Column	1

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand; however, in the field, its texture may be classified as a course gravel/sand.
(b) References: 1 = Gee and Campbell 1980.

3.0 Neptunium

Thermodynamic data suggest that Np exists as a monovalent cation, NpO_2^+, in solution under oxidizing conditions, except above a pH of approximately 9 where it forms a neutral species with bicarbonate, $NpO_2HCO_3^0$ (Rai and Zachara 1984; Ames and Rai 1978). The data also indicate that Np is capable of complexing with other common anions (chloride, sulfate, fluoride, and nitrate). However, complexed species should not occur at significant concentrations under ambient low ionic strength in Hanford groundwater conditions (Rai and Zachara 1984; Ames and Rai 1978). Assuming that NpO_2^+ is the dominant aqueous species, Np should sorb via cation exchange. Routson et al. (1976) and Benson (1961) indicated that Np does not compete successfully with common cations, such as calcium and magnesium, for sorption sites. Routson et al. (1976) reported a Np-K_d value of 2.37 mL/g for a Burbank sand in contact with a solution with a calcium concentration of 2 x 10^{-3} M (Table 3.1). Using a subsample of the Burbank sand from Routson's laboratory, Sheppard et al. (1976) reported an appreciably greater Np-K_d value, 15.4 mL/g, than Routson et al. (1976). This difference is likely due to the lower ionic strength of the distilled water system of Sheppard et al. (1976) than the 0.002 M Ca system of Routson et al. (1976). The latter system is more relevant to the PA. Serne et al. (1993) determined Np-K_d values ranging from 13.5 ± 3 to 29.5 ± 3.6 mL/g (an average of 21.7 ± 8.6 mL/g) for Hanford sediment, Trench 8, in Np-spiked Hanford groundwater. Based on these data, the best estimate for a Np-K_d value in the far field is approximately the mean, 15 mL/g (Table 1.1). As observed with the I-K_d data, Np-K_d values decreased in high ionic strength solutions (Tables 3.1 and 3.2). Experiments were conducted with only one soil type, Burbank sand, with a wide range of ionic strength solutions. The best estimate for a Np-K_d value at high ionic strength solutions is equal to both the mean and median of the available data (Table 1.3). Neptunium is likely to precipitate from solution under reducing conditions. With these conditions the best estimate of a K_d value (incorrectly used to describe a solubility phenomena) is likely to be appreciably greater, 200 mL/g (Tables 1.2 and 1.4).

Table 3.1. Np-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

	Experimental Conditions				
Np-K_d (mL/g)	^{237}Np (nCi/mL)	Aqueous Phase	Solid Phase$^{(a)}$	Expt'l Method	Reference$^{(b)}$
21.7	100	Hanford groundwater	Trench 8 loamy sand	Batch	1
2.4	1480	0.002 M Ca	Burbank loamy sand	Batch	2
20.2	25	Distilled water	Ritzville silt loam	Batch	3
15.4	25	Distilled water	Burbank loamy sand	Batch	3

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand; however, in the field, its texture may be classified as a course gravel/sand.

(b) References: 1 = Serne et al. 1993; 2 = Routson et al. 1976; 3 = Sheppard et al. 1976.

Table 3.2. Np-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, High-Ionic Strength (1.0 M > x > 0.01 M) Conditions

	Experimental Conditions				
Np-K_d (mL/g)	^{237}Np (nCi/mL)	Aqueous Phase	Solid Phase[(a)]	Expt'l Method	Reference[(b)]
1.0	1480	0.02 M Ca	Burbank loamy sand	Batch	1
0.8	1480	0.05 M Ca	Burbank loamy sand	Batch	1
0.7	1480	0.1 M Ca	Burbank loamy sand	Batch	1
0.3	1480	0.2 M Ca	Burbank loamy sand	Batch	1
3.8	1480	0.03 M Na	Burbank loamy sand	Batch	1
3.7	1480	0.3 M Na	Burbank loamy sand	Batch	1
3.7	1480	0.75 M Na	Burbank loamy sand	Batch	1
3.1	1480	0.015 M Na	Burbank loamy sand	Batch	1

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand, however, in the field, its texture may be classified as a course gravel/sand.

(b) References: = Routson et al. 1976.

4.0 Selenium

Selenium is expected to sorb to Hanford sediments very weakly for a number of reasons. It is likely to exist as an anion in waste form leachates and groundwater (Rai and Zachara 1984; Singh et al. 1981; Frost and Griffin 1977; Leckie et al 1980). Specific adsorption onto geologic materials is not likely to play an important role in controlling Se mobility because the near-field plume (pH ≈ 11) and natural groundwater (pH ≈ 8) will have a sufficiently high pH to greatly reduce the number of anion exchange sites. Additionally, the plume is expected to have high concentrations of sulfate (total S levels in the plume may be 1 mM (Rawlins et al. 1994), which are known to compete with Se for mineral adsorption sites (Hingston et al. 1971).

Selenate SeO_4^{2-} sorption experiments on Hanford sediment CGS-1 had similar results when the background solution was a natural Hanford groundwater (pH 8.3) or a process waste stream (pH 4.5). K_d values were 0.78 ± 2.17 and -1.32 ± 1.96 mL/g, respectively (Serne et al. 1993) (Table 4.1). The negative K_d values are a physical impossibility and reflect analytical or laboratory error. Nearly identical K_d values were obtained with Hanford sediments TBS-1 and Trench 8. For the TBS-1 sediment, the Se-K_d value in groundwater was -3.44 ± 1.37 mL/g and in simulated process waste stream 4.6 ± 0.7 mL/g. For the Trench 8 sediment, the Se-K_d value in groundwater was 2.1 ± 1.2 mL/g. These are exceptionally low K_d values compared to those reported in other sediments (Rai and Zachara 1984). The presence of small concentrations of Al or Fe oxides in the solid phase may result in substantially higher Se sorption (Leckie et al. 1980). The effective range of Se-K_d values in ambient vadose zone sediments is 0 to 0.78 mL/g (Table 1.1). The best estimate Se-K_d value for low and high ionic strength conditions is 0 mL/g. Under reducing conditions, Se precipitation is not likely to occur under the environmental conditions under consideration (Rai and Zachara 1984; Singh et al. 1981; Frost and Griffin 1977; Leckie et al. 1980; Benjamin and Bloom 1981).

Table 4.1. Se-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

	Experimental Conditions				
Se-K_d (mL/g)	Se (mg/L)	Aqueous Phase	Solid Phase(a)	Expt'l Method	Reference(b)
0.78	0.1	Hanford groundwater	CGS-1 sand	Batch	1
-3.44(c)	0.1	Hanford groundwater	TBS-1 loamy sand	Batch	1
-2.4	0.1	Hanford groundwater	Trench 8 loamy sand	Batch	1

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand; however, in the field, its texture may be classified as a course gravel/sand.
(b) References: 1 = Serne et al. 1993.
(c) Negative K_d values are physically impossible and represent analytical or laboratory error.

5.0 Technetium

Experimental work on Hanford sediments and soils in oxidized environments indicate that Tc exists as an anion, TcO_4^-, and does not readily complex with other chemical species (Bondietti and Francis 1979; Gu and Schulz 1991; Hanke et al. 1986). Consequently, Tc is considered to be relatively non-adsorbing in most environments, especially in low organic matter, low oxide mineral, and high pH systems, as exist in the Hanford far field. Serne et al. (1993) determined the Tc-K_d values of three Hanford sediments: sediment TBS-1 had a K_d of 0.1 ± 0.5 mL/g; sediment CGS-1 had a K_d of 0.1 ± 0.7 mL/g; sediment from Trench 8 had a K_d of 0.1 ± 0.5 mL/g (Table 5.1). Using two different sediments from the Hanford Site, Gee and Campbell (1980) determined K_d values by batch technique ranging from -2.77 to 0.5 mL/g (Table 5.1); at higher ionic strength solutions, they determined through column studies that Tc-K_d values ranged from -3 to 0.04 mL/g (Table 5.2). K_d values determined with Hanford sediment (Trench 8) and leachates derived from solid waste (wood, cardboard, and paper), which contained considerable concentrations of total organic carbon (407 to 440 mg C/L), showed no significant difference from those determined without organic materials present (data not tabulated because of high organic carbon content). Significant sorption may occur in soils that contain considerable amounts of natural organic matter, which tends to sorb anionic species and may reduce Tc to its +4 oxidation state, causing precipitation or sorption (Gu and Schulz 1991).

The quality of the Tc-K_d data is considered to be good because of the strong agreement between the Hanford Site values and those generated from other sediments (Rai and Zachara 1984; Gu and Schulz 1991). The best estimate for Tc-K_d values in low and high ionic strength, oxic conditions is 0 mL/g (Tables 1.1 and 1.3), which is essentially the mean and median value of the tabulated data. Under reducing conditions, Tc(VII) may convert to Tc(IV), resulting in the formation of the sparingly soluble precipitate TcO_2 (Bondietti and Francis 1978; Gu and Schulz 1991; Hanke et al. 1986). The best estimate of Tc-K_d values (incorrectly used to describe a solubility phenomena) in high or low ionic strength solutions and under reducing conditions is 200 mL/g (Tables 1.2 and 1.4).

Table 5.1. Tc-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

	Experimental Conditions				
Tc-K_d (mL/g)	^{95}Tc (nCi/mL)	Aqueous Phase	Solid Phase[(a)]	Expt'l Method	Reference[(b)]
0.1	100[(c)]	Hanford groundwater	CGS-1	Batch	1
0.1	100[(c)]	Hanford groundwater	Touchet Bed, loamy sand	Batch	1
0.1	100[(c)]	Hanford groundwater	Trench 8 loamy sand	Batch	1
-2.77[(d)]	2	0.003 M carbonate/ sulfates	Ringold loam	Batch	2
-1.13	2	0.0006 M carbonates/sulfates	Ringold loam	Batch	2
-0.04	2	0.014 M carbonate/sulfate	Ringold loam	Batch	2
0.57	2	0.003 M nitrate/chloride	Ringold loam	Batch	2
0.54	2	0.0006 M nitrate/chloride	Ringold loam	Batch	2
-0.51	2	0.014 M nitrate/chloride	Ringold loam	Batch	2
0.07	2	0.003 M carbonate/ sulfates	Rupert sand	Batch	2
-1.62	2	0.0006 M carbonates/sulfates	Rupert sand	Batch	2
-0.31	2	0.014 M carbonate/sulfate	Rupert sand	Batch	2
0.06	2	0.003 M nitrate/chloride	Rupert sand	Batch	2
0.52	2	0.0006 M nitrate/chloride	Rupert sand	Batch	2
0.38	2	0.014 M nitrate/chloride	Rupert sand	Batch	2

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand; however, in the field, its texture may be classified as a course gravel/sand.
(b) References: = Serne et al. 1993; 2 = Gee and Campbell 1980.
(c) ^{99}Tc (not ^{95}Tc) was used in experiment.
(d) Negative K_d values are physically impossible and represent analytical or laboratory error.

Table 5.2. Tc-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, High-Ionic Strength (1.0 M > x > 0.01 M) Conditions

	Experimental Conditions				
Tc-K_d (mL/g)	Tc (nCi/mL)	Aqueous Phase	Solid Phase(a)	Expt'l Method	Reference(b)
0.03	2	0.16 M nitrate/chloride	Rupert sand	Column	1
0.04	2	0.16 M nitrate/chloride	Ringold loam	Column	1
-3.0(c)	2	0.16 M nitrate/chloride	Rupert sand	Batch	1

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand; however, in the field, its texture may be classified as a course gravel/sand.

(b) References: 1 = Gee and Campbell 1980.

(c) Negative K_d values are physically impossible and represent analytical or laboratory error.

6.0 Uranium

Under oxidizing (oxic) conditions typical for Hanford vadose zone sediments and the upper unconfined aquifer, dissolved U is predicted to exist as a cation, UO_2^{2+}, up to a pH of approximately 6; as a neutral hydroxide species, $UO_2(OH)_2^0$, from a pH of 6 to 8; and as an anionic carbonate, $UO_2(CO_3)_3^{4-}$, above a pH of 8 (Rai and Zachara 1984; Rai et al. 1990). Uranium may also complex with other anions (e.g., fluoride, chloride, and phosphate) to form neutral or anionic species, but such complexes should not exist in significant concentrations under ambient Hanford groundwater conditions. These estimated species distributions suggest that U would sorb, via cation exchange, under acidic conditions and sorb very poorly under neutral and basic conditions.

Recent laboratory studies indicate that U sorption onto Hanford sediments is generally quite weak under ambient Hanford groundwater conditions (Serne et al. 1993) (Table 6.1). U-K_d values for a Trench 8 sediment and Hanford groundwater were 1.9 ± 1.4 mL/g at 5 days and 2.4 ± 0.6 mL/g at 44 days of contact (the overall average of these values, 2.3 mL/g, is reported in Table 6.1). Adsorption of U onto Hanford Site Trench 8 sediment from solid waste (wood and paper) leachates containing significant concentration of organic matter was significantly decreased over that of ambient groundwater; K_d values were essentially 0 mL/g. The average U-K_d value for Hanford groundwater and CGS-1 sediment was 1.7 ± 4.2 mL/g and TBS-1 sediment was 79 ± 26 mL/g. The appreciably higher K_d value of the TBS-1 is clearly not an outlier, the results were reproduced nine times (three contact times that were triplicated). The cause of the high K_d value is presumably due to the high silt and clay content (20 percent <2-mm fraction) in the sample. It is important to note that lenses of fine-grade material are know to form thin layers in the Hanford lithologic formation underlying the 200 East Area (DOE 1993). These lenses may greatly limit U transport though this geological strata.

In 1994, C.W. Lindenmeier, R. J. Serne, J. C. Conca, and A. T. Owen conducted a number of UO_2^{2+} column studies under saturated and unsaturated conditions for Trench 8 sediment (Table 6.1) at Pacific Northwest Laboratory. In total, they reported 10 column studies and determined the K_d values to range from 0.08 to 2.81 mL/g. There were three primary variables in these U column studies: retention time (residence time of the solute in the sediment column), pore-water velocity, and volumetric water content. The factor having the greatest effect on the U-K_d values was the volumetric water content; its correlation coefficient with U-K_d was 0.691 (degrees of freedom = 8, $P \leq 0.01$). Retention time and pore-water velocity were not significantly correlated ($P \geq 0.05$) to U-K_d values. It is important to note that all three of these parameters are concomitant, and therefore, a true statistical measure of their significance on the dependent variable, U-K_d value, requires covariant analysis. This analysis is not possible with the nonreplicated data set. However, this study represents the first in-depth study of U transport in unsaturated conditions and, in addition to providing K_d values under unsaturated conditions, identifies a number of experimental variables related to unsaturated column studies that have an effect on measured K_d values. Using the same sediment and batch techniques, Serne et al. (1993) reported K_d values within this range, 2.4 ± 0.6 mL/g. Higher values are often obtained by the batch than the column technique because of greater residence time and greater mixing of sediment and aqueous phase.

General indications are that U sorption values will be low under most ambient Hanford circumstances because U tends to form neutral or anionic carbonate species at neutral or basic pH values. The best estimate of U-K_d values under oxic, low ionic strength conditions is equal to the median value, 0.6 mL/g (Table 1.1). This value weighs the column studies more than the batch studies. It also weighs coarse grain sediment (Trench 8 and CGS-1) more than finer grain sediment (TBS-1). It is important to note that the presence of a layer of fine sediment in the flow path of the contaminant plume may have a profound effect on the extent of U transport. Under anoxic conditions, U(VI) may precipitate to form a sparingly soluble complex (Carnahan 1988; Bruno et al. 1988); thus the best estimate value under reducing conditions is appreciably higher, 100 mL/g

(Tables 1.2 and 1.4). Although the quality of the U-K_d data is fairly good, it is highly variable and the PA calculations show that dose is very sensitive to the U-K_d parameter. When U-K_d values below 0.5 mL/g were input into the transport code, U was the key dose contributor (Piepho et al. 1994).

Table 6.1. U-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

	Experimental Conditions				
U-K_d (mL/g)	U (ng/L)	Aqueous Phase	Solid Phase[(a)]	Expt'l Method	Reference[(b)]
1.98	100	Hanford groundwater	Trench 8 loamy sand	40% saturated column	1
0.49	100	Hanford groundwater	Trench 8 loamy sand	40% saturated column	1
2.81	100	Hanford groundwater	Trench 8 loamy sand	38% saturated column	1
0.62	100	Hanford groundwater	Trench 8 loamy sand	22% saturated column	1
0.45	100	Hanford groundwater	Trench 8 loamy sand	30% saturated column	1
0.54	100	Hanford groundwater	Trench 8 loamy sand	23% saturated column	1
0.62	100	Hanford groundwater	Trench 8 loamy sand	25% saturated column	1
0.40	100	Hanford groundwater	Trench 8 loamy sand	17% saturated column	1
0.10	100	Hanford groundwater	Trench 8 loamy sand	7% saturated column	1
0.08	100	Hanford groundwater	Trench 8 loamy sand	7% saturated column	1
1.7	50	Hanford groundwater	CGS-1 sand	Batch	2
2.3	50	Hanford groundwater	Trench 8 loamy sand	Batch	2
79.3	50	Hanford groundwater	TBS-1 loamy sand	Batch	2

(a) Solid phase texture indicates the <2-mm fraction used in the K_d analysis and not that existing in the field. For example, the <2-mm fraction may have a texture of a sand; however, in the field, its texture may be classified as a course gravel/sand.

(b) References: 1 = Results of studies in 1994 by C. W. Lindenmeier, R. J. Serne, J. L. Conca, and A. T. Owen at Pacific Northwest Laboratory; 2 = Serne et al. 1993.

(c) Negative K_d values are physically impossible and represent analytical or laboratory error.

7.0 References

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Appendix A

Sediment Characteristics

Appendix A
Sediment Characteristics

Sediment Name	Hanford Site Location	Field Texture	> 2 mm fraction %	$CaCO_3$ %	pH	C.E.C.(a) meq/100 g	Ref.(b)
TBS-1 (Touchet)	Army Loop Rd (S of 200 E)	Sand	<1.1	1.9	8.2	6.0	1
MSG-1	Grout Vault (S of 200-E)	Medium Course Sand	2.2	0.8	8.1	7.8	1
CGS-1	Quarry (W of 200-E)	Course Gravel Sand	82.5	1.0	7.7	2.6	1
Trench-8	W-5 Burial Ground Side Wall	Medium Course Sand	9.7	2.0	7.9	5.2	1
Ringold	Ringold Formation, 200 Area	--	--	1.4	8.8	12.0	2
Rupert Sand	Eolian Origin, Surface Sediment, 200 Area	--	--	0.1	8.2	5.0	2
Burbank	200 Area	Sand	--	0.8	7.0	4.9	3 and 4

(a) C.E.C. represents cation exchange capacity.
(b) References:

1 = Serne, R. J., J. L. Conca, V. L. LeGore, K. J. Cantrell, C. W. Lindenmeier, J. A. Campbell, J. E. Amonette, and M. I. Wood. 1993. *Solid-Waste Leach Characteristics and Contaminant-Sediment Interactions. Volume 1: Batch Leach and Adsorption Tests and Sediment Characterization.* PNL-8889, Vol 1, Pacific Northwest Laboratory, Richland, Washington.

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