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K_d Values for Agricultural and Surface Soils for Use in Hanford Site Farm, Residential, and River Shoreline Scenarios

Technical Report for Groundwater Protection Project -- Characterization of Systems Task

R. Jeff Serne

August 2007

Prepared for Fluor Hanford, Inc. and
the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Richland, Washington 99352

Summary

This report provides best estimate K_d values and a minimum and maximum range of K_d values to be used for agricultural soils and Columbia River bank sediments that exist today or would exist in the future when portions of the Hanford Site are released for farming, residential, and recreational use after the U.S. Department of Energy (DOE) completes clean up of defense waste on the site. The K_d values should be used to determine the fate and transport rates of contaminants and their availability for plant and animal uptake in selected non-groundwater scenarios included in Hanford Site environmental impact statements, risk assessments and specific facility performance assessments.

This report describes scenarios such as a small farm where drilling of a well inadvertently goes through buried waste and brings waste to the surface, allowing the tailings to become available for direct human exposure or incorporation into garden crops and farm animals used for food by the farm family. The K_d values recommended in this report can also be used to calculate sediment-water partitioning factors used to predict plant and animal uptake from interaction with the contaminated soil.

The values recommended herein should not be used to predict exposure to contaminants in drinking water or from water used for irrigation. For the waterborne scenarios, K_d values recommended in Last et al. (2004) and Krupka et al. (2004) should be used because they better predict the distribution and mobility of contaminants in water.

Other non-groundwater scenarios include (1) recreational or native American use of the Columbia River bank that exposes humans to contaminants adsorbed to the river bank sediments, (2) contaminants transferred from sediments to plants or animals that reside along the Columbia River bank, and (3) any other scenario that requires estimation of soil-to-plant or soil-to-animal transfer parameters that are often calculated based on soil K_d values.

For these non-groundwater exposure scenarios, choosing a larger K_d value from available values will be “conservative” in the sense that large K_d values keep the contaminant available longer (retains more contaminants in the soils) such that exposure to humans or incorporation into animals and plants is maximized. Therefore, the K_d values in this report are slightly biased to keep the contaminants near the ground surface for longer time periods. Upon exposure to water or by direct contact, the contaminated near-surface soils allow more opportunity for direct exposure and incorporation into plant matter and animals.

The following contaminants of concern were the focus of the search: americium, bismuth, carbon-14, carbon tetrachloride, cesium, chlorine-36, chromium, cobalt, iodine, lanthanides (especially europium), lead, neptunium, nickel, nitrate/nitrite, plutonium, polonium, protactinium, radium, selenium, strontium, technetium, tritium, and uranium.

Acknowledgments

The author wishes to thank George Last and Bruce Napier at PNNL for providing funds to perform this work, and delineating the list of contaminants for which K_d values should be tabulated and for providing technical comments on the draft report. The author thanks Ken Krupka and Kirk Cantrell, both in the Applied Geology and Geochemistry Group at PNNL, for their technical comments on the draft report, which improved the final version significantly. The author thanks Launa Morasch (PNNL) for performing the final document editing.

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1.0 Introduction

This report tabulates K_d values for a selected list of contaminants of interest isolated within near-surface agricultural soils and Columbia River bank near-surface sediments that exist today or would exist in the future when portions of the Hanford Site are released for farming, residential, and recreational use after the U.S. Department of Energy (DOE) waste cleanup activities are completed. The soil and sediment types considered are surface soils that are capable of sustaining large agricultural farms or small family farms that produce typical food crops and surface soils typically found at rural/suburban residences. In addition, the K_d values tabulated in this report can be used to evaluate the risks of exposure to near-shore Columbia River bank sediments that may have been contaminated by Hanford activities and are currently found along the riverbank today and into the future. The K_d values can be used to determine the fate and transport rates and availability for plant and animal uptake of these contaminants in selected scenarios included in Hanford Site environmental impact statements, risk assessments, and specific facility performance assessments, such as:

1. Small farm development that includes inadvertently drilling a well through buried waste, bringing solid/sediment-bearing waste to the surface. These tailings then become available for direct human exposure or incorporation into garden crops and farm animals used for food by the farm family. The K_d values recommended in this report can be used to calculate direct human exposure to the sediments bearing small masses of tailings and to determine sediment-water partitioning factors used in predicting plant and animal uptake from interaction with the contaminated soil. The K_d values recommended herein should not be used to predict migration of contaminants to the groundwater or for direct exposure to contaminants in the drinking water. For such water-borne scenarios, K_d values recommended in Last et al. (2004) and Krupka et al. (2004) should be used because they better predict the distribution and mobility of contaminants in water. There is one scenario involving irrigation water that can use the K_d values from this report. The uptake of contaminants from contaminated irrigation water by the future agricultural and Columbia River bank sediments can use the tabulated K_d values because future soil and riverbank sediments have the potential to adsorb more contaminants than the current sediments based on the assumption that future soil will contain more organic matter and have a slightly lower overall mean particle size due to soil amendments and farming practices.
2. Recreational or Native American use of the Columbia River bank. In this scenario, humans may be exposed to contaminants adsorbed to the riverbank sediments or contaminants transferred from sediments to plants or animals that reside along the Columbia River bank.
3. Any other scenario that requires estimation of soil-to-plant or soil-to-animal transfer parameters that are often calculated based on near-surface soil K_d values.

To re-iterate, the K_d values reported herein should not be used for fate and transport modeling for water-borne contaminant scenarios except when the modeling is used to estimate accumulation of contaminants by future surface soils from irrigation practices. Scenarios for which the K_d values tabulated herein should not be used include transport of contaminants through the vadose zone to the groundwater or human exposure from well waters. For the groundwater/well water scenarios, K_d values have been extensively collated, reviewed, and documented in reports such as Cantrell et al. (2003), Last et al. (2004) and Krupka et al. (2004). The K_d values recommended in Last et al. (2004) are intended to provide best estimate values and a range in uncertainty for scenarios where liquid waste was disposed to subsurface disposal facilities with emphasis on adsorption. Values tabulated in Krupka et al. (2004) are slightly biased toward values that promote contaminant mobility via water pathways.

For the non-groundwater exposure scenarios, choice of a larger K_d value will be conservative in the sense that large K_d values keep the contaminant available longer (retains more contaminants in the soils) so that incorporation or exposure to humans, animals, and plants is maximized for the non-groundwater scenarios. Therefore, the K_d values in this report are slightly biased to keep the contaminants near the ground surface for longer time periods than generally assumed for past liquid waste disposal operations. Upon exposure to water or by direct contact, the contaminated near-surface soils allow more opportunity for direct exposure and incorporation into plant matter and animals. On the other hand, for groundwater/ well water scenarios, choosing lower K_d values leads to more rapid transport of contaminants from the near surface to the groundwater and, thus, more exposure or risk to the receptor who imbibes the water or bathes in the contaminated water.

One goal of this study was to capture the fact that agricultural soils often contain more organic matter and silt- and clay-size particles than generally exists in the native Hanford formation sands and aeolian deposits that lie near ground surface today. Both the increased organic matter content and increased fine-particle content tend to increase the adsorption properties of most contaminants (see EPA 1999a for discussion on the impact of organic matter and particle surface area); this would lead to larger K_d values than one should use for modeling contaminant adsorption on typical Hanford sand sediments found today.

A recent screening survey of soil pH values at highly vegetated upland sites and along the Columbia River shoreline at locations designated informally as “Vernita Riparian,” “100B/C Riparian,” “Downstream Riparian,” and “Upland Reference” show slightly acidic pH values (6.2 to 7.8) likely caused by decaying plant material.[1] These are the pH measurements and general soil descriptions used in this report for vegetated near-surface and Columbia River bank soils. The slightly acidic nature of these soils is an indication that future soils may have lower than ambient pH. This influenced the range of K_d values recommended in this report because pH is a key parameter that influences the adsorption properties of many of the contaminants and hydrous oxide coatings/adsorbents present in soils. It is not clear whether the lowered pH values signify the presence of any potential strong radionuclide chelating agents, such as fulvic and humic acids, or whether the slightly acidic pH values are caused by less strong complexing moieties, such as simple organic acids. In fact, the slightly acidic pH values measured on these few samples have not been shown to be caused by decay of organics, it is only a supposition at this time. The possibility that decaying organic matter has produced significant concentrations of strong chelators such as fulvic and humic acids should be considered if further measurements of pH and other parameters are made on soil along the Columbia River or upland at highly vegetated sites.

One might also assume that sediments along a river shoreline include more fine-grained sediments and organic matter than sediments/soils from inland areas based on the deposit of suspended fine sediments and organic debris from the river water and the higher potential for plant growth because of the high water content. However, at present there is no evidence that the sediments along the Columbia River banks within the Hanford Site show significantly smaller grain size or larger organic content than the

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[1] Personal communication from JL Downs (Pacific Northwest National Laboratory, Richland, Washington) to RJ Serne (PNNL) via email, dated March 4, 2005.

native vegetated Hanford formation/aeolian sediments inland (Patton and Crecelius 2001).[2] The data in Patton and Crecelius (2001) show total organic carbon contents for sediments along the Columbia River banks at three sloughs range from <0.5 to 1.5% by weight and that the silt and clay content is generally <10% by weight. These particle-size values are similar to agricultural soils expected at the Hanford Site in the future and not much different from the existing sediments in burial grounds at present Serne et al. (1993) and Horton et al. (2003). The observed organic carbon content in the sloughs is somewhat greater than subsurface sediments near burial grounds that generally have organic carbon at or less than 0.1%.[3] However, values between 0.5 and 1.5% organic carbon are not high compared to highly productive agricultural soils, which typically reach values of several %wt (Tan 2000 and Tan 2003). Therefore, I recommend that the K_d values reported in this report be used for all Columbia River shoreline fate/risk calculations and for Hanford agricultural soil/Hanford family farm, recreational and residential land use scenarios that do not involve ingestion of contaminated well water by humans. The K_d values can also be used to estimate plants/animal uptake factors, which are generally related to concentrations of contaminants in the surface soils, for irrigation with contaminated well water.

The K_d values in this tabulation should also not be used to represent release of contaminants directly from solid waste tailings inadvertently brought to the surface. If large masses of waste reach the surface, release or leach values need to be measured or estimated on a case-by-case basis with keen attention to using appropriate conditions and test protocols. Rather, the K_d values tabulated herein represent adsorption and desorption from the near surface soils and river bank sediments themselves. One exception is found in the discussion of appropriate K_d values for uranium (see Section 3.23) where some consideration of waste-induced influence is assumed.

The source of contaminants in the scenarios of interest is assumed to be either:

1. residual adsorbed or precipitated contaminants in sediments deeper than those removed during clean up (often sediments from the near ground surface to a depth of 15 ft are removed from inactive disposal sites and clean fill is emplaced in the excavation to bring the contours back to grade) that are inadvertently brought to the surface and mixed with near-surface soils

   or

2. solidified waste (e.g., grout, glass, solid waste) that is inadvertently exhumed from depths below 15 ft during well drilling activities. The exhumed waste is assumed to be mixed with near surface soils around the well location or wherever the Hanford farmer would place the well drilling tailings. It should be noted that the volume of waste brought to the ground surface by well drilling should be rather limited. Because of the assumed small mass of waste, except for the contaminant uranium, this tabulation does not account for any significant changes in the solution composition or physical attributes of the soils/river bank sediments in the selection of K_d values.

   or

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[2] Personal communication from GW Patton and RE Peterson (Pacific Northwest National Laboratory, Richland, Washington) with RJ Serne (PNNL), June 2005.

[3] Data taken from a Hanford Site database available at \\wd40960\AGGData\AGGPublic\COS Min Database\Database Project\Master Database.xls or available from GV Last (Pacific Northwest National Laboratory, Richland, Washington).

3. re-deposited waste solids that had been dispersed into the atmosphere by some activity or waste brought near the surface by burrowing animals and mixed with the surface soils. Again the assumption is that the mass or percentage of waste that gets mixed with the near-surface soils/ sediments is small and does not significantly alter the soil and pore water attributes, except for uranium.

These mixed waste/soils are contacted by natural precipitation and irrigation waters and also can impact humans by direct exposure and be transferred to plants (e.g., vegetables, grasses, fruit trees), farm animals (via forage and direct ingestion), and fish (via food chain dynamics and direct ingestion) taken from the Columbia River. The pore waters in the agricultural soils and Columbia River bank sediments are assumed to be similar to natural precipitation, irrigation water, or Columbia River water that exhibits near-neutral pH values (i.e., between 7 and 8) and low ionic strength as opposed to being highly saline and either caustic- or acidic-pH bearing chemical processing waste streams that were present during the re-processing activities needed to obtain plutonium. It also has been assumed that any solid waste tailings brought to the ground surface are of limited mass that does not cause wide excursions in pH or the dissolved solids content of waters that come in contact with the tailings, excepting for uranium, which is hyper-sensitive to carbonate/pH variations.

A final process that has not been considered in this study is the impact from addition of fertilizers and other possible soil amendments to increase agricultural productivity. The study assumed that fertilizers and other soil amendments would not dramatically alter the soil pore water chemical composition or soil adsorption properties aside from promoting increased organic matter build up and a slight lowering of the mean particle-size distribution, both of which generally increase adsorption tendencies for most contaminants.

Past K_d tabulations performed for Hanford uses did not focus on agricultural soils or river bank sediments because they are not located on the Hanford Site or proximal to the majority of past and current burial grounds. Therefore, K_d literature was reviewed to locate applicable studies for low ionic strength "natural" solutions, such as groundwater, river and lake water, and rainwater contacting agricultural soils and river bank sediments. Sorption data for chemical waste streams were used only if no other data were available. Hanford Site-specific data was relied on when it was available and other general literature data was factored in to a lesser extent. From the strategies used in other Hanford K_d tabulations, only K_d value used to describe far-field or waste un-impacted conditions were considered based on the assumption that any waste brought to the surface during drilling or burrowing animal scenarios are of limited mass. The one exception was uranium, which at Hanford appears to be quite sensitive to two key variables, pH and dissolved inorganic carbon content, that both are highly influenced by common solid waste to be left in shallow-land burial facilities once the Hanford Site is released for public use.

As a service to other K_d compilers, this study contains more recent literature and provides a brief synopses of the new literature in the hope that it will be easier to update the Hanford K_d compilations in the future.

1.1 Where Did the K_d Values Come From

This critical review and update of K_d values for non-groundwater scenarios (i.e., that do not involve direct ingestion of contaminated well water by humans or animals) uses some past data compilations performed by PNNL staff and published compilations by Coughtrey and Thorne (1983a,b) and Coughtrey et al.(1983, 1984a, 1984b, 1985) and by Thibault et al. (1990) as the main source of information.

The list of contaminants to review was prepared by Bruce Napier (PNNL) and was based on his risk assessment experience for the Hanford Site over the last ~30 years. The contaminants that have been important risk drivers for past Hanford Site assessments are listed in Section 1.2. The list is somewhat reduced from the list originally used in Napier and Snyder (2002) based on preliminary exposure dose calculations performed in the cited report. That is, some contaminants were "de-listed" based on their low inventories and/or low dose impacts.

A previous effort to tabulate K_d values for agricultural soils for non-groundwater scenarios was provided to Bruce Napier in unpublished letter reports,[4] which are attached as Appendix A to this report, from geochemists at Hanford. The geochemists had performed a limited review of available literature that focused on finding K_d values to use for groundwater scenarios, where conservative values are those biased to low numbers. The geochemists took the ranges in the available literature, with emphasis on Hanford Site-specific data when available, and simply chose the high end of the range as appropriate for non-groundwater scenarios. For the non-groundwater scenarios, conservatism was achieved by choosing high K_d values. These past letter reports were summarized as a table in Napier and Snyder (2002, Table 2).

This report, recommends using K_d values that take into consideration the nature of projected Hanford agricultural soils and river bank sediments as described in the introduction as opposed to just recommending the highest value reported in the literature. The K_d values recommended in this report should be more realistic, but remain reasonably conservative for non-groundwater scenarios.

In addition, three electronic databases were searched: on-line documents at DOE-OSTI at http://www.osti.gov/bridge/search.easy.jsp , *Web of Science* for journal manuscripts from 1980 to 2004, and On-line Computer Library Center's *First Search (*http://www.oclc.org/firstsearch/). Each database was searched using a combination of key words such as the element of interest and K_d or adsorption. When the matches were large the search was further refined with adding soil(s) or sediment(s) as a qualifier. From these searches, approximately 50 published references were found with useful data since the past compilations by Coughtrey and Thorne (1983a, 1983b), Coughtrey et al. (1983, 1984a, 1984b, 1985), and Thibault et al. (1990). All the new materials were read and useful data extracted to aid in developing the database. Because no sorption literature was found for some of the contaminants of interest, some expert judgment was used to populate the database in this report. The nuclides for which relevant data were non-existent or too sparse to select actual Hanford Site-specific K_d values were bismuth, carbon tetrachloride, chlorine-36, nitrate/nitrite, polonium, protactinium, radium, and tritium.

1.2 List of Contaminants Reviewed

The following contaminants of concern were the focus of the search: americium, bismuth, carbon-14, carbon tetrachloride, cesium, chlorine-36, chromium, cobalt, iodine, lanthanides (especially europium), lead, neptunium, nickel, nitrate/nitrite, plutonium, polonium, protactinium, radium, selenium, strontium, technetium, tritium, and uranium. The review was limited to these contaminants, per Mr. Napier's

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[4] On October 3, 1997 Dr. Dan Kaplan prepared a letter report, *Selected K_d Values for Agricultural Soils*. This letter report was revised/updated by R. Jeff Serne on May 17, 1999.

request that is based on his experience and other published Hanford risk and performance assessments (Mann et al. 2001; Kincaid et al. 1998, 2000; and DOE 2004), which have shown that these are potential key risk drivers for groundwater, Hanford farm, Native American or recreational scenarios. Contaminants included in Napier and Synder (2002) that were not reviewed further include actinium (Ac), curium (Cm), iron (Fe), manganese (Mn), potassium (K), rhenium (Re), radon (Rn), ruthenium (Ru), antimony (Sb), tin (Sn), thorium (Th), and yttrium (Y). In Napier and Snyder (2002, Table 2), maximum K_d values and ranges where available are listed for these contaminants. Also the same information is found in Appendix A of this report.

1.3 Additional Discussion

This report does not include much discussion on key geochemical characteristics of the contaminants of interest, soils and sediments and the low ionic strength waters that influence the myriad of reactions that control the magnitude of adsorption. Neither should it be considered a primer on the conceptual models that are commonly used to quantify the processes that sequester contaminants in soils and sediments or the processes that promote the mobility of contaminants by keeping them in the aqueous fluids that percolate through sediments. Adsorption-desorption processes, K_d tabulations, and what parameters control the degree of adsorption for each contaminant are discussed in detail elsewhere. For example, there is a recent three volume set published by the Environmental Protection Agency (EPA 1999a, 1999b, and 2004). Other compilations, such as Ames and Rai (1978), Krupka et al. (2002), and Krupka et al. (2004), discuss the geochemistry and solution speciation, using Eh-pH or Pourbaix diagrams, for many of the contaminants reviewed herein.

It is also assumed that the K_d values do not depend on the isotope number for a given element and that all isotopes of a given element have the same K_d value. That is, the K_d for ^{152}Eu, ^{154}Eu, and ^{155}Eu are the same for a given soil/water system and geochemical conditions; and K_ds for ^{234}U, ^{235}U, and ^{238}U are the same, etc. The assumed lack of sensitivity to isotope number for K_ds is not necessarily upheld for other parameters such as soil to plant, plant to animal or human uptake factors and toxicity, which sometimes do depend on the isotope number.

The last discussion point is adsorption-desorption hysteresis, which is a term used to describe a commonly observed phenomenon where measured K_d values differ depending on whether the measurement was made by putting the contaminant in solution and contacting “clean” solids or taking previously contaminated solids and contacting them with clean water. Quite often the K_d value measured by the former method yields a lower value than when the K_d value is measured by leaching or desorption off the solid. There has been significant discussion of this hysteresis phenomenon and several process level or mechanistic explanations have been offered. More discussion is found in EPA (1999a) but knowledge of the cause(s) is not complete. For this K_d value, it was assumed that the key scenarios of interest are biased toward desorption processes; this is yet another reason to favor higher K_d values than tabulations that are concerned mostly with how fast contaminants might reach the groundwater. Table 1.1 notes which elements seem to be more sensitive to adsorption-desorption hysteresis. But in general, most of the K_d values chosen in this tabulation and shown in the left hand columns in Tables 1.2 and 1.3 have been increased somewhat to account for some desorption hysteresis.

Table 1.1. Qualitative Ranking of Adsorption-Desorption Hysteresis

Qualitative Degree of Sorption Hysteresis				
	Non-Groundwater Scenarios			
Constituent	**High**	**Moderate**	**Low**	**Comment**
Americium	**X**			Precipitation rxn usually cause
Bismuth	**X**			Precipitation rxn usually cause
Carbon tetra chloride		**?**		"Aging" effects often discussed
^{14}Carbon as carbonate			?	Could be moderate when isotope exchange into crystal lattics sites is significant
^{36}Chlorine			X	
Cesium		**X**		Special case where clays with frayed edge sites dominate
Cr as chromate		x	X	Redox induced precipitation rxn can move to moderate
Cobalt		X		Sorption of ferric oxides not displaced by competitive common cations; requires drop in pH
Iodide		**X**		Common observation some suggest organic matter is key
Lanthanides	**X**			Precipitation rxn usually cause
Lead (Pb)	**X**			Precipitation rxn usually cause
Neptunium		**X**		Redox induced precipitation rxn usually cause
Nitrate/Nitrite			X	
Nickel		X		Sorption of ferric oxides not displaced by competitive common cations; requires drop in pH
Plutonium	**X**			Redox induced precipitation rxn usually cause
Polonium	**X**			Precipitation rxn usually cause
Protactinium	**?**			Precipitation rxn might be cause
Radium			X	
Selenium		**X**		Can be redox sensitive and selectively adsorbed by ferric oxides
Strontium			X	Isotope exchange into carbonate minerals can generate hysteresis
Technetium		**x**	X	Can be redox sensitive and reduced form high insoluble
Tritium (^{3}H)			X	
Uranium (VI) (short times)		**X**		Precipitation rxn usually cause
Uranium (VI) (long times)		**x**	X	Can be redox sensitive and reduced form high insoluble

Bold X = strong consensus amongst geochemists.
X= most geochemists favor this choice; most likely tendency.
Small x = also possible but less likely; hysteresis usually occurs with redox change to reducing conditions.
? = literature is not available or contradictory (experts in active debate) or placed in category that author favors.

Table 1.2. Comparison of Recommended K_d Values from this Activity to Past Tabulations for Non-Groundwater Scenarios

	K_d Values (mL/g)			
	Non-Groundwater Scenarios			
	This Work		**Napier and Snyder (2002)**	**Napier and Snyder(2002)**
Constituent	**Best**	**Range**	**Best**	**Range**
Americium	**500**	**60 to 5000**	1500	67 to >1200
Bismuth	**400**	**100 to 5000**	900	NA
Carbon tetra chloride	**1.0**	**0.1 to 5**	NA	NA
^{14}Carbon as carbonate	**7**	**0.5 to 100**	7	0.03 to 4.56
^{36}Chlorine	**0.5**	**0 to 2**	1	-0.008 to -0.13
Cesium	**2000**	**200 to 5000**	2000	>200 to 10,000
Cr as chromate	**3**	**0.3 to 10**	NA	NA
Cobalt	**50**	**10 to 1000**	NA	NA
Iodide	**3**	**0 to 15**	15	0.05 to 15
Lanthanides	**400**	**50 to 3000**	1500	1000 to >2000
Lead (Pb)	**600**	**270 to 10,000**	80,000	13,000 to 79,000
Neptunium	**25**	**2 to 50**	25	2.4 to 21.7
Nitrate/Nitrite	**0.5**	**0 to 2**	NA	NA
Nickel	**200**	**50 to 1500**	2400	50 to 2350
Plutonium	**600**	**200 to 5000**	5000	80 to 4300
Polonium	**400**	**150 to 1100**	1100	196 to 1063
Protactinium	**25**	**150 to 10,000**	3600	NA
Radium	**200**	**5 to 500**	500	214 to 467
Selenium	**15**	**3 to 30**	2	-3.4 to 0.78
Strontium	**50**	**5 to 200**	180	5 to 173
Technetium	**0.5**	**0 to 2**	2	-3.4 to 0.57
Tritium (^{3}H)	**0.2**	**0 to 1**	0.7	0 to 0.7
Uranium (VI) (short times)	**30**	**5 to 50**	7	0.08 to 3.5
Uranium (VI) (long times)	**5**	**0 to 20**	7	0.08 to 3.5

NA = Not available or not discussed.

Table 1.3. Recommended K_d Values and Values for Waterborne Scenarios Used in Other Tabulations

Constituent	**Non-Water Scenarios**		**K_d Values (mL/g)**									
			Water Scenarios									
			Last et al. (2004)		**Krupka et al. (2004)**		**Thibault et al. (1990)**		**Coughtrey and Thorne (1983a,b), Coughtrey et al. (1983, 1984a, 1984b, 1985)**		**Ames and Rai (1978)**	**Onishi et al. (1981)**
	This Work		**Last**	**Last**	**Krupka**	**Krupka**	**Thibault***	**Thibault***	**Coughtrey**	**Coughtrey**	**Ames & Rai**	**Onishi**
	Best	**Range**	**Best**	**Range**	**Best**	**Range**	**Sand**	**Loam**	**Best**	**Range**	**Range**	**Range**
Am	**500**	**60 to 5000**	NA(1)	NA	300	60 to 1300	1900	9600	2000	1200 to 8700	125 to 800	85 to 900
Bi	**400**	**100 to 500**	NA	NA	NA	NA	100(2)	450	NA	NA	NA	NA
Carbon tetra chloride	**0.6**	**0.1 to 1**	0.2	0.1 to 0.6	NA	NA	NA	NA	NA	NA	NA	NA
^{14}C	**7**	**0.5 to 100**	unsuitable	unsuitable	5	0.5 to 1000	5	20	NA	NA	NA	NA
^{36}Cl	**0.5**	**0 to 1**	NA	NA	0	0 to 0.6	NA	NA	NA	NA	NA	NA
Cs	**2000**	**200 to 5000**	2000	200 to 10,000	2000	500 to 4000	280	4600	1000	100 to 10,000	>200 to 2000	50 to >1000
Cr	**3**	**0.3 to 10**	0	0 to 0.3	0	0 to 0.6	70	30	NA	NA	NA	NA
Co	**50**	**10 to 1000**	NA	NA	2000	1000 to 12,500	60	1300	10 to 15	NA	30 to 1000	
I	**3**	**0 to 15**	0.2	0 to 2	0.25	0 to 15	1	5	6	NA	NA	<5
Lanthanides	**400**	**50 to 3000**	200	10 to 1000	300	60 to 1300	400-500	800-1500	NA	NA	>1900	400 to 3000
Pb	**600**	**270 to 10,000**	NA	NA	10,000	8000 to 80,000	270	16,000	NA	NA	NA	NA
Np	**25**	**2 to 50**	10	2 to 30	15	2 to 25	5	25	50	0.2 to 929	2.4 to 30	2.4 to 35
Nitrate/Nitrite	**0.5**	**0 to 1**	NA	NA	0	0 to 0.6	NA	NA	NA	NA	NA	NA
Ni	**200**	**50 to 1500**	NA	NA	300	50 to 2500	400	300	20	NA	NA	NA
Pu	**600**	**200 to 5000**	600	200 to 2000	150	50 to 2000	550	1200	5000	18 to 10,000	>2000	>500
Po	**400**	**150 to 1100**	NA	NA	NA	NA	150	400	ND	ND	NA	NA
Pa	**25**	**150 to 10,000**	NA	NA	15	2 to 25	550	1800	NA	NA	NA	NA
Ra	**200**	**5 to 500**	NA	NA	14	5 to 200	500	36,000	NA	NA	NA	200 to 500
Se	**15**	**3 to 30**	5	3 to 10	7	3 to 15	150	500	>9	NA	NA	NA
Sr	**50**	**5 to 200**	22	10 to 50	14	5 to 200	15	20	NA	NA	12 to 70	15 to 30
Tc	**0.5**	**0 to 1**	0	0 to 0.1	0	0 to 0.6	0.1	0.1	0.11	ND	0	0
^{3}H	**0.2**	**0 to 1**	0	0	0	0	0.06	20	NA	NA	0	0
U (short times)	**30**	**5 to 50**	0.8	0.2 to 4	1	0.1 to 80	35	15	NA	NA	NA	NA
U (long times)	**5**	**0 to 20**	0.8	0.2 to 4	1	0.1 to 80	35	15	NA	NA	NA	NA

(1) NA = not available or not discussed.
(2) Values in red type were estimated from plant uptake factors.
(3) Unsuitable = Authors do not feel that carbon-14 sequestration in sediments can be quantified by K_d construct.

2.0 Recommended K_d Values

Table 1.2 presents the new recommended K_d values and ranges, which should replace those given in Napier and Snyder (2002, Table 2), that should be used for scenarios involving humans, animals and plants getting exposed in the future getting exposed to contaminants present in agricultural soil and river bank sediments. The table compares the new recommendations to K_d values listed in Napier and Snyder (2002, Table 2) that were generally biased high to assure conservatism.

In addition, Table 1.3 presents the data available in past compendiums that focused on tabulating K_d values for waterborne scenarios, where low K_d values assure conservatism in risk predictions of contaminant transport in water. More discussion on the past compendiums follows. To reiterate, the terms non-groundwater scenario and waterborne scenario are used to differentiate the scenarios being considered in this report from the traditional groundwater scenarios, which are most often the key risk driving scenarios. Non-groundwater scenarios are those described in the Section 1.0 and include the adsorption of contaminants in irrigation water by the near-surface agricultural soils and river bank sediments. Briefly, non-groundwater scenarios include human, animal, and plant exposure to contaminants attached to solids, such as soil or waste particles. The sources of contaminants found attached to the solids include liquid waste that once percolated through the solids and left contaminants via adsorption/precipitation processes, and particles of solidified waste/contaminated sediments brought to the ground surface via drilling, land contouring, animal burrowing, or wind erosion.

This report does not discuss probability distribution functions for K_d values for any elements except cesium and strontium. This is because there is an adequate data population of K_d values for other elements wherein enough laboratory or field measurements have been made in which all the parameters that affect the K_d value are kept similar enough to claim that the observed K_d values do in fact come from the same population. That is, one cannot take K_d values for a given element that represent measurements on many different adsorbents or many different water types and claim that the values are from the same statistical population and the main variation is caused by inherent natural heterogeneity. The numerical value measured is so dependent on the type of adsorbent, its available surface area, the type of solution bearing the contaminant and the concentrations of all the aqueous species in the solution that most K_d datum represent a distinct statistical population. Seldom is the same adsorbent and same solution studied with adequate replication to establish a true statistical population that can be used to develop probability distribution functions.

However, the Characterization of Systems Project has decided to use a mathematical protocol to take "best" K_d values and K_d value ranges and generate probability distribution functions. The details are presented in Last et al. (2006) and are repeated herein as a quote. Words in parentheses were added for clarity.

> 'Probability distribution functions for the K_d values were generated according to the following set of rules and derived from the minimum, maximum, and best estimate K_d values (provided by the expert geochemists for scenarios of interest).
>
> Case 1: Where the minimum estimate, best estimate, and maximum estimate were all greater than zero, a lognormal distribution was assumed. The best estimate was assigned

> to the median value. The minimum estimate was assigned to the lower 1% tail of the distribution, and the maximum estimate was not used in defining the distribution.
>
> Case 2: Where the minimum estimate was zero, but the best estimate and maximum estimate were greater than zero. A lognormal distribution was used, with the best estimate assigned to the median value, the lower 1% tail of the distribution assigned to the value 0.001, and the maximum estimate used to define a probability truncation limit for the upper tail of the distribution (if less than 0.99 probability, otherwise truncation was set to 0.99).
>
> Case 3: Where the minimum and best estimates were zero, but the maximum estimate was greater than zero. A composite distribution was used. The value zero was assigned a 50% probability. The other portion of the distribution was assigned a triangular distribution where the minimum and mode were both zero and the maximum was assigned to the upper tail estimate.
>
> Case 4: Where the best estimate is “unsuitable” or not provided, a uniform distribution is assumed between the minimum and maximum values.
>
> In those cases where a lognormal distribution was assumed, the lognormal distributions were truncated at the 1% and 99% levels, thereby preventing the generation of values that could fall below the minimum estimate.’

This paradigm allows for stochastic risk predictions to be performed and, if stochastic risk predictions are required for scenarios addressed in this report, the same paradigm is acceptable. For cesium and strontium K_d values, Case 1 would be used if one follows Last et al. (2006), which relies on a log normal distribution.

2.1 K_d Values Available in Other Compendiums Germane to This Activity

Thibault et al. (1990) and Sheppard and Thibault (1990) generated a compendium of K_d values for four types of soils: clay-dominated, loam, sand-dominated, and organic-dominated. Based on their generic descriptions, a future Hanford Site “agricultural soil” would remain in the sand-dominated category. The clay dominated category contains >35% clay and the organic-dominated category contains >30% organic matter such as bog and peat materials. To allow some perspective of the variation in K_d in the general literature for adsorption of radionuclides to different soil types, I provide the values for both the sand-dominated and loam categories used by Thibault et al. (1990) in Table 1.3. Their definition of a loam is equal distributions of sand-silt-clay or greater than 80% silt and clay (by default <20% sand). Some of the contaminants’ K_d values from Thibault et al. (1990) as listed in Table 1.3 were derived from soil-plant transfer factors because no data were found in the literature when they performed their review. The values that were estimated from plant uptake factors are italicized and in red type. Besides the contaminants of interest herein, Thibault et al. (1990) provide values for antimony (Sb), arsenic (As), barium (Ba), cadmium (Cd), calcium (Ca), copper (Cu), curium (Cm), iron (Fe), lithium (Li), manganese (Mn), molybdenum (Mo), tellurium (Te), terbium (Tb), thorium (Th), tin (Sn), zinc (Zn), and zirconium (Zr).

Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1984b, 1985) published a six volume compendium of K_d, plant-soil transfer factors, animal-food transfer factors and information on human assimilation of radionuclides from plants and animals in both terrestrial and aquatic environments. Parameters to quantify these transfer or uptake factors between soils, waters, plants and animals for the following elements are tabulated in the Coughtrey et al.'s six volume compendium: americium (Am), antimony (Sb), bromine (Br), cadmium (Cd), cesium (Cs), cerium (Ce), chloride (Cl), chromium (Cr), cobalt (Co), curium (Cm), iodine (I) iron (Fe), lanthanides, manganese (Mn), molybdenum (Mo), neptunium (Np), nickel (Ni), niobium (Nb), plutonium (Pu), rubidium (Rb), ruthenium (Ru), selenium (Se), silver (Ag), sodium (Na), strontium (Sr), sulfur (S), technetium (Tc), tellurium (Te), tin (Sn), zinc (Zn), and zirconium (Zr). The K_d values listed for terrestrial soils in the six volume compendium are shown in Table 1.3.

The Hanford Site-specific K_d recommended values and ranges from Last et al. (2004), which emanates from the tabulations and discussions found in Cantrell et al. (2003), and from the Hanford Site integrated disposal facility (IDF) geochemical data package, Krupka et al. (2004) are also shown in Table 1.3. Cantrell et al. (2003) tabulate K_d values for most of the contaminants of interest to this report. Krupka et al. (2004) also tabulate K_d values for the following elements/constituents not discussed in this report: actinium (Ac), niobium (Nb), ruthenium (Ru), tin (Sn), thorium (Th), and zirconium (Zr).

2.2 Recommended Best K_d and Range for Agricultural Soils and Columbia River Bank Sediments

The second and third columns from the left in Table 1.2 list the recommended best K_d value and range to use for risk and performance assessment scenarios (excluding the "water-borne" scenarios) mentioned in Section 1.0. Table 1.2 also includes values recommended in the past (Napier and Synder 2002) for the same non-groundwater scenarios of interest herein.

Table 1.3 lists the K_d values tabulated for water-borne scenarios or pathways that are being or have been used to perform risk and performance assessments at the Hanford Site and other nuclear waste disposal sites. A brief rationale for the choice of the best range for each contaminant is provided in Section 3 along with some discussion on literature recently obtained and not used in past Hanford K_d tabulations such as Last et al. (2004) and Krupka et al. (2004).

As mentioned in the Section 1, adsorption-desorption hysteresis is a commonly observed phenomenon in laboratory batch K_d studies. Table 1.1 ranks the degree, which is a qualitative measure of both frequency and magnitude of the difference (K_d desorption value > K_d adsorption value), to which each contaminant appears to be effected. The qualitative ranking categories are high, moderate, and low degrees of impact. These qualitative rankings are based on geochemical literature discussions and expert opinion/experience of the author. For those contaminants with a high degree of impact some further discussion is often found in the individual element discussions in Section 3.

3.0 Description of Literature Reviewed to Update Non-Groundwater Scenario K_d Values for Agricultural Soils and River Bank Sediments

This section contains details on literature reviewed to augment past Hanford K_d tabulations, which in general have ignored agricultural soil and Columbia River bank sediments, and justification for the K_d values chosen in Table 1.2 columns two and three from the left.

3.1 Americium

Americium is found predominately in solution in the +3 valence state and in low ionic strength groundwater or vadose-zone pore waters with circum-neutral pH (see EPA 2004). At pH values between 6 and 8 americium aqueous complexes with hydroxide and carbonate dominate and these complexes have cationic charges (see Figure 5.1 in EPA (2004). Americium is also only moderately soluble at circum-neutral pH. There is adequate Hanford Site-specific adsorption data (see Cantrell et al. 2003 and Krupka et al. 2004 for primary references) that are consistent with the general literature that indicates americium adsorbs fairly strongly adsorption to soils and sediments. Some of the generic literature that shows K_d values larger than several thousand mL/g may reflect that precipitation/co-precipitation occurred during the adsorption measurements. The reader is referred to Cantrell et al. (2003) and Krupka et al. (2004) for details on the Hanford Site-specific information.

Roussel-Debe (2005) determined the K_d for six agricultural soils near French nuclear reactors. The soil closest in physical and chemical properties to Hanford sands yielded a K_d value of 2000 mL/g. Using statistical correlations such as particle size and slurry pH, Roussel-Debe (2005) recommends an average K_d for a sandy soil with alkaline pH of 2800 mL/g. In general, the French soils contained significantly more silt and clay than Hanford sands so I recommend the value be set at 500 mL/g for Hanford-relevant agricultural soils and river bank sediments.

Tanaka and Muraoka (1999) measured the adsorption of americium onto <1 mm-size particles of several sand sediments, soils and crushed tuff and sandstone rocks that surround the proposed Shimokita low-level waste disposal site in Japan. Trace concentrations of Am(III) were placed in distilled water and contacted with the sorbents for 7 days. The adsorbed americium was desorbed using a sequence of ever more aggressive chemical reagents to ascertain how strong the adsorption was. The pH of the sand slurries, which most closely resemble Hanford sands varied from 6.4 to 7.5 whereas Hanford soil pH currently vary between 7.5 to 8.5 but with increased organic content might lower to the values in this study. The americium K_d values for the sands that most closely resemble Hanford sands varied between 1000 and 7000 mL/g and the desorption K_d values into 0.5 M salt solutions were greater than 10,000 mL/g, thus showing significant irreversibility. Based on the selective chemical reagent extractions, Tanaka and Muraoka suggested that americium was associated with hydrous iron and manganese phases in the sands. The adsorption K_d values in Tanaka and Muraoka (1999) are a factor of two to fourteen times greater than the value I recommend, but my range covers most of the range in this study. I also believe that desorption of sorbed americium often shows hysterisis (desorption can be more difficult to effect than adsorption) as found in this study but I set the upper range K_d value at 5000 mL/g to account for the fact that Hanford sands contain a high percentage of particles >1 mm.

Livens et al. (1994) measured the K_d value of americium in three fine-grained river estuary sediments proximate to Sellafield reprocessing plant. The americium K_d value was determined from extracting the pore fluids from the saturated sediments and measuring the activity in each phase. The K_d values were 3360, 3450, and 3480 mL/g for the three sediments. Because the Columbia River bank sediments are not as fine-grained, a lower K_d value of 500 mL/g was chosen, but the range was extended to values even larger than Livens et al. (1994) found.

Emery et al. (1974) and Emery and Garland (1974) studied pond sediments and pond water at U Pond on the Hanford Site in 200 West Area. On average, the pond sediments contained 53.9 pCi/g Am-241 and the overlying waters contained 1.1 pCi/L. It was assumed that the pond sediments and water represent an equilibrium condition; the in-situ K_d for americium is 49,000 mL/g. Emery and Garland (1974) and Emery et al. (1974) also extracted the pond sediments with several chemical reagents in a sequential fashion wherein each extract used became more vigorous in reaction. Over 90% of the americium was not extractable from the pond sediments even with strong reagents. These authors also subjected the pond water to filtration through a serial reactive resin bed and found that the dissolved americium was 30% cationic and 70% anionic in nature. The source of the americium reaching the U Pond was likely from plutonium purification and finishing processes, and it is possible that some of the americium reached the pond sediments as suspended particulates. Thus, the observed in-situ K_d value may be inflated over situations where the source is dissolved contaminants in waste waters. Emery and McShane (1978) describe detailed limnological and ecological studies at several Hanford ponds and ditches however only data on gross alpha and gross beta are presented for concentrations in sediments and waters. Thus, no specific element K_d values could be calculated. Emery et al. (1974) and Emery and Garland (1974) found no evidence that radiation dose at these facilities were limiting the algal, plant, or animal communities present even though calculated maximum dose rates reached 1 R per week and maximum gross beta levels reach 10^4 pCi/L in the water.

A value of 500 mL/g was chosen as a representative best K_d value based on the discussion in the following paragraphs. The recommended range of 60 to 5000 mL/g excludes the extremely high values >5000 mL/g reported in the literature because these data may be biased by precipitation reactions. Americium desorption is often hysteretic such that K_d desorption is greater than K_d adsorption e.g., see Tanaka and Muraoka (1999) and Topcuoglu et al. (2002). A value of 500 mL/g was used for the best or most probable K_d, which is larger that the value 300 mL/g recommended in Krupka et al. (2004) for water-borne pathways, but smaller than the values chosen by Thibault et al. (1990) and Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1984b, and 1985). The value of 500 mL/g is near the mid point of ranges recommended in the two early Hanford tabulations by Ames and Rai (1978) and Onishi et al. (1981). Note that Last et al. (2004) do not list K_d data for americium. The recommended value of 500 mL/g is lower than the value (1500 mL/g) chosen in Napier and Snyder (2002), which was based on less critical analysis of past tabulations and general literature.

Thus, because the source of americium in the Hanford farm, native American, and recreational land use scenarios will be solids containing americium that must desorb to migrate, the "best" K_d value should be greater than values chosen to represent the adsorption of americium from contaminated waters (i.e., groundwater scenario data). Because desorption is slower and more difficult than adsorption processes, a higher K_d value is recommended for the non-groundwater scenarios. If K_d sensitivity predictions are performed, they should weight values higher than 500 mL/g more strongly than K_d values lower than 500 mL/g. If using an americium K_d value of 500 mL/g does not generate significant risks in any non-groundwater scenario of interest, it is likely that this contaminant will not be an important risk driver for future Hanford conditions. The available data, however, is too sparse to select quantitative probability distribution functions but as mentioned weighting values above 500 mL/g up to the upper range value of 5000 mL/g is supportable because of the desorption hysteresis.

3.2 Bismuth

Bismuth is generally assumed to exist in natural environments in the +3 valence state and when in solution at circum-neutral pH would be present as hydroxide complexes; predominately cationic and neutrally charged (Ames and Rai 1978). Bismuth is slightly soluble at circum-neutral pH. There is no available sorption-desorption data for bismuth adsorbing to Hanford Site-specific sediments under Hanford geochemical conditions, and little general literature exists. Bismuth is a daughter product of uranium decay and is generally found to associate with sediments in comparison with uranium when slurries of bottom sediments from the ocean or lakes are analyzed (see references listed in Onishi et al. 1981).

Ulrich and Degueldre (1993) determined the influence of the ionic strength and of pH on the adsorption/desorption processes of lead, bismuth, and polonium on montmorillonite clay. The solution composition had no influence on the sorption/desorption of both bismuth and polonium. The K_d values for ^{210}Bi and ^{210}Po ranged from 10^4 to 10^7 mL/g (note that the values on the high end might reflect precipitation). When adsorption and desorption coefficients are compared, the K_d values for bismuth and polonium adsorption were several orders of magnitude lower than those obtained for desorption. The concentrations of these two elements in the liquid phase are limited by the formation of bismuth- and polonium-colloids prior to the sorption step. This fact could explain the differences in the K_d values between the adsorption and desorption processes and the extremely high adsorption K_d values. Only very small amounts of bismuth and polonium could be desorbed from the montmorillonite suggesting quasi-irreversible adsorption is occurring. The difference in the K_d values between adsorption and desorption is approximately two orders of magnitude in the case of bismuth, which would classify bismuth as highly impacted by sorption hysteresis.

Based on these measurements on montmorillonite clay, bismuth is either removed from solution by strong adsorption and/or precipitation processes. The value of 400 mL/g was chosen arbitrarily as the best K_d value and a range of 100 to 5000 mL/g was chosen based mainly on expert judgment. As shown in Table 1.3 no other tabulations are available for bismuth K_d values. The value of 400 mL/g was chosen to reflect that bismuth may be slightly more soluble than americium and thus might have a lower effective K_d. The recommended bismuth K_d value is less than the value of 900 mL/g tabulated in Napier and Snyder (2002) in order to use a bit less conservatism. The range was increased from 100 to 5000 to better reflect the lack of measured data. If predicted doses from bismuth contamination in Hanford scenarios ever rise large enough to merit concern, some Hanford Site-specific adsorption-desorption testing using appropriate soils/sediments and pore waters would be needed to validate the K_d values recommended herein.

3.3 Carbon-14

As discussed in Cantrell et al. (2003), carbon-14 released at Hanford is not well understood and how to describe its fate is problematical. Carbon-14 when in soil and sediment pore waters of interest to this report is assumed to be present predominately as the bicarbonate anion. Bicarbonate in Hanford waters will react with calcium carbonate minerals that are ubiquitous in the Hanford sediments. Through isotope exchange reactions, any $H^{14}CO_3^-$ ions in solution will react with stable carbonate ions bound in carbonate-bearing minerals in the soil such that the $^{14}CO_3$ species will be incorporated into the solids until equilibrium is reached. At equilibrium, the ratios of ^{14}C to ^{12}C in the solids, in the surrounding pore water, and in the vadose zone air present in the pores will all be the same. Further, there is a constant supply of ^{14}C being formed by cosmic ray bombardment of nitrogen in the atmosphere such that a global supply is present that will mingle with any ^{14}C that was generated during fuel irradiation at Hanford and directly released or released during fuel reprocessing. Plants also utilize carbon during photosynthesis and do discriminate between the various isotopes; ^{12}C, ^{13}C, and ^{14}C such that the isotopic dynamics are quite complicated. Isotope exchange can be considered as a specialized form of adsorption-desorption that is not readily described by the K_d construct.

The author recommends a K_d value of 7 mL/g and a range of 0.5 to 100 mL/g for carbon-14 based on very limited Hanford specific data (see Krupka et al. 2004) and general literature that is hard to formulate directly as K_d values. The K_d value was increased from 5 to 7 mL/g to reflect the fact that non-groundwater scenario K_d values should be larger than values chosen for groundwater scenarios. The recommended value is the same as Napier and Synder (2002) and slightly larger than the value recommended by Thibault et al. (1990). No other tabulations offered carbon-14 K_d values. Last et al. (2004) specifically avoid offering K_d values based on the isotopic exchange conundrum. The range shown in Table 1.2 was chosen to be larger than Thibault et al. (1990) but smaller than the range discussed in Krupka et al. (2004), which considered carbon-14 sequestration in large masses of cement waste forms and concrete structures.

Should scoping risk assessments suggest that carbon-14 is a significant risk driver and if actual measurements of the amount of carbon-14 present in Hanford waste, sediments, and waters become available some of the more mechanistic isotope exchange reactions between air, soil, water, and plant material might be used for specific scenarios to improve risk predictions. More discussion of these ideas can be found in Martin (1996) and references therein.

3.4 Carbon Tetrachloride

Very little literature was found that describes the interactions of carbon tetrachloride with soils and sediments in a quantitative fashion. Most of the literature describes the interaction only in qualitative terms and shows that carbon tetrachloride does not interact strongly with soils and sediments.

Harmon et al. (1992) determined the K_d for trichloroethylene, carbon tetrachloride and vinyl chloride using a gravelly sand aquifer sediment taken from Moffet Field, California. Batch K_d tests were performed at 1:1 sediment to solution ratios in glass ampoules. Radiotraced (^{14}C-organic species) were added to simulated groundwater and tests performed for 1, 3, 5, 10, and 30 days of contact. The K_d value for carbon tetrachloride was found to be 1.1 ± 0.2 mL/g.

Other quantitative data relevant to the Hanford Site is reviewed in Cantrell et al. (2003). There are some traditional laboratory batch K_d test results specific to Hanford, and it is common knowledge, based on the large plume of carbon tetrachloride in the upper unconfined aquifer at Hanford (see Hartman et al. 2004) that carbon tetrachloride is quite mobile in the Hanford subsurface. The range of carbon tetrachloride K_d values reported by Cantrell et al. (2003) was expanded slightly to 0.1 to 5 mL/g and the best value was raised to 1 mL/g to keep carbon tetrachloride from being rapidly removed from the near surface environment. This increased K_d value should be appropriately conservative for the non-water scenarios.

The amount and concentration of carbon tetrachloride that would be found along the Columbia River shoreline or in areas outside the 200 Area exclusion zone are likely very small. Also the mass of carbon tetrachloride that has been solidified in solid waste forms and destined to be buried at near-surface Hanford burial grounds that might be inadvertently drilled into after Hanford lands are released is also likely very minute.

A best K_d value of 0.6 mL/g carbon tetrachloride was chosen based on the high-end range selected by Last et al. (2004) for groundwater scenarios. However, non-groundwater scenarios should be assigned larger K_d values. The lower value in Last et al. (2004) tabulation was maintained for the lower range and the upper range was increased to 1 mL/g. Napier and Synder (2002) did not tabulate K_d values for carbon tetrachloride.

3.5 Cesium

There is a wealth of Hanford Site-specific as well as general literature on cesium adsorption-desorption interactions, but not much on projected Hanford agricultural soils and river bank sediments. Cesium exhibits fairly simple aqueous speciation in natural waters where it is predominately found as the free monovalent cation, Cs^+ (see EPA 1999b). It reacts with soils and sediments by ion exchange and some unique specific-adsorption reactions with certain types of clay minerals, which are found in good abundance in Hanford soils and sediments. Illite, biotite, and vermiculite minerals are highly selective to adsorbing cesium and once adsorbed, the cesium is only partially released with difficulty from the aforementioned clay minerals

Shimada et al. (1996) determined K_d values, transfer coefficient by direct foliar absorption (K), and transfer coefficients for root uptake for strontium-90 and cesium-137 for several Japanese soils. The K_d values were found to be 10 to ~300 for strontium-90 and 10 to 2000 mL/g for cesium-137.

Chang and Hsu (1993) determined the adsorption of trace amounts of cesium-137 onto bentonite clay (<75-micron-size fraction) and reagent grade sand (silica 250-micron-size fraction) using distilled water with contact times of three days in batch tests. The sand slurry pH was 4.9 and the bentonite slurry pH was 6.1, which are both lower than Hanford Site conditions although in general the sorption of cesium is not strongly sensitive to pH. The cesium K_d for the bentonite was 6200 mL/g and for the silica sand was 29 mL/g.

Lima and Mazzilli (1994) measured the adsorption of cesium onto oven-dried river sediments from the Pinheiros River in Brazil using actual river water with pH 5.5 to 6.0. The river sediments were quite fine grained; 49% fine sand, 10% coarse sand, and 40% silt/clay with a total cation exchange capacity of 11 meq/100 g, which is ~2.5 to 3 times larger than the total cation exchange capacity of typical Hanford sand sediments. Batch adsorption tests as a function of pH, solid to solution ratio, and contact time were performed. Lima and Mazzilli (1994) found quite low K_d values for cesium (21 to 33 mL/g) and no sensitivity to pH (between 4 and 8) and that equilibrium was reached within a few days. They felt that oven drying the sediment had collapsed the clays and lowered the true adsorption capabilities of the sediments.

Pulford et al. (1998) studied the geochemical associations of Sellafield waste radionuclides in saltmarsh sediments from south-west Scotland. The radionuclides are transported to this environment in association with particulate material and cesium-137 was found to be predominantly (80 to 98%) non-extractable with saline waters. The relatively aggressive nature of the chemical extractants required to remove the radionuclides from the sediments suggests that they were in a form that was unlikely to result in their being released into the aquatic environment or taken up by plants. Plutonium had a greater potential mobility or bioavailability than cesium. The K_d values for desorption of cesium-137 from the sediment by freshwater, groundwater, and seawater were all approximately 10^5 mL/g, confirming its immobility in this environment. The desorption K_d values for stable cesium-133 were all approximately 10^6 mL/g, so the stable cesium did not have a significant influence on the radiocesium in this sediment.

The low ionic strength circum-neutral fluids expected in the near surface sediments at Hanford promote strong adsorption of cesium. Therefore, the recommended K_d value was chosen as 2000 mL/g in agreement with the two recent Hanford tabulations (Cantrell et al. 2003; Krupka et al. 2004) and the Napier and Synder (2002) tabulation. The cesium K_d range is 200 to 5000 mL/g based on averaging the ranges cited in the two recent Hanford tabulations for water-borne scenarios. There is enough cesium adsorption data for Hanford sediments and in general literature to suggest that a probability distribution function that is log normal around the best value and which incorporates the low and high range as limits is reasonable.

3.6 Chlorine-36

The atmospheric nuclear bomb testing in the 1950s to 1960s spread chlorine-36 around the world. It is often used as a tracer to estimate water recharge in near-surface sediment (Lo Russo 2003; Prych 1998; Gee et al. 2005 and references therein) and has been used to infer fracture flow geometry at the Yucca Mountain proposed repository (Campbell et al. 2003). Chlorine-36 is a long-lived radionuclide (half life of 301,000 years) and exhibits high mobility in the vadose zone and groundwater. It is also produced in reactor operations as an activation product of chlorine-35 and also results from a small amount of natural atmospheric and subsurface production. Although chlorine-36 in the vadose zone and groundwater may be present due to atmospheric fallout and a small subsurface production rate, the bulk of chlorine-36 in groundwater at several DOE sites has been shown to result from reactor operations (Beasley et al. 1992, 1993).

The fate of chlorine-36 from the Hanford fuel re-processing is unknown. Chlorine chemistry is similar to iodine, so it is likely that a fraction of the chlorine-36 was released to the atmosphere with the iodine-129 and -131 radionuclides. Beasley et al. (1993) postulate that chlorine-36 is released as chlorine gas and as NOCl gas during fuel decladding and fuel dissolution. Because of the long half-life of chlorine-36, it is likely that breakthrough occurred on the silver scrubbers used at Hanford to remove iodine-131 from the gases released through stacks. If this is the case, then chlorine-36 would have been released to the atmosphere through the entire period of Hanford fuel reprocessing operations. Chlorine-36 is expected mainly to stay in the aqueous waste streams and, thus, much chlorine-36 may have remained with the high activity fission products in the Hanford tank waste streams and a smaller fraction should be present in other aqueous waste streams that went to cribs, ponds and trenches. Pre-bomb pulse chlorine-36:chloride ratios[1] in the atmosphere at Hanford are estimated to be 735 to 876 x 10^{-15}

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[1] Chlorine-36 to chloride ratios are atoms of chlorine-36 divided by atoms of total chloride in a given medium such as air, suspended particles, water samples or sediments/rocks.

atoms/atoms with atmospheric bomb pulse levels increasing to approximately 4000 to 8000 x 10^{-15}. Deep groundwater in the Columbia River Basalts has reported ratios of 7 to 1130 x 10^{-15} (Gifford et al. 1985). Some chlorine-36 measurements in groundwater samples from the Hanford Site were measured in 1995 [see Dresel et al. (2002) for details]. The 1995 sampling confirms the presence of measurable amounts of chlorine-36 in 17 out of 18 water samples studied. The maximum chlorine-36 concentration detected, 94.3 pCi/L in well 199-F8-1 at the 100-F Area, corresponds to a potential ingestion dose of less than 0.2 mrem/yr. The variability in chlorine-36 concentrations and in chlorine-36:chloride ratios in these 17 groundwater samples suggests that further sampling and analysis for chlorine-36 would be productive to evaluate origins. The chlorine-36:chloride ratios for the 17 groundwater samples ranged from 604 to 216,000,000 x 10^{-15}. These shallow groundwater samples range from low levels consistent with pre-bomb pulse background to levels over 4 orders of magnitude greater than the bomb-pulse ratios. The higher ratios clearly show a large Hanford Site contribution. By contrast, the ratios reported at the Savannah River Site ranged from 61 to 4452 x 10^{-15} (Beasley et al. 1992). Ratios reported at Idaho National Environmental Engineering Laboratory ranged from 539 to 1,560,000 x 10^{-15} (Beasley et al. 1993). The Hanford production and release of chlorine-36 appears to have been significantly greater than at these other two DOE sites.

The limited Hanford groundwater data suggest that chlorine-36 is found in the large-volume liquid releases to cribs during Hanford fuel re-processing. The chlorine-36 activity in the 17 groundwater samples correlates poorly with tritium, indicating that these two mobile species provide complementary information on groundwater contamination.

There is very little data available on the interactions of chloride with sediments at the Hanford Site or in general literature. Most scientists consider chloride and other halides, such as bromide, to be conservative tracers that do not adsorb to soils/sediments. In fact, some studies have observed (from chloride breakthrough curves for pulse inputs of chloride into packed soil/sediment columns) that chloride actually migrates faster than tritium. The explanation is that anions such as chloride exhibit anion exclusion properties whilst percolating through sediments that contain a negative net surface charge. The anions are repulsed by the sediment surfaces and remain in the middle of the pores in the packed columns and travel slightly faster than the some of the water molecules that interact slightly with the sediment surfaces. Gee and Campbell (1980) compared chloride and tritium breakthrough curves from typical Hanford sand sediments and found anion exclusion that could be quantified as a negative K_d with values varying between -0.008 and -0.13 mL/g. A detailed discussion of anion exclusion is found in Jurinak et al. (1987).

Seaman et al. (1995, 1996) ran bromide breakthrough curves on four sandy soils from the Upper Coastal Plain (Georgia) and found that bromide breakthrough was retarded in comparison to tritium. Bromide retardation was sensitive to whether the companion cation was sodium (NaBr) or magnesium ($MgBr_2$). At low ionic strength (0.001 N), the bromide retardation factor varied from 0.95 to 2.51, where a value less than 1 signifies anion exclusion and a value greater than 1 signifies adsorption. For the one soil studied at three ionic strengths, the bromide retardation factor dropped from 2 to 1.35 to 1.08, respectively as ionic strength was increased from 0.001 to 0.01 and 0.1 N. Because the pH of these coastal sediments was buffered by the solids to 3.9 to 5.0, they contained more positively charged sites on hydrous oxides present in the soils than would be found in Hanford Site soils. Because of the net positive charge on the hydrous oxide surfaces, the anion bromide should show some interaction, however at higher pH values more relevant to Hanford Site conditions the net charge on hydrous oxides should be more negative and anion repulsion could occur. For Hanford sediments the pH is never as low as in the studies of Seaman et al. (1995, 1996) so the phenomenon of anion sorption is not as accentuated at Hanford as for acidic soils.

Thus, the author recommends a conservative K_d value for chlorine of 0.5 mL/g for use in the agricultural, river bank, and Native American scenarios germane to this report to reflect that chloride may interact somewhat with sediments in these non-groundwater scenarios. An appropriate range would be 0 to 2 mL/g. The recommended values for chloride are the same values chosen for technetium-99, another highly mobile, predominately anionic species in Hanford near-surface sediments and soils.

A second option for chlorine-36 would be to use the values chosen for iodine-129 where the "best" value of 3 mL/g was chosen and a range of 0 to 15 mL/g. However, trace concentrations of iodide react with Hanford formation sediments in more complicated and as yet unexplained ways, such that it is not the most appropriate analog for chloride. The recommended K_d value for chlorine-36 of 0.5 mL/g is one-half the value tabulated in Napier and Snyder (2002) and the range, 0 to 2 mL/g, used in this report is more consistent than theirs. Napier and Snyder (2002) used a range of -0.008 to -0.13 mL/g, which is not compatible with choosing a best value of 1 mL/g. Last et al. (2004) did not discuss chlorine-36 and Krupka et al. (2004) chose a best value of 0 mL/g and a range of 0 to 0.6 mL/g for groundwater scenarios.

3.7 Chromium

Chromium is found in two valence states in the natural environment, as a rather immobile Cr(III) form and as a mobile Cr(VI) form. In low ionic strength solutions, only Cr(VI) form as the chromate anion [CrO_4^{2-}] is found in oxidizing and circum-neutral pH conditions. At moderately to highly reducing conditions, chromium exists as Cr(III) at circum-neutral pH and is not soluble and readily precipitates with other cations, especially Fe(III) to form insoluble oxides (see Krupka et al. 2004 and EPA 1999b for more details on chromium geochemistry). At the Hanford Site, oxidized chromium as [CrO_4^{2-}] was used as a corrosion inhibiter in reactor cooling water and in some of the fuel processing activities. Several [CrO_4^{2-}] groundwater plumes are found in the 100 Areas near the Columbia River from the usage in reactor cooling water and in the 200 Areas from usage in fuel processing to extract plutonium (see Hartman et al. 2004). Thus, there is field evidence of chromate mobility through the vadose zone and upper unconfined aquifer at Hanford.

Also, the toxicity of chromium to animals (humans and especially fish such as young salmon smolts) and plants depends on the valence state of the chromium. The oxidized chromate form is much more toxic than the reduced Cr(III) form. When chromate interacts with some types of sediments it can be reduced to Cr(III) species, which are sparingly soluble and readily precipitate. However, at the Hanford Site, laboratory tests using Cr(VI)-spiked circum-neutral pH low-ionic strength waters suggest that Hanford sediments do not appear to reduce and immobilize significant amounts of chromate over time spans of days to a month (see references in Cantrell et al. 2003). It is not clear whether significant amounts of chromate have been reduced in the Hanford vadose zone over the 40 to 50 years of operations and cleanup efforts, but based on the observed chromate groundwater plumes it would appear not. Enriched organic sediments and the decay of plant matter might raise the probability of some reduction of chromate; however, there does not appear to be any quantitative data on this process and whether future Hanford agricultural soils will effectively reduce past chromium contamination is not known.

Carey et al. (1996) measured the K_d for Cr(VI) spiked into a 0.01 M calcium nitrate background electrolyte as a function of pH on several soils from New Zealand, two of which are predominately sands with low organic carbon contents, and 0.1 wt% hydrous iron oxides, and a total cation exchange capacity of 5 meq/100 g. These characteristics are very similar to Hanford Site Hanford formation sands. The slurry pH of the New Zealand sand soils was 6.0 and 6.5, which is slightly more acidic than the Hanford formation sand pH values. Batch K_d tests were performed at solid to solution ratio of 1 g per 10 mL of spiked background electrolyte for one day at various pH values. Based on extracting values of a plot of sorption isotherms at pH values between 5 and 7, estimates of the Cr(VI) K_d varies from <1 to <5 mL/g for the two New Zealand soils that are similar to Hanford formation sands. The K_d increases significantly when the system pH drops to 4 or lower reflecting the expected tendency for increased anion adsorption onto variably-charged surface sites that become progressively more positively charged as pH drops. This study of New Zealand sand soils suggests that future Hanford soils might sequester some chromate if they become somewhat enriched with organic matter so that pH values can drop below neutrality.

Based on an adequate Hanford Site-specific database and the literature cited, a "best" K_d value of 3 mL/g was chosen as a reasonable value to keep [CrO_4^{2-}] in the surface sediments and waste solids inadvertently brought to the surface during the drilling scenario or to account for some potential chromate reduction by decay of organic matter. This K_d value is larger than the recommended value for water-borne scenarios where the concern is rapid movement to groundwater and accessibility to residents that drink the water and to the Columbia River (see Last et al. 2004 and Krupka et al. 2004). Napier and Synder (2002) did not provide K_d values for chromium. In contrast the K_d value recommended in this report of 3 mL/g is much lower than the values of 30 to 70 mL/g recommended by Thibault et al. (1990), which were heavily weighted to adsorption of chromate onto acidic soils with high hydrous ferric oxide contents from the Savannah River Site. The pH and hydrous ferric oxide contents of Hanford sediments are not similar to Savannah River soil values, so the chromium K_d values chosen by Thibault et al. (1990) and Sheppard and Thibault (1990) are not relevant to the Hanford Site.

The K_d range for chromium was selected as 0.3 to 10 mL/g to allow risk sensitivity predictions. This range is larger than ranges chosen by Last et al. (2004) and Krupka et al. (2004) which both used a range of 0 to <1 mL/g. The range of Cr K_d values was expanded to account for the possibility of the future agricultural soils to be more acidic and contain more organic matter (potential chromate reductant) and hydrous oxides (potential for more adsorption onto positively-charged surface sites).

3.8 Cobalt

Cobalt can be found in nature in two valence states, Co(II) and Co(III); however, the Co(II) is by far more prevalent (Smith and Carson 1981). In low ionic strength and circum-neutral pH waters relevant to Hanford Site, cobalt aqueous speciation is predominately complexes with hydroxide and carbonate but precipitation/co-precipitation processes can limit the dissolved concentrations of cobalt (see Krupka and Serne 2002 for more cobalt geochemistry discussion and Eh-pH diagrams). There is an adequate Hanford Site-specific database of laboratory studies of the adsorption of cobalt onto Hanford sediments that have low organic matter content and natural slightly alkaline pH conditions in waters that contain no organic complexants. Most of the adsorption studies show moderate to strong adsorption occurs if the water is low ionic strength and does not include man-made chelating and complexing agents (e.g., ethylene diamine tetraacetic acid [EDTA], cyanide) used in fuel processing activities and waste-stream recovery operations to capture specific isotopes such as uranium, cesium-137, and strontium-90. There are no Hanford-specific studies of cobalt adsorption onto agricultural soils or river bank deposits so the following literature was reviewed.

Lima and Mazzilli (1994) measured the adsorption of cobalt onto oven-dried river sediments from the Pinheiros River in Brazil using actual river water with pH 5.5 to 6.0. The river sediments were quite fine grained, i.e., 49% fine sand, 10% coarse sand and 40% silt/clay with a total cation exchange capacity of 11 meq/100 g, a value ~2.5 to 3 times larger than typical Hanford sand sediments. Batch adsorption tests as a function of pH, solid-to-solution ratio, and contact time were performed. Lima and Mazzilli (1994) found that the cobalt K_d was quite sensitive to pH and varied from 47 to 1660 mL/g as the pH varied between 4 and 8. The cobalt K_d also did not reach an equilibrium value over 15 days of contact. Thus there was a slow process that continued to remove cobalt from solution.

Bidoglio et al. (1994) studied the adsorption of cobalt (10^{-10} M) onto quartz sand from a bicarbonate-rich groundwater at pH 8.3 with and without the presence of 10 ppm humic acid. Ninety pore volumes of the groundwater were percolated through the quartz columns. Very little of the cobalt migrated through the 20-cm-long quartz column. In some tests, a 5-cm layer of manganese oxide was placed 5 cm from the inlet end. The profile of adsorbed cobalt inside these columns showed that the MnO_2 layer caused a large build up of cobalt in the manganese oxide layer. The manganese oxide could have oxidized Co(II) to Co(III) and effectively sequestered the cobalt irreversibly. The 10 ppm humic acid-laden groundwater did allow more cobalt to breakthrough out of the quartz column. However, no quantitative K_d information was presented in the manuscript thus it can only be stated that cobalt mobility is sensitive to the presence of dissolved organic matter in the pore water. See further discussions on the impacts of man-made organic ligands such as EDTA on cobalt adsorption in Krupka and Serne (2002). In general, the manmade chelating agents and, by inference from field studies, natural organic matter decay products, such as fulvic and humic acids, impact cobalt adsorption more so at basic pH conditions than acidic conditions.

Napier and Snyder (2002) and Last et al. (2004) did not review cobalt adsorption data so Coughtrey and Thorne (1983a, 1983b), Coughtrey et al. (1983, 1984a, 1984b, and 1985), Thibault et al. (1990) and Ames and Rai (1978) provide the primary cobalt K_d references used for this report. The most probable value found in Krupka et al. (2004), 2000 mL/g, in retrospect seems too large. However, the choice of a K_d for cobalt was not important in groundwater risk scenarios used for the Integrated Disposal facility (IDF) performance assessment because the only cobalt radionuclide is ^{60}Co, which has a relatively short half life of 5.7 years. Thus, little critical analyses was put into choosing cobalt's most probable K_d value for the IDF tabulation and its projected risk would have been very low even if a much smaller K_d value would have been chosen. Coughtrey and Thorne (1983a, 1983b), Coughtrey et al. (1983, 1984a, 1983b, and 1985), Thibault et al. (1990) and Ames and Rai (1978) recommend cobalt K_d values of 10 to 15, 60, and 30 to 1000 mL/g, respectively. For use in the scenarios of interest in this report a cobalt K_d value of 50 mL/g was chosen as a reasonable "best" value and a range of 10 to 1000 mL/g. The most probable value is slightly smaller than the one found in Thibault et al. (1990), which again is dominated by Savannah River sediment data that contains more hydrous ferric oxides that are very strong adsorbers of cobalt. Agricultural soils might contain higher dissolved organic complexants than the studies used by all these tabulations; however, most of the non-groundwater scenarios also require cobalt desorption to occur to remove the cobalt from the exposure pathway. Desorption hysteresis may counteract any tendency for enhanced cobalt mobility via organic complexants. Thus, these two competing processes should off set each other.

3.9 Iodine

Iodine can exist in sediment-natural water environments in three oxidation states -1 (iodide I^-), +5 (iodate IO_3^-), and 0 (I_2). However, the former two are the most prevalent at pH conditions found at the Hanford Site. Both iodide and iodate are mono-valent anions and are not expected to adsorb strongly to Hanford Site sediments that have a negative net surface charge at pH conditions of interest. Iodine aqueous species do adsorb more than chloride or pertechnetate and their adsorption tendencies are sensitive to the presence of organic matter. See Krupka et al. (2004) and EPA (2004) for more details on iodine geochemisty and speciation diagrams as a function of pH and Eh. There are no Hanford-specific studies on adsorption of iodine species on agricultural soils or river bank deposits so the following literature was reviewed.

Bird and Schwartz (1996) measured the K_d value for iodide spiked into Winnipeg River water (low ionic strength solution that is similar to rain water) onto four lake sediments. One of the lake sediments was sand that is similar to Hanford Site surface sediments. Under oxic conditions, the observed iodide K_d value was 0.2 mL/g.

Yoshida et al. (1998) used batch adsorption tests to measure the K_d value for trace amounts of iodide and iodate (added as ^{125}I tracers in distilled water) onto 69 different near-surface soils in Japan. The tests were performed at 1:10 wet soil to distilled water ratio and the contact time was 14 days. Within the group of soils (three different soils were in this group), the one with the most Hanford-relevant properties was labeled sand-dune regosols in their paper. The sand-dune regolith soils had mean cation exchange capacity of 1.4 meq/100 g and an organic carbon content of 0.06 % but the mean soil slurry pH was 6.3, which is slightly lower than at the Hanford Site. The mean K_d value for iodide on these sand-dune soils was 16 mL/g and for iodate was 14.1 mL/g. The K_d values for the three samples in the sand-dune group must have varied significantly because the standard deviations were 19.9 and 16.1 mL/g for the iodide and iodate, respectively. When the soils were autoclaved for 70 minutes at 121°C and the adsorption tests repeated, the K_d values dropped significantly to 0.7 ± 0.9 and 3.0 ± 3.1 mL/g for iodide and iodate, respectively. Yoshida et al. (1998) suggest that 86% of iodide and 50% of iodate sorption in the Japanese surface soils are attributable to microbial activities that are destroyed by the autoclaving.

At the Hanford Site, sub-surface sediments with very low organic content adsorb iodide anions with an average K_d value that ranges between 0.2 and 0.4 mL/g (see Um et al. 2004; Um and Serne 2005). Kaplan et al. (2000) found that the mineral illite could adsorb trace concentrations of iodide to yield a K_d as large as 15 mL/g. To accommodate the influence of greater organic matter content, increased micro-biological activity, and possibility of enriched silt/clay content in agricultural soils, the value chosen for the iodine K_d value is 3 mL/g and a range of 0 to 15 mL/g for sensitivity studies for scenarios of interest to this report. The recommended "best" value differs from Napier and Snyder's (2002) value of 15 mL/g and the best values from Last et al. (2004) (0.2 mL/g) and Krupka et al. (2004) (0.25 mL/g). In the former case, a value of 15 mL/g is based heavily on the iodide adsorption on illite and in the latter two tabulations the emphasis was on groundwater scenarios. Thibault et al. (1990) and Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1983b, and 1985) recommend a most probable iodine K_d value between 1 and 6 mL/g, depending on grain size. Thus, the recommendation of 3 mL/g is mid-way between most of the other non-Hanford focused tabulations and slightly larger than the Hanford groundwater scenario K_d values. The recommended range, 0 to 15 mL/g, is similar to the range chosen by Napier and Snyder (2002) and Krupka et al. (2004) but is larger that the range chosen by Last et al.

(2004), which was 0 to 2 mL/g. The increased K_d value range and the "best" K_d value were to accommodate the observed increased adsorption of iodide caused by organic matter and microbiological activity. Agricultural soils and river bank sediments may contain higher organic matter content and more microbiological activity than the sediments generally studied by Hanford Site geochemists.

3.10 Lanthanides (especially Europium)

In general, all the lanthanide elements exist in natural pore waters in the trivalent oxidation state at circum-neutral pH and mildly oxidizing conditions prevalent in the near-surface sediments at the Hanford Site. Aqueous speciation calculations suggest that inorganic complexes with hydroxide, and to a lesser extent inorganic anions such as carbonate, sulfate, and chloride, dominate over the free tri-valent cation in solution. The lanthanides are also rather insoluble such that hydrous oxide-hydroxide precipitates can be expected to keep solution concentrations quite low. Between precipitation and adsorption reactions, lanthanides show strong preferences for binding with sediments with high K_d values almost universally found. Krupka and Serne (2002) provide more details on europium geochemistry and aqueous speciation and solubility plots versus pH and Eh.

Although adsorption data for lanthanides at relevant Hanford Site conditions are sparse, data is adequate to objectively choose most probable and ranges in K_d values for the scenarios of interest. Cantrell et al. (2003) and Krupka and Serne (2002) provide discussion and observed K_d values. Wang et al. (2004) evaluated the interactions of europium (III) onto bentonite, used for backfill material in nuclear waste disposal, using computer modeling and literature information. Their literature review concluded that the K_d for europium varies as a function of pH and europium concentration present but almost universally is found to range from 100 to 10,000 mL/g. This recent study/review has the same conclusions as past Hanford-relevant studies of lanthanide adsorption onto sediments.

Recent adsorption studies by Wenming et al. (2001) and Xiangke et al. (2001) present data for some pure minerals and natural sediments (but at more acidic pH conditions than are found at Hanford Site) that point out the key controlling adsorption processes/tendencies for europium. Clay minerals such as smectites, which are the dominant clay mineral in Hanford Site sediments, strongly adsorb europium. In addition, aluminum oxides and ferric oxides also are strong adsorbents for europium. The K_d value for europium is quite sensitive to pH conditions and above pH 5 adsorption of europium is almost 100%. Wenming et al. 2001 and Xiangke et al. 2001 suggest that europium hydroxide aqueous species form "bridged" complexes with adsorbent surfaces. Desorption of previously adsorbed europium shows considerable hysteresis meaning that desorption K_d values are larger than the adsorption K_d values. Dissolved natural organic acids such as fulvic acid can lower the sorption of europium, but natural organic acids are quite low in pore waters at the Hanford Site today however may increase in the agricultural soils of the future. In selecting the recommended values the impacts of higher concentrations of organic acids and lower pH, which lower K_d values, were considered.

Given the past laboratory studies relevant to Hanford and the recent literature, which continues to support our past understanding, a "best" K_d value of 400 mL/g and a range of 50 to 3000 mL/g were chosen for the lanthanides for use in agricultural soil and river bank sediments interacting with low ionic strength and neutral pH natural waters. In a similar fashion to americium and bismuth, the desorption K_d value for lanthanides is generally larger than the adsorption K_d values so most sensitivity calculations should weight values >400 mL/g as more common. The current recommendation for the most probable or "best" K_d value for lanthanides, 400 mL/g, is lower that the value listed in Napier and Snyder (2002) and larger than the values chosen by Last et al. (2004) and Krupka et al. (2004) for groundwater scenarios, 200 and 300 mL/g, respectively. The recommended value was raised to 400 mL/g to accommodate desorption hysteresis mechanisms. This value is similar to the Thibault et al. (1990) range for sands, 400 to 500 mL/g. The chosen range is slightly wider than those chosen by Last et al. (2004) and Krupka et al. (2004) to accommodate the desorption hysteresis impacts that are more important for the non-groundwater scenarios of interest to this K_d value tabulation.

3.11 Lead

Lead in natural pore waters is generally found only as the divalent free cation or complexed with various inorganic anions such as carbonate, sulfate and chloride. Lead is not soluble in Hanford Site pore waters in contact with sediments. Phosphate solids are especially insoluble, but hydroxides, oxides, carbonates and sulfates are also rather insoluble. There have been some studies of lead adsorption onto Hanford sediments given the fact that lead-lined nuclear reactor cores from decommissioned submarines are buried at Hanford (see summary in Rhoads et al. 1992). The K_d for lead is very large (>10,000 mL/g) in Hanford geochemical systems. See further discussions on lead geochemistry and aqueous complex speciation plots in EPA (1999b). There are no Hanford-specific studies on the adsorption of lead onto agricultural soils or river bank deposits so the following literature was reviewed.

Ulrich and Degueldre (1993) determined the influence of the ionic strength and of pH on the adsorption/desorption processes of lead on montmorillonite clay. For lead, a strong dependence of the adsorption and desorption processes on the ionic strength was observed at $pH < 7$, whereas, at higher pH values, this dependence totally disappears. Large K_d values were measured for lead. The K_ds range from 10^3 to 10^5 mL/g. When adsorption and desorption coefficients are compared, the lead K_d values are similar for both adsorption/desorption. Montmorillonite clays dominate the small wt % fraction of Hanford sediment clay-sized particles, however, it is difficult to correlate the K_d values in this paper with the bulk sand sized Hanford sediments.

Napier and Snyder (2002) list the most probable K_d value for lead as 80,000 mL/g while Last et al. (2004) do not discuss lead and Krupka et al. (2004) recommend a lead K_d value of 10,000 mL/g. Thibault et al. (1990) recommend a lead K_d value of 270 mL/g for a sand sediment and 16,000 for a loam soil. The author chose 600 mL/g as the "best" K_d value to represent non-groundwater scenarios of interest to this report and a range of 270 to 10,000 mL/g. Lead sorption is often described as being affected by sorption hysteresis, and the large K_d value found in most tabulations likely are indicative of a significant level of desorption difficulty. For any scenario where lead might be present in large enough amounts to be predominantly precipitated in discrete solids values larger that 1000 to values as high as 10,000 mL/g should be used in sensitivity calculations.

3.12 Neptunium

Neptunium is a redox sensitive element that can exist in five valence states (from +3 to +7) but in natural environments found at the near surface Hanford Site only the +4 and +5 oxidation states are relevant. The Np(V) aqueous species is the neptunyl oxy-cation (NpO_2^+) that can form carbonate complexes at pH values above 8.5 or when dissolved carbonate is significantly higher than normal. Most Np(V) solids are quite soluble so that precipitation is not expected. Under slightly reducing conditions neptunium is present in the environment as Np(IV) species, which are very insoluble. In circum-neutral pH solutions, the reduced neptunium is generally fully hydrolyzed as $Np(OH)_4^0(aq)$ species that readily co-precipitates or adsorbs. Some minerals in natural sediments can reduce Np(V) aqueous species and cause Np(IV) species to bind tightly to the sediment surfaces. The Np(V) aqueous species do not form as strong of complexes with common anions such as carbonate, sulfate, and chloride as other actinides so the net charge on Np(V) aqueous complexes in often +1 leading to moderate to strong adsorption tendencies dependent upon pH of the soil/sediment water system. If Np(V) aqueous species get reduced to Np(IV) species adsorption/precipitation increases significantly. More discussion of neptunium geochemistry, adsorption tendencies, and diagrams of aqueous speciation calculations and solubility tendencies as a function of pH and Eh are found in Krupka et al. (2004) and EPA (2004). There are no Hanford-specific studies of neptunium onto agricultural soils or river bank deposits so the following literature was reviewed.

Sakamoto et al. (1990) measured the K_d for Np(V) onto four Japanese soils two of which are quartz and feldspars dominated sands that are quite similar to Hanford formation sediments at the Hanford Site. Batch adsorption tests using a 0.01 M sodium perchlorate background electrolyte spiked with 1×10^{-5} M Np(V) were performed at 30°C for 7 days. The solid to solution ratio was 1 g to 5 mL. The Np(V) K_d for the two sands was sensitive to the pH of the system. At pH values below 7, the K_d was less than 5 mL/g and as low as 1 mL/g. At pH values between 7 and 9, the neptunium (V) K_d value rose sharply to values between 7 and 15 mL/g for the two sands. Between pH values of 9 to 11 the Np(V) K_d values were fairly constant at 7 and 15 mL/g for the two sands.

Recently Niitsu et al. (1997) measured the adsorption on Np(V) species added to 0.1 M sodium perchlorate background electrolyte onto kaolinite clay as a function of pH and with and without the presence of dissolved humic acid (up to 5 g/L). In the pH range of interest to Hanford Site, pH 7 to 8.5, the K_d for neptunium varied from 2 to 20 mg/L and humic acid did not have an appreciable effect on the sorption.

Tanaka and Muraoka (1999) measured the adsorption of Np(V) onto <1 mm-sized particles of several sand sediments, soils, and crushed tuff and sandstone rocks that surround the proposed Shimokita low-level waste disposal site in Japan. Trace activities of Np(V) were placed in distilled water and contacted with the sorbents for 7 days. The adsorbed neptunium was desorbed using a sequence of ever more aggressive chemical reagents to ascertain how strong the neptunium adsorption was. The pH of the sand slurries, which most closely resemble Hanford Site sands, varied from 6.4 to 7.5 similar to the recent measurements of highly vegetated surface soils at the Hanford Site. The Np(V) K_d values for the sands that most closely resemble Hanford Site sands varied between 2.3 and 10 mL/g and the desorption K_d values into 0.5 M salt solutions ranged from 12 to 25 mL/g, thus showing some irreversibility. Based on the selective chemical reagent extractions, Tanaka and Muraokao (1999) suggested than Np(V) adsorption was associated with ion exchangeable sites in the sands.

Pratopo et al. (1991) measured the adsorption of Np(IV) present in 0.1 M carbonate solution onto quartz. The tests were performed in an argon filled glove box to keep the Np(IV) from oxidizing to the much more soluble Np(V) species (NpO_2^+). The observed K_d values were large, >130 mL/g, at pH values near 8 and up to 1000 mL/g at pH values greater than 12. Pratopo et al. (1993) measured the adsorption of Np(IV) aqueous species present in 0.1 M carbonate solution onto bentonite clay. The tests were performed in an argon-filled glove box to keep the Np(IV) from oxidizing to the much more soluble neptunyl species (NpO_2^+). The observed K_d values were large >100 mL/g at pH values near 5 and up to 10,000 mL/g at pH values greater than 8.

For near-surface environments of interest to the future agricultural and river bank sediments on the Hanford Site, neptunium may exist in the oxidized Np(V) state in bulk solution, but some of the dissolved Np(V) species may adsorb via a reduction mechanism, perhaps facilitated by decaying organic matter, to Np(IV) species that form strong bonds with sediment surfaces. Also, the desorption hysteresis phenomenon, which causes release of bound neptunium to be more difficult than the original adsorption process, was observed in some of the cited literature. Taking into consideration the possible reduction of some Np(V) and desorption hysteresis, a "best" K_d value of 25 mL/g with a range of 2 to 50 mL/g was chosen for the non-groundwater scenarios of interest. The best value is about two times higher than values recommended (see Last et al. 2004; Krupka et al. 2004) for water-borne scenarios used in other Hanford Site risk and performance assessment activities. For non-groundwater scenarios, desorption hysteresis is quite important and based on the results of Niitsu et al. (1997) natural organic complexes do not increase neptunium mobility; thus, most data suggest that neptunium K_d values for non-groundwater scenarios should be larger than values for groundwater/any waterborne scenarios. The recommended K_d value for neptunium is the same as used by Napier and Snyder (2002), the same as Thibault et al. (1990) for a loam soil, one-half the value recommended by Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1984b, and 1985) and on the upper end of ranges tabulated by Ames and Rai (1978) and Onishi et al. (1981). The recommended range is somewhat wider than the ranges recommended by Last et al. (2004) and Krupka et al. (2004) but smaller than the range reported in Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1984b, and 1985). The neptunium K_d range was increased over the range chosen by the other compilations for "groundwater" scenarios to emphasize that desorption hysteresis/reduction should be active in the "non-groundwater" scenarios of interest.

3.13 Nickel

Nickel is a transition metal, which is somewhat similar to cobalt, generally found in aqueous pore waters as the divalent free cation and as complexes with common inorganic anions such as carbonate, sulfate, chloride and hydroxide. Nickel sorption data suggests that it adsorbs fairly strongly to sediments when the pH of slurries is near neutral. In general, nickel seems to adsorb more strongly than cobalt in most sediments, including Hanford Site sands. It appears that nickel is also less prone to forming strong organic complexes with chelating agents used at Hanford Site and more prone to co-precipitating with other trace metals such as iron, manganese and aluminum when acidic wastes are neutralized. None of the aqueous speciation tabulations that we have been relying upon discuss nickel geochemistry but Baes and Mesmer (1976) and Pourbaix (1966) discuss aqueous speciation in the simple water-H^+-OH^- system and state that nickel aqueous speciation is quite similar to cobalt. There are no Hanford-specific studies of nickel adsorption onto agricultural soils or river bank deposits so the following literature was reviewed.

Stauton (2004) determined the K_d value for nickel on 13 soil types from France as a function of pH, dissolved organic carbon content, background electrolyte ($CaCl_2$ between 0.001 and 0.1 M), and competing trace metals. Most of the soils were finer grained than Hanford Site sands and had slurry pH values slightly more acidic than Hanford conditions. The K_d for the 0.01M $CaCl_2$ background solution condition, which is more saline than expected conditions in the near surface sediments at the Hanford Site ranged from 65 to 1830 mL/g. For the soils most similar to Hanford Site projected agricultural soils, the K_d ranged from 130 to 780 mL/g.

Christensen et al. (1996) measured the K_d value for nickel on 12 Danish sandy aquifer sediments using a 0.001 M $CaCl_2$ solution and found the K_d value to vary between 3 and 7250 mL/g. Most of the variability correlated with the slurry pH of the system. Low K_d values were found for acidic conditions, and the highest K_d values were found for alkaline conditions. The Danish sediments with characteristics similar to Hanford Site sands, the K_d values ranged from 250 to 440 mL/g.

Martino et al. (2004) studied the adsorption of ^{63}Ni onto estuarine suspended solids from brackish waters from the Mersey Estuary, England. The equilibrium sediment to water distribution coefficient for nickel-63 (^{63}Ni) ranged from about 200 mL/g in the upper estuary to about 1200 mL/g in the marine end-member, and ^{63}Ni added to riverine sediment exhibited little tendency to desorb when re-suspended in saline water.

Brown et al. (1994) performed sorption and transport experiments to determine how vadose-zone microbes affect sorption of Ni(II) and Cd(II) on crushed volcanic tuff and transport of Ni^{2+} in the same medium. Sorption of Ni to the tuff was less in samples inoculated with microbes than in sterile samples. Since Ni(II) sorption was the same both in the presence and absence of microbes when a buffer was used, microbes appear to decrease sorption by decreasing solution pH. Nickel isotherms were linear up to initial concentrations of 10 mg/L and the average K_d was 184 mL/g for sterile sediment. This study suggests that the presence microbes do not significantly change the adsorption tendencies of nickel to soils; however the microbes can change pH conditions and pH is a key variable that controls nickel adsorption to soils.

Based on past Hanford studies of cobalt and nickel adsorption as summarized in the tabulations noted and the new literature reviewed, 200 mL/g was chosen as a “best” K_d value and a range of 50 to 1500 mL/g for non-groundwater Hanford Site scenarios of interest herein. The recommended best K_d value is considerably lower than the value presented in Napier and Snyder (2002), which simply chose a maximum value from limited Hanford data. The recommended K_d value is also lower than the most probable Ni K_d value (300 mL/g) presented in Krupka et al. (2004) and Thibault et al. (1990; [300 and 400 mL/g for loam and sand sediments, respectively), but higher than the value (20 mL/g) chosen by Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1983b, and 1985). A best K_d value of 200 mL/g was chosen as a reasonable value to show greater adsorption than cobalt but to allow for some decreased sorption as soil pH may decrease as soils retain more organic matter and to allow for the possibility of some complexing with dissolved organic matter. The recommended range is similar to ranges shown in Tables 1.2 and 1.3. The nickel K_d may exhibit some hysteresis during desorption such that values higher than 200 mL/g should be weighted more heavily in sensitivity calculations. This weighting would capture any risk sensitivity to the choice of K_d value. If sensitivity calculations suggest nickel is a key dose contributor, site specific laboratory adsorption-desorption are recommended.

3.14 Nitrate/Nitrite

No sorption experiments have been run using trace concentrations of nitrate or nitrite contacting Hanford Site sediments. Most of the Hanford Site literature and groundwater monitoring measurements show that nitrate is quite mobile and does not interact strongly with Hanford Site sediments. The fact that large groundwater plumes of nitrate exist below many of the disposal facilities at Hanford Site attests to its mobility (see Hartman et al. 2004). It has been found that much of the nitrite present in single-shell tank supernatant solutions that have leaked into the vadose zone gets oxidized to nitrate (see Serne et al. 2002b, 2002c).

There is a large mass of agricultural literature that explains a detailed microbiological-chemical cycling of the various forms of nitrogen present in the environment (N_2 gas, nitrate (NO_3^-), nitrite (NO_2^-), and ammonia (NH_3, NH_4^+). The valence state of the nitrogen species are N(0), N(V), and N(III) for the gas, nitrate, and nitrite/ammonia species. Significant quantities of nitrogen-bearing fertilizers are applied to agricultural soils around the Hanford Site such that if Hanford Site lands are released back to the public for farming, more detailed conceptual models of how to assess the nitrogen cycle in risk assessments may be warranted.

Seo and Lee (2005) used time reflectometry probes to measure the migration of nitrate, chloride, and phosphate spiked into 0.005 M $CaCl_2$ solution that was percolated through intact cores. The test conditions were column dimensions 10 cm diameter by 15 cm long. The columns were filled with a sandy loam (71% sand, 19% silt, and 10% clay) from the Knoxville, Tennessee Agricultural Experiment Station. The natural soil pH is 5.7 and the organic carbon content was 0.89%. Although no K_d values were reported, Seo and Lee (2005) state that the migration of nitrate and chloride through the columns was almost identical. Phosphate, however, was significantly retarded. Both the chloride and nitrate were assumed to be non-interacting solutes and thus would be assigned a K_d value of zero.

Qafoku et al. (2000) measured the retardation of nitrate percolating through 13 different subtropical and tropical subsoils that contained hydrous oxide-rich minerals with variably charged surface sites. The native pH for the subsoils ranged from 4.82 to 6.40 and their anion exchange capacity (AEC) ranged from 0.01 to 1.86 cmol/kg (meq/100 g). A relationship was found that related the K_d for nitrate (present in 5 mmol $Ca(NO_3)_2$ solution) versus the anion exchange capacity of these soils for pH values within the range of the native sediments. The relationship is

$$K_d = -0.1325+1.1505*AEC$$

where the units of K_d are mL/g and of anion exchange capacity are $cmol_c$/kg (centimoles of charge per kilogram of soil; same as old convention units meq per 100 g soil).

It is not clear whether this relationship would be applicable to the more alkaline relatively un-weathered Hanford formation sand sediments present at the Hanford Site. Also, it is not known what the anion exchange capacity is for Hanford Site sediments because it has not been measured. Anion exchange capacity is measured by the procedure found in Zelzny et al. (1996). If the Hanford Site surface soils and/or Columbia River bank sediments become enriched in decaying organic matter, the pH will drop and some of the minerals and particle coatings will exhibit net positive surface charges and thus anions such as nitrate, chloride (chlorine-36), selenate, selenide, iodide, iodate, and pertechnetate might adsorb more strongly than is generally found today. This phenomenon was accommodated in the K_d value recommendations for all anionic contaminants in this report.

At present, K_d values are provided for nitrate/nitrite assuming no microbiologic interaction and account for only inorganic adsorption reactions onto Hanford Site near-surface sediments. However, the "best" K_d value of 0.5 mL/g should be used for nitrate in a similar fashion as for chloride and technetium and the range should be set at 0 to 1 mL/g for sensitivity calculations. In the other tabulations, only Krupka et al. (2004) discusses nitrate K_d values. For groundwater scenarios, they recommended 0 mL/g for the most probable K_d value and a range of 0 to 0.6 mL/g.

The recommendations in this report for nitrate are based on the assumption that Hanford Site derived nitrate contamination that will be present in the future will mainly be solidified waste forms (mainly cementitious) that are inadvertently brought to the surface during well drilling activities or present at low concentrations in near-surface sediments as remnants of past liquid disposals . Because most of highly contaminated near-surface contaminated sediments will be removed to a depth of 15 feet and backfilled with clean sediments before the Hanford Site lands are released, high nitrate concentrations of DOE-Defense-waste origin probably will not be found in the near surface soils. In fact, most of the near surface nitrate- or nitrogen-containing sediments in future agricultural soils would likely come from application of fertilizers or less likely use of previously contaminated groundwater for irrigation.

3.15 Plutonium

The geochemistry of plutonium in near-surface environments germane to the Hanford Site can be quite complex. The four valence states +3, +4, +5, and +6 are all possible for plutonium species in solution or in solid precipitates. However, it is likely that dissolved plutonium will be dominated by the oxycations PuO_2^+ [Pu +5] and PuO_2^{2+} [Pu +6] in near-surface Hanford sites of interest. At circum-neutral pH and mildly oxidizing conditions both forms of plutonium would likely be present in solution as complexes with inorganic anions (hydroxide and carbonate), although similar to Np(V), the free cation PuO_2^+ [Pu(V)] can dominate speciation in the weak acid to neutral pH region of low ionic strength waters. Also similar to neptunium, aqueous plutonium species can be reduced when interacting with the surfaces of many common minerals found in Hanford Site sediments. Reduced plutonium, Pu(III) and Pu(IV), are relatively insoluble at neutral pH conditions and also strongly sorbed onto solids. For more details on plutonium geochemistry and adsorption tendencies see the summaries presented in EPA (1999b), Ames and Rai (1978), Onishi et al. (1981) and Rai and Serne (1978). Based on these publications in general, no matter what the redox conditions at circum-neutral pH, plutonium exhibits strong adsorption tendencies in most environments. There are no Hanford-specific studies on plutonium adsorption onto agricultural soils or river bank deposits so the following literature was reviewed.

Roussel-Debe (2005) measured the K_d values for plutonium onto six agricultural soils taken from near French nuclear reactors. The plutonium was added to the lab tests in an oxidized form, but no valence state measurements of the ending state of the plutonium were performed. The soil that was most similar to Hanford Site sands yielded a K_d value of 540 mL/g. Based on statistical correlations that showed the plutonium K_d value was most sensitive to particle size, Roussel-Debe (2005) recommended a default value of 2200 mL/g for plutonium sorbing to agricultural soils in France. However, the French soils in general had much more silt and clay content than Hanford Site sands.

Tanaka and Muraoka (1999) measured the adsorption of Pu(IV) onto <1 mm-size particles of several sand sediments, soils and crushed tuff and sandstone rocks that surround the proposed Shimokita low-level waste disposal site in Japan. Trace activities of Pu(IV) were placed in distilled water and contacted with the sorbents for 7 days. The adsorbed plutonium was desorbed using a sequence of ever more aggressive chemical reagents to ascertain how strong the adsorption was. The pH of the sand slurries, which most closely resemble Hanford Site sands varied from 6.4 to 7.5 similar to the recent measurements of highly vegetated surface soils at the Hanford Site. The Pu(IV) K_d values for the sands that most closely resemble Hanford Site sands varied between 250 and 700 mL/g and the desorption K_d values into 0.5 M salt solutions ranged from 2000 to 5000 mL/g, thus showing high irreversibility. Based on the selective chemical reagent extractions, Tanaka and Muraoka (1999) suggested that plutonium was associated with hydrous iron and manganese phases in the sands.

Livens et al. (1994) measured the K_d value of plutonium on three fine-grained river estuary sediments proximate to Sellafield reprocessing plant. The plutonium K_d was determined from extracting the pore fluids from the saturated sediments and measuring the activity in each phase. The K_d values varied significantly with values 166, 730, and 1010 mL/g for the three sediments. No plutonium valence state measurements were made on the pore fluids. No explanation was offered for the wide range but it was noted that plutonium exhibited less adsorption than americium for these three river fine-grained sediments.

Lu et al. (2003) measured the adsorption of ^{239}Pu(V) at 2.33 x 10^{-7} M spiked into groundwater from the Yucca Mountain repository site onto fine colloids of hematite, montmorillonite, and silica as a function of time, temperature, ionic strength and colloid concentration. For all three adsorbents, the plutonium K_d was large 7 x 10^{+5}, 1 x 10^{+4}, and 7 x 10^{+3} mL/g for hematite, montmorillonite, and silica colloids, respectively. Desorption of ^{239}Pu from ^{239}Pu-loaded colloids was considerably slower than the adsorption process. The findings relevant to our sorption evaluation is the high K_d values for plutonium onto all the solids and the desorption hysteresis.

Linsalata and Cohen (1980) measured the K_d for Pu(IV) spiked into Hudson River estuary water [pH 7.4] onto clay-sized river bottom sediments and silty fine-sand river bottom sediments. The K_d values were determined after 10 days of equilibration and as a function of dissolved salt content, pH, variable dissolved carbonate, sulfate and humate concentrations. However, no plutonium valence state determinations were made on the final solutions. For clay-size river sediments, the measured plutonium K_d ranged from 200,000 to 600,000 mL/g regardless of the values of the dependent variables. For the silty fine-sand, the plutonium K_d values dropped to 60,000 mL/g. Increasing salinity to 24‰ (parts per thousand) or increasing the sulfate and carbonate concentrations to values 4 times larger than the estuary normal concentrations did not cause a lowering in K_d. The dissolved humate and total organic carbon content of the sediments did alter the K_d but Linsalata and Cohen (1980) do not state what the effects were. This study shows that plutonium K_d values, at least when starting species is Pu(IV), are rather insensitive to dissolved salt content but sensitive to available surface area (particle size) of the adsorbent and in general the K_d for plutonium is large.

Skipperud et al. (2000) measured the K_d for several plutonium species [Pu(III,IV), Pu(V,VI), and Pu(II,IV)-organic complex] spiked into seawater onto marine sediment from a fjord using 2 g of solids (dry weight basis) and 15 mL of seawater. The batch tests were run for 0.5 hour up to 180 days. The adsorption reactions were slow and even after 180 days had not reached steady state. The K_d values for the various types of spiked Pu as a function of time are shown in Table 3.1.

The K_d values shown in Table 3.1 increase with time and no pseudo-equilibrium was reached even after six months. Thus, the contact time between contaminated sea water and sediments should be taken into account in dose assessment models. The results indicate that the distribution coefficient, K_d, for plutonium depends on the plutonium species that is present in the experiments. Thus, sediments act as a sink for Pu(III, IV) (high K_d), while Pu(III, IV)-organic and Pu(V, VI) should be considered more mobile (lower K_d). Furthermore, the Pu-solids interaction results obtained from sequential extraction depends less on Pu-species than on time of contact (binding strength increases with time). Desorption K_d values indicate that the observed initial adsorption of plutonium to sediments reflects and is dependent on

Table 3.1. K_d Values (mL/g) for Various Plutonium Species as a Function of Time

Species	1 Week	1 Month	6 Months	Desorption After 6 Months
Pu(V, VI)	60 ± 17	199 ± 20	550 ± 160	1355 ± 15%
Pu(III,IV)	780 ± 160	2015 ± 950	6180 ± 2300	1930 ± 40%(b)
Pu(III,IV)-organic(a)	60 ± 15	135 ± 30	210 ± 60	1410 ± 30%

(a) Organic complex was 0.033 mg/L ethylene diamine tetraacetic acid (EDTA).
(b) Desorption value seems low and questionable.

plutonium speciation in the water soluble phase and that later, stronger sediment-plutonium interactions are rather independent of the original speciation. In terms of the long-term changes, transformation processes must be studied in order to assess rates of transfer between inert and mobile fractions due to contact time.

Emery et al. (1974) and Emery and Garland (1974) studied pond sediments and pond water at U Pond on the Hanford Site in 200 West Area. On average, the pond sediments contained 390 pCi/g total plutonium and the overlying waters contained 0.01 pCi/L total plutonium. If it is assumed that the pond sediments and water represent an equilibrium condition, then the in-situ K_d for plutonium is 3.9 x 10^{+7} mL/g. Emery et al. (1974) also extracted the pond sediments with several chemical reagents in a sequential fashion wherein each extract used became more vigorous in reaction. Over 73 to 93 % of the plutonium was not extractable from the pond sediments even with strong reagents. Emery and Garland (1974) and Emery et al. (1974) suggest that only 9% of the plutonium associated with the sediments would be available to the pond's food web. Emery and Garland (1974) and Emery et al. (1974) also subjected the pond water to filtration through a serial reactive resin bed and found that the dissolved americium was 37% non-ionic (removed by aluminum oxide resin, 60 to 80% cationic and 5% anionic in nature. The source of the plutonium reaching the U Pond was from plutonium purification and finishing processes, and it is possible that some of the plutonium reached the pond sediments as suspended particulates. Thus, the observed in-situ K_d value may be inflated over situations where the source is dissolved contaminants in waste waters. Emery and McShane (1978) describe detailed limnological and ecological studies at several Hanford ponds and ditches however only data on gross alpha and gross beta are presented for concentrations in sediments and waters. Thus no specific element K_d values could be calculated. Emery and McShane (1978) found no evidence that radiation dose at these facilities were limiting the algal, plant or animal communities present even though calculated maximum dose rates reached 1 R per week and maximum gross beta levels reach values of 10^4 pCi/L in the water.

Based on the past tabulations of plutonium K_d value s and the cited literature, it is recommended that the "best" or most probable plutonium K_d value of 600 mL/g be used and that the range be set at 200 to 5000 mL/g. The plutonium K_d exhibits hysteresis during desorption so values higher than 600 mL/g should be weighted more heavily in sensitivity calculations. The new recommended "best" K_d value and range are lower than and similar to the values tabulated in Napier and Snyder (2002), respectively. Napier and Snyder (2002) used a "best" value of 5000 mL/g. Last et al. (2004) and Krupka et al. (2004) recommended "best" values for plutonium K_d at 600 and 150 mL/g, respectively for groundwater scenarios. Both of these tabulations also used a high range value of 2000 mL/g. The range for agricultural soils was widened to 5000 mL/g to reflect desorption hysteresis being quite common. Desorption will be the key process that changes the concentration of contaminants in the non-groundwater scenarios of interest to this report. Thibault et al. (1990) recommended a plutonium K_d value for sands at

550 mL/g and Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1984b, and 1985) recommended a value of 5000 mL/g. Some of the tabulations may include studies where plutonium precipitation was inflating the measured K_d values; therefore, the author recommends a "best" K_d value of 600 mL/g and use of a wide range to cover desorption hysteresis and the likelihood that plutonium precipitation could have been present during the original waste solidification or sediment contamination events.

3.16 Polonium

No studies of polonium adsorption onto Hanford Site sediments are available in the literature and very little general literature exists regarding polonium adsorption. There is some literature on the distribution of ^{210}Po between solution and particulate matter in freshwater lakes, estuaries and the ocean and all of the references suggest that the bulk of polonium is found adsorbed or co-precipitated into the sediments as opposed to being in solution.

For example, Swarzenski et al. (1999) found 50 to 70% of the total ^{210}Po in marine waters in a Norwegian fjord was present in filterable solids. Although the actual concentration of suspended particles in the fjord water was not specified, but the mass was not large; therefore the in-situ K_d must be quite large. Wieland et al. (1991) studied the residence and settling times of particles and particle-reactive nuclides (evaluated from in situ tracer studies which can be used as diagnostic indicators of trace element pathways in lakes). Natural ^{210}Pb and ^{210}Pb fluxes through Lake Zurich (at 50 and 130 m depth) from 1983 to 1987 allowed the calculation of nuclide residence times as well as particle settling and transit velocities in Lake Zurich. The residence time of ^{210}Pb and stable lead in the lake is approximately 1 month. ^{210}Po (daughter of ^{210}Pb) was removed from Lake Zurich with removal times of 10 to 26 months indicating slow removal processes or efficient recycling in the lake water. Balistrieri et al. (1995) also studied the cycling of stable lead, ^{210}Pb, and ^{210}Po in Lake Sammanish, Washington, that exhibits seasonal changes from having the lower water column oxic and anoxic. The key finding that is relevant to our interest is that dissolved polonium was removed from the water column by suspended and bottom sediments less quickly than lead. Thus, in freshwater lakes, the K_d value for ^{210}Po should be less than that for ^{210}Pb.

Vaaramaa et al. (2003) determined the percentage of ^{210}Po that was particulate versus in a dissolved state using pressure filtration of five potable groundwaters taken from wells in Finland. Three of the waters exhibited low ionic strength, neutral pH, calcium bicarbonate dominated fluids common in granite aquifers. Two of the waters were slightly higher ionic strengths and dominated by sodium chloride. The granite waters are quite similar to Hanford Site pore waters. In all five waters, the chemical concentration of polonium was lower than or equal to 2 x 10^{-16} mol/L. The adsorption of polonium species on colloids or larger particles of other elements is the probable cause for the extremely low solution concentrations. For the three granitic waters, the percentage of ^{210}Po that is not bound to particles ranges from 32 to 65%. In the more saline waters <10% of the ^{210}Po is not bound to particles. Vaaramaa et al. (2003) suggest that most of the ^{210}Po in the groundwater is bound in particles having high concentrations of Fe, Mn, and humus. In other studies, they found that iron and aluminum colloids and suspended particles may adsorb polonium. In comparison to radium and uranium, the polonium is most prone to be associated with particles suspended in the groundwater.

Ulrich and Degueldre (1993) determined the influence of the ionic strength and of pH on the adsorption/desorption processes of lead, bismuth, and polonium onto montmorillonite clay. The ionic medium had no influence on the sorption/desorption of both bismuth and polonium. For all of these nuclides, large K_d values were measured. The K_d values range from 10^4 to 10^7 mL/g for ^{210}Bi and ^{210}Po. When adsorption and desorption coefficients are compared, the bismuth and polonium adsorption K_d values were several orders of magnitude lower than those obtained for desorption. The chemical activities of free bismuth and polonium in the liquid phase are limited by the formation of bismuth and polonium-colloids prior to the sorption step. While the adsorption of lead was reversible, only very small amounts of bismuth and polonium could be desorbed from the montmorillonite (quasi-irreversible adsorption). The difference in the K_d values between adsorption and desorption is approximately one order of magnitude in the case of polonium. This suggests that polonium exhibits a high degree of sorption hysteresis.

Finally, Baes et al. (1984) (one of the few tabulations of environmental transport parameters that does discuss polonium; however limited to Oak Ridge National Laboratory conditions) and Thibault et al. (1990) suggest that polonium would adsorb fairly strongly to sediments. Napier and Snyder (2002) use a best value of 1100 mL/g for the polonium K_d value and a range of 196 to 1063 mL/g. Therefore, the "best" or most probable K_d value should be set at 400 mL/g, similar to its parent in the U decay chain, bismuth, and the range be set at 150 to 1100 mL/g based on Baes et al. (1984) and references in Napier and Syder (2002). These recommended K_d values are smaller than those recommended for lead. If doses from polonium ever rise large enough to merit concern some Hanford Site-specific polonium adsorption-desorption testing using appropriate sediments and pore waters would be needed to validate these recommended values.

3.17 Protactinium

No studies of protactinium adsorption onto Hanford sediments are available and very little general literature exists regarding protactinium adsorption. Thibault et al. (1990) relied on soil-to-plant transfer factors to estimate K_d values for protactinium. Because so little is known about the aqueous chemistry and adsorption properties of protactinium, geochemists working in nuclear waste disposal and risk assessment often choose low K_d values because protactinium exhibits a +5 valence state similar to neptunium. For conservatism, neptunium is often used as an analog for protactinium when choosing sorption fate (see for example Krupka et al. 2004). But for the non-groundwater scenarios of interest to this report, conservative K_d values would be larger values than for groundwater scenarios. The recent electronic databases query found the following publications that mention protactinium in subsurface environments.

There is data on the fate of protactinium in marine environments that show very strong adsorption onto fine-grained particles. For instance a recent article, Geibert and Usbeck (2004), measured the K_d value for protactinium adsorbing from several seawaters onto smectite clay, biogenic opal, calcite, and manganese oxides. The protactinium K_d values onto the fine-grained minerals ranged from 1.6 x 10^{+6} mL/g for smectite, 6.1 x 10^{+7} mL/g for manganese oxide, 1.7 x 10^{+5} mL/g for calcite and 5 x 10^{+5} mL/g for biogenic opal. It is difficult to determine how to use the information from the marine studies where it is generally found that daughter products of uranium and thorium, such as protactinium, readily adsorb onto the fine-grained suspended particles and rapidly descend to the ocean floor (for example see the references cited in the Onishi et al. 1981 tabulation). Based on Onishi et al. (1981) observed K_d values for most daughter products of uranium and thorium (and thorium itself) onto fine-grained particles are always large, $>10^{+4}$ mL/g. At the Hanford Site, the particle size distribution is much larger than for marine sediments, so adsorption may be less because of the much smaller available surface area. However, it would appear that the concern that protactinium might form a weak sorbing aqueous species PaO_2^+ similar to the neptunyl species is not supported strongly by the marine experiments. Whatever the aqueous species of protactinium that is present in circum-neutral pH regimes (note seawater has a pH of 8.1), sorption appears to be quite large.

The "best" or most probable protactinium K_d value should be set at 400 mL/g, similar to the values for other daughter products in the uranium and thorium decay chains such as bismuth and polonium (see Table 1.2 or 1.3). Napier and Snyder (2002) list a "best" protactinium K_d value of 3600 mL/g, but do not list a range. The protactinium K_d range should be set at 150 to 10,000 mL/g to account for its seemingly very high association with solids (at least in marine environments) and to allow for wide ranging sensitivity calculations. If doses from protactinium ever rise large enough to merit concern some Hanford Site-specific adsorption-desorption testing using appropriate sediments and pore waters would be needed to validate the recommended "best" value and to better constrain the range. Geibert and Usbeck (2004) present a useful methodology for obtaining protactinium tracer for laboratory adsorption testing based on milking a short-lived tracer, ^{233}Pa from its parent ^{237}Np.

3.18 Radium

No sorption work has been done for radium using Hanford Site specific sediments or waters; however, there is adequate general literature to suggest that radium is fairly strongly adsorbed onto most sediments in contact with low ionic strength solutions with circum-neutral pH values. In solution, radium is found to exist solely as a free divalent cation or complexed to common inorganic anions, especially sulfate. Radium readily co-precipitates with calcium sulfate, gypsum, in brackish environments (Langmuir and Reise 1985). These co-precipitation processes also occur in waste disposal or natural environments that have elevated dissolved sulfate concentrations. Sulfate is the second most abundant dissolved anion in the natural groundwaters underlying the Hanford Reservation (see data in Hartman et al. 2004). These summary statements and more discussion on radium geochemistry can be found in EPA (2004) and Ames and Rai (1978).

Zhang et al. (2001) measured the adsorption of barium (an analog for radium) onto montmorillonite clay and found the bulk of the barium to adsorb onto the fixed charge sites on the basal plane of the clay sheet structure and to be rather insensitive to changes in pH and sensitive to cation competition (ionic strength) similar to expectations from classical ion exchange processes. However, a small fraction of barium also adsorbed on the montmorillonite edge sites, forming a strong inner-sphere surface complex through sharing of oxygen atom(s) from de-protonated –OH groups of the aluminum octahedral layer. This type of adsorption is considered quite strong and fairly irreversible. Perhaps this inner-sphere complex reaction leads to relatively strong adsorption for large-size divalent cations, such as radium.

Baraniak et al. (1999) studied the adsorption of ^{226}Ra spiked into groundwater onto sand, clay, and lime marl sediments from the Saxon Elbe river valley. Batch K_d tests were performed using from 1 to 10 grams of crushed solids into 5 to 50 mL of groundwater for contact times of up to 8 weeks at 14°C. The groundwater was a calcium-bicarbonate/sulfate dominated water with similar concentrations as Hanford Site groundwater and a pH of 7.1. For a crushed sandstone made up predominately of quartz, the radium K_d ranged from 62 to 178 mL/g. In long contact tests (up to 21 weeks), the radium K_d slowly increased but ~95% of the radium that adsorbed did so in the first 24 hours of contact. The very slow reaction that incorporates small additional amounts of radium was hypothesized by Baraniak et al. (1999)

as being radium incorporation/substitution into crystals of barite (barium sulfate a very insoluble compound). In fact, the K_d for radium is found to increase if additional barium (7×10^{-7} to 2.5×10^{-5} M) and sulfate (1.25×10^{-4} to 1×10^{-1} M) is added to the groundwater. Thus, it appears that two sequestering processes, classical surface adsorption and co-precipitation/replacement with barite, are active for radium.

Sun and Torgersen (2001) measured the distribution of ^{224}Ra between sediments and their pore waters at three locations off Long Island, New York. Although this is a marine system and not directly correlatable to Hanford Site, the K_d values for radium ranged from 4 to 85 mL/g with the median value 22 and an average of 24 mL/g. This range of K_d values and the mean suggest that radium sorption in some environments can be relatively low and more similar to strontium than divalent transition metals in waters with high concentrations of competing cations. This report relied on the Baraniak et al. (1999) and Sun and Torgersen (2001) to choose the range for the radium K_d for this compilation.

Aguado et al. (2004) sampled riverbed sediments from an estuary historically affected by waste discharged by several phosphate fertilizer plants, which contained enriched uranium-series radionuclides. A selective extraction technique, which relied on a sequence of more and more aggressive reagents, was used to determine the binding energy for ^{226}Ra to the sediment one year after anthropogenic discharges had ceased. The results obtained revealed that one year after ^{226}Ra inputs had stopped, the radionuclide was associated mostly with the more refractory portions of the sediment. Consequently, it was concluded that there was little potential for remobilization of ^{226}Ra contamination from sediments to the aqueous phase in the future under normal environmental conditions.

Brenner et al. (2004) used gamma spectroscopy to measure ^{226}Ra activities in sediment cores from 20 lakes and a wetland in Florida. Shallow sediments from two lakes (Round and Rowell) possess very high (>20 dpm/g) ^{226}Ra activities that exceed total ^{210}Pb activities, clearly illustrating disequilibrium between ^{226}Ra and supported ^{210}Pb. Supported ^{210}Pb activity is generally thought to come from in situ, ^{226}Ra containing detrital mineral particles, and is typically assumed to be in secular equilibrium with ^{226}Ra activity. Since 1966, Round Lake has been augmented hydrologically with ^{226}Ra-rich (~6.2 dpm/L) groundwater pumped from the local deep aquifer. Adsorption of dissolved ^{226}Ra to recent Round Lake sediments probably accounts for the high measured ^{226}Ra activities in the sediments and the pronounced disequilibrium between ^{226}Ra and supported ^{210}Pb in topmost deposits. They suspect that many Florida water bodies receive some ^{226}Ra-rich runoff and seepage from groundwater pumped for irrigation, residential use, industrial applications, and mining. This may account for increases in ^{226}Ra activity measured in shallow sediment cores from some Florida lakes. Significant groundwater pumping began within the last century, and there has been insufficient time for supported ^{210}Pb to come into equilibrium with adsorbed ^{226}Ra in uppermost deposits. One can, thus, estimate that ^{226}Ra present in wastewater is prone to adsorbing quite favorably to the freshwater sediments.

Sarkar et al. (1999) studied the adsorption of radium onto soils in Florida from highly saline “reject” water [total dissolved solids (TDS) = 6500 mg/L] from an electrodialysis reversal water treatment plant. Flow-through column results indicated that ^{226}Ra accumulated throughout the soil profile, and that only small amounts of ^{226}Ra escaped out of the column. Equilibrium geochemical modeling, using the computer code MINTEQA2, suggested that radium adsorption on sand was the primary mechanism responsible for ^{226}Ra retention in the soil column. In the absence of competing Ca, 100% of the ^{226}Ra was predicted to adsorb on sandy soil at neutral pH. The presence of Ca decreased overall retention of ^{226}Ra, due to competition for similar surface sites. The ^{226}Ra concentration in the system was too low to induce direct precipitation of radium salts. Radium adsorption on sand was influenced by pH of the soil solution, with more radium adsorbed at higher pH.

Benes et al. (1997) collected 134 suspended and bed sediment samples from rivers and reservoirs in Czechoslovakia. They then determined the radioactive equilibrium between parents (^{238}U and ^{232}Th) and their daughters (^{226}Ra and ^{228}Ra), respectively in the solids. Benes et al (1997) found that the two radium isotopes were either in equilibrium with their parents or in the case of ^{228}Ra was often present in excess, which suggests that radium is readily removed from fresh water solution by adsorption reactions.

Willett and Bond (1995) studied the adsorption of ^{226}Ra onto soils from the Alligator Rivers Region of the Northern Territory in Australia. All sorption studies were conducted with a 1/5 ratio of soil (oven-dry equivalent) to background electrolyte of 0.0025 M $MgSO_4$. This solution was used because it was similar in composition to the retention pond water at the Ranger uranium mine. The reaction period for the batch sorption tests was 24 hours. For a sand soil with low cation exchange capacity (1 to 2.5 meq/100 g), organic carbon content of 0.1 to 0.7 % and a natural pH of 5.6 to 5.7, the ^{226}Ra K_d values ranged from 3000 to 5000 mL/g. Once adsorbed only <1% of the sorbed radium was displaced on re-suspension of the samples in the background electrolyte.

Kadko et el. (1987) measured the ^{226}Ra concentration in both the marine sediments and its pore water in eastern equatorial Pacific Ocean waters just north of the Galapagos Islands off the western coast of Central America. The ^{226}Ra activity in pore waters ranged from 0.5 to 7 dpm/kg of seawater and the ^{226}Ra content in the sediments ranged from 10 to 70 dpm/g. These two measurements can be combined to get an estimate of the ^{226}Ra K_d by assuming that a kilogram of seawater has a density of 1.03 g/mL. The calculated radium K_d range is $1.4 \times 10^{+3}$ to $1.4 \times 10^{+5}$ mL/g, but these values are likely too high for Hanford Site use because some of the ^{226}Ra in the sediments is bound in crystalline mineral lattices and not truly part of the exchangeable portion of the sediment. Thus, any radium present in Hanford wastes is not likely to become incorporated into crystalline lattice sites in surface soils or Columbia River bank sediments in the time periods of interest to performance assessments.

Napier and Snyder (2002) recommended a "best" radium K_d value of 500 mL/g and a range of 214 to 467 mL/g. Thibault et al. (1990) recommends a K_d value of 500 mL/g for sand. For groundwater scenarios Krupka et al. (2004) recommends a value of 14 mL/g and a range of 5 to 200 mL/g. Onishi et al. (1981) is the only other reference listed in Table 1.3 that mentions radium and they provide a range from 200 to 500 mL/g. From the mentioned tabulations and all the generic radium studies just described, a radium K_d value of 200 mL/g was chosen for the "best" or most probable value and a range of 5 to 500 mL/g for sensitivity analyses for the agricultural and river bank soil scenarios described earlier. A lower "best" K_d value was selected than recommended by Napier and Snyder (2002) and Thibault et al. (1990) to honor the lower K_d values found by Baraniak et al. (1999) and Sun and Torgersen (2001) and to stay consistent with classical ion exchange as being a dominant sequestration mechanism for radium. Because the future Hanford agricultural soils and river bank sediments will remain rather coarse in particle size, the K_d might be lower than many contaminants that adsorb more so by variable charged hydrous oxides, or co-precipitate/adsorb with high pH sensitivity such as transition metals, lanthanides, and most lower valence state actinides. Selecting the wide range (5 to 500 mL/g) would allow sensitivity studies to determine whether radium is in fact a significant dose contributor for the agricultural soil and river bank sediment substrates and non-groundwater scenarios of interest. If sensitivity studies show radium may be a significant dose contributor, then laboratory studies using appropriate radium-spiked soils and sediments should be performed to validate radium K_d values.

3.19 Selenium

Adequate Hanford specific adsorption literature exists for selenium because of recent work performed for the low-activity glass waste project, (i.e., renamed IDF) in which selenate species were used (Um and Serne 2005). Krupka et al. (2004) and Cantrell et al. (2003) review the Hanford specific literature, including the recent studies published in Um and Serne (2005). In natural waters of interest to the Hanford Site selenium is found in two forms and valence states, selenate (SeO_4^{2-}) [Se(VI)] and selenite (SeO_3^{2-}) and/or ($HSeO_3^{2-}$) [Se(IV)]. Interestingly, elemental selenium can be a solubility control across most pH values for moderately reducing redox conditions (Krupka et al. 2004, Figure 3.8). Mixed iron hydroxide-selenium oxides may also be controlling the concentrations of dissolved selenium in natural pore waters if high concentrations are present. A recent review of the aqueous speciation thermodynamics reports that the stability constants for several aqueous species in older literature are quite suspect and new recommendations are offered (Seby et al. 2001). However, adsorption reactions likely are the main controller of selenium in the scenarios of interest to this report based on the likely very low concentrations of selenium and the key radionuclide of interest, ^{79}Se. More detailed discussions on selenium geochemistry, including aqueous speciation diagrams as a function of pH and Eh, and adsorption properties are found in Krupka et al. (2004) and Ames and Rai (1978). Although not completely determined, the selenate form of selenium is the most likely form present in the Hanford near-surface soils and sediments based on our geochemical experience.

Dhillon and Dhillon (1999) measured the K_d for eight surface soils from India that are much finer-grained than Hanford sands. The form of selenium used was selenite traced with ^{75}Se radioisotope that was not specified as to species. The range of K_d values was 1 to 241 mL/g with the higher values for pH conditions in the acidic range 4 to 6. The soil that resembled Hanford sands most closely yielded a K_d value that varied from 1 to 8.3 mL/g.

Singh et al. (1981) determined the Langmuir adsorption constants for five Indian surface soils using both sodium selenite and sodium selenate spiked into a very dilute sodium chloride solution. None of the soils are as coarse grained or exhibit as low of cation exchange capacity as the Hanford soils. For the Indian soil that has characteristics closest to the Hanford Site sands, the K_d for selenite varied from 5 to 20 mL/g as the selenite equilibrium concentration drops from 7.4 to 0.2 mmol/L. For this same soil the K_d for selenate varied from 7 to 33 mL/g as the selenate equilibrium concentration drops from 9 to 0.13 mmol/L. Contrary to studies discussed in the following paragraphs, selenate adsorbed more than selenite in Singh et al. (1981) studies. No explanation for this apparent discrepancy is available.

Dong et al. (1999) studied the adsorption of selenite spiked into 0.01 M calcium chloride background electrolyte onto an iron oxide reach acidic soil from China. The soil contained 12% by weight iron oxides, had 38% clay content, 0.78% organic matter, a slurry pH of 4.78 and a cation exchange capacity of 10.1 meq/100 g. Batch adsorption tests were carried out in the background electrolyte at pH 6.3 to 6.8 for about 60 hours at solution to solid ratios of 150 to 500 mL/g. The average selenite K_d for the soil was 477 mL/g, but if the soil was treated with citrate dithionite to remove the iron oxides the K_d dropped to between 11 and 16 mL/g. There was also a slight bit of hysteresis in that the desorption K_d values were 10 to 30% larger that the adsorption K_d values. The selenite K_d values for the iron oxide removed soil are similar to values found for Hanford coarse sands that were contacted with trace concentrations of selenate (see review discussion in Cantrell et al. 2003) suggesting that there may not be significant difference between adsorption of selenite and selenate once most of the iron oxides are removed.

Del Debbio (1991) measured the adsorption of selenite (SeO_3^{2-}) onto alluvium, interbed sediment and crushed basalt rock from the Idaho National Laboratory, Idaho Falls, Idaho. The alluvium and interbed sediment are somewhat similar to the near surface soils at the Hanford Site in terms of calcium carbonate content and mineralogy, but they are more fine grained and would be designated as sandy loam and silt as opposed to Hanford Site materials being categorized as sand or gravelly sands. The Idaho solids were contacted with a groundwater that is very similar in chemical composition to Hanford Site groundwaters, at a ratio of 1 g solid to 20 mL groundwater. The batch contact time was six days. The adsorption of selenite was found to vary significantly with the concentration of selenite present in the water. As the initial concentration of selenite varied from $\sim 10^{-7}$ to 6×10^{-5} M, the K_d for selenite dropped from 17 to 4 and 63 to 6 mL/g for the interbed sediment and alluvium, respectively. The K_d values for the low end of the selenite concentrations studied are close to the value recommended in this report, 15 mL/g, especially if the Idaho values decrease because they are for finer grained particles than the Hanford solids of interest. Again, this compilation relies on the apparent similarity between K_d values for selenite and selenate when hydrous oxide contents of the solids are low to make this comparison.

Soils in the alluvial fan in the western San Joaquin Valley, California, have been extensively studied to understand the geochemistry of selenite and selenate species of selenium. Dissolution and leaching of soil salts by irrigation water is a primary source of selenium to shallow groundwater in the western San Joaquin Valley. The following references present a very detailed geochemical understanding of selenium that is transferable to most environments: Fio and Fujii (1990), Fio et al. (1991), Neal et al. (1987a, 1987b), Neal and Sposito (1989), Sposito et al. (1988), Sposito et al. (1991), and Wright (1999). Within these references sorption studies showed that selenate is not adsorbed to the alluvial fan soils, whereas selenite is rapidly adsorbed. The data show that >50% of the soluble Se(IV) added to the soils in a rate study was adsorbed after 8 hours, and maximum adsorption was essentially reached after about 24 hours. The similar adsorption rates and maximum partitioning under sterile and un-sterile conditions indicate that biological activity probably did not alter the concentration of Se(IV). Selenite adsorption by the alluvial fan soils decreased uniformly with increasing pH in the range 4 to 9 and was independent of soil series above pH 6. Considerable hysteresis was found between selenite adsorption and desorption. Desorption was measured by replacing 100 mL of the solution in the centrifuge bottles after the adsorption step with 100 mL of 0.005 M $CaSO_4$ solution. The centrifuge bottles were returned to the shaker and the concentration of Se was measured after 0.5, 1, 4, 8, and 24 hours. About 88% of the Se(IV) remained adsorbed at the end of the desorption experiment. No discernible change in the amount of selenite adsorbed was found as a result of increased chloride concentration or through the addition of 16 mmol Na_2SO_4. In contrast, an initial concentration of phosphate comparable to that of selenite resulted in a decrease of selenite adsorbed by approximately one-half. Selenite is resistant to leaching and therefore can represent a potential long-term source of Se to groundwater.

In contrast, selenate behaved as a conservative constituent under alkaline and oxidized conditions and was easily leached from the alluvial fan soils. Selenate adsorption in contrast to the results obtained for selenite, was not detected over the pH range of 5.5 to 9 in either the NaCl or $NaClO_4$ background electrolyte. The authors concluded from their results that Se(VI) behaves similarly to SO_4, and forms weakly bonded, outer-sphere surface complexes. In general, outer sphere complexes are weaker in nature than inner sphere complexes and thus more amenable to reversible adsorption-desorption.

Theoretical calculations, presented in the suite of San Joaquin references, for the oxidation of selenium by NO_3^- and oxygen show favorable differences in Gibbs free energies for the oxidation, indicating that nitrate can act as an electron acceptor for the oxidation of selenium. Management of nitrogen fertilizer applications might help to control the oxidation and mobilization of Se and other trace constituents into the environment. The results for the San Joaquin Valley soils indicated that, at native levels of NO_3, effective microbial catalysis of SeO_4^{2-} reduction, occurred in the soil under the conditions of the experiments in agreement with isolation of bacterial species that can respire SeO_4^{2-} while oxidizing organic acids typical of suboxic soil environments.

Goldberg and Glaubig (1988) studied selenite and selenate sorption on a calcareous, montmorillonitic soil as a function of solution pH (2-11) at two initial total selenium concentrations. Selenate sorption was not observed at any pH value for either initial selenium concentration. Selenite sorption exhibited a maximum near pH 3, a sharp decline to pH 6, and a sorption plateau above pH 7. Selenite sorption as a function of pH was studied on reference minerals representative of the dominant mineral constituents of the studied soil, which included montmorillonite, kaolinite, and calcite. Selenite sorption on the clay minerals increased at low pH, exhibited a peak near pH 5, and decreased at higher pH. Selenite sorption on calcite increased from pH 6 to 8, peaked between pH 8 and 9, and decreased above pH 9. The soil sorption plateau above pH 7 virtually disappeared after removal of calcite, indicating that calcite plays an important role in selenite sorption onto calcareous soils. The lack of selenate adsorption onto the soil studied even at pH values as low as 2 is likely caused by a lack of any significant amounts of variable charged hydrous oxides in the soil. Many soils with measurable quantities of hydrous oxides do show measurable anion adsorption at acidic pH values.

Chao and Sanzolone (1989) concluded that in soils developed through intensive leaching and weathering, selenium tends to be associated with oxide minerals and a great proportion of the selenium is resistant to chemical dissolution. For soils with high pH and low content of oxide minerals, selenium is present as mobile selenate and can be easily extracted.

Despite the fact that aqueous selenium in low ionic strength circum-neutral waters are anionic, Hanford specific adsorption on coarse sand sediments (see Um and Serne 2005 and data compiled in Cantrell et al. 2003) studies show moderate adsorption is occurring. Unlike pertechnetate, nitrate, and iodine aqueous species, selenium sorbs with K_d values greater than 5 mL/g. For the “non-water” scenarios of interest to this report, a K_d value of 15 mL/g was chosen to represent the “best” or most probable value and a range of 3 to 30 mL/g was chosen for selenium adsorption onto future Hanford agricultural soils and river bank sediments. The high end of the range has not been observed in Hanford specific studies but will allow for some conservatism in the sensitivity calculations to ascertain whether selenium is a significant risk contributor. The “best” K_d value and range differ (are higher and wider, respectively) from those tabulated in Napier and Snyder (2002), which did not have access to the most current IDF project selenate adsorption studies. The selenium adsorption value estimates (150 to 500 mL/g dependent on particle size) in Thibault et al. (1990) are not measurement and are based on soil-to-plant transfer factors. The Thibault et al. (1990) values are quite large and do not seem reasonable, certainly not for Hanford specific conditions. Coughtrey and Thorne (1983 a, 1983b) and Coughtrey et al. (1983, 1984a,b, and 1985) recommend a selenium K_d value of >9 mL/g. Last et al. (2004) and Krupka et al. (2004) recommend most probable K_d values of 5 and 7 mL/g and ranges of 3 to 10 and 3 to 15 mL/g, respectively for groundwater scenarios. This study recommends a slightly higher “best” K_d value than these two groundwater scenario based tabulations to account for some potential desorption hysteresis and potential for lower pH conditions in organic rich soils, which would favor more anion adsorption. The K_d range was expanded to 30 mL/g to allow some conservatism to be introduced into the predictions. If sensitivity studies show selenium may be a significant risk contributor, then laboratory studies using appropriate selenium-spiked soils and sediments should be performed to validate the K_d values.

3.20 Strontium

There is a large Hanford-specific database for adsorption of strontium, which has been tabulated and discussed in Cantrell et al. (2003), Krupka et al. (2004) and Ames and Rai (1978). General literature on strontium adsorption is also very large. All available literature suggest that strontium adsorbs moderately to sediments and follows quite simple ion exchange relationships for most circumstances. At Hanford in some selected environments it is suggested that radio-strontium may also exchange for stable strontium and calcium in carbonate minerals in a similar fashion as ^{14}C exchanges for ^{12}C species in the carbonate solids. The sources of ^{90}Sr in the scenarios of interest for this report are likely solidified waste forms and residual contaminated near-surface sediments. Both sources of ^{90}Sr contamination might engage in these isotope exchange reactions with carbonate minerals present in the near-surface soils and with carbonate that forms on cementitious waste inadvertently brought to the surface.

Only one new study was found in the literature review for near-surface or agricultural soils. Shimada et al. (1996) determined K_d values, transfer coefficient by direct foliar absorption (K) and the transfer coefficients for root uptake for ^{90}Sr and ^{137}Cs for several Japanese soils. The strontium K_d values were found to be 10 to ~300 mL/g for ^{90}Sr. Another study with data for deep sediments similar to Hanford's is also discussed. Del Debbio (1991) measured the adsorption of strontium [Sr^{2+}] onto alluvium, interbed sediment and crushed basalt rock from the Idaho National Laboratory, Idaho Falls, Idaho. The alluvium and interbed sediment are somewhat similar to the near surface soils at the Hanford Site in terms of calcium carbonate content and mineralogy but they are more fine grained and would be designated as sandy loam and silt as opposed to Hanford Site materials being categorized as sand or gravelly sands. The Idaho solids were contacted with a groundwater, which is very similar in chemical composition to Hanford-site groundwaters, at a ratio of 1 g solid to 20 mL groundwater. The batch contact time was six days. The adsorption of strontium was not found to vary significantly with the concentration of strontium present in the water. As the initial concentration of strontium varied from $\sim10^{-6}$ to 5×10^{-4} M, the K_d for strontium ranged from 190 to 110 and 52 to 35 mL/g for the interbed sediment and alluvium, respectively. The K_d values for the high end of the strontium concentrations studied by Del Debbio (1991) are close to the value recommended in this report, 50 mL/g.

Based primarily on the large Hanford strontium K_d database, the "best" or most probable K_d value was chosen to be 50 mL/g, which is larger than values chosen by Last et al. (2004, 22 mL/g) and Krupka et al. (2004, 14 mL/g) and which represent surface adsorption onto sediments for waterborne scenarios. To keep the ^{90}Sr available longer for the non-groundwater scenarios of interest to this report and to account for the hysteresis in K_d caused by the isotope exchange into carbonate minerals, the higher value of 50 mL/g is recommended. For a K_d range to use in sensitivity calculations, 5 to 200 mL/g was chosen, which is essentially the same as the ranges recommended by Napier and Snyder (2002) and Krupka et al. (2004). There is adequate strontium adsorption data such that Last et al. (2004) recommend using a log normal probability distribution on the range of K_d values around the mean value. However, while this choice might be acceptable for strontium and cesium adsorption data, the adsorption data population for other contaminants is not large enough to defend choosing a probability distribution function.

In comparison to the other data tabulations for Sr K_d values shown in Table 1.3, the most probable or "best" value chosen in this report is larger than the value recommended by Thibault et al. (1990), which offers 15 mL/g. Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1984b, and 1985) did not tabulate strontium K_d data.

3.21 Technetium

There is a large Hanford-specific database for adsorption of technetium in its oxidized form, pertechnetate [TcO_4^-, Tc(VII)]. General literature on pertechnetate adsorption is also very large. Both types of adsorption data suggest that pertechnetate does not adsorb to most sediments and soils and adsorbs weakly to some. Hanford-specific data is reviewed in Cantrell et al. (2003) and Krupka et al. (2004). Technetium is redox sensitive and can be reduced to a very insoluble form by strong reductants such as sulfide often present in natural reducing environments. Ferrous iron can also reduce pertechnetate but not as readily as it reduces chromate. Reducing conditions probably will not be found in the near-surface scenarios of interest to this report. However, to allow technetium to be available longer for the scenarios of interest, the possibility of some technetium reduction was considered, perhaps by decaying organic matter in the agricultural soil and river bank sediments. General technetium geochemistry and adsorption tendencies are summarized in much more detail in Krupka and Serne (2002), Krupka et al. (2004) and EPA (2004).

Based on the Hanford-specific technetium adsorption database and plausible geochemical differences caused by addition of more organic matter to the soils and sediments of interest and allowing for the source of much of the technetium to be in waste tailings inadvertently brought to the ground surface, a "best" or most probable K_d value of 0.5 mL/g and a range of 0 to 1 mL/g for sensitivity calculations is recommended. Napier and Snyder (2002) selected a most probable technetium K_d value of 2.0 mL/g and a range of -3.4 to 0.57 mL/g, which does not include their recommended value.

The recommended "best" K_d value and range are the same as the values chosen for chloride (^{36}Cl), and nitrate. The recommended "best" K_d value is larger than the value of 0 mL/g recommended for groundwater scenarios found in Last et al. (2004) and Krupka et al. (2004) because of the need to account for the possibility of some technetium reduction and resistance to desorption out of disaggregated solid wastes. Thibault et al. (1990), Coughtrey and Thorne (1983a, 1983b) and Coughtrey et al. (1983, 1984a, 1983b, and 1985) recommended technetium K_d values of 0.1 and 0.11 mL/g, respectively. The recommended K_d range, 0 to 1 mL/g, is also somewhat larger than used in the groundwater scenario recommendations for the same reasons as a higher most probable K_d value was chosen.

3.22 Tritium

Tritium is often used as a tracer for water molecules in column breakthrough testing and is assumed to define the zero K_d condition. Alternatively, one could assume that the anions often used as water tracers define the zero K_d condition and ignore the concept of anion exclusion. The column experiments performed by Gee and Campbell (1980) used both tritium and chloride as tracers and they found chloride broke through the columns slightly earlier than tritium. This observation was used to quantify the anion exclusion process by assuming that tritium was the true conservative (non-interacting tracer; $K_d = 0$). It is conceivable that tritium as the water molecule (THO) or hydroxyl species (OT^-) where $T= {}^3H$ can enter into isotope exchange reactions with water molecules adsorbed to solids (as waters of hydration) or with hydroxyl groups on the surfaces of hydrous oxides present on the surfaces of solids. If significant, such isotope exchange reactions can keep tritium available in the near surface agricultural and river bank sediments of interest herein.

A "best" K_d value of 0.2 mL/g was chosen arbitrarily, the lowest of all values chosen in this report, to describe tritium's isotope exchange potential. Also, the K_d range to use for sensitivity analyses was chosen as 0 to 1 mL/g, consistent with the other very mobile constituents (chloride-36, nitrate, and technetium) in this report. With a half-life of 12.34 years, tritium should not be a significant risk factor for very long once all activities cease at Hanford. Napier and Snyder (2002) selected a most probable tritium K_d value of 0.7 mL/g with a range of 0 to 0.7 mL/g. The only other K_d tabulation that discusses tritium was Thibault et al. (1990); for a sand they recommend a tritium K_d value of 0.06 mL/g and 20 mL/g for loam.

3.23 Uranium

There is adequate Hanford Site specific data on the adsorption of uranium onto Hanford sediments from circum-neutral pH and low ionic strength waters to develop technically defensible K_d values to use for the scenarios of interest where the original source of uranium was some liquid waste stream contacting Hanford sediments (Cantrell et al. 2003; Krupka and Serne 2002; and Krupka et al. 2004). There is also a large quantity of general literature that complements and corroborates the Hanford-specific data (EPA 1999b). In the near-surface sediments of interest for the agricultural and river bank sediments and scenarios being considered, uranium geochemistry should be dominated by oxidized uranium [U(VI)]. The key aqueous species are carbonate complexes of the uranyl [UO_2^{2+}] cation such as $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$. The uranyl complexes are anionic or at pH values below 6 neutrally charged (Krupka et al. 2004, Figure 3.12). Although fairly soluble, uranyl minerals do exist and are stable in Hanford near-surface environments as evidenced by ongoing studies in the 300 Area at Hanford (Serne et al. 2002a; Zachara et al. 2005). Plausible controlling solids for uranyl forms of uranium include alkali and alkaline-earth uranyl silicates such as boltwoodite, uranyl silicate (uranophane), alkali and alkaline earth oxy hydroxides such as clarkeite, and mixed uranyl carbonates. When uranium is present at very low concentrations (low ppm to sub ppm) adsorption may be the only retardation mechanism that occurs between sediments and natural pore waters. When this is the case, uranium is moderately mobile in Hanford sediments because the uranyl carbonate complexes do not adsorb appreciably at pH conditions above 7.

If the source of the uranium in the future near-surface environments of interest comes from cementitious or glass waste forms or from contaminated sediments in which discrete uranium bearing precipitates are present that are inadvertently brought to the surface, then uranium fate should be treated differently from all the other contaminants discussed in this report. Uranium geochemistry and uranium leaching and/or release from environments with a combination of low dissolved carbonate and high pH are highly dependent on the interrelated dissolved carbonate and pH. Upon water leaching, fresh or young cement and vitrified borosilicate glasses both generate solutions with higher than ambient pH values. Cement leachate also increases dissolved calcium concentrations in the nearby pore waters and glass leachate contains high sodium concentrations that further react with Hanford sediments to exchange sodium for calcium on sediment surface exchange sites. The net pore water/sediment reactions in the near vicinity of leaching cement or glass promote the precipitation of calcium carbonate such that the residual pore water exhibits low dissolved carbonate, low dissolved calcium, and higher than ambient pH.

This geochemical state promotes sequestration of uranium within the sediments surrounding the impacted pore water. The effects are quite dramatic such that even small amounts of solidified waste (cement or glass) can influence uranium fate. Thus, even though this K_d value tabulation was assuming that there would be insignificant mass of solidified waste brought to the surface to impact contaminant K_d values, uranium is the one exception. In addition, based on the common occurrence of co-precipitated or discrete uranyl solid phases in near-surface Hanford sediments in the 300 Area (and likely other disposal sites), that are not influenced by cement or glass, uranium deserves special treatment. Finally, with the knowledge that simplified risk assessments for the "non-groundwater" scenarios of interest use only K_d values to quantify all retardation (as opposed to separating solubility and adsorption processes into two distinct retardation processes), the "best" or most probable and range of K_d values to use for the non-groundwater scenarios of interest for uranium in this compilation are split into two categories.

For situations where the uranium source was discrete uranium-bearing soils, cement, or glass waste, a desorption K_d greater than 10 mL/g is justified. Cement and waste glass secondary minerals are very good at sequestering uranium. High uranium sequestration is observed until all the free calcium hydroxide in hydrated cement is converted to calcite (calcium carbonate see further discussion in Serne et al. 1996) and until the pH of fluids surrounding the weathering glass drops below 9, because the geochemical conditions during the early phases of leaching favor calcite precipitation and low aqueous concentrations of carbonate. Recall that uranium complexes with dissolved carbonate are the key reason that uranium remains mobile in pore fluids interacting with sediments. If the dissolved carbonate is kept low because of calcite precipitation, there is less tendency for the anionic uranyl carbonate aqueous complexes to dominate. Fresh or young cement has a large source of soluble calcium that can reduce the carbonate concentrations in pore fluids to such low values that the strong and very soluble uranyl-carbonate complexes dissociate and allow the free uranyl cation to adsorb/co-precipitate. The same phenomenon occurs when glass weathering releases high concentrations of sodium, which exchanges with calcium on the surrounding soil cation-exchange sites resulting in high dissolved calcium that forces calcite to precipitate. The glass weathering reactions also increase the pH of the near by pore fluids, which also promotes calcite precipitation and the lowering of dissolved carbonate concentrations.

However, for both cement and glass waste forms after long time periods, the excess calcium (in the cement waste) and sodium (in the glass) is depleted and there will be a net influx, via partially air-filled pores, of carbon dioxide to the vadose zone. The carbon dioxide causes pore fluid pH values to drop to values near 8.3 and replenishes the dissolved carbonate (bicarbonate is the dominant species at this pH) in the pore fluids. The increasing dissolved carbonate can then start to complex and enhance the mobility of previously sequestered uranium. The same time-dependent reactions can re-mobilize uranium that was originally precipitated as discrete minerals in contaminated sediments or adsorbed to sediment surfaces under conditions when there was less carbonate present in solution. Thus, at long time periods uranium will become more mobile and likely percolate deeper into the sediment profile and become a potential threat to the groundwater pathways. Although only a crude knowledge of the time-dependent dynamics for the weathering of cement and glass waste forms exists, the reactions may be completed in a matter of a few hundred years for small masses of solid waste forms brought to the surface and mixed with near-surface sediments. Depending on the assumed mass of uranium present in a cementitious or glass waste form that is intercepted by drilling activities and inadvertently brought to the ground surface, it would be more technically defensible to run two time periods wherein the first few hundred years use uranium desorption K_d values between 10 and 50 mL/g; then for later time periods, the desorption K_d should be reduced to values that range from 0 to 10 mL/g for the non-water surface exposure of drilling tailings from drilling through solidified waste scenario and for contaminated sediments that contain discrete uranium mineral precipitates.

Tables 1.2 and 1.3 list two recommended "best" K_d values and ranges for uranium. For the short-time period (arbitrarily set at 200 years from the time when the wastes are brought to ground surface), a "best" uranium K_d value of 30 mL/g and a range of 5 to 50 mL/g is recommended. For the longer time period (arbitrarily set at >200 years from the time the uranium-bearing wastes were brought to the ground surface), a "best" uranium K_d value of 5 mL/g and a range of 0 to 20 mL/g is recommended.

These recommendations differ from those used by Napier and Synder (2002), where the "best" or most probable K_d value for uranium was set at 7 mL/g and the range was 0.08 to 3.5 mL/g, even though it does not even include the most probable value. The two-category recommended uranium K_d values and ranges have higher most probable values than the two groundwater-scenario based tabulations of Last et al. (2004, 0.8 mL/g) and Krupka et al. 2004, 1 mL/g). The recommended uranium K_d ranges are much wider than the range chosen by Last et al. (2004, 0.2 to 4 mL/g) but narrower than the range chosen by Krupka et al. (2004, 0.1 to 80 mL/g). Krupka et al. (2004) did consider some of the waste form reactions that lower dissolved carbonate concentrations in choosing their range. The only other K_d value tabulation that listed values for uranium was Thibault et al. (1990). They recommended a uranium K_d value of 35 mL/g for sands and 15 mL/g for loams.

3.24 Additional Information

A few other relevant observations were found during the review of recent literature. A study of the vertical transport of ^{60}Co, ^{137}Cs, and ^{226}Ra in agricultural soils placed in a 1 m^2 cross section lysimeter facility with crop rotation and plowing to depths of 20 cm each year showed that after nine years there was very little vertical migration of these three contaminants below the 20-cm depth of plowing (Shinonaga et al. 2005). Shinonaga et al. (2005) concluded that using large K_d values for these three contaminants such as those tabulated by Sheppard and Thibault (1990) are justified. The Sheppard and Thibault (1990) K_d values are the same as those found in Thibault et al. (1990) and listed in Table 1.3. The recommended K_d values in this report for these three radionuclides are also large and not too different from Sheppard and Thibault (1990).

Echevarria et al. (2005) measured the adsorption of niobium, a fission product for which very little data has been reported in the literature. Some risk assessments have predicted that niobium may be a significant risk driver because a small K_d value is often chosen in order to accommodate the known Nb(V) valence state. The assumption is often made that Nb(V) forms NbO_2^+ ions similar to neptunyl (NpO_2^+) that under some conditions does show low sorption tendencies. Echevarria et al. (2005) measured niobium adsorption onto three sediments from France. For the sediment with characteristics most similar to Hanford sands, they found a niobium K_d value of 1980 mL/g. The other two sediments, both dominated by silt and clay, yielded niobium K_d values ~3000 mL/g. The niobium adsorption was slow with solution concentrations still decreasing after three days of contact such that the authors suggested it might take up to 30 days to reach a true equilibrium. They also suggest that the reaction may be slow hydrolysis and precipitation of niobium hydroxides. It thus appears that niobium may not be as mobile as some risk assessments have assumed based on potential similarities to neptunium. This information is provided in the hope that future risk predictions for "exotic" contaminants, for which little experimental data are available, at least have this one source of measured data.

For readers interested in plant and animal transfer factors from soils there is a recent report that compiles such factors, Robertson et al. (2003).

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Appendix A

Past Letter Reports on Agricultural Soil K_d Values

Selected K_d Values for Agricultural Soils

Daniel I. Kaplan
October 3, 1997
Updated by R. Jeff Serne
May 17, 1999

K_d Value Selection Process: A systematic process was used to select K_d values for the radionuclide transport simulation (Table A.1). In each case the largest value within the range of likely values was selected. Whenever possible, selected K_d values were based on data generated from experiments conducted with Hanford sediments and aqueous phases of low ionic strength, neutral to basic pH levels, and oxidizing conditions. The selected K_d values of C, Cl, I, Np, Pb, Ra, Se, Sr, Tc, tritium, and U were based on Hanford Site data (see Appendix B in this report). Of these selected K_d values, Cl, I, Ru, and Tc K_d values were increased to account for the likely increase in adsorption to the organic matter that is likely to be present in the solid phase. The K_d values for which there were no Hanford Site data available were based on non-Hanford Site reported data for agricultural soils, silt textured soils or freshwater sediments (Bi, Fe, Mn, K, Pa, Po, Re, Rn, and Th).

Table A.1. Selected K_d Values (mL/g) for Agricultural Soils

Rad.	Selected Conservative K_d Values for Agricultural Soils(a) (mL/g)	Reported K_d Range (mL/g)	Number of Reported K_d's	Rational	Ref(b)
Ac	1500	--	0	No Hanford soil data available. Estimate based on Am-K_d data. Am is close to Ac on the periodic chart and both have +3 oxidation states. As reviewed by Kaplan et al., reported Am-K_d's in Hanford sediments range from 67 to >1200.	1
Am	1500	--	2	As reviewed by Kaplan et al. reported Am-K_d's in Hanford sediments range from 67 to >1200.	1
Bi	900	--	0	No Hanford soil data available. For silt soils, Thibault et al. (1990) estimated a geometric mean K_d of 450.	2
C	7.0[c]	0.03 to 4.56	6	Hanford sediment data. ^{14}C exists as anionic $H^{14}CO_3^-$ or $^{14}CO_3^{2-}$ species. Some loss of aqueous ^{14}C-carbonate will occur due to partitioning to gas phase (CO_2[g]) and isotopic exchange with calcite, which controls ^{12}C concentrations through solubility in Hanford sediments.	3
Ce	1500	3 to >2000 but always >1000 for all but acid conditions	>5	Hanford sediment data. Ce exists as hydrolyzed species that are very sensitive to pH of the porewater. Sorption is very high in neutral pH solutions (K_d > 1500). Benson (1960) found Ce sorbed to Hanford soils would exchange with other cations when pH was greater than 7.4.	16
Cl	1	-0.008 to -0.013	2	Hanford sediment data. Soluble anion. Selected K_d value increased to account for the likely presence of organic matter in the solid phase.	4
Cm	1500	--	0	No Hanford soil data available. Estimate based on Am-K_d data. Cm chemical behavior is very close to that of Am, both have +3 oxidation states and are close to each other on the periodic chart. As reviewed by Kaplan et al. reported Am-K_d's in Hanford sediments range from 67 to >1200.	1
Cs	2000	>200 to 10,000	>20	Hanford sediment/groundwater system, 540 to 3180 mL/g (5 and 8). K_d consistently has median value ~2000 for Hanford sediments and low-ionic strength neutral pH solutions. Mica/illite minerals "fix" Cs and make it difficult to exchange off sediment.	5, 8, 22
Fe	3500	--	0	No K_d tests per se have been made on Hanford sediments. But Fe present in reactor cooling water was found to be bound (98%) to small particulates (<0.3 µm). The iron was not cation exchangeable using high NaCl solution [Roberstson and Perkins (1975), Roberstson et al. 1973]. Iron in Hanford sediments is likely precipitated and bound strongly. Under oxidizing conditions Fe is present as ferric, which quite insoluble at neutral pH. K_d's for marine sediments are >10,000 ml/g. (Onishi et al. 1981).	23

Table A.1. (contd)

Rad.	Selected Conservative K_d Values for Agricultural Soils[(a)] (mL/g)	Reported K_d Range (mL/g)	Number of Reported K_d's	Rational	Ref[(b)]
I	15	0.05 to 15	21	Hanford sediment data. Soluble anion that has been shown to be sorbed to Hanford sediments, perhaps to Fe-oxides or carbonates in these soils. Selected K_d value increased to account for the likely presence of organic matter in the solid phase.	4, 5, 6
K	10	2 to 9	10	No Hanford soil data available. For agricultural soils, Baes and Sharp estimated a geometric mean K_d of 5.5 based on reported K_d range of 2 to 9.	10
Mn	2400	--	0	No Hanford K_d data per se are available. Studies of Mn in reactor cooling water show that Mn differs from Fe and is mostly soluble and cationic as opposed to particulate. Mn adsorbed to Columbia River sediments does desorb appreciably when immersed in seawater [Roberstson and Perkins (1975), Evans and Cutshall 1973]. We suggest that Mn fate be considered similar to Ni.	
Np	25	2.4 to 21.7	10	Hanford sediment data. Exists primarily as NpO_2^+.	5, 6, 8, 9
Ni	2400	50 to 2350	>10	Hanford sediment data. 440 to 2350 mL/g (5) Broad range of sediments, including those from Hanford, 50 to 230 mL/g (18)	5, 18
Pa	3600	--	0	No Hanford soil data available. For silt soils, Thibault et al. (1990) estimated a geometric mean Pa-K_d of 1800. Pa exists in the periodic chart between Th and U. In nature it generally has a +5 oxidation state. It does not behave like Np(V) in solution, nor like Th or U.	2
Pb	80,000	13,000 to 79,000	>50	Hanford sediment data. Pb^{2+} can (co)precipitates readily and sorbs strongly to sediments	7
Po	1100	196 to 1063	6	No Hanford soil data available. For agricultural soils, Baes and Sharp estimated a geometric mean K_d of 540 based on reported K_d range of 196 to 1063.	10
Pu	5000	80 to 4300	7	Hanford sediment data. Pu(V, VI): Hanford subsoils, pH 4 to 12, 80 to >1980 mL/g (19). Washington A Sediment (Hanford) K_d = 100 mL/g (21), Washington B Sediment (Hanford) K_d = 4300 mL/g (21).	19, 21
Ra	500	214 to 467	4	Hanford sediment data. Like alkaline earth (K_d: Ra>Ba>Sr>Ca>Mg)	11
Re	80	--	0	No Hanford soil data available. For silty soils. Thibault et al. (1990) estimated a geometric mean Re-K_d of 40.	2
Rn	0.1	--	0	No Hanford soil data available. Rn is essentially an inert gas. Precipitation and adsorption is not important.	12

Table A.1. (contd)

Rad.	Selected Conservative K_d Values for Agricultural Soils[(a)] (mL/g)	Reported K_d Range (mL/g)	Number of Reported K_d's	Rational	Ref[(b)]
Ru	500	40 - 750	5	Rhodes (1957a) has Hanford specific data. K_d is highest for solutions with neutral pH and drops off when there are acid conditions or basic conditions above 9. Ru in the presence of high nitrate forms mobile nitroso compounds. At Hanford chemical processing wastes disposed to ground allowed rapid migration of Ru. However Ru not in the presence of nitrate does exhibit moderate to strong adsorption (K_d = 40 to 400 ml/g). Roberstson and Perkins (1975) found that Ru in reactor cooling water was mainly bound to fine-grained particulates that were filtered out before reaching groundwater or the Columbia River.	16
Se	2	-3.4 to 0.78	3	Hanford sediment data. Soluble anion, SeO_4^{2-}	5
Sb	5000	180 to >4000	4	No Hanford soil data available. Sb-125 K_d values at the Savannah River Site burial grounds were 180, 2300, 3800, and >4000 mL/g	20
Sr	180	5 to 173	>25	Hanford sediment data. Na System, 173 mL/g, 49 to 50 mL/g (15) Ca System, 8 to 13 mL/g, 5 to 19 mL/g (15) 5 to 120 mL/g (16) 19.1 to 21.5 mL/g (5) Na System, pH 7 to 11, 14.9 to 25.1 mL/g (17)	15, 16, 5, 17
Sn	900	--	0	No Hanford soil data available. For silty soils. Thibault et al. (1990) estimated a geometric mean Re-K_d of 450.	2
Tc	2	-3.4 to 0.57	45	Hanford sediment data. Soluble anion, TcO_4^-. Selected K_d value increased to account for the likely presence of organic matter in the solid phase.	4, 5, 6
Th	600,000	2000 to 510,000	17	No Hanford soil data available. For agricultural soils, Baes et al. (10) estimated a geometric mean K_d of 150,000 based on reported K_d range of 2000 to 510,000. It is apparent that the higher Th-K_d's included in their data set represented precipitation reactions. Thus, a lower Th-K_d than the upper limit is suggested for this simulation.	10
^3H	0.7	0.00 to 0.7	>13	Hanford sediment data. Typically, a K_d = 0 is used for ^3H. ^3H may substitute for ^1H in water on clays and other hydrated soil constituents. The upper limit of the reported ^3H-K_d values is hard to rationalize. Thus, a somewhat lower ^3H-K_d than the upper limit is suggested for this simulation.	13

Table A.1. (contd)

Rad.	Selected Conservative K_d Values for Agricultural Soils[(a)] (mL/g)	Reported K_d Range (mL/g)	Number of Reported K_d's	Rational	Ref[(b)]
U	7	0.08 to 3.48[(d)]	51	Hanford sediment data. UO_2^{2+} is highly mobile in carbonate systems. The suggested value is greater than the upper limit because the sediments used to generate this range generally had quite low organic matter concentrations. It is anticipated that the agricultural soils will have relatively high organic matter concentrations.	5, 6, 14
Y	1500	--	0	No Hanford soil data available. Baes et al. (1984, Table 2.13) estimated that the geometric mean for Y-K_d values for agricultural soils and clays of pH 4.5 to 9 is 510 mL/g, with a range of 160 to 1640 mL/g.	10

(a) Estimated largest K_d value likely to exist in a hypothetical agricultural soil located on the Hanford Site.

(b) References: 1 = Kaplan et al. 1995; 2 = Thibault et al. 1990; 3 = Martin 1996; 4 = Gee and Campbell 1980; 5 = Serne et al. 1993; 6 = Kaplan et al. 1996; 7 = Rhoads et al. 1992; 8 = Routson et al. 1976; 9 = Sheppard et al. 1976; 10 = Baes et al. 1984; 11 = Ames and Rai 1978; 12 = Tanner 1980; 13 = Jones et al. 1980; 14 = Lindenmeier et al. 1995; 15 = Routson et al. 1978; 16 = Rhodes 1957b; 17 = Nelson 1959; 18 = Serne and Relyea 1983; 19 = Rhodes 1957a; 20 = Stone et al. 1984, 21 = Glover et al. 1976; 22 = Kaplan et al. 1998, 23 = Robertson and Perkins 1975.

(c) The mobility of ^{14}C in surface soils may not be governed by adsorption processes, as described by the K_d metric. Instead, partitioning into the gas phase and isotopic dilution with ^{12}C in the dissolved phase and in the solid phase (e.g., calcite) may be more important processes controlling aqueous ^{14}C concentrations.

(d) This range of data omits one value, U-K_d = 79.4, that is being treated as an outlier.

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Appendix B

K_d Values of Hanford Sediments

Last Revision: May 18, 1999

K_d VALUES of HANFORD SEDIMENTS
Last revisions are shown in red and were made on May 18, 1999.

Table B.1. C-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

C-K_d (mL/g)	Experimental Conditions				
	C[(a)] (nCi/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[(b)]
4.06	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 30 days	1
4.37	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 30 days	1
3.8	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 30 days	1
3.47	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 30 days	1
4.56	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 30 days	1
3.74	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 30 days	1
0.05	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 2.69 d	1
0.05	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 2.44 d	1
0.07	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 8.37 d	1
0.12	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 8.21 d	1
0.04	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 22.3 d	1
0.03	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 21.2 d	1
0.09	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 3.8 d	1
0.07	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, Residence time = 3.7 d	1

(a) ^{14}C spike was placed in Hanford groundwater containing about 70 mg/L total alkalinity (as $^{12}CO_3^{2-}$).
(b) Reference: 1 = Martin 1996.

Table B.2. Cl-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

Cl-K_d (mL/g)	**Experimental Conditions**				
	Cl (Mole/L)	**Aqueous Phase**	**Solid Phase**	**Expt'l Method**	**Ref[(a)]**
-0.013	0.01	Synthetic groundwater	Hanford surface soil (Soil B), pH 8.2, CEC 5 meq/100 g	Column	1
-0.008	0.01	Synthetic groundwater	Hanford surface soil (Soil B), pH 8.2, CEC 5 meq/100 g	Column	1
(a) References: 1 - Gee and Campbell 1980					

Table B.3. I-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

I-K_d (mL/g)	**Experimental Conditions**				
	^{125}I (nCi/mL)	**Aqueous Phase**	**Solid Phase**	**Expt'l Method**	**Ref[(a)]**
0.7	200	Hanford groundwater	CGS-1, sand, pH 7.7, CEC = 2.6 meq/100 g	Batch	1
1.4	200	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	1
1.3	200	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	Batch	1
7	200	0.003 M carbonate/sulfate	Rupert, sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
8	200	0.0006 M carbonates/sulfates	Rupert, sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
11	200	0.014 M carbonate/sulfate	Rupert, sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
15	200	0.003 M nitrate/chloride	Rupert, sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
10	200	0.0006 M nitrate/chloride,	Rupert, sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
4	200	0.014 M nitrate/chloride	Rupert, sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
0.2	100	Hanford groundwater, Contact time = 14 d	Trench 94, course sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
2.1	100	Hanford groundwater, Contact time = 200 d	Trench 94, course sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
3.2	100	Hanford groundwater, Contact time = 260 d	Trench 94, course sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
9.8	100	Hanford groundwater, Contact time = 330 d	Trench 94, course sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
2.0	100	Hanford groundwater, Contact time = 14 d	Trench AE-3, silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
2.1	100	Hanford groundwater, Contact time = 200 d	Trench AE-3, silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3

Table B.3. (contd)

I-K_d (mL/g)	Experimental Conditions				
	^{125}I (nCi/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[(a)]
3.1	100	Hanford groundwater, Contact time = 260 d	Trench AE-3, silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
4.1	100	Hanford groundwater, Contact time = 330 d	Trench AE-3, silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
1.1	100	Hanford groundwater, Contact time = 14 d	TBS-1, Sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
0.05	100	Hanford groundwater, Contact time = 200 d	TBS-1, Sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
10.5	100	Hanford groundwater, Contact time = 260 d	TBS-1, Sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
6.8	100	Hanford groundwater, Contact time = 330 d	TBS-1, Sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
0.04 ± 0.07 (20 analyses)	100	Hanford groundwater, Contact time =14 d	ILAW Borehole [299-E17-21] Sediment, pH 8.75 ± 0.15; CEC 6.18 ± 2.18 meq/100 g (20 analyses)	Batch	4
(a) References: 1 = Serne et al. 1993; 2 = Gee and Campbell 1980, 3 = Kaplan et al. 1996, 4 = Kaplan et al. 1998.					

Table B.4. Pb-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

Pb-K_d (mL/g)	Experimental Conditions				
	Pb (µg/L)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[(a)]
13,000 to 79,000	0.001 to 1	Hanford groundwater (Well 6-S3-25)	218-E-12B Burial Ground Hanford Sediment, Sand, pH 8.35, Smectite, illite, plagioclase dominated, CEC = 5.1 meq/100 g	>50 Batch Tests, contact time = 7 to 10 days	1
~13,000 to ~30,000	0.001 to 1	Hanford groundwater (Well 6-S3-25)	218-E-12B Burial Ground Hanford Sediment, Sand, pH 8.35, Smectite, illite, plagioclase dominated, CEC = 5.1 meq/100 g	>8 Batch Tests, contact time = 30 days	1
(a) References: 1 = Rhoads et al. 1992.					

Table B.5. Np-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

Np-K_d (mL/g)	Experimental Conditions				
	^{237}Np (nCi/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref$^{(a)}$
21.7	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	Batch	1
2.4	1480	0.002 M Ca	Burbank, loamy sand, pH 7, CEC = 4.9 meq/100 g	Batch	2
20.2	25	Distilled water	Ritzville, silt loam	Batch	3
15.4	25	Distilled water	Burbank, loamy sand, pH 7, CEC = 4.9 meq/100 g	Batch	3
14.5	100	Hanford groundwater, contact time = 7 d	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	4
20.0	100	Hanford groundwater, contact time = 77 d	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	4
2.6	100	Hanford groundwater, contact time = 7 d	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	4
14.7	100	Hanford groundwater, contact time = 77 d	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	4
3.7	100	Hanford groundwater, contact time = 7 d	Trench TBS-1, Sand, pH 8.2, CEC = 6 meq/100 g	Batch	4
4.4	100	Hanford groundwater, contact time = 77 d	Trench TBS-1, Sand, pH 8.2, CEC = 6 meq/100 g	Batch	4

(a) References: 1 = Serne et al. 1993; 2 = Routson et al. 1976; 3 = Sheppard et al. 1976; 4 = Kaplan et al. 1996.

Table B.6. Ra-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

Ra-K_d (mL/g)	Experimental Conditions				
	Ra (mg/L)	Aqueous Phase	Solid Phase	Expt'l Method	Ref$^{(a)}$
354	7	Simulated river water	Utah, sandy soil, 2-5% calcite, quartz and feldspar, pH 7.9	Batch	1
289	7	Simulated river water	Utah, sandy soil, 2-5% calcite, quartz and feldspar, pH 7.6	Batch	1
467	7	Simulated river water	Utah, sandy soil, 2-5% calcite, quartz and feldspar, pH 7.8	Batch	1
214	7	Simulated river water	Utah, sandy soil, 2-5% calcite, quartz and feldspar, pH 7.7	Batch	1

(a) References: 1 = Ames and Rai (1978) referenced Serne (1974) = Personal communication with R.J. Serne, PNNL.

Table B.7. Se-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

Se-K_d (mL/g)	Experimental Conditions				
	Se (mg/L)	**Aqueous Phase**	**Solid Phase**	**Expt'l Method**	**Ref(a)**
0.78	0.1	Hanford groundwater	CGS-1, sand, pH 7.7, CEC = 2.6 meq/100 g	Batch	1
-3.44	0.1	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	1
-2.4	0.1	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	Batch	1
6.73 ± 1.90 (20 analyses)	100nCi/ml	Hanford groundwater, Contact time = 14 d	ILAW Borehole [299-E17-21] Sediment, pH 8.75 ± 0.15; CEC 6.18 ± 2.18 meq/100 g (20 analyses)	Batch	2

(a) References: 1 = Serne et al. 1993, 2 = Kaplan et al. 1998.

Table B.8. Tc-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

Tc-K_d (mL/g)	Experimental Conditions				
	^{95}Tc (nCi/mL)	**Aqueous Phase**	**Solid Phase**	**Expt'l Method**	**Ref(a)**
0.1	100(b)	Hanford groundwater	CGS-1, sand, pH 7.7, CEC = 2.6 meq/100 g	Batch	1
0.1	100(b)	Hanford groundwater	Touchet Bed, loamy sand	Batch	1
0.1	100(b)	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	Batch	1
-2.77	2	0.003 M carbonate/ sulfates	Ringold, loam, pH 8.8, CEC = 12 meq/100 g	Batch	2
-1.13	2	0.0006 M carbonates/sulfates	Ringold, loam, pH 8.8, CEC = 12 meq/100 g	Batch	2
-0.04	2	0.014 M carbonate/sulfate	Ringold, loam, pH 8.8, CEC = 12 meq/100 g	Batch	2
0.57	2	0.003 M nitrate/chloride	Ringold, loam, pH 8.8, CEC = 12 meq/100 g	Batch	2
0.54	2	0.0006 M nitrate/chloride	Ringold, loam, pH 8.8, CEC = 12 meq/100 g	Batch	2
-0.51	2	0.014 M nitrate/chloride	Ringold, loam, pH 8.8, CEC = 12 meq/100 g	Batch	2
0.07	2	0.003 M carbonate/ sulfates	Rupert ,sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
-1.62	2	0.0006 M carbonates/sulfates	Rupert ,sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
-0.31	2	0.014 M carbonate/sulfate	Rupert ,sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
0.06	2	0.003 M nitrate/chloride	Rupert ,sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
0.52	2	0.0006 M nitrate/chloride	Rupert ,sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2
0.38	2	0.014 M nitrate/chloride	Rupert ,sand, pH 8.2, CEC = 5.0 meq/100 g	Batch	2

Table B.8. (contd)

Tc-K$_d$ (mL/g)	Experimental Conditions				
	^{95}Tc (nCi/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref(a)
-0.16	0.33	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.05	0.67	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.01	1.0	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.04	3.3	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.06	6.67	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.05	10	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.05	33	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.04	66.7	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.03	100	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.06	0.33	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
0.12	0.67	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.11	1.0	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.05	3.3	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.01	6.67	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.02	10	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.03	33	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.03	66.7	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.01	100	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.02	100	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.02	100	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
0.08	100	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
0.11	100	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch	3
-0.01	100	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.19	100	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3

Table B.8. (contd)

Tc-K_d (mL/g)	Experimental Conditions				
	^{95}Tc (nCi/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[(a)]
0.03	100	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
0.11	100	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
-0.03	100	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
-0.19	100	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
0.07	100	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
0.07	100	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	3
-0.01 ± 0.01 (20 analyses)	100(b)nCi /ml	Hanford groundwater, Contact time = 14 d	ILAW Borehole [299-E17-21] Sediment, pH 8.75 ± 0.15; CEC 6.18 ± 2.18 meq/100 g (20 analyses)	Batch	4

(a) References: 1 = Serne et al. 1993; 2 = Gee and Campbell 1980; 3 = Kaplan et al. 1996, 4 = Kaplan et al. 1998.
(b) ^{95m9}Tc (not ^{95}Tc) was used in experiment.

Table B.9. ^{3}H-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.05 M) Conditions

^{3}H-K_d (mL/g)	Experimental Conditions				
	^{3}H (ng/L)	Aqueous Phase	Solid Phase[(a)]	Expt'l Method	Ref[(b)]
0.05			Rupert sand, 91% sand, 7% silt, 2% clay	Column	1
0.1			Rupert sand, 91% sand, 7% silt, 2% clay	Batch	1
0.5			Rupert sand, 91% sand, 7% silt, 2% clay	Batch	1
0.04			Rupert sand, 91% sand, 7% silt, 2% clay	Column	1
0.1			Rupert sand, 91% sand, 7% silt, 2% clay	Batch	1
0.3			Rupert sand, 91% sand, 7% silt, 2% clay	Batch	1
0.06			Ringold Formation, loamy sand, 81% sand, 15% silt, 4% clay	Column	1
0.1			Ringold Formation, loamy sand, 81% sand, 15% silt, 4% clay	Batch	1
0.7			Ringold Formation, loamy sand, 81% sand, 15% silt, 4% clay	Batch	1
0.1			Ringold Formation, loamy sand, 81% sand, 15% silt, 4% clay	Batch	1
0.4			Ringold Formation, loamy sand, 81% sand, 15% silt, 4% clay	Batch	1
0.1			Hanford sand, pH 8.0	Column	1
0.01			Hanford sand, pH 8.0	Batch	1

(a) References: 1 = Jones et al. 1980.

Table B.10. U-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

U-K_d (mL/g)	Experimental Conditions				
	U (ng/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[a]
1.98	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	40% saturated column	1
0.49	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	40% saturated column	1
2.81	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	38% saturated column	1
0.62	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	22% saturated column	1
0.45	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	30% saturated column	1
0.54	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	23% saturated column	1
0.62	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	25% saturated column	1
0.40	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	17% saturated column	1
0.10	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	7% saturated column	1
0.08	100	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	7% saturated column	1
1.7	50	Hanford groundwater	CGS-1, sand, pH 7.7, CEC = 2.6 meq/100 g	Batch	2
2.3	50	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	Batch	2
79.3[b]	50	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	2
1.1	200	Hanford groundwater, pH 8.5	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
1.5	200	Hanford groundwater, pH 8.3	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
1.4	200	Hanford groundwater, pH 8.4	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
2.1	200	Hanford groundwater, pH 9.1	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch	3
0.8	200	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, 40% Saturated	3
1.5	200	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, 75% Saturated	3
0.9	200	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, 100% Saturated	3
0.42	50	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	Column, 34% Saturated	3
0.51	50	Hanford groundwater	Trench-8, loamy sand, pH 7.9, CEC = 5.2 meq/100 g	Column, 39.4% Saturated	3
2.48	50	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, 52% Saturated	3
1.96	50	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, 63% Saturated	3

Table B.10. (contd)

U-K_d (mL/g)	Experimental Conditions				
	U (ng/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[(a)]
1.24	50	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, 81% Saturated	3
0.91	50	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Column, 100% Saturated	3
4.6	50	Hanford groundwater	McGee Ranch, Silty loam, pH 8.2	Column, 60% Saturated	3
1.58	50	Hanford groundwater	McGee Ranch, Silty loam, pH 8.2	Column, 100% Saturated	3
0.5	350	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 7 d	3
0.8	350	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 260 d	3
0.9	350	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 330 d	3
1.1	350	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact time = 398 d	3
1.25	350	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch, Contact time = 7 d	3
1.6	350	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch, Contact time = 260 d	3
2.1	350	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch, Contact time = 330 d	3
2.05	350	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch, Contact time = 398 d	3
1.63	350	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact time = 7 d	3
3.23	350	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact time = 260 d	3
3.4	350	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact time = 330 d	3
3.48	350	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact time = 398 d	3
0.21	5	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact Time =30 d	3
0.16	7	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact Time =30 d	3
0.1	22	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact Time =30 d	3
0.18	52	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact Time =30 d	3
0.11	81	Hanford groundwater	Trench 94: Course Sand, pH 8.2, CEC = 5.3 meq/100 g	Batch, Contact Time =30 d	3
0.25	2	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact Time =30 d	3
0.64	4	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact Time =30 d	3
0.52	7	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact Time =30 d	3

Table B.10. (contd)

U-K_d (mL/g)	Experimental Conditions				
	U (ng/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[(a)]
0.48	23	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact Time =30 d	3
0.35	47	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact Time =30 d	3
0.53	67	Hanford groundwater	Trench AE-3: Silty loam, pH 8.3, CEC = 6.4 meq/100 g	Batch, Contact Time =30 d	3
0.62 ± 0.12 (20 analyses)		Hanford groundwater, Contact time = 14 d	ILAW Borehole [299-E17-21] Sediment, pH 8.75 ± 0.15; CEC 6.18 ± 2.18 meq/100 g (20 analyses)	Batch	4

(a) References: 1 = Lindenmeier, et al. 1995; 2 = Serne et al. 1993; 3 = Kaplan et al. 1996, 4 = Kaplan et al. 1998.
(b) This K_d values appears too large and will be treated as an outlier.

Table B.11. Cs-K_d Values Under Neutral-to-High pH, Low Organic Material Concentrations, Oxic, Low-Ionic Strength (≤0.01 M) Conditions

Cs-K_d (mL/g)	Experimental Conditions				
	U (nCi/mL)	Aqueous Phase	Solid Phase	Expt'l Method	Ref[(a)]
2055±597 (20 analyses)	25	Hanford groundwater, Contact time = 14 d	ILAW Borehole [299-E17-21] Sediment, pH 8.75 ± 0.15; CEC 6.18 ± 2.18 meq/100 g 20 analyses)	Batch	1
2190 ± 870 (9 analyses)	50	Hanford groundwater	CGS-1, sand, pH 7.7, CEC = 2.6 meq/100 g	Batch	2
7610 ± 4690 (9 analyses)	50	Hanford groundwater	TBS-1, loamy sand, pH 8.2, CEC = 6 meq/100 g	Batch	2

(a) References: 1 = Kaplan et al. 1998. 1995; 2 = Serne et al. 1993.

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