SRNL
SAVANNAH RIVER NATIONAL LABORATORY

INTER-OFFICE MEMORANDUM

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Re: **Distribution Coefficients for Various Elements of Concern to the Tank Waste Performance Assessment**

The distribution coefficients (K_d values) for Ag, Al, As, Cd, Ce, Cr, Cu, F, Fe, Hg, K, Mn, Mo, N, Na, Pd, Sb, and Zn for the tank closure performance assessment are presented in Table 1. More specifically, K_d values were selected from the SRS literature, and when not available, from the open literature. This report does not include any new laboratory results. K_d values were selected and justification for their selection are presented for four chemical/mineralogical environments: 1) far-field sandy sediment, 2) far-field clayey sediment, 3) oxidizing concrete, and 4) reducing-grout. A detailed description of these four environments is presented in Kaplan (2006). These elements are generally not the key dose contributors to the performance assessment, but various isotopes of these elements are included in the decay chain of important dose contributors. The inclusion of these isotopes in the performance assessment calculation is important to provide an accurate calculation of risk. This memorandum is intended to provide early guidance for the performance assessment. This data will be added to an existing geochemical data package (Kaplan, 2006), and this data package will be reissued, along with other data, later in the year.

Table 1. K_d values appropriate for SRS sediment and cementitious conditions.

Element	Assumed Species	Analog	Best Sandy	Best Clayey	Concrete Oxidizing$^{(a)}$	Concrete Reducing$^{(a)}$	Comments
Ag	Ag^+		60	150	1/1/0.1	1/1/0.1	There have not been any Ag sorption studies conducted with SRS sediments. Consequently, literature values were used to select K_d values for this table. In a survey of literature K_d values, Thibault et al. (1991) reported sand K_d values of 20, 60, 75 and 200 mL/g and silt/clay Ag-K_d values of 100, 170, 200, 200, and 300 mL/g. Concrete Ag K_d values from Bradbury and Sarott (1995).
Al	Low pH: Al^{3+} $Al(OH)^{2+}$, $Al(OH)_3^0$ High pH: $Al(OH)_4^-$, $Al(OH)_4^{2-}$	In concrete = Am^{3+}	1300	1300	5000/5000 /1000	5000/5000 /1000	Crapse et al (2004; Table 33) made some in situ measurements of Al solubility in a wide pH range from sediments collected from D-Area. At pH2.8, Al porewater concentrations were at a maximum at pH 2.8, ~20,000 ppb, and then the concentration decreased to ~10 ppb at pH 7, and then at higher pH levels the Al concentrations started to rise again. At the pH of interest for the hypothetical Sandy and Clayey sediment, pH 5.5, the Al concentrations (solubility) were ~300 ppb. A comparable Kd value was 1300 mL/g (Al-solid = 402ppm [Table 25 in Crapse et al. 2004) Al-liquid = 0.3ppm [Fig. 33 Crapse et al. 2004]). Theoretically, Al sorption is actually quite large in alkaline environments. Assumed to behave in concrete like Am; concrete K_d values taken from Bradbury and Sarott (1995).
As	AsO_4^{3-}		100	200	1000/1000 /100	1000/1000 /100	Crapse et al. (2004) measured in situ As K_d values in D-Area, SRS, by collecting porewater/sediment paired samples along a pH gradient. The As Kd values varied by 3 orders of magnitude along the transect and did not vary in a consistent manner with pH. At pH 5.5, the K_d values varied from about 30 to 20,000 mL/g (Figure 45; Crapse et al. 2004). As(V) (AsO_4^{3-}) behaves like PO_4^{3-}. As(V) sorbs in soils generally more than As(III) (Krupka and Serne 2004). As(V) is more expected in oxidizing vadose zone environments. As(V) sorption at pH <7 is independent of pH; at pH >7, As sorption decreases with pH. Baes and Sharp (1983) suggested a single default K_d value of 6.7 mL/g for As(V) based on 37 As K_d values from 1.9 to 18 mL/g for agricultural soils and clays in the pH range of 4.5 to 9.0. Importantly, few of these soils are typical of SRS subsurface sediments. Buchter et al. (1989) in a survey of 11 soils, reported that a soil ("Windsor": pH = 5.8, OM =

Ele-ment	Assumed Species	Analog	Best Sandy	Best Clayey	Concrete Oxidizing[(a)]	Concrete Reducing[(a)]	Comments
							0.67%, silt+clay = 25%) similar to that of a clayey SRS sediment had an As K_d value of 105 mL/g. In the same study, the sediment most like a "sandy SRS sediment" ("Spodosal": pH 4.3; OM = 1.98%, silt+clay = 10%) had an As K_d value of 19 mL/g. Mariner et al. (1996) measured As(V) K_d values between 0.3 to 6.5 mL/g in a contaminated aquifer ranging in pH from 8.4 to 11. The selection of K_d for the SRS subsurface was largely based on the results of Crapse et al. (2004). Phosphate, an analog for AsO_4^{3-}, is known to be strongly bound to concrete.
Cd	Cd(II)	Soft divalent cation in cementitious material, Pd^{2+}. See "Comments".	4	10	100/100 /10	1000/100 /10	There have not been any Cd sorption studies conducted with SRS sediments. Consequently, literature values were used to select K_d values for this table. Cd in groundwater with pH <8.2 tends to exist as the uncomplexed Cd^{2+} species. In systems of pH >8.2, it exists as $CdCO_3^0$. Sulfate tends to form strong complexes with Cd and keeps aqueous Cd concentrations low. $CdCO_3$ (otavite) limits Cd solution concentration in alkaline soils. In low redox systems and where sulfide is present, such as expected in the reducing grout, Cd may precipitate. Precipitation with carbonate is increasingly important as the pH increases above 8, such as would be expected in cementitious environments. At low concentrations and in the pH of SRS sediment, cation exchange and surface complexation onto iron oxides are expected to be important. Buchter et al. (1989) in a survey of 11 soils, reported that a soil ("Windsor": pH = 5.8, OM = 0.67%, silt+clay = 25%) similar to that of a clayey SRS sediment had a Cd K_d value of 14.4 mL/g). In the same study, the sediment most like a "sandy SRS sediment" ("Spodosal": pH 4.3; OM = 1.98%, silt+clay = 10%) had a Cd K_d value of 5.5 mL/g. Krupka et al. (1999) recommended that for sediments between the pH 5 to 8 to use Cd K_d values in the range of 8 to 4000 mL/g. This recommendation was based upon an extensive survey of the literature; 174 K_d values; however most of these K_d values were measured in surface soils and under conditions unlike the SRS. The suggested sediment K_d values reported here are largely based upon the findings of Buchter et al. (1989) and basic principles of Cd geochemistry. It is assumed that in cementitious environments, Cd will coprecipitate with $CaCO_3$ and under reducing and high S concentrations conditions in grout that Cd will precipitate as CdS, a sparingly soluble precipitate. Adapted the palladium K_d from

Element	Assumed Species	Analog	Best Sandy	Best Clayey	Concrete Oxidizing[(a)]	Concrete Reducing[(a)]	Comments
							Bradbury and Sarott (1995) for the Cd oxidized cementitious K_d values. For reducing cementitious K_d values, increased K_d by an order of magnitude to reflect presence of CdS species.
Ce	Ce(III)	For concrete = Am^{3+}	1000	1500	5000/5000 /1000	5000/5000 /1000	In geologic environments, Ce forms weak complexes (Ames and Rai 1976). A ranking of complexes from least to greatest in natural oxidizing environment is $CeCl_2^+$, $CeNO_3^{2+}$, Ce^{3+}, and $CeSO_4^+$. Ames and Rai (1976) also reference several studies that indicate that Ce is strongly bound to soils in a non-exchangeable fraction. They also reference a non-linear relationship with Ce sorption and pH. Rhodes (1957) studied the sorption of Ce on a Hanford soil as a function of pH. The K_d values rose steadily from 3 mL/g at pH 1.6 to >1980 mL/g at pH 6.1. The K_d then decreased to a minimum of ~100 mL/g at pH 10 and increased again to >1980 mL/g at pH 12 and above. Bensen (1960) reported that the soil uptake of Ce added as Ce(III) was identical to that added as Ce(IV). Ames and Rai (1976) noted Ce K_d of 1050 and 1300 mL/g on Casaccia soil (no characterization data of the soil or solution were provided). Assumed to behave in concrete like Am; concrete K_d values taken from Bradbury and Sarott (1995).
Cr	CrO_4^{2-} in soils and Cr(III) in reducing concrete	For oxidizing concrete = Cl^-; reducing concrete = Am^{3+}	4	10	20/20/2	5000/5000 /1000	There have not been any Cr sorption studies conducted with SRS sediments. Consequently, literature values were used to select K_d values for this table. Important geochemical properties of Cr include 1) Cr exists as Cr(VI) in oxidizing soils and Cr(III) in reducing soils, 2) increase pH results in decreased Cr(VI) K_d values; 3) presence of competing anions reduce Cr(VI) adsorption (reviewed in Krupka et al. 1999). Krupka et al. (1999), in a review of Cr K_d values suggested that a soil with low Fe concentration and a pH between 5.1 to 6, should have a K_d values in the range of 20 to 34 mL/g. At pH values > 7.1, the K_d decreases to a range of 0 to 7 mL/g. In concrete, assume to behave like Cl^-, and in reducing concrete assumed to precipitate as Cr(III) and therefore that K_d is taken from Am(III) (Bradbury and Sarott 1995).
Cu	Cu^{2+} = oxidized Cu^+ = reduced	For concrete = Ag^+	50	70	1/1/1	1/1/1	Cu exists primarily in the +2 state through pH/Eh region of interest. Cu^+ exists only under reducing & pH >6. CuS formed under reducing conditions. In oxidizing conditions, above pH 7, Cu(OH)2 dominates; below pH 7, Cu2+ dominates. Did not find any K_d values in literature in acidic or circum-neutral sediments. There have not been any K_d

Element	Assumed Species	Analog	Best Sandy	Best Clayey	Concrete Oxidizing[(a)]	Concrete Reducing[(a)]	Comments
							values measured in SRS subsurface sediments. In a fine sand (pH 8.3; OM. = 1.4%) Cu Kd = 206 mL/g (Wong et al. 1983); this value is probably the upper limit of the K_d we may expect in the SRS subsurface due to the elevated organic matter concentrations and pH. Buchter et al. (1989) in a survey of 11 soils, reported that a soil ("Windsor": pH = 5.8, OM = 0.67%, silt+clay = 25%) similar to that of a clayey SRS sediment had a Cu Kd value of 77.1 mL/g. In the same study, the sediment most like a "sandy SRS sediment" ("Spodosal": pH 4.3; OM = 1.98%, silt+clay = 10%) had a Cu K_d value of 56 mL/g. Assumed to behave in concrete like Ag^+; concrete K_d values taken from Bradbury and Sarott (1995).
F	F^-	For soil = anions; for concrete = Cl^-	0	0	20/20/2	20/20/2	There have not been any F sorption studies conducted with SRS sediments. Consequently, it was assumed to behave like other monovalent anions, which tend to have K_d values of 0 or near 0 mL/g. Assumed to behave in concrete like Cl^-; concrete K_d values taken from Bradbury and Sarott (1995).
Fe	Fe^{3+} in oxidizing and Fe^{2+} in reduced	For reducing concrete = Ni; oxidizing concrete = Am	200	400			Knox and Kaplan 2003 measured Fe K_d values of a subsurface sediment collected from TNX. They conducted a sorption isotherm (K_d as a function of dissolved Fe concentrations at ambient pH levels, pH 5.8. They noted a rather flat response of K_d to Fe_{aq} concentrations from 0.1 to 0.21 µg/L; above this concentration Fe precipitation obviously occurred because the K_d values increased precipitously. The Fe K_d values at the low concentrations were 400 mL/g. Assumed to behave in oxidizing concrete like Am^{3+} and in reducing concrete like Ni; concrete K_d values taken from Bradbury and Sarott (1995).
Hg	Hg^{2+} in oxidizing and Hg^0 in reduced	Concrete = Soft divalent cation, Pb^{2+}	800	1000	500/500 /50	500/500 /50	There has been two Hg^{2+} sorption study conducted with SRS sediments (Bibler and Marson 1992; Kaplan and Iverson 1999). The study by Bibler and Marson (1992) will not be discussed because they only used limited particle size fractions (40 to 60 mesh or 80 to 100 mesh sediment fractions) and they conducted their test using extremely high background ionic strength solutions, non-environmentally relevant levels, which would be expected to greatly influence sorption measurements. Kaplan and Iverson (1999) measured Hg^{2+} values of an SRS vadose zone sand and clay sediment samples, meant as end-members of the types of sediments encountered at the SRS. The sand had K_d values ranging from 956 to 2452 mL/g, while the clay sample had Kd values 1296 to 8517 mL/g. K_d values measured in the clay

Element	Assumed Species	Analog	Best Sandy	Best Clayey	Concrete Oxidizing$^{(a)}$	Concrete Reducing$^{(a)}$	Comments
							sediment did not vary systematically with pH: at pH 3.1, 4.1, and 9.3 the Hg-K_d = 58, 1296 and 429 mL/g, respectively. Elevating Cl, a Hg-complexing ligand known to reduce Hg sorption, concentrations by 0, 2, or 20 mg/L Cl^-, caused only marginal decreases in Hg K_d: 1296, 773, and 967 mL/g, respectively. Based on these data, Kaplan and Iverson (1999) concluded that the mostly likely K_d for this system was 800 mL/g for the sand and 1000 mL/g for the clay. The corresponding reasonably conservative K_d values for the sand and clay were 600 and 700 mL/g, respectively. In a survey of 11 non-SRS sediments, Buchter et al. (1989) reported that a soil ("Windsor": pH = 5.8, OM = 0.67%, silt+clay = 25%) similar to that of a clayey SRS sediment had a Hg K_d value of 130 mL/g. In the same study, the sediment most like a "sandy SRS sediment" ("Spodosal": pH 4.3; OM = 1.98%, silt+clay = 10%) had a Hg K_d value of 86 mL/g. Assumed to behave in concrete like Pb^{2+}; concrete K_d values taken from Bradbury and Sarott (1995).
K	K^+	See comments	10	60	1/2/2	1/2/2	There have not been any K sorption studies conducted with SRS sediments. Consequently, literature values were used to select K_d values for this table. K is a monovalent cation in the same group in the periodic chart as Cs^+. It differs from Cs^+ in that it is more strongly hydrated and therefore would be expected to sorb less. Also, it does not strongly bind to mica/illite clays and stable (natural) K^+ exists in 1-5 ppm concentrations in uncontaminated groundwater, >4 orders of magnitude greater than stable Cs. Thus stable K^+ would compete with radioactive K^+ for sorption sites onto geological surfaces. For these reasons, we set the K-K_d values to about one quarter that of Cs^+, an element we have a lot of sorption data. These values were also selected somewhat lower than actually anticipated to account for the uncertainty associated with these K K_d estimates. For the cementitious material K_d values, the K K_d values were set to 1/10 or less of the value of Cs-K_d values by Bradbury and Sarott (1995).
Mn	Mn^{2+} for reduced conditions; Mn^{4+} for oxidized		15	200	100/100 /10	100/100 /10	Thibault et al. (1990) report Mn K_d values for sand textured sediments of 14, 18, 24, 29, 30, 50, 70, 71,150, 2000 mL/g, and for clay sediments of 250, 2100, and 10,000 mL/g. In a highly weathered soil and low in organic matter, as we have ion the SRS, Willett and Bond (1995) reported K_d values at pH 5, 6, and 7 of 4, 5, and 9.5 mL/g.

Ele-ment	Assumed Species	Analog	Best Sandy	Best Clayey	Concrete Oxidizing[(a)]	Concrete Reducing[(a)]	Comments
	conditions.						The increase Mn sorption with pH was attributed to the tendency for Mn to hydrolyze at elevated pH levels. Their soil had lower Fe-oxide concentrations than SRS sediments. Cementitious K_d values taken from Mn K_d values reported in Bradbury and Sarott (1995).
Mo	Mo(VI) as MnO_4^{2-} in oxidizing environ.; $CaMoO_4^0$ in cementitious environ.; Mo(IV) in reducing environ.		6	120	0.1/0.1 /0.1	0.1/0.1 /0.1	There have not been any Mo sorption studies with SRS sediments conducted. Estimates were based on literature values. Mo will exist as an anion in sediments and as a neutral species in cement environments. Mo does not form strong complexes in environmental conditions. Mo sorbed strongly by Al oxides and especially by Fe-oxides by a strong sorbing mechanism, ligand exchange (reviewed by Goldberg et al. 1996). This results in an inner sphere complex with the Fe-oxides. Consequently, this is a pH dependent process, where lower the pH, the greater the Mo sorption, with a maximum at pH ~5 and essentially no sorption at pH >8. Mo adsorption on calcite, one of the phases formed on cement surfaces, is very low, indicating that $CaCO_3$ is not a significant sink for Mo. Thibault et al. (1990) report Mo K_d values for sand textured sediments of 1, 1.8, 8, and 20 mL/g, and for clay sediments of 13, 40, 40, 140, 160, 200, and 400 mL/g.
N	NO_3^- & NO_2^-	Anion	0	0	0/0/0	0/0/0	
Na	Na^+	See comments	5	25	0.5/1/0.5	0.5/1/0.5	There have not been any Na sorption studies conducted with SRS sediments. Estimates were based on literature values. Because Na is highly hydrated, it tends to sorb weakly to sediments and concrete.
Pd	Pd-metal (Pd^0) in soil & cement; PdS in reducing grout	Ni^{2+}	7	30	100/100 /10	100/100 /10	There have not been any Pd sorption studies conducted with SRS sediments. Estimates were based on literature values. Palladium is stable as elemental Pd or PdS or $Pd^{II}S_2$ under reducing environments (Brookins 1987). As a sulfide, it would be expected to be sparingly soluble. No Pd – soil or Pd –cement K_d values were found in the literature. The estimates selected here assume that Pd exists like a soft metal, such as Ni^{2+}; soil K_d values taken from Ni K_d values reported by Kaplan (2006). Assumed to behave in concrete like Am^{3+}; concrete K_d values taken from Bradbury and Sarott (1995).
Sb	Oxidizing: Sb(III) as	For concrete = Am^{3+}	2500	2500	5000/5000 /500	5000/5000 /500	Sb sorption studies have been conducted with SRS burial ground sediments with deionized water or groundwater (Hoeffner 1985). They

Element	Assumed Species	Analog	Best Sandy	Best Clayey	Concrete Oxidizing[a]	Concrete Reducing[a]	Comments
	$HSbO_2^0$ and $Sb(OH)_3^0$						indicate Sb sorbs strongly to our sediment, with a near linear decrease in $\log K_d$ values and pH. The decrease in sorption at the more basic pH levels may be a result of formation of anionic Sb complexes, such as $Sb(OH)_4^-$ and SbO_2^-. Between pH 5 and 6, the five measured K_d values were between 3000 and 4000 mL/g. At pH 10.5, the Sb-K_d value was 22 mL/g. SRS lysimeter studies indicate that a small amount of Sb is in a mobile anionic form (Hoeffner 1985). The literature indicates there have been very few Sb sorption studies conducted. Ames and Rai (1978) reviewed Sb sorption and concluded that over the pH range of 4 to 8 that Sb exists as a neutral and complexed species. In high pH and high ionic strength systems, Sb K_d values were very low, 0 to 2 mL/g (Ames and Rai 1976). When Ca concentrations were high, such as in a concrete system, Sb appeared to coprecipitate with $CaCO_3$, yielding K_d values in the range of 17 to 122 mL/g. Beneath leaking high level waste tanks in Hanford, Sb was found to migrate rapidly through the subsurface sediment (at the same rate as Co and ruthenium) (Ames and Rai 1978). The high mobility may be attributed to the tendency of Sb to form complex or hydrolyzed species which are neutral or negatively charged. Assumed to behave in concrete like Am^{3+}; concrete K_d values taken from Bradbury and Sarott (1995).
Zn	Zn^{2+}	Pb^{2+}			100/100 /10	100/100 /10	There have been no Zn sorption studies with SRS sediments conducted. Estimates were based on literature values. Thibault et al. (1991) in their review of Zn Kd values reported K_d values for sandy sediments of 20, 90, 200, 2000 mL/g and for clayey sediments, K_d values of 80, 1200, 1900, and 5100 mL/g. Adapted the palladium K_d from Bradbury and Sarott (1995) for the Zn cementitious K_d values.

[a] Concrete K_d values are places in order of Young Cement/Moderately Aged Cement/Aged Cement. These terms are defined in Kaplan (2006) and reflect the age of the concrete in terms of the number of pore volumes of water that pass through them: Young cement is <50 pore volumes, Moderately Aged Cement is 50 to 500 pore volumes, and Aged Cement is >500 pore volumes. These pore volume definitions are based on experimental work that correlate to changes in mineral composition of the concrete.

OM = organic matter

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