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Recommended Distribution Coefficients, Kd Values, for Special Analysis Risk Calculations Related to Waste Disposal and Tank Closure on the Savannah River Site (U)

D. I. Kaplan

August 31, 2005

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1.0 ABSTRACT

The purpose of this document is to provide a technically defensible list of distribution coefficients, or Kd values, for use in performance assessment (PA) and special analysis (SA) calculations on the SRS. Only Kd values for radionuclides that have new information related to them or that have recently been recognized as being important are discussed in this report. Some 150 Kd values are provided in this report for various waste-disposal or tank-closure environments: soil, corrosion in grout, oxidizing grout waste, gravel, clay, and reducing concrete environments. Documentation and justification for the selection of each Kd value is provided.

2.0 OBJECTIVE AND SCOPE

The objective of this task was to provide distribution coefficients, or Kd values[1], for future used in Special Analyses and Performance Assessments conducted to evaluate risk associated with waste disposal or tank closure activities on the Savannah River Site. Only Kd values for radionuclides that have new information related to them or that have recently been recognized as being important to the special analysis are discussed. It is meant that this new information will be added to the existing database of Kd values presented by McDowell-Boyer *et al*. (2000). As such, similar underlying assumptions and references were made in the selection of this data set as was done in McDowell-Boyer *et al.* (2000). Foremost, all attempts were made to maintain a consistent conservatism throughout the entire data set. Following is a brief description of the different Kd values; additional details are presented in McDowell-Boyer *et al*. (2000; page 4-16).

- Soil Kd: Values were selected to represent sandy textured sediments; used for slit trench waste forms and for backfill, and native soil.
- Corrosion in Grout Kd: Values based on experimental or calculated estimates to iron oxides and represent radionuclide sorption to iron oxides that form as a result of oxidation of metal in the waste form, in the waste itself, or rebar in concrete; used for low-activity waste (LAW) vault and cement-stabilized encapsulated waste.
- Oxidizing Grouted Waste Kd: Values used for intermediate level vault, intimately-mixed cement-stabilized encapsulated, and naval reactor waste forms.
- Gravel Kd: Gravel was used to engineer the disposal facility; these Kd values were selected based on the assumption that the sediment had a sandy texture and were set equal to the soil Kd values, *i.e.*, “Soil Kd” = “Gravel Kd.”
- Clay Kd: Natural clay layers exist in the subsurface and clay layers are added as engineered barriers; these values are to be used where clay textured sediment or engineered barriers exist.
- Reducing Concrete Kd: These values are used in reducing concrete environments, *i.e.*, in concretes were the reductant, slag, is purposely introduced into the mixture to create a reducing environment.

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[1] A distribution coefficient is a ratio of the concentration of a radionuclide on the sediment to that in the groundwater in contact with the sediment: e.g., $Pu_{sediment}/Pu_{groundwater}$ = Kd(Pu)

Importantly, the scope of this task did not include a systematic check of the assumptions or of the previously used Kd values reported by McDowell-Boyer *et al.* (2000).

3.0 RESULTS

The new Kd values are presented in bold font in Table 1. The historical Kd values, in regular (non-bold) font, (*i.e.*, those previously reported by McDowell-Boyer *et al.* 2000) are left in the table to provide perspective as to the relative magnitude of the new values. Explanation for the selection of the new Kd values are presented in the “Comments” column or as footnotes.

Table 1. New (Red Font) and Historic (Black Font; McDowell *et al.* 2000) Distribution Coefficients for Various Subsurface Environments.

Element	**Soil Kd (mL/g)**	**Corrosion in Grout Kd (mL/g)**	**Oxidizing Grouted Waste Kd (mL/g)**	**Gravel Kd (mL/g)**	**Clay Kd (mL/g)**	**Reducing Concrete Kd[a] (mL/g)**	**Comments**
Ac	450	3700	5000	450	2400	5000	Ac(III) “Reducing Concrete Kd” value is based on using Am(III) as an analogue. Changed the “Corrosion in Grout Kd” to be consistent with Appendix E (pp. E-5) in McDowell et al. 2000) and “Am(III) analogue” assumption.
Ag	20	80	1	20	80	1	Soil Kd: 6 Sand sediment Kd values range from 20 to 1000, Avg = 317 ± 370 mL/g. Clay Kd = 6 clayey sediment Kd values ranged from 80 to 1000 mL/g, Avg = 317 ± 350 mL/g.[b] Assumed that Corrosion Kd equaled Clay Kd. Assumed Gravel Kd = Sand Kd as reported in Thibault *et al.* (1990).
Al	3700	3700	5000	3700	3700	5000	[e] See footnote at bottom of table.
Am	1900	3700	5000	1900	8400	5000	
Ar	0	0	0	0	0	0	Ar is a noble gas. As such it does not interact with other elements or surfaces.
At	0.6	0.6	2	0.6	1	2	At is a halogen. At “Reducing Concrete Kd” value is based on using iodine as an analogue. The values on this line differ greatly from those previously specified. Old values are presented below in footnote [f].
Ba	10	3	1	10	110	1	Set Ba Kd values equal to those of Sr.
Bi	450	3700	5000	450	12000	5000	According to Pourbaix (1974), Bi(III) is dominant oxidation state. Bi(III) “Reducing Concrete Kd” value is based on using Am(III) as an analogue. Corrected the “Corrosion in Grout Kd” to be consistent with Appendix E (pp. E-5) in McDowell et al. 2000) and “Am(III) analogue” assumption. Corrected the “Corrosion in Grout Kd” to be consistent with Appendix E (pp. E-5) in McDowell et al. 2000).
Bk	1900	3700	5000	1900	8400	5000	Bk(III/IV); No data is available. Bk(III/IV) “Reducing Concrete Kd” value is based on using Am(III) as an analogue.
C	2	0	5000	2	1	5000	Concrete is a very effective “getter” for CO_3^{2-}. Selected a high Kd to produce this effect. [h]
Ca	10	3	1	10	110	1	Ca(II) “Reducing Concrete Kd” value is based on using Sr(II) as an analogue.
Cd	600	600	5000	600	3300	5000	Cd(II) “Reducing Concrete Kd” value is based on using Sr(II) as an analogue.
Cf	510	3700	5000	510	8400	5000	Cf(III) “Reducing Concrete Kd” value is based on using Am(III) as an analogue.
Cl	0	0	20	0	0	20	Cl(-I) is an anion. In high pH environments, such as in concrete, it will sorb poorly. No sorption data available to SRS sediments. Conservative estimate of 0 mL/g. Bradbury and Sarott (1995) provide a “reducing concrete Kd” = “Oxidizing Grouted Waste Kd” = 20 mL/g.
Cm	4000	3700	5000	4000	6000	5000	
Co	8	1200	100	8	96	100	Co(II) “Reducing Concrete Kd” value is based on using Ni(II) as an analogue. “Corrosion in Grout Kd” value is based on using Ni(II) as an analogue. “Gravel Kd” = “Soil Kd” = Kd’s representative of a sandy soil.
Cs	330	330	20	330	1900	20	
Eu	1900	3700	5000	1900	8400	5000	Eu(III) “Reducing Concrete Kd” and “Corrosion in Grout Kd” are based on using Am(III) as an analogue. “Gravel Kd” = “Soil Kd” = Kd’s representative of a sandy soil.
Fr	330	330	20	330	1900	20	No or few sorption tests have been conducted with Fr. Based Kd values on using Cs(I) as a chemical analogue.
Gd	1900	3700	5000	1900	8400	5000	Gd(III); No sorption data is available. Based selection of Kd values on using Am(III) as a chemical analogue.

Element	Soil Kd (mL/g)	Corrosion in Grout Kd (mL/g)	Oxidizing Grouted Waste Kd (mL/g)	Gravel Kd (mL/g)	Clay Kd (mL/g)	Reducing Concrete Kd$^{(a)}$ (mL/g)	Comments
H	0	0	0	0	0	0	
I	0.6	0	2	0.6	1	2	Changed the “Corrosion in Grout Kd” to be consistent with Appendix E (pp. E-5) in McDowell et al. 2000).
K	3	5	2	3	5	2	K(I) “Reducing Concrete Kd” value set equal to the “Oxidizing Grouted Waste Kd “ value. “Corrosion in Grout Kd” value = “Clay Kd” value. “Gravel Kd” = “Soil Kd” = Kd’s representative of a sandy soil.
Kr	0	0	0	0	0	0	Kr is a noble gas. As such it does not interact with other elements or surfaces.
Mo	3	0.6	1	3	13	0.4	Mo is +6 in oxidizing conditions and +4 in reducing conditions. Mo exists primarily as an anion in the oxidized form and presumably as an cation in the reduced form. (Pourbaix 1974). “Corrosion in Grout Kd” is based on using iodine as an analogue. “Gravel Kd” = “Soil Kd” = Kd’s representative of a sandy soil.
Na	3	5	2	3	5	2	Na(I) Kd values selection were based on using K(I) as an analogue.
Nb	160	160	500	160	900	500	
Ni	400	1200	100	400	650	100	
Np	5	750	5000	5	55	5000	
Pa	550	550	5000	550	2700	5000	
Pb	270	270	500	270	550	500	
Pd	55	55	100	55	270	100	
Po	150	150	500	150	3000	5000	Po(IV) “Reducing Concrete Kd” value is based on using Th(IV) as an analogue.
Pu(III/IV)	370	370	5000	370	6500	5000	(g)
Pu(V/VI)	15	15	5000	15	50	5000	(g)
Pu(colloid)	0	0	0	0	0	0	(g)
Ra	500	60	50	500	9100	50	
Rb	55	55	20	55	270	20	Rb(I) “Reducing Concrete Kd” value is based on using Cs(I) as an analogue.
Re	0.1	0	1	0.1	0.1	1000	Tc is an excellent analogue for Re. Used Tc as a chemical analogue to selected Kd values.
Rn	0	0	0	0	0	0	Rn is a noble gas. As such it does not interact with other elements or surfaces.
S	150	170	0.1	5	76	0.1	Used Se as a chemical analogue to select Kd values.
Sb	450	450	5000	450	12000	5000	Sb may exist in the +3 or +5 oxidation states. In oxidizing concrete as +5, otherwise as +3 (Pourbaix 1974). Use Bi as a chemical analogue to select Sb Kd values.
Sc	1900	3700	5000	1900	8400	5000	Used Am(III) as a chemical analogue to select Kd values.
Se	150	170	0.1	5	76	0.1	
Sm	1900	3700	5000	1900	8400	5000	Sm(III) “Reducing Concrete Kd” value is based on using Am(III) as an analogue.
Sn	130	55	1000	130	670	1000	
Sr	10	3	1	10	110	1	
Tc	0.1	0	1	0.1	0.1	1000	Changed the “Corrosion in Grout Kd” to be consistent with Appendix E (pp. E-5) in McDowell et al. 2000).

Element	**Soil Kd (mL/g)**	**Corrosion in Grout Kd (mL/g)**	**Oxidizing Grouted Waste Kd (mL/g)**	**Gravel Kd (mL/g)**	**Clay Kd (mL/g)**	**Reducing Concrete Kd(a) (mL/g)**	**Comments**
Te	0	0	0	0	0	0	Te exists as +4, +5, +6, and +7 oxidation states in the Eh and pH range expected in the LAW vault and surround environment. In all oxidation states it will exist as an anion. Because of the lack of data, the conservative assumption that no sorption occurs was made.
Th	3200	2200	5000	3200	5800	5000	
Tl	20	80	1	20	80	1	Tl(I) in dominant oxidation state (Tl(III) would not be common). Ag$^+$ is used as an analogue for the selection of Kd values; like Ag(I), Tl(I) is a soft metal.
U	800	6000	2000	800	1600	5000	U(IV); Note difference Between "Reducing Concrete Kd" and "Concrete Kd"
W	0	0	0	0	0	0	(c) See footnote at bottom of table.
Y	1900	3700	5000	1900	8400	5000	Used Am(III) as a chemical analogue to selected Kd values for Y(III).
Zr	600	600	5000	600	3300	5000	

(a) Data from Bradbury and Sarott 1995, Table 2 - Region II, unless otherwise stated in "Comments."

(b) Thibault et al. 1990.

(c) Tungsten will exist predominantly as WO_4^{2-} in the Eh and pH range expected in the LAW vault and surround environment. The only conditions that it would be likely to exist as a strongly sorbing species is under strongly acidic and reducing conditions, conditions that are not expected under the modeling scenario. Because of the lack of data, the conservative assumption that no sorption occurs was made.

(d) Reardon 1992.

(e) Aluminum sorption to sediments is controlled largely by solubility and to a lesser extent by adsorption. To accommodate the model, a Kd (which describes sorption, rather than solubility) will be provided. To estimate a soil Kd, ground water Al concentration and soil Al concentration were used. Al groundwater concentrations very in E-area between 0.0178 to 0.216 mg/L (EGIS; http://egis1.srs.gov/Website/sgcp_wasteunits/viewer.htm). An average Al concentration of sediments on SRS is 8,060 mg/kg for 168 samples (Looney et al. 1990; Table 6.1). Dividing the soil Al concentration by the groundwater Al concentration, yields a ratio of >37,000 mL/g. A value 1/10 this, 3,700 mL/g, was enter as "soil," "gravel," and "clay" Kd values into the table to provide some level of conservatism. In concrete environments, several phases, some of which are gel phases, would be expected to control Al aqueous concentrations. Reardon (1992; Table 6) measured 0.22 mmol/L Al in contact with a concrete he described as being in a moderately advanced stage of aging (Phase II out of III stages) that contained 5,528 mmol/kg of CSH (Calcium-silica-hydrogen gel). Using these concentrations to make a Kd value yields, 25,000 L/kg; the conservative value entered in the table is 5,000 mL/g. Kaplan 2004 (WSRC-RP-2004-00267) offered different Al Kd values. These previous assumptions did not use field data, instead relied upon the use of Cm(III) as an analogue. The actual values do not vary greatly between those proposed here and those proposed previously, however, the justification has improved. Kaplan (2004) suggested Al Kd values for soil, grout, clay and concrete to be 40, 5000, 12000, and 5,000 mL/g, respectively. Finally, a more sophisticated handling of Al solubility in contaminant transport modeling on the SRS is presented by Powell et al. (2005; page 89). It includes a true aluminum solubility, as opposed to distribution coefficients, and adjusts it as a function of pH.

(f) For At, The original Soil Kd was 40 mL/g, Oxidizing Grouted Waste Kd = 5000 mL/g, Clay Kd = 12,000 mL/g, and Clay Kd = 5000 mL/g. There values are anywhere from 1 to 4 orders of magnitude greater than those listed above.

(g) Kaplan (2004)

(h) Khoury et al. 1985; 1992.

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