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L'ENERGIE ATOMIQUE
DU CANADA, LIMITEE

**SOIL NUCLIDE DISTRIBUTION COEFFICIENTS
AND THEIR STATISTICAL DISTRIBUTIONS**

**COEFFICIENTS DE REPARTITION DES NUCLIDES DANS LE SOL
ET LEUR REPARTITION STATISTIQUE**

**Marsha I. Sheppard, Donald I. Beals, Denis H. Thibault
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**Etablissement de recherches
nucléaires de Whiteshell**

**Pinawa, Manitoba R0E 1L0
December 1984 décembre**

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RÉSUMÉ

Les évaluations écologiques de l'évacuation des déchets de combustible nucléaire dans les formations de roche plutonique nécessitent une analyse de la migration des nuclides qui passent de l'enceinte d'évacuation à la biosphère. Pour l'analyse de la migration des nuclides à travers l'enceinte d'évacuation, les matériaux tampons et de remblayage, la roche plutonique et les morts-terrains consolidés et non consolidés, par l'entremise de l'eau souterraine, on se sert de modèles nécessitant des coefficients de répartition (K_d) pour décrire l'interaction des nuclides et des matériaux géologiques et artificiels. Ce rapport présente des coefficients de répartition dans le sol particuliers à certains éléments et leur répartition du point de vue statistique, à partir d'une étude bibliographique en détail. Les éléments radioactifs considérés furent les suivants: actinium, américium, bismuth, calcium, carbone, cérium, césium, iode, plomb, molybdène, neptunium, nickel, niobium, palladium, plutonium, polonium, protactinium, radium, samarium, sélénium, argent, strontium, technétium, terbium, thorium, étain, uranium et zirconium. Les éléments stables considérés furent les suivants: antimoine, bore, cadmium, tellure et zinc. Lorsque la disponibilité des données le permet, les coefficients de répartition et leur répartition sont indiqués pour les sols sablonneux, limoneux, argileux et organiques. L'utilisation de nos valeurs est recommandée pour les évaluations effectuées dans le cadre du programme canadien de gestion des déchets de combustible nucléaire.

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ABSTRACT

Environmental assessments of the disposal of nuclear fuel waste in plutonic rock formations require analysis of the migration of nuclides from the disposal vault to the biosphere. Analyses of nuclide migration via groundwater through the disposal vault, the buffer and backfill, the plutonic rock, and the consolidated and unconsolidated overburden use models requiring distribution coefficients (K_d) to describe the interaction of the nuclides with the geological and man-made materials. This report presents element-specific soil distribution coefficients and their statistical distributions, based on a detailed survey of the literature. Radioactive elements considered were actinium, americium, bismuth, calcium, carbon, cerium, cesium, iodine, lead, molybdenum, neptunium, nickel, niobium, palladium, plutonium, polonium, protactinium, radium, samarium, selenium, silver, strontium, technetium, terbium, thorium, tin, uranium and zirconium. Stable elements considered were antimony, boron, cadmium, tellurium and zinc. Where sufficient data were available, distribution coefficients and their distributions are given for sand, silt, clay and organic soils. Our values are recommended for use in assessments for the Canadian Nuclear Fuel Waste Management Program.

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1. INTRODUCTION

Canada has selected geological containment in a vault deep in plutonic rock in the Precambrian Shield as the most promising method for disposal of its nuclear fuel waste (Boulton, 1978). A stable granitic pluton will most likely be the host rock.

Assessment of the integrity of geological containment requires pathways analysis to determine the travel time from the vault to the biosphere of all the nuclides associated with the waste (Mehta, 1982). The travel time and the predicted nuclide concentrations in the biosphere will depend upon the interaction of the nuclides with their surroundings as they migrate from the vault. Traditionally, this interaction has been described using a distribution coefficient, K_d , for rock, unconsolidated regolith and soil (Wuschke et al., 1981). The objective of this report is to document these K_d values, separating them according to the major soil types found on the Precambrian Shield. These parameter values are required for the soil model in the assessment code used in the Canadian Nuclear Fuel Waste Management Program.

Further, since the assessment code is stochastic, the distributions of the K_d values are also needed. Preliminary work with the K_d values indicates that they are lognormally, as opposed to normally, distributed. The lognormal distribution parameters (\log_{10}) are reported here. These parameters directly represent the data presented where two or more values were found, and have not been adjusted toward conservatism for assessment purposes. Sections 4, 5 and 6 list the soil K_d values, and their appropriate distributions, for the actinides, the radionuclides produced from nuclear fission and the stable nuclides, respectively, that are expected to be present in 100-year cooled nuclear fuel (Mehta, 1982). A reference list is included for each nuclide.

2. DISTRIBUTION COEFFICIENT, K_d

The processes of solute migration pertinent to radionuclide migration in soil and unconsolidated geological materials have been discussed and reviewed extensively (Wheeler, 1976; Onishi et al., 1981; Miller, 1983; Gillham and Cherry, 1979; and Arnold et al., 1982). Many computer models have been developed to predict nuclide migration through soil (Murali and Aylmore, 1981; Yeh and Ward, 1981; Oster, 1982; Miller, 1983; Wong et al., 1983; van Genuchten, 1978; Duguid and Reeves, 1976; and Sheppard, 1981). These models vary in their complexity and purpose. The simplest model of the solute transport process, expressed in one-dimensional form, is

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - \bar{V} \frac{\partial C}{\partial x} \quad (1)$$

where C is the solute concentration in solution, i.e., mass of solute per unit volume of soil ($\text{g}\cdot\text{cm}^{-3}$),
 t is time (s),
 D is the dispersion coefficient ($\text{cm}^2\cdot\text{s}^{-1}$),
 x is the space coordinate (cm), and
 \bar{V} is the average linear pore-water velocity ($\text{cm}\cdot\text{s}^{-1}$).

Since Equation (1) does not account for the interaction of the solute and the solid phase, the distribution coefficient, K_d , has been introduced to describe this interaction. The distribution coefficient is defined as the concentration of solute in the adsorbed phase (mass of solute per unit mass of soil) divided by the concentration of solute in the solution phase (mass of solute per unit volume of soil pore water). The units of K_d are usually mL/g. The K_d value for each nuclide represents the partitioning of the solute between the solid and solution phases and is applicable to equilibrium reactions, such as ion exchange.

Typical radionuclide interactions with soil include other geochemical processes, such as precipitation, coprecipitation, hydrous metal oxide

complexation, organic matter complexation, colloid formation, and microbial effects. Empirically determined K_d values may or may not include these processes.

Ion exchange is one of the most common mechanisms of radionuclide adsorption on geological materials (Ames and Rai, 1978). Thus, the K_d value depends upon several factors, including the cation exchange capacity (CEC), and the species and concentration of both the ion being exchanged and the competing ions. If the nuclide is present in smaller concentrations than the competing ions, then the K_d value will be independent of the concentration of the nuclide, and it will be constant if all other factors remain constant (Johnston and Gillham, 1980).

To incorporate the K_d concept into the solute transport process described by Equation (1), the dispersion coefficient (D) and the pore-water velocity coefficient (\bar{V}) become the effective dispersion coefficient (D') and velocity coefficient (\bar{V}'), respectively, where

$$\begin{aligned} D' &= \frac{D}{R} \\ \bar{V}' &= \frac{\bar{V}}{R} \end{aligned} \quad (2)$$

and R is the retardation factor, defined as

$$R = 1 + \frac{\rho_b}{n} K_d \quad (3)$$

where ρ_b is the bulk density of the soil (g/cm³)
 n is the porosity (cm³/cm³), and
 K_d is the distribution coefficient (mL/g).

The K_d concept is restricted to equilibrium reactions in which the concentrations in the solution and solid phases are related. K_d was initially defined by Mayer and Tompkins (1947) as

$$K_d = \left(\frac{C_o}{C} - 1 \right) \frac{V}{M} \quad (4)$$

where C_0 is the tracer concentration in the solution before adding the sorbent,

C is the tracer concentration in the liquid phase of a sorbent-water suspension,

V is the volume of liquid, and

M is the mass of solid.

Despite the fact that the K_d concept strictly applies only to simple cation-exchange, K_d values are reported that describe more complex reactions. This is in response to the need for input to simple migration models. This report does not review the soil chemistry of the nuclides considered; this has been done adequately elsewhere (Jenson, 1980; Johnston and Gillham, 1980; Allard et al., 1977; Friedman, 1976, Swedish Nuclear Fuel Supply Co. Ltd., 1983; Ames and Rai, 1978). The report does list all of the K_d values by predominant soil type for the Precambrian Shield (sand, silt, clay and organic (Beals, 1984)) and includes other pertinent information found in the literature. Table 1 lists K_d distribution parameter estimates for some nuclides combining all soils, as reported by Baes and Sharp (1981).

TABLE 1
ESTIMATES OF DISTRIBUTIONS OF K_d * VALUES

Nuclide	μ^{**}	σ^{***}	$\exp(\mu)^+$ (mL/g)	K_d Range (mL/g)
Am	2.9	1.3	810	1.0 to 47 000
Ce	3.0	0.6	1100	58 to 6000
Cs	3.0	0.8	1100	10 to 52 000
Np	1.0	1.0	11	0.16 to 929
Pb	2.0	0.7	99	4.5 to 7600
Po	2.7	0.3	540	200 to 1100
Pu	3.3	1.0	1800	11 to 300 000
Sr	1.4	0.9	27	0.37 to 400
Tc	-1.5	0.5	0.03	0.003 to 0.28
Th	4.8	0.6	60 000	2000 to 510 000
U	1.6	0.6	45	11 to 4400

* From Baes and Sharp (1981)

** Mean of the logarithm (to base 10) of K_d

*** Standard deviation of the logarithm (to base 10) of K_d

+ Median value of K_d with a 0.5 cumulative probability

3. DISTRIBUTION COEFFICIENTS FOR THE ACTINIDES

3.1 ACTINIUM

Nothing was found in the literature on K_d values or the soil chemistry of actinium. We recommend using the K_d values for americium because of their chemical similarity.

3.2 AMERICIUM

Americium has been studied extensively because of weapons testing in the 1950s. The summary on americium geochemistry presented by Johnston and Gilham (1980) indicates that

- (1) the most stable form of americium in aqueous solutions is Am^{3+} ;
- (2) the soil sorption of americium is correlated to cation exchange capacity, clay content, and concentration and type of the competing ions in solution, indicating that the principal retardation mechanism is ion exchange;
- (3) at high K_d values, americium adsorption is sensitive to the concentration of americium in solution.

Table 2 lists the K_d values reported by various investigators and includes soil information (texture, pH, competing ions, etc.) pertinent to the sorption data. The recommended K_d value means, standard deviations, ranges and distribution parameters by soil type for americium, based on Table 2, are given in Table 3.

TABLE 2

K_d VALUES FOR AMBITION: LITERATURE SURVEY SUMMARY

Soil Type	Y (%)	Z (%)	Z (%)	Z (%)	pH	P ₁ (V)	OC (mg/100 g)	Z Free Iron (mM)	Competing Cation	K _d (mL/g)	Soil Location or Description	Reference
Silt	11.0	14.5	1.0	2.4	-	5.1 (4.39)	-	1.65	-	9.83x10 ¹	Malbis (Louisiana)	Mahita et al., 1979
	11.0	14.5	1.0	2.4	-	5.3 (5.71)	-	1.65	-	8.03x10 ¹	Malbis (Louisiana)	Mahita et al., 1979
	11.0	14.5	1.0	5.7	-	5.0 (4.58)	-	1.52	-	1.54x10 ¹	Lynn (Maine)	Mahita et al., 1979
	11.0	14.5	1.0	5.7	-	5.0 (5.17)	-	1.52	-	1.82x10 ¹	Lynn (Maine)	Mahita et al., 1979
	11.0	14.5	1.0	8.4	-	6.0 (5.71)	-	5.29	-	2.18x10 ¹	Alkem (California)	Mahita et al., 1979
	11.0	14.5	1.0	8.4	-	6.0 (6.72)	-	5.29	-	1.06x10 ¹	Alkem (California)	Mahita et al., 1979
	11.0	14.5	1.0	-	-	7-8	-	-	Na (0.02 sat. solution)	4x10 ²	(Netherlands)	Baars & Verkerke, 1977
	21.2	2.8	0.43	-	8.1	-	5.94	-	-	7.14x10 ²	(Richland, Washington)	Ames & Rai, 1978
	9.12	7.8	1.0	1.19	-	4.0	-	2.01	-	4.76x10 ²	Riquay (Barnwell, SC) 0-5 cm	Ames & Rai, 1978
	9.16	5.4	3.0	0.99	-	6.7	-	1.79	-	4.17x10 ²	Riquay (Barnwell, SC) 5-15 cm	Ames & Rai, 1978
	9.45	1.6	3.8	0.21	-	5.2	-	0.89	-	2.49x10 ²	Riquay (Barnwell, SC) 15-50 cm	Ames & Rai, 1978
	65.2	27.0	5.8	0.45	-	8.1	-	6.14	-	1.25x10 ²	Hanford A	Ames & Rai, 1978
	81.6	12.6	3.8	0.17	-	8.4	-	4.95	-	8.33x10 ¹	Hanford B	Ames & Rai, 1978
	62.6	39.4	18.0	0.60	-	8.6	-	15.04	-	3.92x10 ¹	Idaho A	Ames & Rai, 1978
	60.5	19.4	20.2	0.18	-	8.4	-	10.64	-	4.35x10 ¹	Idaho B	Ames & Rai, 1978
Silt	83.4	8.8	7.8	0.16	-	8.4	-	6.38	-	3.7x10 ¹	Idaho C	Ames & Rai, 1978
	59.2	28.4	22.4	0.98	-	7.7	-	18.36	-	1.09x10 ¹	Idaho D	Ames & Rai, 1978
	44.0	29.0	36.0	2.4	0.4	5.7	0.41	20.9	-	2.5x10 ² ± 210 ¹	Colorado A (Rocky Flats)	Glover et al., 1976
	66.0	14.0	22.0	3.4	0.3	5.6	0.52	17.5	-	6.0x10 ² ± 24 ¹	Colorado B (Sugar Loaf)	Glover et al., 1976
	44.0	24.0	32.0	0.2	7.0	8.3	0.41	13.8	-	3.0x10 ² ± 10 ¹	Idaho B	Glover et al., 1976
	66.0	11.0 ¹	23.0	0.3	5.2	8.0	0.47	8.2	-	8.2x10 ² ± 43 ¹	Idaho C	Glover et al., 1976
	34.0	32.0	30.0	0.1	0.0	7.5	0.45	17.5	-	1.0x10 ¹ ± 1.5x10 ¹	Idaho D	Glover et al., 1976
	74.0	12.0	14.0	0.3	0.6	8.0	0.43	6.4	-	1.2x10 ² ± 7 ¹	Washington A (Hanford)	Glover et al., 1976
	74.0	12.0	14.0	0.1	0.0	8.2	0.44	5.8	-	2.3x10 ² ± 5 ¹	Washington B (Hanford)	Glover et al., 1976
	74.0	2.0	20.0	0.7	0.2	5.4	0.54	2.9	-	8.2x10 ¹ ± 1 ¹	S. Carolina (Barnwell)	Glover et al., 1976
	48.0	34.0 ¹	18.0	0.7	0.2	6.4	0.49	7.0	-	4.0x10 ² ± 11 ¹	New Mexico (Los Alamos)	Glover et al., 1976
	42.0	9.0	9.0	0.6	0.7	4.8	0.57	3.8	-	1.9x10 ² ± 20 ¹	Arkansas B	Glover et al., 1976
	silt	clay	loam	2.8	5.9 (5.41)	-	20	1.29	-	2.98x10 ¹	Sharpsburg (Iowa)	Mahita et al., 1979
	silt	clay	loam	2.8	5.9 (6.56)	-	20	1.29	-	1.78x10 ¹	Sharpsburg (Iowa)	Mahita et al., 1979
Clay	16.0	16.0	16.0	2.5	6.7 (6.12)	-	25	2.41	-	7.38x10 ¹	Yolo (California)	Mahita et al., 1979
	16.0	16.0	16.0	2.5	6.7 (6.98)	-	25	2.41	-	2.02x10 ¹	Yolo (California)	Mahita et al., 1979
	16.0	50.0	14.0	0.4	1.72	7.8	0.44	15.5	-	5.9x10 ¹ ± 230 ¹	Idaho A	Glover et al., 1976
	9.0	54.0	37.0	2.3	0.6	2.3	0.57	16.2	-	1.8x10 ¹	Arkansas C	Glover et al., 1976
	31.0	53.0	16.0	3.6	0.7	3.6	0.56	17.4	-	1.6x10 ¹ ± 190 ¹	Illinois	Glover et al., 1976
	clay	clay	clay	0.6	7.8 (7.12)	-	30	1.20	-	3.56x10 ¹	Holtville	Mahita et al., 1979
	clay	clay	clay	0.6	7.8 (8.04)	-	30	1.20	-	4.72x10 ¹	Holtville	Mahita et al., 1979
	clay	clay	clay	-	7-8	-	-	-	Na (0.02 sat. solution)	5x10 ¹	(Netherlands)	Baars & Verkerke, 1977
	5.0	31.0	64.0	0.7	2.4	7.9	0.42	29.6	-	5.2x10 ¹ ± 970 ¹	Colorado C (Rocky Flats)	Glover et al., 1976
	12.0	32.0	56.0	1.0	0.0	4.8	0.49	20.3	-	2.5x10 ¹ ± 470 ¹	Tennessee (Oak Ridge)	Glover et al., 1976
	15.0	32.0	54.0	2.7	0.0	5.4	0.45	16.0	-	9.2x10 ¹ ± 79 ¹	New York (West Valley)	Glover et al., 1976
	17.0	34.0	54.0	3.2	0.9	6.2	0.57	34.4	-	2.9x10 ¹ ± 1800 ¹	Arkansas A	Glover et al., 1976
	clay	clay	clay	-	-	2.7	-	-	0.64 m/L NaCl	25.1	--	Brickman, 1980
	clay	clay	clay	-	-	0.9	-	-	0.64 m/L NaCl	4.0x10 ¹	--	Brickman, 1980
Organic	organic	organic	40.8	-	7.2 (7.14)	-	60	1.57	-	7.26x10 ¹	Egbert	Mahita et al., 1979
	organic	organic	40.9	-	7.2 (7.54)	-	60	1.57	-	5.32x10 ¹	Egbert	Mahita et al., 1979

* When the value is too small, it is the 10¹ of the extract.

** The distribution coefficient values for ambition were calculated and within an competing ions over two orders of magnitude are reported in Baars et al., 1975.

*** Values determined with initial ambition concentration of 10⁻¹⁰ mol/L.

**** Sand, silt and clay percentages exceed 100% in original report.

TABLE 3
K_d FOR AMERICIUM

Soil Type	\bar{K}_d^* (mL/g)	S.D. ⁺	n	K _d Range (mL/g)	Lognormal Distribution [‡] μ σ
Sand	6.146×10^3	1.1159×10^4	27	82 to 4.35×10^4	3.105 0.8172
Silt	1.4351×10^4	1.1282×10^4	7	1.6×10^3 to 2.98×10^4	3.946 0.5382
Clay	6.0501×10^4	1.29×10^5	9	25.1 to 4.0×10^5	3.832 1.23
Organic	6.398×10^3	1.228×10^3	2	5.529×10^3 to 7.266×10^3	3.802 0.0839

* \bar{K}_d = mean of K_d values

+ S.D. = standard deviation of K_d values

‡ Base 10 logarithms here and in all subsequent tables

Baes and Sharp (1981) suggested a mean value of 2.9 for the $\log_{10} K_d$ for americium, combining all soil types, and a corresponding standard deviation of 1.3 (see Table 1). Allard et al. (1977) reported K_d values for clay/mud of 2×10^2 to 1.6×10^4 mL/g and for granite of 5.0×10^3 to 1.6×10^4 mL/g. Vandergraaf (1982) recommended a range of 1×10^3 to 2×10^4 mL/g for granite. The values for granite should be similar to those for coarse-textured soil (sand).

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3.3 BISMUTH

Nothing was found in the literature on soil K_d values for bismuth, but bismuth should behave similarly to polonium because of their proximity in the Periodic Table.

3.4 LEAD

Lead is a heavy-metal cation of general environmental concern in most industrial areas. Consequently, considerable information exists about its environmental behaviour (Gerritse et al., 1982; Wolf et al., 1977; Soldatini et al., 1976; Abd-Elfattah and Wada, 1981). Unfortunately, not much K_d information is available (see Table 4).

The recommended K_d value means, standard deviations, ranges and distribution parameters for lead by soil type, based on Table 4, are given in Table 5.

* Unpublished, unpublished report available from SNO. Atomic Energy of

TABLE 4

K_d VALUES FOR LEAD : LITERATURE SURVEY SUMMARY

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCl ₂	pH Saturated Paste	E _c (V)	CEC (cmq/100 g)	% Free Iron Oxides	Competing Cation	K _d (ml/g)	Soil Location or Description	Reference
Sandy	-	-	0	3.5	-	4.5-5.0	-	22	-	[Ca ²⁺] = 0-0.015 mol/L	2.8x10 ²	Soil C	Gerritse et al., 1982
	-	-	0	3.5	-	4.5-5.0	-	22	-	[Ca ²⁺] = 0-0.015 mol/L	1.3x10 ³	Soil C	Gerritse et al., 1982
	-	-	20	2.5	-	7.5-8.0	-	16	-	[Ca ²⁺] = 0-0.015 mol/L	3.5x10 ³	Soil D	Gerritse et al., 1982
Organic	unpolluted organic soil			90	-	4.5	-	-	-	-	2.32x10 ⁶	Soil A	Gerritse et al., 1982
	unpolluted peat			990	-	4-5	-	-	-	[Ca ²⁺] = 0-0.015 mol/L	1.8x10 ²	Peat A	Gerritse et al., 1982
	unpolluted peat			990	-	4-5	-	-	-	[Ca ²⁺] = 0-0.015 mol/L	6.3x10 ⁴	Peat A	Gerritse et al., 1982
	polluted peat			990	-	6.2	-	-	-	-	2.34x10 ⁶	Soil B	Gerritse et al., 1982
	sphagnum peat			-	-	4-5	-	-	-	-	8x10 ⁴	-	Wolff et al., 1977
	sphagnum peat			-	-	4-5	-	-	-	0.025 meq Ca ²⁺ /ml	2x10 ²	-	Wolff et al., 1977

TABLE 5

K_d FOR LEAD

Soil Type	K _d (mL/g)	S.D.	n	K _d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	1693	1646	3	280 to 3500	3.035	0.5527
Organic	2.7845x10 ⁴	2.6024x10 ⁴	4	180 to 6.3x10 ⁴	3.954	1.150

Baes and Sharp (1981) suggested a mean value of 2.0 for the log₁₀K_d for lead, with a corresponding standard deviation of 0.7 (see Table 1). Lead should behave similarly to polonium and K_d values for polonium can be found in Section 3.6.

Lead References

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3.5 NEPTUNIUM

The summary on neptunium geochemistry presented by Johnston and Gillham (1980) indicates that

- (1) neptunium should exist as Np^{5+} in the form of NpO_2^+ in an oxidizing soil environment; however, it is not evident whether NpO_2^+ is also the dominant species under reducing conditions;
- (2) neptunium colloids have been reported in some soil-solution experiments (Sheppard et al., 1976) and absent in others (Routson et al., 1977).

The recommended K_d value means, standard deviations, ranges and distribution parameters for neptunium by soil type, based on Table 6, are given in Table 7.

TABLE 7

K_d FOR NEPTUNIUM

Soil Type	K_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	37.6	94.57	17	0.16 to 390	0.6782	0.9728
Silt	47.41	35.88	6	1.27 to 95	1.426	0.6925
Clay	1327.	1529.	4	41 to 3200	2.619	0.9222
Organic	857.5	101.1	2	786 to 929	2.932	0.0513

Baes and Sharp (1981) suggested a mean value of 1.0 for the $\log_{10}K_d$ for neptunium, with a corresponding standard deviation of 1.0 (see Table 1). Allard et al. (1977) reported K_d ranges of 10 to 16 mL/g for clay/mud and 25 to 50 mL/g for granites. Vandergraaf (1982) recommended a K_d of 40 to 100 mL/g for granites.

Neptunium References

Allard, B., H. Kipatsi and J. Rydberg. 1977. Sorption of long-lived radionuclides in clay and rock. Part 1. Determinations of Distribution Coefficients. KBS Technical Report 55.

TABLE 6

K_d VALUES FOR NEPTUNIUM : LITERATURE SURVEY SUMMARY

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCO ₃	pH Saturated Paste	E _p (%)	CFC (meq/100 g)	% Free Iron Oxides	Competing Cation	K _d (ml/g)	Soil Location or Description	Reference
Sand	fine sandy loam			2.4	-	5.3 (4.59)	-	15	1.65	-	3	Malbis (Louisiana)	Nishita et al., 1979
	fine sandy loam			2.4	-	5.3 (5.57)	-	15	1.65	-	18	Malbis (Louisiana)	Nishita et al., 1979
	fine sandy loam			5.7	-	5.0 (4.42)	-	15	1.52	-	3	Lyman (Maine)	Nishita et al., 1979
	fine sandy loam			5.7	-	5.0 (6.06)	-	15	1.52	-	32	Lyman (Maine)	Nishita et al., 1979
	light loam			8.4	-	6.0 (5.56)	-	15	5.29	-	26	Aiken (California)	Nishita et al., 1979
	light loam			8.4	-	6.0 (5.57)	-	15	5.29	-	108	Aiken (California)	Nishita et al., 1979
	sand			-	-	2.5 - 3.1	-	-	-	0.002 mol/L Ca	2.37	Burbank (Washington)	Routson et al., 1977
	sand			-	-	2.5 - 3.1	-	-	-	0.2 mol/L Ca	0.36	Burbank (Washington)	Routson et al., 1977
	sand			-	-	2.5 - 3.1	-	-	-	0.015 mol/L Na	3.9	Burbank (Washington)	Routson et al., 1977
	sand			-	-	2.5 - 3.1	-	-	-	3.0 mol/L Na	3.2	Burbank (Washington)	Routson et al., 1977
	sandy clay			-	-	2.5 - 3.1	-	-	-	0.002 mol/L Ca	0.25	South Carolina	Routson et al., 1977
	sandy clay			-	-	2.5 - 3.1	-	-	-	0.2 mol/L Ca	0.16	South Carolina	Routson et al., 1977
	sandy clay			-	-	2.5 - 3.1	-	-	-	0.015 mol/L Na	0.7	South Carolina	Routson et al., 1977
	sandy clay			-	-	2.5 - 3.1	-	-	-	3.0 mol/L Na	0.4	South Carolina	Routson et al., 1977
	75.0 21.2 3.8			0.43	-	8.1	-	5.94	-	-	15.4	Burbank (Richland, Washington)	Ames & Rai, 1978
	96.6 1.6 1.9			0.21	-	5.2	-	0.69	-	-	32.4	Puquay (5-50 cm)	Ames & Rai, 1978
Silt	sand			0.39	-	8.1	0 sat.	-	-	-	390 ± 16	N.E. Irish Sea Sediment	Fowler & Aston, 1982
	silty clay loam			2.8	-	5.9 (5.83)	-	20	1.29	-	35	Sharpsburg (Iowa)	Nishita et al., 1979
	silty clay loam			2.8	-	5.9 (5.85)	-	20	1.29	-	95	Sharpsburg (Iowa)	Nishita et al., 1979
	loam			2.5	-	6.7 (6.13)	-	25	2.41	-	52	Yolo (California)	Nishita et al., 1979
	loam			2.5	-	6.7 (6.83)	-	25	2.41	-	81	Yolo (California)	Nishita et al., 1979
	12.6 65.8 21.6			3.61	-	5.3	-	16.88	-	-	1.27	Muscovine	Ames & Rai, 1978
Clay	32.0 59.0 12.0			0.84	-	6.5	-	10.76	-	-	20.2	Ritaville	Ames & Rai, 1978
	clay			0.6	-	7.8 (7.29)	-	30	1.20	-	41	Holtville	Nishita et al., 1979
	clay			0.6	-	7.8 (8.28)	-	30	1.20	-	117	Holtville	Nishita et al., 1979
	clay			0.86	-	8.1	0 sat.	-	-	-	1950 ± 310	NW Mediterranean sea sediment	Fowler & Aston, 1982
	clay			0.29	-	8.1	0 sat.	-	-	-	3200 ± 890	NE Atlantic sea sediment	Fowler & Aston, 1982
Organic	organic			40.8	-	7.2 (6.24)	-	60	1.57	-	786	Egbert	Nishita et al., 1979
	organic			40.8	-	7.2 (7.25)	-	60	1.57	-	929	Egbert	Nishita et al., 1979

* When value is bracketed it is extract pH.

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3.6 POLONIUM

Polonium information in the literature is extremely scarce. Bismuth should behave similarly to polonium, and also lead and polonium should behave similarly. All of the K_d information on polonium reported here (see Table 8) comes from one research program (Hansen, 1970; Hansen and Watters, 1971). The chemical form of natural polonium in soils, resulting from the decay of radium, may be similar to that of selenium (Hansen, 1970). Tellurium is also a member of Group VIA of the Periodic Table and may behave similarly to polonium. Polonium compounds with +2 and +4 oxidation states have been reported, with the preferred oxidation state being +4 (Hansen, 1970). Polonium in air is generally found as polonium dioxide (PoO_2)

* Unrestricted unpublished report, available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

TABLE 8

K_d VALUES FOR PLUTONIUM: LITERATURE SURVEY SUMMARY

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCl ₂ Sat. Sol.	HI	P _h	CFC (mg/100 g)	% Free Iron Oxides	Competing Cation	K _d (mL/g)	Soil Location or Description	Reference
41	42	32	26	0.1	-	7.6	-	22.8	-	-	120 ± 54	Munn silty clay loam (A1) (Colorado)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	203 ± 22	Munn silty clay loam (A2) (Colorado)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	310 ± 41	Munn silty clay loam (B ₁) (Colorado)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	766 ± 148	Munn silty clay loam (B ₂) (Colorado)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	1213 ± 186	Munn silty clay loam (B ₃) (Colorado)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	643 ± 85	Munn silty clay loam (C) (Colorado)	Hansen & Mattern, 1971
42	31	39	-	-	-	7.9	-	-	-	-	723 ± 83	Dinadale silty clay loam (C) (Iowa)	Hansen & Mattern, 1971
43	42	13	2.3	-	-	6.5	-	5.4	-	-	192 ± 26	Lapeer loam (Ap) (Wisconsin)	Hansen & Mattern, 1971
44	22	24	-	-	-	6.7	-	-	-	-	206 ± 11	Lapeer loam (B ₁) (Wisconsin)	Hansen & Mattern, 1971
45	27	21	-	-	-	5.5	-	-	-	-	508 ± 34	Lapeer loam (B ₂) (Wisconsin)	Hansen & Mattern, 1971
46	15	23	-	-	-	5.7	-	-	-	-	414 ± 42	Lapeer loam (B ₃) (Wisconsin)	Hansen & Mattern, 1971
47	14	10	-	-	-	7.8	-	-	-	-	275 ± 9	Lapeer loam (C) (Wisconsin)	Hansen & Mattern, 1971
48	0	5	-	-	-	5.9	-	3.0	-	-	26 ± 2	Adamsville (A ₁) (Florida)	Hansen & Mattern, 1971
49	6	10	-	-	-	5.4	-	2.6	-	-	32 ± 3	Elarton (A ₁) (Florida)	Hansen & Mattern, 1971
50	2	3	-	-	-	5.5	-	1.8	-	-	25 ± 2	Lakeland (A ₁) (Florida)	Hansen & Mattern, 1971
51	1	2	-	-	-	5.5	-	1.5	-	-	17 ± 1	Leon (A ₁) (Florida)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	15 ± 0.6	Leon (A ₂) (Florida)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	55 ± 17	Leon (B ₁) (Florida)	Hansen & Mattern, 1971
-	-	-	-	-	-	-	-	-	-	-	77 ± 29	Leon (C) (Florida)	Hansen & Mattern, 1971
56	2	2	-	-	-	5.6	-	4.6	-	-	17 ± 1	Muskie (A ₁) (Florida)	Hansen & Mattern, 1971
57	30	13	-	-	-	5.5	-	-	-	-	13 ± 2	Darling gravelly sandy loam (B ₂) (Colorado)	Hansen & Mattern, 1971
74	17	8	-	-	-	5.7	-	-	-	-	30 ± 7	Darling gravelly sandy loam (B ₃) (Colorado)	Hansen & Mattern, 1971
80	16	4	-	-	-	6.0	-	-	-	-	66 ± 9	Darling gravelly sandy loam (C) (Colorado)	Hansen & Mattern, 1971
74	22	4	-	-	-	6.0	-	-	-	-	75 ± 8	Darling gravelly sandy loam (C ₂) (Colorado)	Hansen & Mattern, 1971
49	33	12	4.0	-	-	6.6	-	16.8	-	-	254 ± 22	Copebic sandy loam (A ₂) (Wisconsin)	Hansen & Mattern, 1971
57	31	13	-	-	-	5.5	-	-	-	-	371 ± 36	Copebic sandy loam (B ₁) (Wisconsin)	Hansen & Mattern, 1971
47	29	24	-	-	-	6.8	-	-	-	-	137 ± 5	Copebic sandy loam (B ₂) (Wisconsin)	Hansen & Mattern, 1971

(continued...)

TABLE 8 (Continued)

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCl ₂ Saturated Paste	pH	P _h (V)	CFC (mg/100 g)	% Free Iron Oxides	Coexisting Oxide	K _d (mL/g)	Soil Location or Description	Reference
Sandy	94	27	5	-	-	5.9	-	-	-	-	242 ± 25	Copple sandy loam (B ₁) (Wisconsin)	Hansen & Matterns, 1971
	96	46	10	2.4	-	6.8	-	5.1	-	-	227 ± 20	Onaway fine sandy loam (Ap) (Wisconsin)	Hansen & Matterns, 1971
	96	43	11	-	-	6.9	-	-	-	-	412 ± 150	Onaway fine sandy loam (B ₁ rh) (Wisconsin)	Hansen & Matterns, 1971
	67	23	13	-	-	8.2	-	-	-	-	248 ± 1200	Onaway fine sandy loam (C ₁) (Wisconsin)	Hansen & Matterns, 1971
	67	20	13	-	-	8.4	-	-	-	-	7020 ± 3600	Onaway fine sandy loam (C ₂) (Wisconsin)	Hansen & Matterns, 1971
	92	5	13	-	-	6.3	-	2.7	-	-	76 ± 11	Andre (A ₁) Alabama	Hansen & Matterns, 1971
	91	0	9	-	-	5.0	-	1.9	-	-	188 ± 15	Interdependence (A ₁) (Alabama)	Hansen & Matterns, 1971
	63	27	10	-	-	5.6	-	3.4	-	-	49 ± 3	Wichman (A ₁) (Alabama)	Hansen & Matterns, 1971
	11	68	21	1.9	-	5.8	-	25.2	-	-	1030 ± 49	Dinsdale silty clay loam (A) (Iowa)	Hansen & Matterns, 1971
	17	55	28	-	-	5.6	-	-	-	-	976 ± 127	Dinsdale silty clay loam (B) (Iowa)	Hansen & Matterns, 1971
Silty	3	73	24	4.5	-	5.5	-	28.4	-	-	1136 ± 118	Muscataine silty clay loam (A) (Iowa)	Hansen & Matterns, 1971
	10	61	29	-	-	5.9	-	-	-	-	968 ± 32	Muscataine silty clay loam (B) (Iowa)	Hansen & Matterns, 1971
	11	65	24	-	-	7.8	-	-	-	-	1830 ± 210	Muscataine silty clay loam (C) (Iowa)	Hansen & Matterns, 1971
	10	80	10	2.1	-	5.9	-	11.2	-	-	970 ± 160	Fayette silt loam (Ap) (Wisconsin)	Hansen & Matterns, 1971
	8	71	21	-	-	6.2	-	-	-	-	122 ± 3	Fayette silt loam (B ₁) (Wisconsin)	Hansen & Matterns, 1971
	5	66	29	-	-	6.1	-	-	-	-	92 ± 3	Fayette silt loam (B ₂₁) (Wisconsin)	Hansen & Matterns, 1971
	2	66	32	-	-	5.6	-	-	-	-	597 ± 55	Fayette silt loam (B ₂₂) (Wisconsin)	Hansen & Matterns, 1971
	5	65	30	-	-	5.3	-	-	-	-	80 ± 2	Fayette silt loam (B ₂₃) (Wisconsin)	Hansen & Matterns, 1971
	5	66	29	-	-	5.5	-	-	-	-	772 ± 29	Fayette silt loam (C ₁) (Wisconsin)	Hansen & Matterns, 1971
	33	-	15	3.8	-	5.1	-	28.9	-	-	24 ± 1	Dorling gravelly sandy loam (A ₁) (Colorado)	Hansen & Matterns, 1971
	27	55	18	-	-	5.5	-	16.4	-	-	405 ± 28	Onyria (A ₁) (Alabama)	Hansen & Matterns, 1971

* All error terms in this table are standard error of the mean (S.E.)

(Hansen, 1970). Hansen also reported that, for pH values of 1 to 5, 7 to 8 and 12 to 14, polonium exists mainly in dissolved forms, while for pH values of 6 to 7 and 10 to 11, most of the polonium exists as a colloid. Thus, in the acidic organic and acidic sandy soils of the Precambrian Shield, polonium may exist only in the dissolved form.

The recommended K_d value means, standard deviations, ranges and distribution parameters for polonium by soil type, based on Table 8, are given in Table 9.

TABLE 9
 K_d FOR POLONIUM

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	504.2	1215	35	13 to 7020	2.188	0.6574
Silt	692.5	535.7	13	24 to 1830	2.607	0.5789

Baes and Sharp (1981) suggested a mean value of 2.7 for the $\log_{10} K_d$ for polonium, with a corresponding standard deviation of 0.3 (see Table 1). The value for μ is higher, but the range of K_d values is narrower, than recommended for lead.

Polonium References

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3.7 PLUTONIUM

Plutonium, like americium, has been studied extensively because of weapons testing in the 1950s. The summary on plutonium geochemistry presented by Johnston and Gillham (1980) indicates that

- (1) Pu^{4+} is considered the most probable oxidation state in the environment because of reduction of Pu^{6+} to Pu^{4+} by organic materials; reduction of Pu^{4+} to Pu^{3+} could occur at pH < 6 under anaerobic conditions;
- (2) plutonium adsorption is a function of oxidation state (Pu^{6+} is adsorbed less than Pu^{4+}), organic matter content and solution pH;
- (3) K_d values for plutonium reported in the literature were often obtained without knowledge of the oxidation state, and caution must be used in interpreting results that use these K_d values.

Most K_d values in the literature apply to aerobic conditions. The recommended K_d value means, standard deviations, ranges and distribution parameters for plutonium by soil type, based on Table 10, are given in Table 11.

TABLE 11
 K_d FOR PLUTONIUM

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution μ σ
Sand	1.041×10^3	1.568×10^3	19	33 to 6.865×10^3	2.663 0.5964
Silt	1.3871×10^4	3.0836×10^4	8	230 to 9.0×10^4	3.474 0.7906
Clay	4.2842×10^4	6.8934×10^4	13	316 to 1.9×10^5	3.706 1.047
Organic	2.2962×10^4	2.8181×10^4	4	1.655×10^3 to 6.2×10^4	3.970 0.7469

Baes and Sharp (1981) suggested a mean value of 2.3 for the $\log_{10} K_d$ for plutonium, with a corresponding standard deviation of 1.0 (see

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- When the value is too small, it is pH of the end-point.
- NaOH titrated with HCl reaction, calculate K_a values using pH for this acid.
- pH value is related with initial plot vs concentration of 10^{-2} (data for 10^{-2} and 10^{-4} mol/L can be found in the reference).
- Graph, pH vs initial concentration, enabled pH in original report.

Table 1). Allard et al. (1977) reported that the plutonium K_d range for both granite and clay/mud is 6.3×10^1 to 1.6×10^2 mL/g. Vandergraaf (1982) recommended a K_d value range for plutonium of 2.8×10^2 to 2.0×10^3 mL/g for granite.

Plutonium References

- Allard, B., H. Kipetsi and J. Rydberg. 1977. Sorption of long-lived radionuclides in clay and rock. Part 1. Determination of distribution coefficients. KBS Technical Report 55.
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- Tamura, T. 1972. Sorption phenomena significant in radioactive waste disposal. In: Underground Waste Management and Environmental Implications. Amer. Assoc. Pet. Geol. Mem. 18, 317-330.
- Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radio-nuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.

3.8 PROTACTINIUM

Nothing on K_d values or soil chemistry for protactinium was found in the literature. We suggest that K_d values for thorium or uranium be used, or even some combination of the values for these elements, such as $\frac{1}{2}(\text{Th}+\text{U})$.

3.9 RADIUM

The interaction of radium with geological materials and soils, and the environmental behaviour of radium have been documented by Gillham et al. (1981b); Nathwanl and Phillips (1979), and Sheppard (1980), respectively. The K_d values for radium vary from 50 to 1000 mL/g (Gillham et al., 1981). Johnston and Gillham (1980) summarized the information relevant to K_d as follows:

- (1) Radium is present as Ra^{++} in the pH range 4 to 8, and does not readily form complex species.
- (2) Radium can be expected to coprecipitate with BaSO_4 , carbonates and ferric hydroxides.
- (3) Cation exchange is an important adsorption mechanism, since K_d values have been correlated to cation exchange capacity (CEC).

* Unrestricted, unpublished report, available from SDBO, Atomic Energy of Canada Limited Research Agency, Chalk River, Ontario K0J 1B0.

The recommended K_d value means, standard deviations, ranges and distribution parameters for radium by soil type, based on Table 12, are given in Table 13.

TABLE 13
 K_d FOR RADIUM

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	1.0435×10^4	2.0845×10^4	3	106 to 3.8×10^4	3.402	1.289
Silt	3.0×10^5	4.3566×10^5	4	2.0×10^4 to 9.5×10^5	5.10	0.6862
Clay	1.5637×10^4	1.7216×10^4	8	696 to 5.6×10^4	3.961	0.5522

The K_d values for strontium may be used as a guide because of the chemical similarity of radium and strontium. Baes and Sharp (1981) suggested a mean value of 1.4 for the $\log_{10} K_d$ for strontium, with a corresponding standard deviation of 0.9 (see Table 1). Allard et al. (1977) reported a K_d range for radium of 40 to 79 mL/g for clay/mud and 63 to 100 mL/g for granite. Vandergraaf (1982) recommended a K_d range of 5 to 5000 mL/g for granite. Since no data were found for organic soils, the radium K_d value for clay, or the strontium K_d value for organic soil, is recommended.

Radium References

- Allard, B., H. Kipatsi and J. Rydberg. 1977. Adsorption of long-lived radionuclides in clay and bedrock. Part 1. Determination of distribution coefficients. KBS Technical Report 55.
- Baes III, C.F. and R.D. Sharp. 1981. Predicting radionuclide leaching from root zone soil for assessment applications. Oak Ridge National Laboratory Report, CONF-810606-44.
- Cochran, J.K. and S. Krishnaswami. 1980. Radium, thorium, uranium and Pb in deep-sea sediments and sediment pore waters from the north equatorial Pacific. Amer. J. Sci. 280, 859-889.

TABLE 12

K_d VALUES FOR RADIUM: LITERATURE SURVEY SUMMARY

Soil Type	Ca ²⁺ (mM)	Mg ²⁺ (mM)	Na ⁺ (mM)	K ⁺ (mM)	pH	Fe ³⁺ (mM)	DOC (mg/100 g)	Z Free Iron Oxides	Gaseating Option	K _d (mL/g)	Soil Location or Description	Reference
Sand	93.0	5.0	2.0	0.01	40.8	7.8 (CaCl ₂)	-	1.19	-	Initial Ra conc. before soil contact 1.6x10 ⁻⁵ eq/L Ca ²⁺ 0.05 mol/L	Leamington (6) medium sand	Gillham et al., 1981b
	93.1	4.8	1.3	3.1	-	5.2	-	10.9	-	no Ca	St. Thomas	Nathani & Phillips, 1979
	93.2	4.4	1.3	3.1	-	5.2	-	10.9	-	no Ca	St. Thomas	Nathani & Phillips, 1979
Clay	35.0	34.0	29.0	0.41	33.5	9.5 (CaCl ₂)	-	8.32	-	Initial Ra conc. before soil contact 4x10 ⁻⁵ eq/L Ca ²⁺ 0.05 mol/L	WRE ² (2) clay loam	Gillham et al., 1981b
	43.7	47.9	45.4	16.2	-	5.4	-	34.7	-	no Ca	Wendover	Nathani & Phillips, 1979
	43.7	47.9	45.4	16.2	-	5.4	-	34.7	-	no Ca	Wendover	Nathani & Phillips, 1979
	43.7	48.0	7.4	1.0	-	4.3	-	10.4	-	Ca ²⁺ 0.05 mol/L	Orlansby	Nathani & Phillips, 1979
	43.7	48.9	7.4	1.0	-	4.3	-	10.4	-	no Ca	Orlansby	Nathani & Phillips, 1979
	31.7	34.7	31.3	0.31	5.2	7.8 (CaCl ₂)	-	31.48	-	Initial Ra conc. before soil contact 3.7x10 ⁻⁵ eq/L Na ⁺ 2M eq/L Ca ²⁺ 75 eq/L	Alberta clay loam	Gillham et al., 1981b
clay	-	-	-	-	7.55*	-	-	-	-	-	clay, mud	Allard et al., 1977
clay and loam	-	-	-	-	-	-	-	-	-	-	clay and loam (Pacific)	Cochran & Krishnaswami, 1980
clay and loam	-	-	-	-	-	-	-	-	-	-	clay and loam (Pacific)	Cochran & Krishnaswami, 1980
clay and loam	-	-	-	-	-	-	-	-	-	-	clay and loam (Pacific)	Cochran & Krishnaswami, 1980
clay and loam	-	-	-	-	-	-	-	-	-	-	clay and loam (Pacific)	Cochran & Krishnaswami, 1980
clay and loam	-	-	-	-	-	-	-	-	-	-	clay and loam (Pacific)	Cochran & Krishnaswami, 1980
clay and loam	-	-	-	-	-	-	-	-	-	-	clay and loam (Pacific)	Cochran & Krishnaswami, 1980

- * WRE = Western Research Establishment, Pinawa, Manitoba
 * Certain values reported and data based on description of deep sea clays.

- Gillham, R.W., H.D. Sharma, M.R. Reddy, E.L. Cooper and J.A. Cherry. 1981b. Barium and radium migration in unconsolidated Canadian geological materials. Atomic Energy Control Board Report, INFO-0048.
- Johnston, H.M. and R.W. Gillham. 1980. A review of selected radionuclide distribution coefficients of geologic materials. Atomic Energy of Canada Limited Technical Record, TR-90*.
- Nathwani, J.S. and C.R. Phillips. 1979. Adsorption of ^{226}Ra by soils in the presence of calcium (2^+) ions. Specific adsorption (II). *Chemosphere* 8, 293-299.
- Sheppard, M.I. 1980. The environmental behaviour of radium. Atomic Energy of Canada Limited Report, AECL-6796.
- Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radionuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.

3.10 THORIUM

Little information is available in the literature on thorium interactions in the environment; however, two brief reviews of thorium chemistry are available (Rancon, 1973; Sheppard, 1980). Johnston and Gillham (1980) summarized the information relevant to K_d as follows:

- (1) The primary thorium adsorption mechanism is ion exchange.
- (2) In non-calcareous soils, thorium adsorption is extremely sensitive to initial thorium solution concentrations. In organic materials, increased pH causes increased humic acid solubility and thorium complexation, resulting in lower K_d values. In calcareous soils, K_d values are high ($> 10 \text{ mL/g}$), regardless of pH or thorium concentration, because of the buffering capacity of the soil and the precipitation of $\text{Th}(\text{OH})_3$.
- (3) K_d values are generally high, ($> 10^3 \text{ mL/g}$), in dilute solutions, indicating limited thorium migration.

The recommended K_d value means, standard deviations, ranges and distribution parameters for thorium by soil type, based on Table 14, are given in Table 15.

TABLE 14

K_d VALUES FOR THORIUM : LITERATURE SURVEY SUMMARY

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCO ₃	pH Saturated Paste	E _h (V)	CTC (meq/100 g)	% Free Iron Oxides	Competing Cation	K _d (mL/g)	Soil Location or Description	Reference
Sed.	45 (S)	-	30	< 1	25 (C car-bonate)	7.0	-	-	-	Th Conc. (1 g/L)	1.5x10 ⁵	Cedarache sediment	Rançon, 1973
	50 (S)	-	50	0	0 (C car-bonate)	3.2	-	-	-	Th Conc. (1 g/L)	8	clay schist	Rançon, 1973
	40 (S)	-	60	0	0 (C car-bonate)	4.8	-	-	-	Th Conc. (0.1 g/L)	1x10 ⁵	clay schist	Rançon, 1973
Oreumite	5 (S)	-	12	60	23 (C car-bonate)	6.7	-	-	-	Th Conc. (1 g/L)	8x10 ⁴	river peat	Rançon, 1973
	5 (S)	-	12	60	23 (C car-bonate)	7.4	-	-	-	Th Conc. (0.1 g/L)	1.5x10 ⁴	river peat	Rançon, 1973
	5 (S)	-	12	60	23 (C car-bonate)	7.4	-	-	-	Th Conc. (0.1 g/L)	1.5x10 ⁴	river peat	Rançon, 1973

TABLE 15
K_d FOR THORIUM

Soil Type	K _d (mL/g)	S.D.	n	K _d Range (mL/g)	Lognormal Distribution	
					μ	σ
Clay	5.0x10 ⁴	7.0710x10 ⁴	2	8.0 to 1.0x10 ⁵	2.95	2.90
Organic	4.75x10 ⁴	4.5962x10 ⁴	2	1.5x10 ⁴ to 8.0x10 ⁴	4.54	0.5141

Baes and Sharp (1981) suggested a mean value of 4.8 for the log₁₀K_d for thorium, with a corresponding standard deviation of 0.6 (see Table 1). Allard et al. (1977) reported K_d ranges from 40 to 316 mL/g and 500 to 1260 mL/g for clay/mud and granite, respectively. Vandergraaf (1982) recommended a K_d value for thorium of 850 mL/g for granite.

Thorium References

- Allard, B., H. Kipatsi and J. Rydberg. 1977. Sorption of long-lived radionuclides in clay and rock. Part 1. Determination of Distribution Coefficients. KBS Technical Report 55.
- Baes III, C.F. and R.D. Sharp. 1981. Predicting radionuclide leaching from root zone soil for assessment applications. Oak Ridge National Laboratory Report, CONF-810606-44.
- Johnston, H.M. and R.W. Gillham. 1980. A review of selected radionuclide distribution coefficients of geologic materials. Atomic Energy of Canada Limited Technical Record, TR-90*.
- Rançon, D. 1973. The behaviour in underground environments of uranium and thorium discharged by the nuclear industry. In: Environmental Behaviour of Radionuclides Released in the Nuclear Industry. International Atomic Energy Agency Report, IAEA-SM-172/5.
- Sheppard, M.I. 1980. The environmental behaviour of uranium and thorium. Atomic Energy of Canada Limited Report, AECL-6795.
- Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radionuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.

* Unrestricted, unpublished report, available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

3.11 URANIUM

Several reviews of uranium chemistry exist (Harnsen and de Haan, 1980; Borovec, 1981; Sheppard, 1980), but few K_d values have been reported in the literature. The summary on uranium geochemistry presented by Johnston and Gillham (1980) indicates that

- (1) in oxidizing environments, U^{6+} compounds are stable and can precipitate, whereas U^{4+} is stable in a reducing environment and would precipitate as UO_2 ; thus the oxidation-reduction status is important;
- (2) soluble uranium (U^{6+}) can be adsorbed or reduced by organic matter; if U^{6+} is reduced to U^{4+} , precipitation can occur;
- (3) UO_2^{2+} can be adsorbed by clay minerals by cation exchange, but may also form complexes with anions such as carbonate or phosphate.

Borovec (1981) indicated that K_d values for uranium for clay minerals range from 50 to 1000 mL/g and for peat from 10^4 to 10^6 mL/g. The recommended K_d value means, standard deviations, ranges and distribution parameters for uranium by soil type, based on Table 16, are given in Table 17.

TABLE 17
 K_d FOR URANIUM

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	8.065	11.22	2	0.13 to 16.0	0.159	1.478
Clay	2.6349×10^5	4.5597×10^5	3	200 to 7.9×10^5	3.543	2.040

Baes and Sharp (1981) suggested a mean value of 1.6 for the $\log_{10} K_d$ for uranium, with a corresponding standard deviation of 0.6 (see

TABLE 16

K_d VALUES FOR URANIUM: LITERATURE SUMMARY

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCl ₂	pH	K _d	CFC	% Free	Competing Cation	K _d	Soil Location or Description	Reference
and (2.5%)	43	-	31	4.1	25 (2 cam-brate)	4.7	-	-	-	-	16	Cadizache sandstone	Rangon, 1973
clay	40	-	60	0	0 (2 cam-brate)	4.7	-	-	-	4.3 ug (D) ₂ ²⁺ /mL	0.13	-	Yamamoto et al., 1973
clay	40	-	60	0	0 (2 cam-brate)	4.7	-	-	-	4.3 ug (D) ₂ ²⁺ /mL	0.25	-	Yamamoto et al., 1973
clay	40	-	60	0	0 (2 cam-brate)	4.7	-	-	-	-	270	altered schist	Rangon, 1973
clay	40	-	60	0	0 (2 cam-brate)	4.7	-	-	-	0.66 mol/L NaCl	200	-	Erickson, 1980
clay	40	-	60	0	0 (2 cam-brate)	4.7	-	-	-	0.66 mol/L NaCl	7.9x10 ⁵	-	Erickson, 1980
clay	40	-	60	0	0 (2 cam-brate)	4.7	-	-	-	-	33	organic peat	Rangon, 1973

Table 1). Allard et al. (1977) reported K_d ranges from 2.5 to 20 mL/g for clay/mud and 4 to 13 mL/g for granites. Vandergraaf (1982) recommended a K_d range of 0.4 to 10 mL/g for granites.

Uranium References

- Allard, B., H. Kipatsi and J. Rydberg. 1977. Sorption of long-lived radionuclides in clay and rock. Part 1. Determination of Distribution Coefficients. KBS Technical Report 55.
- Baes III, C.F. and R.D. Sharp. 1981. Predicting radionuclide leaching from root zone soil for assessment applications. Oak Ridge National Laboratory Report, CONF-810606-44.
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- Harmsen K. and F.A.M. de Haan. 1980. Occurrence and behaviour of uranium and thorium in soil and water. Neth. J. Agric. Sci. 28, 40-62.
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- Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radionuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.
- Yamamoto, T., E. Yunoki, M. Yamakawa and M. Shimizu. 1973. Studies on environmental contamination by uranium. 3: The effects of carbonate ion on uranium adsorption to and desorption from soils. J. Radiat. Res. 14, 219.

* Unrestricted, unpublished report, available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

4. DISTRIBUTION COEFFICIENTS FOR FISSION PRODUCTS

4.1 CALCIUM

K_d values for calcium reported by Graham (1973) and Graham and Silva (1979) vary from 1×10^{-3} to 9.8 mL/g; however, there is some confusion about the units. Wong et al. (1983) reported K_d values of 1117 and 1900 mL/g for a sand and a muck soil, respectively. It is recommended that the K_d values for strontium be used for calcium (see Section 4.11).

Calcium References

- Graham, E.R. 1973. Selective distribution and labile pools of micronutrient elements as factors affecting plant uptake. *Soil Sci. Soc. Amer. Proc.* 37, 70-74.
- Graham, E.R. and C.G. Silva. 1979. Labile pools and distribution coefficients for soil calcium, magnesium, and potassium determined with exchange equilibria and radioisotopes. *Soil Science* 128, 17-22.
- Wong, K.V., S. Sengupta, D. Dasgupta, E.L. Daly, Jr., N. Nemerow and H.P. Gerrish. 1983. Heavy metal migration in soil-leachate systems. *Bio-cycle* 24, 30-33.

4.2 CARBON

Allard et al. (1981) studied the sorption of $H^{14}CO_3^-$ on some solids using the batch technique. The sorption of ^{14}C was generally low, but appeared to increase with increasing calcium content of the solid. Retardation factors of up to 3 (i.e., three times slower transport of ^{14}C than of water) were measured for calcite. Concrete will probably retain most of the ^{14}C , and a retardation factor >10 might be expected for a bentonite-quartz mixture ($K_d = 2.2 \times 10^{-6}$ mL/g). Due to the paucity of information, a conservative retardation factor of 1, or a K_d of 0 mL/g, is recommended.

Carbon Reference

Allard, B., B. Torstenfelt and K. Andersson. 1981. Sorption behaviour of ^{14}C in groundwater/rock and in groundwater/concrete environments. Report Prav 4.27.

4.3 CESIUM

The work of Gillham et al. (1981a) is the most extensive on K_d values for cesium for Canadian soils. Their study showed that for 15 Canadian soils, K_d values for cesium ranged from 1×10^2 to 2×10^4 mL/g, but there was no significant correlation between the K_d value and measured soil properties such as CEC, major cation concentration, clay mineral composition, organic matter content and pH. In more than half of the samples, however, the K_d values were related significantly to the natural exchangeable cesium content of the soil, and this must be accounted for in sorption studies.

The recommended K_d value means, standard deviations, ranges and distribution parameters for cesium, based on Table 18, are given in Table 19.

TABLE 19
 K_d FOR CESIUM

Soil Type	K_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	2163	3226	24	10 to 1.0×10^4	2.668	0.9332
Silt	1.1395×10^4	7899	20	650 to 3.0×10^4	3.912	0.4227
Clay	8379	1.3613×10^4	5	65 to 3.15×10^4	2.945	1.216

Baer and Sharp (1981) suggested a mean value of 3.0 for the $\log_{10} K_d$ for cesium, with a corresponding standard deviation of 0.8 (see Table 1). Allard et al. (1977) reported K_d ranges from 6 to 32 mL/g and 32

K. VALUES FOR COSTS : LITERATURE SURVEY SUMMARY

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* This list is not intended to be all-inclusive.

to 794 mL/g for clay/mud and granite, respectively. Vandergraaf (1982) recommended a K_d range of 40 to 1000 mL/g for granite.

Cesium References

- Aleksakhin, R.M. 1965. Radioactive contamination of soils and plants. USAEC Report AEC-tr-6631.
- Allard, B., H. Kipatsi and J. Rydberg. 1977. Sorption of long-lived radionuclides in clay and rock. Part 1. Determination of Distribution Coefficients. KBS Technical Report 55.
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- Gillham, R.W., L.E. Lindsay, W.D. Reynolds, T.J. Kewen, J.A. Cherry and M.R. Reddy. 1981a. Studies of cesium and strontium migration in unconsolidated Canadian geological materials. Atomic Energy Control Board Report, INFO-0049.
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- Rançon, D. 1972. Practical utilization of the distribution coefficient for the measurement of the radioactive contamination of minerals in rocks, soil and subterranean water. Cadarache Nuclear CEA Research Center, AEL-trans-931, Report-R-4274.
- Rhodes, D.W. 1957. The effect of pH on the uptake of radioactive isotopes from solution by a soil. Soil Sci. Amer. Proc. 21, 389-392.
- Rogowski, A.S. and T. Taira. 1965. Movement of ^{137}Cs by runoff, erosion and infiltration on the alluvial Columbia silt loam. Health Phys. 11, 1333-1340.
- Routson, R.C. 1973. A review of studies on soil-waste relationships on the Hanford Reservation from 1944 to 1967. BNWL-1464, UC-70.

- Serne, R.J., D. Rai and S.J. Phillips. 1978. Monitoring and physical characterization of unsaturated zone transport: Laboratory analysis. In: Nuclear Waste Management Quarterly Progress Report Oct.-Dec. 1977, Pacific Northwest Laboratory Report, PNL-2377-4, UC-70.
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- Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radionuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.
- Zelazny, L.W., D.C. Martens, A.M. El-Prince and C.I. Rich. 1978. Effect of temperature and hydroxy-aluminum interlayers on cesium selectivity and fixation in river suspensions and soils. ORO-4851-2.

4.4 IODINE

The most extensive study of iodine adsorption on soil was that of Wildung et al. (1974). Johnston and Gillham (1980) have summarized the known soil chemistry of iodine as follows:

- (1) the most stable form of iodine in both oxidizing and reducing environments is iodide, I^- . Because the predominant iodine species is an anion, ion exchange would not be important in soil adsorption, particularly at neutral or high pH values.
- (2) organic matter appears to be a significant factor in iodine adsorption.
- (3) K_d values for iodine range from 0.1 to 50 mL/g, depending on the form of the iodine and the pH of the solution. The maximum K_d value would be obtained for I^- at a pH of 4 to 6.

* Unrestricted, unpublished report, available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

Since no K_d values for specific soils were found in the literature, the multiple-regression equations (based on iodide (I^-) and methyl iodide (CH_3I) interactions with 22 soils) reported by Wildung et al. (1974) were applied to soils already described in this report. The K_d values were calculated only for soils whose properties were within the range of the soils used to generate the multiple-regression equations. The equations for iodide K_d and methyl iodide K_d differ, and are

$$K_{d \text{ iodide}} = 0.33 X_1 + 0.09 X_3 \quad (5)$$

$$K_{d \text{ methyl iodide}} = 0.027 X_2 + 0.10 X_3 \quad (6)$$

where X_1 is the silt content (range for equation development is 17.6 to 58.0%),
 X_2 is the clay content (range for equation development is 5.8 to 46.6%), and
 X_3 is the organic carbon content (range for equation development is 0.23 to 28.8%).

The recommended K_d value means, standard deviations, ranges and distribution parameters by soil type (using Equation (6) since it will predict the lowest K_d values because methyl iodide is more highly mobile), based on Table 20, are given in Table 21.

TABLE 21
 K_d FOR IODINE

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution μ	σ
Sand	0.5514	0.3595	7	0.2 to 1.210	-0.3404	0.2929
Silt	0.9145	0.3201	11	0.18 to 1.50	-7.99x10 ⁻²	0.2351
Clay	1.293	0.3697	4	1.03 to 1.83	9.952x10 ⁻²	0.1146

TABLE 20

K_d VALUES FOR IODINE : LITERATURE SURVEY SUMMARY

Soil Type	% Silt	% Clay	% Organic	Calculated K _d (mL/g)	Soil Location or Description	Reference
<u>Sand</u>	31	7	0.38	0.23	Soil #11 (WNRE)	Gillham et al., 1981a
	19	9	0.33	0.28	Soil #12 (BNPD)	Gillham et al., 1981a
	22	18	2.05	0.69	Soil #16 (Alberta)	Gillham et al., 1981a
	29	5.8	0.45	0.20	Hanford A	Ames & Rai, 1978
	19.4	18.0	0.60	0.55	Idaho A	Ames & Rai, 1978
	28.4	22.4	0.98	0.70	Idaho D	Glover et al., 1976
	20.0	16.0	2.4	1.21	Colorado A	Glover et al., 1976
	50	34	0.8	1.00	Idaho A	Glover et al., 1976
<u>Silt</u>	54	37	2.3	1.23	Arkansas C	Glover et al., 1976
	53	16	3.6	0.79	Illinois	Glover et al., 1976
	15	29	0.43	0.83	Soil #1 (WNRE)	Gillham et al., 1981a
	36	29	0.41	0.82	Soil #2 (WNRE)	Gillham et al., 1981a
	35	31	0.40	0.88	Soil #3 (WNRE)	Gillham et al., 1981a
	41	31	1.27	0.96	Soil #5 (Lemington)	Gillham et al., 1981a
	55	33	0.35	0.93	Soil #9 (North Bay)	Gillham et al., 1981a
	34	32	0.85	0.94	Soil #14 (Alberta)	Gillham et al., 1981a
	-	29.1	7.1	1.50	Brookston silt (average profile)	Juo & Barber, 1970
	50	6	0.23	0.18	Ritzville silt	Routson, 1973
	34	35	0.81	1.03	Soil #15 (Alberta)	Gillham et al., 1981a
	32	36	1.0	1.07	Tennessee (Oak Ridge)	Glover et al., 1976
<u>Clay</u>	32	36	2.7	1.24	New York (West Valley)	Glover et al., 1976
	34	56	3.2	1.83	Arkansas A	Glover et al., 1976

The K_d range reported for methyl iodide in soil was 0.1 to 3.1 (Wildung et al., 1974). The multiple-regression equations were developed for mineral soils. A single value of > 30 mL/g for charcoal is pertinent to organic soil (Nowak, 1981). Vandergraaf (1982) recommended a K_d value for iodine of 0 mL/g, until a cationic species of iodine is identified.

Iodine References

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4.5 MOLYBDENUM

No data were found in the literature for molybdenum; however, because of its position in the Periodic Table, the K_d values for technetium can be used for molybdenum.

4.6 NICKEL

Little information exists for soil adsorption of nickel. Swanson (1981) reported a range of K_d values for nickel of 5.2×10^1 to 1.2×10^3 mL/g for a selected size fraction of Hanford soil (75 to 150 μ m) with a soil-to-solution ratio of 0.010 g/mL. Contradictory results and the use of organic complexants in the Swanson experiments indicate that values for specific samples should not be used.

* Unrestricted, unpublished report, available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

Gerritse et al. (1982) suggested that the K_d value range for nickel is 1×10^2 to 1×10^3 mL/g. They reported two sandy mineral soil values of 6×10^1 and 3.4×10^2 mL/g and four peat soil values of 3.6×10^2 , 6×10^2 , 9.9×10^2 and 4.7×10^3 mL/g. Wong et al. (1983) reported K_d values of 604 and 1437 mL/g for a sand and a muck soil, respectively. The recommended values of μ and σ for the K_d distribution for nickel, based on this information and distribution information for the other nuclides, are given in Table 22.

TABLE 22
RECOMMENDED VALUES OF μ AND σ FOR NICKEL

Soil Type	Lognormal μ	Distribution σ
Sand	1.5	1.0
Silt	2.0	1.0
Clay	3.0	1.0
Organic	3.0	1.0

Nickel References

- Gerritse, R.G., R. Vriesema, J.W. Dalenberg and H.P. De Roos. 1982. Effect of sewage sludge on trace element mobility in soils. J. Environ. Qual. 11, 359-364.
- Swanson, J.L. 1981. Effect of organic complexants on the mobility of low-level waste radionuclides in soils: Status report. Pacific Northwest Laboratory Report, PNL-3927, UC-70.
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4.7 PALLADIUM

No specific information was found on soil adsorption of palladium. We suggest that the K_d values for nickel be used for palladium. Vandergraaf/

(1982) reported a range of 0 to 28 mL/g for the K_d of palladium for granite. He recommended that a K_d value of 11 mL/g be used.

Palladium Reference

Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radionuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.

4.8 RARE EARTHS - TERBIUM, SAMARIUM AND CERIUM

Terbium, samarium and cerium are fission products, and it is convenient to discuss these three rare-earth elements together because of their chemical similarity. Cerium was the only one of these elements for which data were found. Vandergraaf (1982) reported that the K_d value for cerium ranges from 250 to 5000 mL/g, and recommended a value of 1000 mL/g. Allard et al. (1977) reported a K_d range for cerium of 100 to 10 000 mL/g for clay/ mud and 1000 to 1.6×10^4 mL/g for granite. Baes and Sharp (1981) suggested a mean value of 3.0 for the $\log_{10} K_d$ for cerium, with a corresponding standard deviation of 0.6, and a K_d range of 58 to 6000 mL/g (see Table 1) for all soils. We recommend using the values of Baes and Sharp for all soil types.

Rare Earths References

Allard, B., H. Kipatzi and J. Rydberg. 1977. Sorption of long-lived radionuclides in clay and rock. Part 1. Determination of Distribution Coefficients. KBS Technical Report 55.

Baes III, C.F. and R.D. Sharp. 1981. Predicting radionuclide leaching from root zone soil for assessment applications. Oak Ridge National Laboratory Report, CONF-810606-44.

Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radionuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.

* Unrestricted, unpublished report, available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

4.9 SELENIUM

Elsokkary (1980) reported selenium adsorption for three soils, which allowed the computation of K_d values of 1.6, 2.2 and 2.5 mL/g on a clay soil and two silty soils, respectively. Frost and Griffin (1977) reported a K_d value of ~ 50 mL/g for HSeO_3^- adsorption on calcium-montmorillonite at a pH of 7. Singh et al. (1981) reported K_d values ranging from 3 to 73 mL/g for selenate adsorption on sandy soils. Since insufficient data are available, we suggest that the values for polonium be used for assessment purposes.

Selenium References

- Elsokkary, I.H. 1980. Selenium distribution chemical fractionation and adsorption in some Egyptian alluvial and lacustrine soils. *Z. Pflanzenernaehr. Bodenk.*, 143, 74-83.
- Frost, R.R. and R.A. Griffin. 1977. Effect of pH on adsorption of arsenic and selenium from landfill leachate by clay minerals. *Soil Sci. Soc. Amer. J.* 41, 53-57.
- Singh, M., N. Singh and P.S. Relan. 1981. Adsorption and desorption of selenite and selenate selenium on different soils. *Soil Sci.* 132, 134-141.

4.10 SILVER

Little information exists for soil adsorption of silver and no specific soil K_d values were found. Consequently, it is suggested that information for copper be used for silver because of their proximity in the Periodic Table. Gerritse et al. (1982) suggested that the K_d value range for silver is 1×10^1 to 1×10^3 mL/g. They reported K_d values of 1.6×10^2 and 5.6×10^1 mL/g for copper on sandy mineral soil, and K_d values of 4.4×10^1 , 1.7×10^2 , 2.2×10^2 and 3.3×10^2 mL/g for copper on peaty soil. Values for copper also ranged from 5.5×10^1 to 1.2×10^3 mL/g for 0 to 0.9 meq Ca^{+2} /g dry peat. Wong et al. (1983) reported K_d values for copper of 206 and 197 mL/g for a sand and a muck soil, respectively. The recommended K_d distribution parameter values for silver, based on this information, on the distribution information for the other nuclides and on Table

ALL INFORMATION CONTAINED HEREIN IS UNCLASSIFIED

Soil Type	Soil No.	Soil Name	Soil Description	Soil pH	Soil E_h (V)	Soil E_h (mV/100 g)	Soil Iron Oxides	Soil Coagulating Cation	Soil E_d (mV/g)	Soil Location or Description	Reference
Silt	-	-	3.5	-	4.5-5.0	-	22	-	1.6x10 ²	Soil C	Gerritsen et al., 1982
	-	20	2.5	-	7.5-8.0	-	16	-	5.6x10 ²	Soil D	Gerritsen et al., 1982
	-	-	-	-	4.8	-	-	0.1 mol/L CaCl ₂	2.7	Florida 1 - sand	Orsham, 1973
	-	-	-	-	6.2	-	-	0.1 mol/L CaCl ₂	33.0	Florida 2 - sand + organic matter	Orsham, 1973
	-	-	-	-	5.0	-	-	0.1 mol/L CaCl ₂	29.0	Missouri 23	Orsham, 1973
Clay	-	-	-	-	7.6	-	-	0.1 mol/L CaCl ₂	200	Missouri 24	Orsham, 1973
	-	-	-	-	4.6	-	-	0.1 mol/L CaCl ₂	333	Missouri 36	Orsham, 1973
	-	-	-	-	4.5	-	-	-	4.4x10 ³	Soil A	Gerritsen et al., 1982
Silt/Clay	-	-	2	-	4 to 5	-	-	-	2.2x10 ³	Soil A	Gerritsen et al., 1982
	-	-	20	-	6	-	-	-	1.7x10 ³	Soil B	Gerritsen et al., 1982
	-	-	20	-	6.2	-	-	-	3.3x10 ³	Soil B	Gerritsen et al., 1982

TABLE 24
RECOMMENDED VALUES OF μ AND σ FOR SILVER

Soil Type	Lognormal Distribution		n
	μ	σ	
Sand	1.726	0.9988	4
Silt	2.090	0.5678	3
Clay	4.0		
Organic	4.184	0.3792	4

Silver References

- Gerritse, R.G., R. Vriesema, J.W. Dalenberg and H.P. De Roos. 1982. Effect of sewage sludge on trace element mobility in soils. J. Environ. Qual. 11, 359-364.
- Graham, E.R. 1973. Selective distribution and labile pools of micronutrient elements as factors affecting plant uptake. Soil Sci. Soc. Amer. Proc. 37, 70-74.
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4.11 STRONTIUM

The most extensive report on strontium K_d values for Canadian soils is the work of Gillham et al. (1981a). For 15 Canadian soils, the K_d values ranged from 2.5 to 1×10^2 mL/g. The study also showed that strontium "in some or possibly many circumstances would migrate at velocities smaller than the groundwater velocity but at velocities which nevertheless could be significant."

The recommended K_d value means, standard deviations, ranges and distribution parameters for strontium by soil type, based on Table 25, are given in Table 26.

K. VALUES FOR STRUCTURE : LITERATURE SURVEY SUMMARY

continued . .

TABLE 25 (Continued)

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCl ₂	pH Saturated Paste	P _h (V)	CFC (meq/100 g)	% Free Iron Oxides	Coexisting Cation	R _d (mL/g)	Soil Location or Description	Reference
Silt	36	35	29	0.43	33.6	8.1 (CaCl ₂)	-	8.4	-	see ref.	2.0x10 ¹	Soil #1 (WNE)	Gillham et al., 1981a
	35	36	29	0.41	33.8	8.1 (CaCl ₂)	-	8.3	-	see ref.	2.0x10 ¹	Soil #2 (WNE)	Gillham et al., 1981a
	36	35	31	0.40	34.1	8.1 (CaCl ₂)	-	8.6	-	see ref.	2.0x10 ¹	Soil #3 (WNE)	Gillham et al., 1981a
	28	41	31	1.27	21.1	7.7 (CaCl ₂)	-	5.9	-	see ref.	1.0x10 ¹	Soil #5 (Lewington)	Gillham et al., 1981a
	12	55	33	0.35	0	6.7 (CaCl ₂)	-	10.2	-	see ref.	1.0x10 ²	Soil #9 (North Bay)	Gillham et al., 1981a
	36	36	32	0.85	5.1	7.7 (CaCl ₂)	-	32.7	-	see ref.	8.0	Soil #14 (Alberta)	Gillham et al., 1981a
	45	44	11	0.14	1.4	8.8 (CaCl ₂)	-	12.0	-	see ref.	1.1x10 ² ± 1	Sediment A (Solution 1)	Serve et al., 1978
	medium loam (0.01 mm)	31.6	2.94	-	6.6	-	-	10.6	-	-	3.0x10 ² ± 10	low ash podzolic	Alekashin, 1965
	medium loam (0.01 mm)	41.4	1.28	-	8.4	-	-	12.2	-	-	1.7x10 ² ± 30	Serveon	Alekashin, 1965
	31	46	0	-	-	-	-	2.4	-	-	1.4x10 ¹	alluvial soil (Cedarache)	Parsons, 1972
	38	62	0	-	-	-	-	2.7	-	-	2.3x10 ¹	alluvial soil (Cedarache)	Parsons, 1972
	18	66	16	-	-	-	-	6.3	-	-	1.8x10 ¹	Vindobanian sed. (Cedarache)	Parsons, 1972
	20	45	15	-	-	-	-	1.8	-	-	1.6x10 ¹	Vindobanian sed. (Cedarache)	Parsons, 1972
	36	52	14	-	-	-	-	4.9	-	-	1.6x10 ¹	Vindobanian sed. (Cedarache)	Parsons, 1972
	45	47	8	-	-	-	-	1.5	-	-	1.4x10 ¹	Vindobanian sed. (Cedarache)	Parsons, 1972
Clay	7	92	1	-	-	-	-	4.2	-	-	2.2x10 ¹	sandy clay sed. (Durance R.)	Parsons, 1972
	18	71	11	-	-	-	-	3.5	-	-	1.6x10 ¹	sandy clay sed. (Durance R.)	Parsons, 1972
	3	96	1	-	-	-	-	5.2	-	-	1.6x10 ¹	sandy clay sed. (Durance R.)	Parsons, 1972
	silt	29.1	7.1	-	6	-	-	39.4	-	3x10 ⁻³ mol/L SrCl ₂	5.0x10 ¹	Brookston silt	Jan & Barber, 1970
	45	50	5	0.23	3.8	-	-	11.0	1.21	0.2 mol/L NaCl	2.47x10 ¹	Pittville silt (avg. profile)	Roussier, 1973
	31	36	33	0.81	5.2	7.8 (CaCl ₂)	-	31.5	-	see ref.	8	Soil #15 (Alberta)	Gillham et al., 1981a
	heavy loam (0.01 mm)	33.42	2.74	-	6.6	-	-	26.1	-	-	5.7x10 ² ± 80	Onestru	Alekashin, 1965
	heavy loam (0.01 mm)	-	-	-	6.7	-	-	30.4	-	-	1.15x10 ³ ± 140	Leached Chernozem	Alekashin, 1965
	heavy loam (0.01 mm)	67.72	4.27	-	8.0	-	-	32.9	-	-	4.3x10 ² ± 30	Southern Chernozem	Alekashin, 1965
	clayey silt (0.01 mm)	50.72	6.46	-	6.8	-	-	32.2	-	-	4.9x10 ² ± 50	Thick Chernozem	Alekashin, 1965
Silt	100 (0.01 mm)	-	-	-	-	-	-	35	-	-	4.7x10 ¹	very fine suspended sediment (Durance River)	Parsons, 1972
	rich	49.8	-	7.0	-	-	-	70.0	-	3x10 ⁻³ mol/L SrCl ₂	1.5x10 ²	Rich	Jan & Barber, 1970

* Data available for coexisting cations sodium, potassium, magnesium, calcium, barium and hydrogen.

TABLE 26
K_d FOR STRONTIUM

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	26.02	30.21	26	2.0 to 114	1.162	0.4964
Silt	49.49	72.44	20	8.0 to 300	1.436	0.4254
Clay	449.2	415.7	6	8.0 to 1150	2.286	0.8239

Baes and Sharp (1981) suggested a mean value of 1.4 for the $\log_{10} K_d$ for strontium, with a corresponding standard deviation of 0.9 (see Table 1). K_d values for strontium determined for various pure clay minerals ranged from 0.2 to 9.0 mL/g (Wahlberg et al., 1965). K_d values determined for various minerals (including clay minerals) ranged from 0 mL/g for quartz to 2.1×10^3 mL/g for alumina (both at pH = 7.5) in a natural water solution and from 1 mL/g for quartz to 1.44×10^3 mL/g for alumina (both at pH = 7.0) in a 0.1 mol/L sodium nitrate solution (Tamura, 1972). Palmer et al. (1981) also reported extensive results for strontium sorption on pure clay, clay/silica and alumina/clay for various solution compositions and pH. Allard et al. (1977) reported a K_d range of 2 to 63 mL/g for clay/mud and a range of 3 to 16 mL/g for granite. Vandergraaf (1982) recommended a K_d range for strontium of 0.6 to 600 mL/g for granite.

Strontium References

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Vandergraaf, T.T. 1982. A compilation of sorption coefficients for radionuclides on granites and granitic rocks. Atomic Energy of Canada Limited Technical Record, TR-120*.

Wahlberg, J.S., J.H. Baker, R.W. Vernon and R.S. Dewar. 1965. Exchange adsorption of strontium on clay minerals. U.S. Geol. Surv. Bull., 1140-C, U.S. Government Printing Office, Washington.

4.12 TECHNETIUM

The environmental behaviour of technetium has recently been reviewed (Turcotte, 1982), as has the chemistry of technetium (Paquette et al., 1980). Technetium migration is generally retarded under reducing conditions (i.e., in geological formations), where it is less soluble. Technetium, however, moves with the groundwater in aerated soils of low organic carbon content. Johnston and Gillham (1980) indicated that

- (1) because the pertechnetate ion, TcO_4^- , is the most stable species of technetium in aqueous solutions, TcO_4^- will not be subject to ion exchange; hence, technetium will show little adsorption to soil;
- (2) in soils with appreciable organic matter, Tc^{4+} may be reduced to Tc^{3+} and adsorbed.

The recommended K_d value means, standard deviations, ranges and distribution parameters for technetium by soil type, based on Table 27, are given in Table 28.

TABLE 28
 K_d FOR TECHNETIUM

Soil Type	K_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution μ σ	
Sand	29.39	100.1	15	1.0×10^{-3} to 388	-1.148	1.565
Silt	1.426	3.269	8	1.0×10^{-3} to 11	-1.332	1.290
Organic	118.4	192.0	3	0.24 to 340	1.029	1.581

* Unrestricted, unpublished report, available from SP90, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

[illegible]

- Negative values will be considered as a zero value.

Baes and Sharp (1981) suggested a mean value of -1.5 for the $\log_{10} K_d$ for technetium, with a corresponding standard deviation of 0.5 (see Table 1). Mousny and Myttenaere (1981) investigated the effect of temperature on the soil adsorption of technetium and reported that, for seven soils investigated (including a podzol soil and a fen soil), K_d ranged from 0.007 to 0.234 mL/g. Baes and Sharp (1981) suggested the K_d range is 0.003 to 0.28 mL/g (see Table 1). Wildung et al. (1974) selected 22 surface soils with the following range of properties:

CEC (meq/100 g)	pH	Carbonate	Organic Carbon	Sand %	Silt	Clay
5.5-90.0	3.6-8.9	0-6.5	0.23-28.8	14.1-73.1	17.6-58.0	3.8-46.6

and suggested that the K_d for technetium ranges from 0.007 to 2.8 mL/g. They also suggested a prediction equation of the form

$$K_d = 0.08 X_3 - 0.09 X_4 \quad (1)$$

where X_3 is the organic carbon content,
 X_4 is the pH.

Vandergraaf (1982) recommended a K_d range for technetium of 0 to 80 mL/g for granites.

Technetium References

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4.13 TIN

Gerritse et al. (1982) suggested that the K_d value for tin ranges from 1×10^2 to 1×10^4 mL/g. We recommend that the K_d distribution information for lead be used for tin.

Tin Reference

Gerritse, R.G., R. Vriesema, J.W. Dalenberg and H.P. De Roos. 1982. Effect of sewage sludge on trace element mobility in soils. *J. Environ. Qual.* 11, 359-364.

4.14 ZIRCONIUM AND NIOBIUM

Rhodes (1957) reported K_d values ranging from 90 mL/g (pH = 6.0) to > 1980 mL/g (pH = 2.7, 3.5, 4.4, 8.4 and 9.3) for zirconium-niobium adsorption on Hanford subsoil. From this information, in the pH range of most interest for surface soil (pH = 5 to 8, a sandy soil), zirconium has an average K_d of 164 mL/g. This appears to agree well with the K_d for niobium of 210 mL/g recommended for granite (Vandergraaf, 1982). Vandergraaf also recommended a K_d range for zirconium of 1000 to 6000 mL/g for granite. Allard et al. (1977) reported a K_d range for zirconium of 50 to 1000 mL/g for clay/mud and 1250 to 6300 mL/g for granite.

Based on this information, the recommended mean of the lognormal distribution for zirconium and niobium is 2.5 with a standard deviation of 1.0. Because information is insufficient to break it down by soil type, one value is recommended for all soil types.

Zirconium and Niobium References

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Rhodes, D.W. 1957. The effect of pH on the uptake of radioactive isotopes from solution by a soil. *Soil Sc. Am. Proc.* 21, 389.

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* Unrestricted, unpublished report, available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ontario K0J 1J0.

5. DISTRIBUTION COEFFICIENTS FOR OTHER NUCLIDES

5.1 ANTIMONY

No specific soil K_d information was found for antimony. The K_d values for lead are recommended for antimony, because of its proximity to lead in the Periodic Table.

5.2 BORON

Little information was found on boron adsorption on soils; however, there is some indication that adsorption is influenced by soil texture and the presence of soluble salts and exchangeable cations (Gupta, 1980). Boron adsorption information reported by Keren and O'Connor (1982) for montmorillonite and illite indicated that the K_d value for boron for these pure clays could be as high as 20 mL/g. That work suggested that the K_d value for soils may be in the range 0 to 10 mL/g. We recommend a value of 1 mL/g for assessment purposes; the lognormal distribution parameter values cannot be given.

Boron References

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- Keren, R. and G.A. O'Connor. 1982. Effect of exchangeable ions and ionic strength on boron adsorption by montmorillonite and illite. *Clays Clay Miner.* 30, 341-356.

5.3 CADMIUM

Most of the work carried out with cadmium has been in response to environmental concerns about the application of sewage sludge to agricultural land. The sorption of cadmium on soils and sediments has been studied by Poelstra et al. (1979), Rendell et al. (1980), Hendrickson and Corey (1981), and Gerritse et al. (1982). The recommended K_d value means, standard deviation based on Table 20

TABLE 29

K_d VALUES FOR CADMIUM: LITERATURE SURVEY SUMMARY

Soil Type	Soil Type	Soil Type	Soil Type	Soil Type	pH	E _h	CaC	% Free	Competing	K _d	Soil Location or Description	Reference
						(V)	(mg/100 g)	Iron Oxide	Cation	(mL/g)		
Sandy	sandy soil	-	-	-	6.5	-	31.5	-	-	66.7 ^a	Sandy soil (Brazosvale) 0-25 cm	Poelstra et al., 1979
	sandy soil	-	-	-	6.5	-	31.4	-	-	47.4 ^a	Sandy soil (Brazosvale) 30-45 cm	Poelstra et al., 1979
	sandy soil	5	1.5	-	4.5 - 5.0	-	22	-	[Ca ²⁺] = 0-0.715 mol/L	2.42x10 ²	Soil C	Gerritse et al., 1982
	sandy soil	20	2.5	-	7.5 - 8.0	-	16	-	[Ca ²⁺] = 0-0.015 mol/L	5.0x10 ²	Soil D	Gerritse et al., 1982
Fine sand	fine sand	1.4	-	-	8.2	-	11	-	-	72	Hallendale (fine sand)	Wong et al., 1983
	fine sand	0.72	-	-	8.4	-	60	1.07	-	76	Imperial (California)	Garcia-Hiragawa, 1980
	fine sand	1.8	-	-	6.0	-	25	1.07	-	9.8	Olivette (California)	Garcia-Hiragawa, 1980
	fine sand	1.5	-	-	5.8	-	24	8.29	-	16	Bonnet (California)	Garcia-Hiragawa, 1980
Silt	silt soil	10	5	trace	7.4	-	-	-	-	62 ^a	(Valburg) 0-30 cm	Poelstra et al., 1979
	silt soil	14.3	-	-	5.2	-	22.8	-	-	23	organic	Garcia-Hiragawa, 1980
	silt soil	31	-	-	5.1 (4.0)	-	-	-	-	37 ^a	(Schroeder)	Poelstra et al., 1979
	silt soil	30	-	-	4.5	-	-	-	[Ca ²⁺] = 0-0.715 mol/L	1.44x10 ³	Soil A	Gerritse et al., 1982
Peat	peat	4	-	-	4 to 5	-	-	-	[Ca ²⁺] = 0-0.715 mol/L	9.0x10 ³	Peat A	Gerritse et al., 1982
	peat	4	-	-	6.2	-	-	-	[Ca ²⁺] = 0-0.715 mol/L	5.76x10 ³	Soil B	Gerritse et al., 1982
	peat	4	-	-	4 to 5	-	-	-	[Ca ²⁺] = 0-0.715 mol/L	1.7x10 ³	Peat	Wolff et al., 1977
	peat	4	-	-	4 to 5	-	-	-	0.025 mol Ca ²⁺ /mL soil	3x10 ⁴	Peat	Wolff et al., 1977
plantation	plantation	47	-	-	7.2	-	34	-	-	341	average of 3 layers	Wong et al., 1983

^a 1 week equilibration

TABLE 30
K_d FOR CADMIUM

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K _d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	189.7	194.1	5	47.6 to 500	2.095	0.4397
Silt	33.93	36.56	3	9.8 to 76	1.359	0.4645
Organic	4246	6110	8	23 to 1.7×10^4	2.880	1.090

Hendrickson and Corey (1981) reported K_d data from several authors, and their plot indicated that the K_d range is 0 to 6 mL/g and is significantly dependent on both the cadmium and calcium contents of the soil. Navrot et al. (1978) reported K_d values for cadmium for five Israeli soils ranging from 1×10^3 to 1×10^4 mL/g, and the K_d value was correlated to specific soil surface area.

Cadmium References

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Wolf, A., K. Bunzl, F. Dietl and W.F. Schmidt. 1977. Effect of calcium ions on the absorption of lead (2+), copper (2+), cadmium (2+) and zinc (2+) by humic substances. *Chemosphere* 6, 207-213.

Wong, K.V., S. Sengupta, D. Dasgupta, E.L. Daly, Jr., N. Nemerow and H.F. Gerrish. 1983. Heavy metal migration in soil-leachate systems. *Bio-cycle* 24, 30-33.

5.4 TELLURIUM

Allard et al. (1977) suggested that the K_d value for tellurium for clay/mud and granite is 1 mL/g. This work suggests that the K_d range may be 0 to 1 mL/g. We recommend a K_d value of 0 mL/g for assessment purposes; no distribution parameter values can be given.

Tellurium Reference

Allard, B., H. Kipatsi and J. Rydberg. 1977. Adsorption of long-lived radionuclides in clay and bedrock. Part 1. Determination of distribution coefficients. KBS Technical Report 55.

5.5 ZINC

Gerritse et al. (1982) suggested that the K_d values for both zinc and cadmium range from 1×10^3 to 1×10^4 mL/g, and their data for sandy and organic soils show that the two elements exhibit very similar sorption behaviour. The recommended K_d value means, standard deviations, ranges and distribution parameters for zinc, based on Table 31, are given in Table 32.

TABLE 32
 K_d FOR ZINC

Soil Type	\bar{K}_d (mL/g)	S.D.	n	K_d Range (mL/g)	Lognormal Distribution	
					μ	σ
Sand	622.0	911.6	5	0.1 to 2120	1.762	1.694
Silt	51.8	68.17	2	3.6 to 100	1.278	1.021
Organic	4092	4909	6	70 to 1.3×10^4	3.185	0.83

TABLE 31

K_d VALUES FOR ZINC: LITERATURE SURVEY SUMMARY

Soil Type	% Sand	% Silt	% Clay	% Organic	% CaCl ₂	pH Saturated Paste	E _p (V)	CFC (mg/100 g)	% Free Iron Oxides	Converting Cation	Z _d (mL/g)	Soil Location or Description	Reference
Sand	100	0	0	0	-	4.5 - 5.0	-	22	-	[Ca ²⁺] = 0-0.015 mol/L	7.0x10 ¹	Soil C	Gerritse et al., 1982
	100	0	0	0	-	7.5 - 8.0	-	16	-	[Ca ²⁺] = 0-0.11 mol/L	2.12x10 ³	Soil D	Gerritse et al., 1982
	-	-	-	-	-	4.8	-	-	-	0.1 mol/L CaCl ₂	0.1	Florida 1	Graham, 1973
	-	-	-	-	-	6.2	-	-	-	0.1 mol/L CaCl ₂	30	Florida 2	Graham, 1973
Fine sand	-	-	-	1.4	-	8.2	-	11	-	-	870	Hallendale fine sand	Wong et al., 1973
	-	-	-	-	-	5.0	-	-	-	0.1 mol/L CaCl ₂	3.6	Missouri 23	Graham, 1973
	-	-	-	-	-	7.4	-	-	-	0.1 mol/L CaCl ₂	100	Missouri 24	Graham, 1973
Organic	-	-	-	90	-	4.5	-	-	-	[Ca ²⁺] = 0-0.15 mol/L	1.89x10 ³	Soil A	Gerritse et al., 1982
	-	-	-	99	-	4 - 5	-	-	-	[Ca ²⁺] = 0-0.015 mol/L	6.3x10 ³	Peat A	Gerritse et al., 1982
	-	-	-	99	-	6.2	-	-	-	[Ca ²⁺] = 0-0.015 mol/L	2.89x10 ³	Soil B	Gerritse et al., 1982
	-	-	-	-	-	4 - 5	-	-	-	-	1.3x10 ⁶	Peat	Wolf et al., 1977
Sphagnum peat	-	-	-	-	-	4 - 5	-	-	-	0.025 mol	7.0x10 ¹	Peat	Wolf et al., 1977
	-	-	-	-	-	7.2	-	36	-	-	412	Plantation peat (average of 3 layers)	Wong et al., 1983

Zinc References

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5. CONCLUSIONS

The paucity of K_d values for organic soil is the most striking observation from our review of the literature. Plutonium, lead, technetium, cadmium and zinc were the only nuclides for which more than two K_d values have been determined for an organic soil (see Table 33). The next most important observation is that very little work has been done with mineral soils for some of the more mobile nuclides with K_d values up to 100 mL/g, such as uranium, technetium, molybdenum, iodine, selenium, carbon, boron, and tellurium. There may be good reasons why more K_d work is not warranted for these nuclides in the Canadian waste management program, such as the formation of precipitates or reduction to an immobile species in the vault or geosphere. Our major recommendation is that effort be directed towards the chemistry (including parameter determination, i.e., K_d determinations) of organic soils, and in particular the reactions of uranium, technetium, iodine, selenium and carbon with organic soils. In spite of the limited data base, it is possible to select reasonable K_d distribution parameter values for most nuclides for long-term waste management assessment purposes.

TABLE 33

AVAILABILITY OF K_d DATA FOR EACH NUCLIDE BY SOIL TYPE

Nuclide	Soil Type			
	Sand	Silt	Clay	Organic
Actinium	X	X	X	X
Americium				X
Antimony	X	X	X	X
Bismuth			X	X
Boron	X	X	X	X
Cadmium			X	
Calcium	X	X	X	X
Carbon	X	X	X	X
Cesium				X
Iodine				X
Lead		X	X	
Molybdenum	X	X	X	X
Neptunium				X
Nickel	X	X	X	X
Palladium	X	X	X	X
Plutonium				
Polonium			X	X
Protactinium	X	X	X	X
Radium				X
Rare Earths	X	X	X	X
Selenium	X	X	X	X
Silver	X	X	X	X
Strontium				X
Technetium			X	
Tellurium	X	X	X	X
Thorium	X	X	X	X
Tin	X	X	X	X
Uranium	X	X		X
Zinc		X	X	
Zirconium & Niobium	X	X	X	X

X denotes 2 or fewer K_d values.

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