R-09-27

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March 2009

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ISSN 1402-3091
SKB Rapport R-09-27

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Abstract

Soil and sediment solid/liquid partition coefficients (Kd) are used to indicate the relative mobility of radionuclides and elements of concern from nuclear fuel waste, as well as from other sources. The Kd data are inherently extremely variable, but also vary systematically with key environmental attributes. For soil Kd, the key variables are pH, clay content and organic carbon content. For sediment Kd, water type (freshwater versus marine) and sediment type (benthic versus suspended) are important.

This report summarized Kd data for soils and sediments computed from indigenous stable element concentrations measured at the Forsmark and Laxemar-Simpevarp sites. These were then compared to several literature sources of Kd data for Ce, Cl, Co, Cr, Cs, Fe, Ho, I, La, Mn, Mo, Nb, Nd, Ni, Np, Pa, Pb, Pu, Ra, Sb, Se, Sm, Sn, Sr, Tc, Th, Tm, U and Yb. The Kd data computed from indigenous stable element concentrations may be especially relevant for assessment of long-lived radionuclides from deep disposal of waste, because the long time frame for the potential releases is more consistent with the steady state measured using indigenous stable elements.

For almost every one of these elements in soils, a statistically meaningful regression equation was developed to allow estimation of Kd for any soil given a modest amount of information about the soil. Nonetheless, the median residual geometric standard deviation (GSD) was 4.3-fold, implying confidence bounds of about 18-fold above and below the best estimate Kd. For sediment, the values are categorised simply by water type and sediment type. The median GSD for sediment Kd as measured at the Forsmark and Laxemar-Simpevarp sites was 2.5-fold, but the median GSD among literature values was as high as 8.6-fold. Clearly, there remains considerable uncertainty in Kd values, and it is important to account for this in assessment applications.

Contents

1 Introduction

Solid/liquid partition coefficients, also known as distribution coefficients and Kd, have been used in safety assessment calculations for nuclear facilities for many years. They endure because of their conceptual simplicity. The Kd is the ratio of the concentration of a nuclide on a solid phase (soil or sediment) divided by the equilibrium concentration in the contacting liquid phase (water). As Kd is used, this implies a linear, zero-intercept relationship between sorbed and non-sorbed species of the element, which is a convenient assumption but certainly not universally true. In most uses, an additional assumption is that the sorption is at equilibrium and is reversible. The conceptual model is most applicable for simple cationic species undergoing ion exchange reactions on simple charged surfaces. However, in practice Kd is measured and used for much more complex systems, and the lack of fit of the simple Kd model to the real system becomes part of the overall uncertainty in the values of Kd.

In a safety assessment, Kd is used as a quantitative indicator of environmental mobility of the element. In general, all isotopes of an element are assumed to have the same Kd value, because sorption is a chemical property generally unaffected by atomic mass or nuclear emissions. In soil, Kd is used to estimate leaching of elements through soil and uptake of elements from soil by plants. In sediments, Kd is used to partition elements between the water column and the sediments, which has implications for removal of the element from the water column by flushing and sedimentation.

Because Kd is a simplification, the values are necessarily empirical and highly dependent on the system where they are measured. The classical method of measuring Kd is to suspend pre-dried and ground samples of the solid (soil or sediment) in a tenfold larger volume of water, add the element of interest, shake the suspension for some time (hours or days), filter off the liquid and measure the concentration of the element in the liquid and on the solid. This is not very representative of an environmental setting, and so there are many variants among the methods.

For soils, the preferred method is to have the element introduced to the soil weeks or months before the measurement or to use contaminated soils collected in the field, the soil is incubated at field-like moisture and temperature before measurement, then the pore water present in the soil is extracted and both porewater and solids are analysed. The extraction requires some force, such as centrifugation. For bottom sediments, the ideal is to collect contaminated sediment in the field and to extract pore water when the sample drains. For suspended sediments, again sampling in the field is ideal and the solids are separated from the sample of the water column by filtration. However, these ideals are rarely met, and it is necessary to consider and interpret Kd measured in a variety of ways.

The calculation of Kd is concentration on the solids (mg kg^{-1} dry solid) divided by concentration in the pore water (mg L^{-1}), giving units of L kg^{-1}. These units are presented because they relate most closely to the units of measure used for the concentrations, and they are the most commonly used units for Kd in the literature. A more correct SI (Le Système International d'Unités) unit would be m^3 kg^{-1}, with values 1,000-fold lower than those in L kg^{-1}.

All values of Kd are assumed to be lognormally distributed based on the Central Limit Theorem, and the assumption of lognormal is generally supported by empirical evidence. Thus, all analysis was done on log-transformed values and summaries are of geometric means (GM) and geometric standard deviations (GSD).

2 Objectives and structure of this report

This report provides a means to estimate Kd values for soils and sediments of the Forsmark and Simpevarp regions. The approach is threefold:

1. To compute Kd values from uncontaminated soils and sediments collected at Forsmark and Laxemar-Simpevarp. For both soils and sediments, the Kd values obtained from this are for desorption of the indigenous elements, and so interpretation is needed in applying these to the case of sorption of contaminant elements. For sediments, both freshwater and marine sediments, and both bottom sediments and suspended sediments were sampled.
2. To compare these site-specific Kd values to other sources of Kd values for similar media, interpret the similarities and differences and make judgments of which values would best represent the assessment scenario for disposal of nuclear waste.
3. To the extent possible, develop a predictive capacity to relate Kd to site properties, so that variation in Kd across the sites can be estimated.

The first sections of the report deal with soils, starting with a description by G. Sohlenius (Sveriges geologiska undersökning, Uppsala) of the sampling done to obtain 7 samples from Forsmark and Laxemar-Simpevarp. The next section by S.C. Sheppard (ECOMatters Inc. Pinawa, MB, Canada) describes the methods used to measure desorption Kd values for indigenous elements in these soils, and sorption Kd values for I and Cl. This includes comparison of the values obtained to those in a data set obtained using the same methods for a broad range of Canadian agricultural soils.

The last section related to soils is a comparison by S.C. Sheppard and B.L. Sanipelli (ECOMatters Inc. Pinawa, MB, Canada) of the above Kd values to the general literature, including the recent compilation by the IAEA program Environmental Modeling for Environmental Safety /EMRAS 2008/, and the development of regression equations to predict Kd as a function of simple soil properties.

Sediments are dealt with in the final section by J. Long, S.C. Sheppard and B.L. Sanipelli (ECOMatters Inc. Pinawa, MB, Canada). The sediment Kd was computed from previously published sediment and water concentrations for samples collected at Forsmark and Simpevarp. These values were for both freshwater and marine samples, and for both bottom- and suspended-sediment samples. These are compared to the general literature, including the recent compilation by /EMRAS 2008/. Apart from the values specific to each sediment type, no further predictive capacity was possible for sediment Kd.

3 Sampling of soils

3.1 The studied sites

SKB performs site investigations for localisation of a deep repository for high level radioactive waste. The site investigations are performed at two sites, Forsmark in Östhammar municipality and Laxemar-Simpevarp in Oskarshamn municipality, see Figure 3-1 and Figure 3-2, respectively. This report presents results from both these sites.

All known regolith in the Forsmark and Laxemar-Simpevarp areas was formed during the Quaternary period and is therefore referred to as Quaternary deposits (QD). After the latest deglaciation, both sites were completely situated below the level of the Baltic Sea. Most of the Forsmark area was situated below the level of the sea as late as 1000 years ago. The Laxemar-Simpevarp site started to emerge from the sea much earlier and most of the area was above the sea level 3000 years ago. The time available for soil forming processes has consequently been much shorter in the Forsmark area compared to the Laxemar-Simpevarp area.

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***Figure 3-1.** Location of the three sampled sites in Forsmark. The properties of the sites are shown in Table 3-1. Site A (AFM001076) is clayey silty till, Site B (PFM006024) is peat, and Site G (PFM002670) is clayey silty till with mull.*

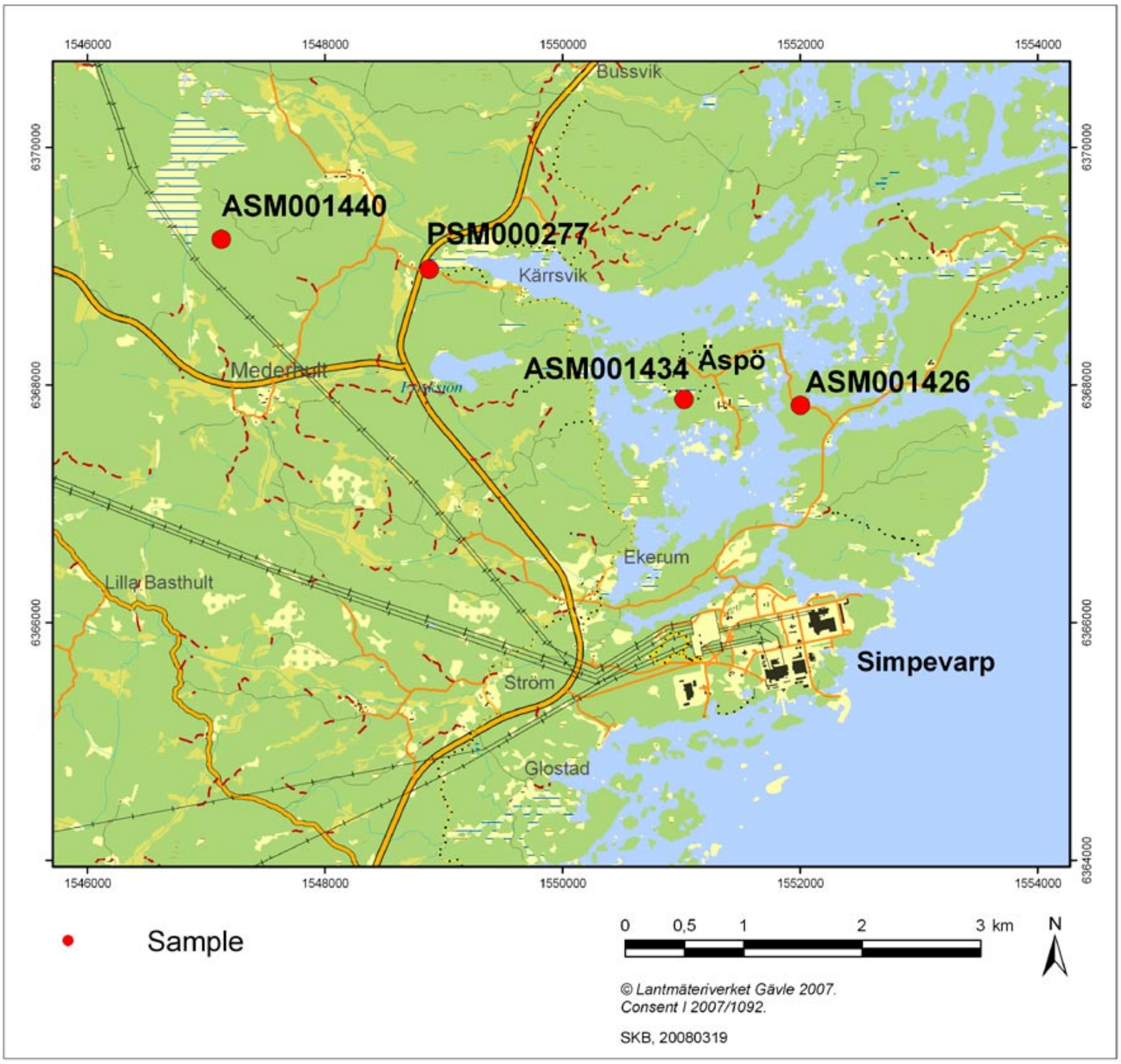

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Figure 3-2. *Location of the four sampled sites in Laxemar-Simpevarp. The properties of the sites are shown in Table 3-1. Site C (ASM001426) is sandy till, Site D (ASM001434) is clay gyttja, Site E (ASM001440) is peat and Site F (PSM000277) is clay gyttja.*

The Forsmark site is characterised by a flat topography gently dipping towards the east. Most of the area is covered by till /Sohlenius et al. 2004/. In parts of the area the till has a high content of clay and is to a large extent used as arable land. The till in Forsmark contains calcite emanating from Ordovician limestone situated north of the area. Calcite is easily dissolved by chemical weathering. However, the area has only been above the sea level for a relatively short period and weathering has consequently not dissolved the calcite present in the till soils.

The Laxemar-Simpevarp area is characterised by a relatively flat bedrock surface. In contrast to the Forsmark area the topography is characterised by a number of pronounced valleys. The highest areas are dominated by till and exposed bedrock, whereas the valleys, often used as arable land, are covered by a relatively thick layer of Quaternary deposits /Rudmark et al. 2005/. The till from the Laxemar-Simpevarp area is generally coarser grained compared to till from the Forsmark area. Clay, sand and peat are the dominating QD in the valleys in the Laxemar-Simpevarp area. A large proportion of these valleys are former wetlands where the groundwater table has been lowered by ditches. The clays from the Laxemar-Simpevarp area often contain considerable amounts of organic matter and are then referred to as clay gyttja (Table 3-1).

Table 3-1. **The sampled sites in the Forsmark and Laxemar-Simpevarp areas.**

Location		Quaternary deposit	Environment	Facility
Site A	AFM001076	clayey silty till	fen, mixed forest	Forsmark
Site B	PFM006024	peat (*Spagnum*)	fen, *Pinus* forest	Forsmark
Site C	ASM001426	sandy till	oak forest	Laxemar-Simpevarp
Site D	ASM001434	clay gyttja	fen, alder forest	Laxemar-Simpevarp
Site E	ASM001440	Peat	former fen, spruce forest	Laxemar-Simpevarp
Site F	PSM000277	clay gyttja	open fen	Laxemar-Simpevarp
Site G	PFM002670	clayey silty till (with mull)	arable land	Forsmark

3.2 Methods

Samples were taken from three and four sites in the Forsmark and Laxemar-Simpevarp areas respectively (Figures 3-1 and 3-2). At each site 10 sub-samples were taken to a depth of 30 cm. The individual sub-samples were "randomly" collected from an area of around 30 m^2. The till samples were taken in spade-dug holes and the other samples were taken by the use of a hand-driven corer. The size of each sub-sample of peat was at least 200 g and the corresponding size of the other sub-samples was at least 100 g. Three additional samples were taken from each site using steel cylinders with known volumes. These latter samples were later used to determine the dry bulk density of the sampled deposits. All samples were placed in plastic bags and stored in a cold room before analyses.

The dry bulk densities were determined in laboratory after drying the samples in the steel cylinders at 105°C. The 10 sub-samples from each site were mixed into one general sample, which was used for further analyses. Grain size analyses, on material < 20 mm, were carried out on samples from five of the sites. The samples from the two peat areas were not analysed for grain size. The grain size distribution of coarse material (grain size 20–0.063 mm) was determined by sieving and fine material (< 0.063 mm) with hydrometer analysis.

The five samples analysed for grain size were also analysed for contents of calcite ($CaCO_3$) and organic material. The $CaCO_3$ analyses were determined (grain sizes < 63µ) using Passon apparatus, /Talme and Almén 1975/ which measures the amount of CO_2 developing during the reaction between $CaCO_3$ and HCl. The content of organic material was measured by determining the weight loss after burning a sample at 550°C. That method is referred to as loss on ignition (LOI).

3.3 Results

The results from the determinations of dry bulk densities are shown in Table 3-2. The QD rich in organic material (clay gyttja and peat) have considerably lower densities than the till deposits, which are dominated by mineral particles.

Table 3-2. Results from the determinations of dry bulk densities.

Site	ID number	Quaternary deposit	Density kg m^{-3}
Site A	AFM001076	clayey silty till	2,140
Site B	PFM006024	peat	120
Site C	ASM001426	sandy till	1160
Site D	ASM001434	clay gyttja	310
Site E	ASM001440	Peat	260
Site F	PSM000277	clay gyttja	310
Site G	PFM002670	clayey silty till	1,520

Results from the analyses of grain size, $CaCO_3$ and organic material (LOI) are shown in Table 3-3. The results clearly reflect the differences between the fine-grained $CaCO_3$–rich till from the Forsmark area (Sites A and G) and the coarser grained $CaCO_3$–free till from the Laxemar-Simpevarp area (Site C).

Table 3-3. Results from the analyses of grain size distribution (material < 20 mm), $CaCO_3$ and organic material for all but the peat soils. All values are shown as % of dry weight. The two clay gyttja samples were never analysed for clay and silt contents separately. For these two samples the sum of the silt and clay contents are presented as one value in the silt column*. The content of organic material is presented as LOI = loss on ignition.

		Quaternary deposit	Gravel % dw	Sand % dw	Silt % dw	Clay % dw	$CaCO_3$ % dw	LOI % dw
Site A	AFM001076	clayey silty till	14.6	42.9	30.3	12.2	25	0.1
Site B	PFM006024	peat	——[na]	——	——	——	——	——
Site C	ASM001426	sandy till	44.3	46.0	6.9	2.9	0.6	3.8
Site D	ASM001434	clay gyttja	0.0	8.7	91.3*	——	0.3	25.6
Site E	ASM001440	peat	——	——	——	——	——	——
Site F	PSM000277	clay gyttja	0.0	3.2	96.8*	——	0.3	33.9
Site G	PFM002670	clayey silty till	17.7	41.3	34.3	6.7	5.3	2.3

[na] Not analysed.

4 Solid/liquid partition coefficients (Kd) for soils sampled at Forsmark and Laxemar-Simpevarp

4.1 Objectives

The objectives in measuring soil solid/liquid partition coefficients (Kd, L kg^{-1}) for 7 soils include contributing to the database of Kd values that might be used in environmental safety assessment of the Forsmark and Simpevarp sites, but more importantly to evaluate if the Kd values of these sites are consistent with the Kds of other soils in the world.

If the Kd values from these 7 soils are consistent with those measured in other soils, then it would be appropriate and arguably preferable to use values drawn from a 'global' database of Kd in safety assessment applications. Conversely, should the 7 soils be unique from Kd values from other soils with similar texture and properties, then a detailed survey of Kd for the assessment sites may be advisable.

4.2 Methods

The methods used have been described by /Sheppard et al. 2007/, and that paper includes comparison of variations in the methods. The preferred method, and method used here (and described in more detail below), was to incubate the soil at least 7 days at field capacity moisture content, and then extract the pore water by centrifugation at about 6,000 m s^{-2} for 1,000 s. Both the soil solids and the pore water were analyzed for elemental composition. The soil samples were kept at the field sampling moisture content prior to incubation, the only drying done was that necessary to get wet samples to a state that could be physically handled (partial grinding and mixing). After the pore water was extracted, the soil samples were dried for analysis and to obtain moisture content data.

The field-moist or partially dried soil was placed in a 60 mL plastic syringe body. Deionized water was added very slowly until the first drop of water dripped from the bottom of the tube. The soil was then considered to be at field capacity moisture. The tube was weighed to determine the amount of water added, and the top and bottom openings were covered with parafilm (parafilm is a laboratory wrap preventing moisture loss). Average soil and water weights for all samples were 67 g and 31 g, respectively.

The sample was left undisturbed for one week, then the parafilm was removed and the tube centrifuged to remove the porewater. The porewater was filtered through a 0.45-μm syringe filter and split. One subsample was acidified to pH < 2 for inductively coupled plasma mass spectroscopy (ICP-MS) analysis of 51 elements, the other subsample was not acidified and was analyzed by ICP-MS for I and by instrumental neutron activation analysis (INAA) for Cl (these unacidified samples were stored frozen if there was a delay before analysis).

Using indigenous elements to measure Kd has the advantage that the soil is more likely at equilibrium even for very slow reactions, including ecological reactions such as cycling through vegetation. However, most Kd values in the literature are from spiking experiments, where the element is added just prior to the measurement of Kd. Thus, spiking was done to measure Kd for I and Cl. The two spiking solutions were a commercial 1,000 mg L^{-1} analytical standard for I and a 1,000 mg L^{-1} solution for Cl prepared from reagent-grade $NaCl_2$. Both solutions were sent for analysis, giving results of 985 mg L^{-1} for Cl and 996 mg L^{-1} for I, agreement within 2%.

For soil samples that were spiked, the soil was weighed into the syringe tube in the same manner described above, and the bottom opening was covered with parafilm. About 5 mL of spiking solution was added to the soil in the tube, and then the soil was left for an hour to absorb the solution. The syringe tubes were weighed to determine the amount of spike added (average of 5.4 g). The soils were then wetted, incubated and centrifuged as above. Not all samples, and in particular the clay-like samples, absorbed the spiking solution well. Thus, after centrifugation, the spiked soil solids were removed and analyzed for I and Cl.

Analysis of the soil solids, including the spiked soils, was by ICP-MS for all elements except I and Cl. For I and Cl, the analysis was by INAA, which gives a true total concentration. Analytical results by ICP-MS were obtained for each soil sample using the Ultratrace 1 method of Activation Laboratories (http://www.actlabs.com/). This method is an aqua regia (acid) partial extraction, which may not extract all elements from mineral matrices, but the more volatile elements should remain in solution. Aqua regia does not decompose all silicate minerals in the soil sample, however, the argument is that the elements in the silicates do not exchange with the pore water so it may be appropriate to exclude them in the analysis of soil elemental composition for Kd.

The concentrations of I in the spiked soils ranged from 40 to 600 mg kg^{-1}, varying because of differences in soil bulk density and the efficiency with which the spike solution was absorbed. The concentrations of Cl in the spiked soils ranged from 100 to 4,600 mg kg^{-1}. These are rather high, as intended to be sure that porewater concentrations were detectable. All samples were analyzed in duplicate, with generally acceptable (small) differences in concentration between the duplicate pairs.

The calculation of Kd is concentration on the solids (mg kg^{-1} dry soil) divided by concentration in the pore water (mg L^{-1}), giving units of L kg^{-1}. For elements with relatively low Kd values, where the Kd is in the same order of magnitude as the soil moisture content, also in units of L kg^{-1} dry soil, then it is important to account for the amount of pore water dried onto the soil solids when the solids are prepared for chemical analysis (this correction has little effect, but is still valid, for elements with high Kd).

Because Kd for Cl is low, and for consistency, this correction was applied to all Kd values here. The correction is $Kd_{(corrected)} = Kd_{(uncorrected)} - MC$, where MC is the soil moisture content (L kg^{-1} dry soil) of the soil when dried for analysis. Because of the properties of these soils, they had high moisture contents after centrifugation, and this correction was quite important for Cl Kd values. The MC values for soils of sites A to G were 0.073, 8.2, 0.016, 3.2, 2.1, 2.8 and 0.071 L kg^{-1}, respectively. This range underscores the wide range in soil properties among these samples, a wider range than many of the soils used to generate the Kd values reported in the literature.

4.3 Results

All of the measured Kd values are shown in Table 4-1 (geometric means (GM) are shown for duplicates). With the exception of the spike values for Cl and I, these are Kd values for elements present in the soil as collected (called native- or indigenous-element Kd values). Native Kd values are typically higher than Kd values from recent element spiking because the retention processes in the soil have had very long times to come to (quasi) equilibrium. More recent additions (spikes) of elements have lower Kds and are implicitly more mobile than native elements. However, in a nuclear waste management context where assessments span millennia, native Kd values may be the most appropriate.

For Cl and I, the native Kds values were almost all larger than the corresponding spike Kds, as expected based on equilibration time. Additionally, the spiked soils were at much higher concentrations: /Sheppard and Evenden 1988/ indicated that Kd for I was linear over 8 orders of magnitude in soil concentration (up to 100 mg kg^{-1}), but /Sheppard et al. 1996/ noted non-linearity above ~1 mg I kg^{-1}. Non-linearity would result in lower Kd at higher soil concentrations, consistent with the finding here.

Sites B and E were peat soils, and in these the Kd value for Cl was relatively high. This is expected because Cl is present in soils as an anion and only reaction with organic matter has an appreciable effect on retention (i.e. causes higher Kd). /Sheppard et al. 2006/ recommended a GM Kd value of 0.1 L kg^{-1} for Cl for all soils, in line with the spike Cl Kd values measured in the SKB soils, but substantially lower than the native Cl Kd values. /Sheppard et al. 2006/ recommended a GSD of at least 10, implying 95th percentile confidence bounds of 0.001 to 10 L kg^{-1}, encompassing most of the measured SKB values.

/Thibault et al. 1990/ gave a range of Kd for I of 1 to 25 L kg^{-1}. Values of Kd are important to assessments only within certain ranges /Sheppard 2005a/, and these values are in the range to be important for all assessments. The reason for the high native Cl Kd values may be because plants growing on the sites will have intercepted Cl from oceanic salt spray, this elevated source may have resulted in greater incorporation of Cl into recalcitrant plant organic materials that then become part of the soil. These would elevate the Cl concentration on soil solids but not affect soil pore-water Cl concentrations, leading to a high native Kd. Site D seems to be an exception, with a lower Kd for native Cl than the Cl spike.

Table 4-1. Solid/liquid partition coefficients (Kd, L kg^{-1}) for Chlorine, Iodine and the suite of elements available from ICP-MS analysis of native elements. GM is the geometric mean of the (up to) 7 values.

Analyte symbol	Site A, clayey till, fen	Site B, peat, fen	Site C, sandy till, oak forest	Site D, clay gyttja, fen	Site E, peat, former fen	Site F, clay gyttja, open fen	Site G, clayey till, arable	GM
Cl native	4.4	37	7.1	0.036	41	5.2	12	5.2
I native	> 90[a]	< 140[a]	60	3,900	1,600	> 8,300[a]	> 210[a]	720
Cl spike	0.017	3.5	0.76	1.1	2	2	0.24	0.65
I spike	0.0041	2.9	0.28	8.8	1.7	2.9	0.069	0.52
Al	4,700	2,600	2,900	7,700	2,800	3,100	11,000	4,300
As	34	340	33	320	150	520	130	140
Ba	34	6.3	30	23	15	54	93	27
Be					290	350		320
Ca	460		460	74	65	24	240	130
Cd	73	480	190	150	220	150	1,200	230
Ce	39,000	4,100	1,700	5,900	3,800	4,400	88,000	8,100
Co	5,000	2,800	2,000	990	200	220	14,000	1,500
Cr	140	900	250	430	370	480	270	350
Cs	28,000		21,000	25,000	57,000	31,000	120,000	38,000
Cu	450	560	160	930	2,100	3,200	700	780
Dy	30,000	2,700	1,700	4,800	3,000	6,500	52,000	6,800
Er	21,000	2,400	1,900	3,700	2,400	6,000	34,000	5,600
Eu	5,600		1,200	2,800	2,000	7,100	11,000	3,800
Fe	25,000	12,000	4,900	34,000	13,000	59,000	160,000	25,000
Ga	2,600	1,300	5,900	9,400	7,800	7,800	14,000	5,500
Gd	22,000	1,800	1,200	3,700	2,400	5,100	51,000	5,200
Ge	380		400	600	680	720	450	520
Hf	650		390	340	520	250	360	400
Ho	37,000		1,800	4,200	2,900	6,700	37,000	7,700
K	790	160	1,200	96	280	1,100	1,200	470
La	44,000	2,600	1,800	5,900	4,000	2,700	80,000	7,200
Li	190			370	220	370		280
Lu	14,000		2,000	2,300	1,500	7,000		3,700
Mg	410	650	1,400	46	140	60	1,400	300
Mn	18,000	1,200	3,600	730	230	130	22,000	1,700
Mo	70	1,200	410	1,200	6,800	3,300	240	810
Na	13	82	19	5.2	17	12	37	19
Nb			1,700	12,000	14,000		36,000	10,000
Nd	46,000	2,100	1,500	5,200	3,200	7,300	76,000	7,500
Ni	3,800		530	460	710	450	3,000	980
Pb	6,000	3,100	2,000	11,000	17,000	44,000	25,000	9,600
Pr	43,000	2,300	1,600	5,500	3,400	6,100	80,000	7,600
Rb	11,000	800	15,000	1,100	3,700	21,000	34,000	6,300
Re				12	29	39		24
Sb	290	180	260	1,200	1,300	1,100	440	520
Sc				330		440		380
Se	10	11	41	56	130	140	14	35
Sm	49,000	1,700	1,300	5,000	3,000	9,000	63,000	7,100
Sr	340	310	1,300	59	68	30	190	160
Tb	38,000		1,500	4,000	2,900	5,900	45,000	7,500
Th	31,000	34,000	5,500	14,000	9,300	34,000	250,000	25,000
Tl		1,900	3,400	1,500	2,000			2,100
Tm			2,600	4,300	2,500	7,600		3,800
U	610	1,500	1,800	5,700	12,000	44,000	3,300	4,000
V	630	430	1,400	610	670	1,800	1,100	840
W		210						
Y	21,000	1,500	2,000	3,700	2,400	3,500	40,000	5,000
Yb	18,000	3,300	1,900	2,800	1,800	7,400	28,000	5,300
Zn	10	2	19	5.4	–1	30	300	16
Zr	480	750	530	730	530	320	440	520

[a] Kd values could not be computed because soil or pore water concentrations were below detection limits, the Kd limit values are shown.

The native Kd values for I are substantially higher than those for Cl, but the spike values are similar. /Sheppard et al. 2006/ suggest GM Kd values for sand, loam, clay and organic soils of 8, 20, 10 and 80 L kg^{-1}, and a GSD of at least 10. These GM values are between the native and spike I Kd values here, but again the 95th percentile confidence bounds encompass most of the measured Kd values. The reason for the high native I Kd values may be the same as for Cl, because I again is reactive with organic materials. Additionally, the sorption of I in soil is redox dependent, in fact toxicity to I can occur in paddy soils that have been drained and are then reflooded /Sheppard and Motycka 1997/.

It seems there is a shift, perhaps temporary, in Kd when soil redox conditions change. The Kd for I is higher for aerated soils relative to the same soils flooded. Because several of the SKB soils appear to have been collected from wet environments, and then partially dried for handling and exposed to air, this may have caused higher Kd values than if Kd had been measured *in situ*. This may appear as an experimental artefact, but the counter argument is that if the soil is used in future for food production (in an assessment scenario), then the aerated Kd is more appropriate.

Thus, this evidence would suggest that if Kd values for Cl and I are important to the assessment, there may need to be special consideration in the choice of values, including perhaps more measurement. Higher Kd values would imply longer accumulation times and higher steady-state soil concentrations.

Table 4-2 is a comparison of the native Kd values for 51 elements from the 7 SKB soils versus those in a database of Kd for Canadian soils measured with the same methods /Sheppard et al. 2007/, and with the review /Thibault et al. 1990/ from which the Kd values in /IAEA 1994/ were obtained. In general, the SKB soils had lower Kd values than the Canadian soils, but were within the ranges observed for the Canadian soils and generally in the ranges from /Thibault et al. 1990/. Table 4-2 shows the 5th and 95th percentiles of the Canadian data, the geometric mean (GM), and geometric standard deviation (GSD) of the Canadian data, and the GM for the SKB soils.

Most meaningful are the last three columns. In these, the position of the SKB data within the Canadian data, expressed as a percentile, is given. Thus, for Al, the GM of the SKB soils was equal to the 13% percentile of the Canadian data: 13% of the Canadian data were lower and 87% were higher. On average, the GM of the SKB soils was at the 27th percentile of the Canadian data. This would normally be considered evidence that they belong to the same population of data.

There are elements where the SKB data were less than the 5th percentile or greater than the 95th percentile (Table 4-2). To put these “outlier” values into context, it is important to consider the elements relevant to nuclear fuel waste, highlighted in bold and colour (Re is highlighted because it was once considered an analogue for Tc). Among these elements, only Se had Kd values outside the 5th and 95th percentiles (they were below the 5th percentile), and the value for SKB soils was below the range from /Thibault et al. 1990/.

It is only speculative to assign a cause to this, but it may be because several of the SKB soils appear to have been collected from wet environments. Selenium is redox sensitive, and the partial drying and exposure to air of the samples of wetland soils may have caused a shift toward oxidized, soluble selenates resulting in a lower Kd value in the samples than in agricultural soils that are almost always well drained. The evidence would suggest that if Kd values for Se are important to the assessment, there may need to be special consideration in the choice of values, including perhaps more measurement. In contrast to Cl and I, the Se values are lower than expected.

Figure 4-1 further illustrates the comparison between the Kd values from the SKB soils and the larger database of Canadian soils. There seems no verifiable reason that the Kd values are below the median of the Canadian data. One difference that may be relevant is that most of the Canadian soils are agricultural, hence with low organic matter contents and subject to fertilization and tillage.

4.4 Conclusions

The Kd values for SKB soils generally fall in the range of Kd values measured in other soils. There were three possible exceptions of potential importance to assessment of nuclear fuel waste disposal. The elements Cl, I and Se had Kd values outside the ranges noted from other soils. The elements Cl and I were found to have high Kd values, which would suggest greater retention in soils. In contrast, the measured Kd value for Se was low, suggesting greater mobility.

Table 4-2. Comparison of native-element Kd values (L kg^{-1}) from 7 SKB sites to values measured using the same methods for 200 Canadian agricultural soils and to the survey by /Thibault et al. 1990/ which was the source for Kd values in /IAEA 1994/.

Element symbol	GM[a] of /Thibault et al. 1990/, the source for /IAEA 1994/	5th percentile of the Canadian soils	95th percentile of the Canadian soils	GM of the Canadian soils	GSD of the Canadian soils	GM 7 SKB sites	GM of 7 SKB soils as percentile of the Canadian soils	GM of sites A,D, F and G (clay tills) as percentile of the Canadian soils	GM of sites B and E (peats) as percentile of the Canadian soils
Al		1,300	510,000	26,000	6.6	4,300	13%	16%	11%
As		130	5,900	980	3.2	140	7%	7%	11%
Ba		40	2,200	210	3.6	27	1%	7%	< 0%
Be	250–3,000	120	1,000	400	3.5	320	33%	36%	31%
Ca	5–90	33	560	120	2.6	130	63%	55%	26%
Cd	40–800	250	4,000	1,400	2.5	230	5%	4%	6%
Ce	500–20,000	4,200	550,000	62,000	4.9	8,100	13%	24%	4%
Co	60–1,300	400	44,000	4,200	4.5	1,500	24%	31%	13%
Cr	30–1,500	460	22,000	3,400	3.5	350	2%	1%	8%
Cs	**270–4,600**	**2,500**	**290,000**	**41,000**	**4.4**	**38,000**	**40%**	**41%**	**53%**
Cu		230	4,700	1,300	2.5	780	27%	37%	40%
Dy		2,700	280,000	25,000	3.8	6,800	17%	30%	7%
Er		2,600	230,000	19,000	3.5	5,600	14%	28%	4%
Eu		1,800	76,000	8,000	3	3,800	24%	41%	7%
Fe	165–800	1,500	260,000	29,000	4.7	25,000	38%	58%	28%
Ga		880	110,000	13,000	4.4	5,500	30%	34%	18%
Gd		2,300	200,000	24,000	3.8	5,200	15%	28%	3%
Ge		180	3,600	1,000	2.6	520	26%	26%	27%
Hf	450–5,400	4,500	200,000	45,000	4.2	400	1%	1%	1%
Ho	250–3,000	2,300	300,000	19,000	3.8	7,700	23%	39%	8%
K		55	7,700	680	5.3	470	41%	44%	25%
La		3,600	280,000	46,000	4.3	7,200	14%	25%	4%
Li		49	3,500	370	4.1	280	45%	49%	40%
Lu		1,200	110,000	12,000	3.9	3,700	20%	28%	6%
Mg		53	2,400	310	3.5	300	55%	40%	56%
Mn	50–750	110	150,000	3,100	12	1,700	49%	52%	32%

Element symbol	GM[a] of /Thibault et al. 1990/, the source for /IAEA 1994/	5th percentile of the Canadian soils	95th percentile of the Canadian soils	GM of the Canadian soils	GSD of the Canadian soils	GM 7 SKB sites	GM of 7 SKB soils as percentile of the Canadian soils	GM of sites A,D, F and G (clay tills) as percentile of the Canadian soils	GM of sites B and E (peats) as percentile of the Canadian soils
Mo	10–125	22	550	110	2.8	810	97%	93%	> 100%
Na		2.1	620	32	6.4	19	48%	40%	58%
Nb	160–2,000	1,400	440,000	32,000	5.9	10,000	25%	40%	34%
Nd		3,400	310,000	39,000	4.3	7,500	15%	30%	3%
Ni	300–1,100	290	6,200	1,700	2.6	980	21%	33%	14%
Pb	**270–22,000**	**2,400**	**140,000**	**19,000**	**3.4**	**9,600**	**26%**	**41%**	**20%**
Pr		4,000	270,000	42,000	4.1	7,600	15%	27%	3%
Rb	55–670	950	61,000	8,200	4.3	6,300	37%	51%	10%
Re		**2.6**	**250**	**29**	**4**	**24**	**41%**	**41%**	**46%**
Sb	45–550	100	1,800	440	2.5	520	57%	63%	53%
Sc		230	1,100	650	2.1	380	15%	15%	
Se	**150–1,800**	**49**	**3,100**	**340**	**3.7**	**35**	**3%**	**2%**	**3%**
Sm	245–3,000	2,800	250,000	31,000	4.1	7,100	16%	34%	4%
Sr	**15–150**	**36**	**2,800**	**250**	**3.7**	**160**	**39%**	**27%**	**37%**
Tb		2,100	250,000	18,000	4.5	7,500	29%	43%	11%
Th	**3,200–89,000**	**3,300**	**260,000**	**38,000**	**4.1**	**25,000**	**34%**	**47%**	**28%**
Tl		1,400	55,000	8,400	3.2	2,100	12%	6%	12%
Tm		1,500	330,000	11,000	4.5	3,800	21%	32%	11%
U	**15–1,600**	**25**	**10,000**	**410**	**7.4**	**4,000**	**83%**	**85%**	**84%**
V		1,600	23,000	5,500	2.3	840	1%	1%	0%
Y		2,000	190,000	20,000	3.8	5,000	17%	29%	4%
Yb		2,300	200,000	17,000	3.6	5,300	15%	33%	6%
Zn	200–2,400	49	4,900	280	4.1	16	0%	2%	< 0%
Zr	600–7,300	36	150,000	2,200	18	520	36%	34%	37%
Average							27%	32%	21%

[a] The values given are the range in GM among sand, silt, clay and organic soils.

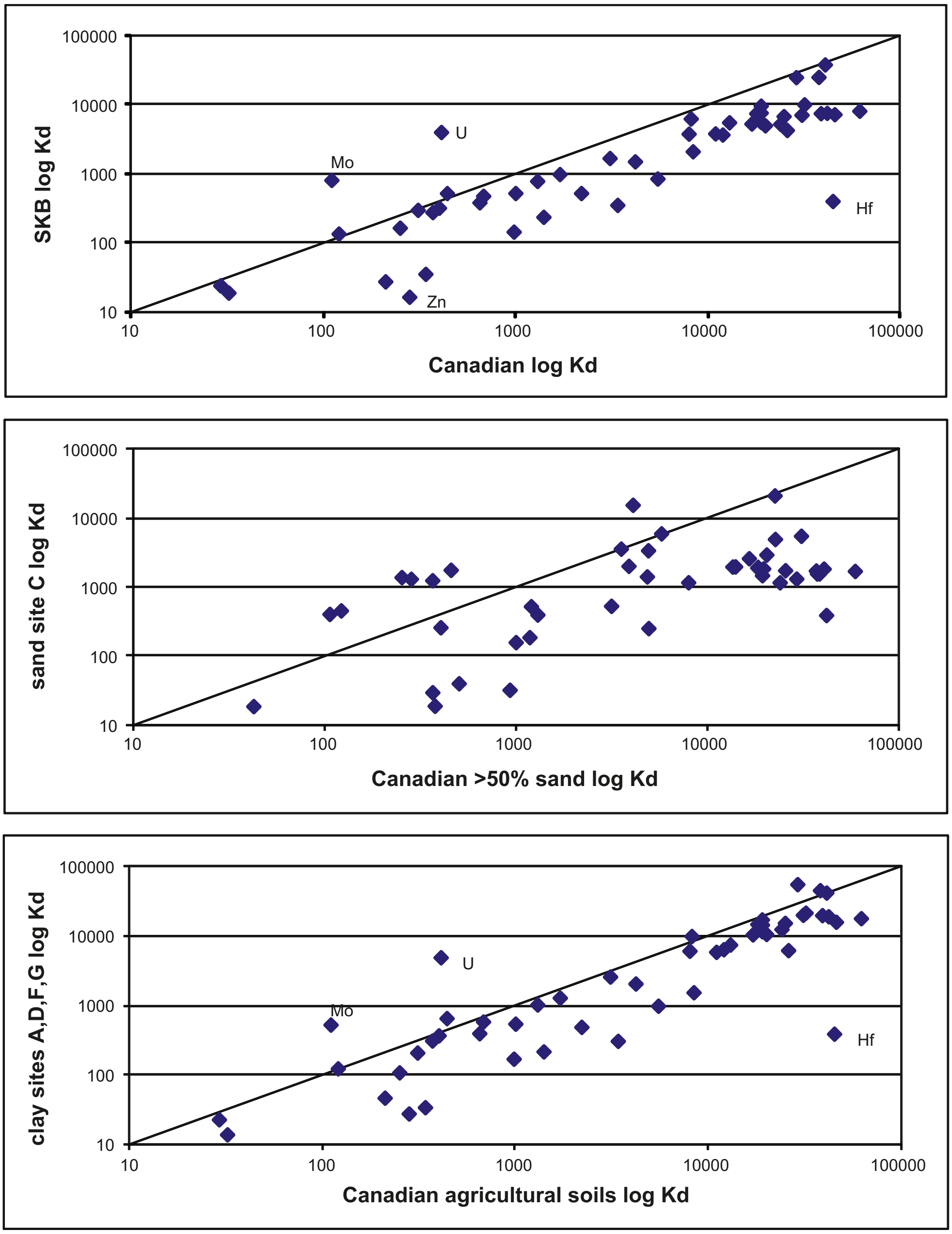

Figure 4-1. *Geometric mean Kd for 51 elements, plotting data from SKB sites versus data measured in the same way on up to 200 Canadian soils. The upper plot is 7 SKB soils versus all the Canadian soils, the middle is the one sandy SKB soil versus the sandy Canadian soils, and the lower is the 4 clay SKB soils versus all the Canadian soils.*

5 Comparison of soil Kd values to literature and equations to estimate soil Kd for use in risk assessment

5.1 Introduction

Soil solid/liquid partition coefficients (Kd, L kg^{-1}) are presented for the elements Ce, Cl, Co, Cr, Cs, Fe, Ho, I, La, Mn, Mo, Nb, Nd, Ni, Np, Pa, Pb, Pu, Ra, Sb, Se, Sm, Sn, Sr, Tc, Th, Tm, U and Yb, and these Kd values are to be representative of the soils present at Forsmark and Simpevarp. Values of Kd for many, but not all, of these elements have been measured in 7 soils from Forsmark and Simpevarp (the previous section), mostly as desorption of indigenous element.

Most of the above elements have very long-lived isotopes, and for the long-term assessment of nuclear fuel waste, it makes sense to select Kd values that represent steady state conditions, such as Kd values measured by desorption of indigenous element. However, some of these elements have only short-lived isotopes and are not progeny of long-lived decay chains. For these, it is relevant to consider short-term reactions, as discussed for each element in the following section.

5.1.1 Limits on effectiveness of Kd values

An additional aspect of soil Kd values is that there are minimum and maximum values that will have a significant effect in an assessment, depending on the time scale of the assessment /Sheppard 2005a/. Values of Kd below about 0.1 L kg^{-1} are indicative of high mobility, and variation of tenfold around this value, and especially lower, will not have a large effect on assessment results because the element is essentially as mobile as water.

Thus, it is not productive to create a detailed predictive model for Kd in this range. Similarly, there is an upper limit Kd beyond which the element can be considered essentially immobile for a specific assessment time frame. Thus, it is again not productive to create detailed predictive models for high values, in the order of 10^4 L kg^{-1} and above. In this report, Kd values below and above these limits are reported, but with less emphasis on predictive equations.

5.1.2 Sources of data

There are several general sources of Kd values to draw upon for application to the SKB sites, each with advantages and disadvantages.

1. For a subset of the elements (Ce, Cl, Co, Cr, Cs, Fe, Ho, I, La, Mn, Mo, Nb, Nd, Ni, Pb, Sb, Se, Sm, Sr, Th, U and Yb), Kd was measured using the indigenous element concentrations in 3 soils from the Forsmark area and 4 from the Simpevarp area (data shown in the previous section). These are obviously ideal, except that 7 soils may not be enough to sufficiently characterise the sites. Additionally, these values represent desorption of indigenous elements. Contaminants from a waste facility will be less strongly sorbed initially (lower Kd), but in the long term (centuries to millennia) the Kd values should converge with those of the indigenous elements as measured in these soils.
2. For two elements (Cl and I), Kd was measured using artificially spiked element concentrations in 3 soils from the Forsmark area and 4 from the Simpevarp area (data shown in the previous section). These are short-term sorption Kd values and were substantially different from those measured by desorption of indigenous elements (see 1, above), and so are of uncertain value.
3. For a series of these elements (Cl, I, Np, Ra and U), recent extensive literature reviews have been completed for Ontario Power Generation /Sheppard et al. 2006/, and the data sets generated (named OPG in this report) for these were available for further interpretation. These Kd values span a wide range in soil properties, but also a wide range in experimental methods.

4. For the same subset of elements as in 1 (above) and measured by the same laboratory, there is a data set (named NTE in this report) of Kd values for about 200 soils. Although the number of soils allows for development of some predictive capability, the soils were almost all production agriculture soils, and so most were mineral soils with relatively low organic carbon contents and near-neutral pH. It can be argued that a critical group or reference individuals will focus food production on such soils, but these types of soils do not dominate spatially at Forsmark and Simpevarp.
5. For Tc, there is a data set of Kd values for 34 soils that were predominantly organic /Sheppard et al. 1990/. Although limited because they include few mineral soils, the results are fairly conclusive that there are two Kd values important for Tc: one very near zero for aerobic soils and a somewhat higher value for anaerobic soils.
6. For most of these elements, soil Kd values are available from older compendia. Perhaps the most widely cited is /IAEA 1994/, which was derived from /Thibault et al. 1990/ and /Sheppard and Thibault 1990/, and the data set from the latter is available for further interpretation. These span a wide range in soil properties, but also a wide range in experimental methods.
7. The EMRAS program of the IAEA will replace the /IAEA 1994/ report with a new report, and will have a revised set of Kd values. A pre-publication list of values is available for comparison here (personal communication, S. Fesenko, IAEA).
8. The ongoing scientific literature is a diffuse source of further Kd values, again these span a wide range in soil properties, but also a wide range in experimental methods. Reference citations are given in each section below.

There is a balance to be achieved between deriving site-specific parameter values and utilizing as much of the larger global population of Kd values as possible. /Sheppard 2005b/ concluded from meta-analysis that as more transfer data become available, with more and more information to categorise these data, an asymptote in uncertainty is approached. This is not a surprising result; in geospatial problems this is analogous to a semi-variogram. For Kd, /Sheppard 2005b/ suggested an asymptotic geometric standard deviation (GSD) of about 5-fold, based on the data from /Thibault et al. 1990/. The implication is that doing detailed site-specific measurements of Kd is not likely to improve on this level of uncertainty, so an argument to focus an assessment on site-derived data to the exclusion of other data is not well founded.

It is very relevant to include off-site literature values in the choice of Kd for a given assessment. However, this does not exclude refining the choice of values to represent soil properties relevant to the site. Thus, in this report, where possible, multiple regression equations were used to summarise Kd values from several sources, and these were subsequently used to derive best estimates for the 7 sites sampled at Forsmark and Simpevarp. It is envisioned that these equations could be used to derive best estimates for other soils at these sites, bearing in mind that there is an inherent variability in Kd values on the order of 5-fold, so that best estimate Kd values within about 25-fold would not be significantly different (for example by t test at infinite degrees of freedom).

5.1.3 Soils from the SKB sites

The soils sampled as described earlier in this report were sent to a laboratory in Canada for measurement of Kd, and as a result were analysed for mineral particle size fractionation, organic carbon content and pH (Table 5-1). Some of these measurements from laboratories in Sweden were previously reported (Table 3-3), and in general the two sets of values agree. The values in Table 5-1 were used for regression analysis because they were done in the same laboratory as the NTE soils.

Table 5-1. Properties of soils from Forsmark and Simpevarp (see Tables 3-1 to 3-3 for further data).

Site	ID number	Quaternary deposit	Environment	Clay (%)[1]	Organic carbon (%)[2]	pH[3]
A	AFM001076	clayey silty till	fen, mixed forest	9	0.29	6.5
B	PFM006024	peat (*Spagnum*)	fen, *Pinus* forest	57	56	3.5
C	ASM001426	sandy till	oak forest	1	1.7	5.0
D	ASM001434	clay gyttja	fen, alder forest	27	21	5.0
E	ASM001440	peat	former fen, spruce forest	29	28	4.0
F	PSM000277	clay gyttja	open fen	26	20	5.0
G	PFM002670	clayey silty till (with mull)	arable land	5	1.8	5.5

[1] determined by ALS Laboratories, hydrometer method on the mineral fraction only.
[2] determined by ALS Laboratories, wet oxidation for sites A, C, and G and loss on ignition for sites B, C, D, and E.
[3] determined by litmus paper on soil water paste allowed to stand for > 30 minutes.

5.2 Methods

5.2.1 Development and use of multiple regression equations

Where possible, regression equations were developed to enable prediction of best-estimate Kd values for a specific set of soil properties. The dependent variable was $\log_{10}$(Kd), with the exception of Kd for Tc and Cl where it was $\log_{10}$ (Kd+1)[1]. The statistical method to derive predictive equations reported here is forward stepwise multiple regression, using Statistix-9 software (Analytical Software, Tallahassee, FL, www.statistix.com).

In this method, the user defines the dependent variable and a series of potential independent variables. The process begins by including the one independent variable that explains the largest portion of the variance in log Kd. It is included only if it's corresponding regression coefficient is statistically significant at $P < 0.05$. In the next step, the next most useful independent variable is included, again only if it's corresponding regression coefficient is statistically significant.

This proceeds until no further independent variables significantly contribute to explaining the variance in log Kd. At each step, if the inclusion of a new independent variable renders the coefficient of a previously included variable non-significant at $P < 0.05$, the non-significant variable is removed from the regression – effectively it has been made redundant. In the end, the regression equation may contain anywhere from none to all the independent variables, with only those that are statistically significant remaining in the equation.

Interactions among independent variables are not implicitly included in the stepwise regression process. For soil Kd, pH and clay content are often the most influential variables, and there is an inherent interaction between them in that acidic soils are often low clay content soils (sands). Experience with the NTE data set showed that it was often useful to consider the clay-content · pH interaction in the regressions for Kd. This was done by creating a new independent variable as the product clay · pH (the units of this product are pH %).

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[1] The reason for the exception with Tc and Cl is that a Kd of zero is among the expected values for aerobic soils, and statistically this means that some measured values may be negative. A negative Kd is theoretically impossible, but if the amount sorbed is estimated by the difference in solution concentration after contact with soil, then because of normal experimental variability on occasion more Tc will be measured in solution than was initially added, and it is statistically correct to represent this as a negative Kd. Thus, using log (Kd+1) allows log transformation of almost all near-zero Kd data. This approach was published by /Sheppard et al. 1990, Sheppard 2003/.

There is potential to include a large number of independent variables, however to be practical these should be limited to parameters commonly measured both in literature studies and in soils at the SKB sites. In all the data sets, the minimum list of independent variables was:

- soil pH (generally using pH determined in distilled water rather than in dilute salts, primarily because it is more commonly reported, the pH in water is usually about 1 pH unit higher than that in dilute salt),
- soil clay content (expressed as a percent of the mass of the mineral content of the soil, which represents mineral soils well but has less value for organic soils that have very low mineral contents),
- soil organic carbon (expressed as a percent of the mass of the whole dry soil, and lower than soil organic matter content by a factor of about 1.72),
- the product of pH · clay-content.

To illustrate the regression process, below is the development of the equations for log(Kd) for Cs. In this example, a 'dummy' variable was used to differentiate sorption data (dummy = 1) from desorption data (dummy = 0). This variable was 'forced', and so the first iteration of the regression process gave the equation:

$\text{Log(Kd)} = 4.62 - 1.40\cdot(\text{dummy})$

where the coefficient –1.40 was significant at $P < 0.0001$.

In the second iteration, the pH · clay interaction was the next most important independent variable and so was added to the equation as:

$\text{Log(Kd)} = 4.29 - 1.28\cdot(\text{dummy}) + 0.0020\cdot(\text{pH})\cdot(\text{clay})$

where both coefficients –1.28 and 0.0020 were significant at $P < 0.0001$.

The third iteration included soil pH as the next most important independent variable, and this iteration did not remove any of the previous independent variables:

$\text{Log(Kd)} = 3.817 - 1.248\cdot(\text{dummy}) + 0.00154\cdot(\text{pH})\cdot(\text{clay}) + 0.0798\cdot(\text{pH})$

where the coefficient –1.248 was significant at $P < 0.0001$, 0.00154 was significant at $P = 0.0004$ and the coefficient 0.0798 for soil pH was just significant at $P = 0.013$.

The fourth interation found no other independent variables that would have statistically significant coefficients, and did not identify any of the previously entered variables that were now redundant. Clay content was the next closest, but the level of P for its coefficient if it was entered was $P = 0.076$, higher than the cut-off value of $P = 0.05$. Thus, the final equation was that derived in the third interation (above). This equation had an $r^2 = 0.41$. As will be discussed below in the section on Cs, it was decided to average the results of sorption and desorption experiments, so the value of 'dummy' was set to 0.5, and the regression constant became $3.817 - 1.248\cdot(0.5) = 3.19$.

Not all the Kd values had these independent variables explicitly measured in the manner required. Where possible, surrogate data were used and interpreted into the required units. For example, the report of a soil being a 'clay' or 'clay-loam' soil implies soil clay content of > 28%. Relative few soils have clay contents > 50%, so when only the text description was available the value used to represent soils reported as clay or clay-loam was 30%. Similarly, soils reported as silts or loams were assumed to have 15% clay, and those reported as sands: 5% clay.

In absence of pH data, a value of 6.5 was assigned, as representative of a global average pH. In absence of soil organic carbon or organic matter data, soils described as mineral were assumed to have 2% organic carbon and those described as organic: 30% organic carbon. In regression analysis, it is preferred to have a distribution of values for the independent variables, not a lot of repeated values. We investigated the impact of these assumed values using Kd for I as the test case.

The ‘OPG’ Kd values were first assigned the assumed values of pH, clay and organic carbon contents based on the soil descriptions in the report /Sheppard et al. 2002/, and regressions were developed. Then the original papers were re-reviewed and the actual pH, clay and organic carbon contents were obtained. Regression equations using the real data were very similar to those using the assumed values in terms of the independent variables included in the final equation and the signs and general values of the regression coefficients. Obviously, using assumed values for independent variables is not ideal, but it is important to not lose Kd data from the regression because Kd data are not abundant, and our experience indicates that the assumed values of pH, clay and organic carbon contents do not mislead.

For some of the data sets, soil silt content and sand content were available, and these were included as independent variables. Although silt and sand contents occasionally have predictive capacity, it must be remembered that soil clay, silt and sand contents sum to 100%, so these variables inherently interact. When the situation arises, it is probably better to use clay content alone than the combination of silt and sand contents, even with a small loss in predictive capacity, just because clay content is more commonly measured and reported.

A best-fit equation is not universally relevant. Ideally, it should only be used to interpolate Kd values, that is to be used for predictions only within the range of the independent variables of the ‘training’ data set (the data set used to derive the equation). Even for interpolation, the relevance may be in question if the frequency distributions and interactions of the independent variables in the training data set differ from those where predictions are needed. However, there is also a need to cautiously extrapolate using regression equations.

For either interpolation or extrapolation, it is important to compare predictions to measurements from a ‘validation’ data set. In this case, the measured Kd values in the 7 SKB soils are a validation data set. If there is agreement, approximately within the 25-fold confidence bounds expected for Kd, then there is argument to concatenate the training and validation data set and develop new overall regression equations that encompass all the available data.

In reporting the regression equations, two statistical quantities are also provided here. First is the coefficient of determination, r^2, as a fraction of the variance explained by the model. Second is the residual GSD, which is the antilog of the square root of the mean square error from the regression model of log(Kd) values. The residual GSD is a measure of the variance in Kd not explained by the independent variables, and would be appropriate for defining the statistical dispersion for stochastic modeling.

5.2.2 OPG data sets

/Sheppard et al. 2006/ summarised several reviews of Kd done for Ontario Power Generation (OPG). These reviews simply reported GM and GSD, with little attempt to modify or categorise the values for soil properties. To apply this to the SKB sites, the underlying literature was revisited and data for soil properties were obtained.

5.2.3 Canadian NTE Kd data set

/Sheppard et al. 2007/ reported the development of a Kd data set for agricultural soils in Canada, and it has expanded since that time to include over 200 soils, called here the ‘NTE’ soils. Because the results are referenced throughout this report, it is appropriate to examine the range of properties of these soils, and the correlations among the independent variables (Figure 5-1). Among these soils, clay content, pH and organic carbon content were all significantly ($P < 0.05$) positively correlated, but the correlation coefficients were all < 0.3. The domains of these variables are fairly broad relative to most agricultural soils, but clearly organic soils are not well represented.

5.2.4 Thibault et al. data set

The data set prepared by /Thibault et al. 1990/ and subsequently published by /Sheppard and Thibault 1990/ was the sole source of Kd data for the /IAEA 1994/ compendium. The IAEA compendium also included interpolation and extrapolation to derive estimates of Kd using plant/soil concentration ratios as a guide. The data set of /Thibault et al. 1990/ has been re-entered into computer files to allow regression analysis for the SKB sites. The OPG data sets included all the Thibault et al. data that were still considered relevant, and so care was taken to not double account for these data in the regression analyses.

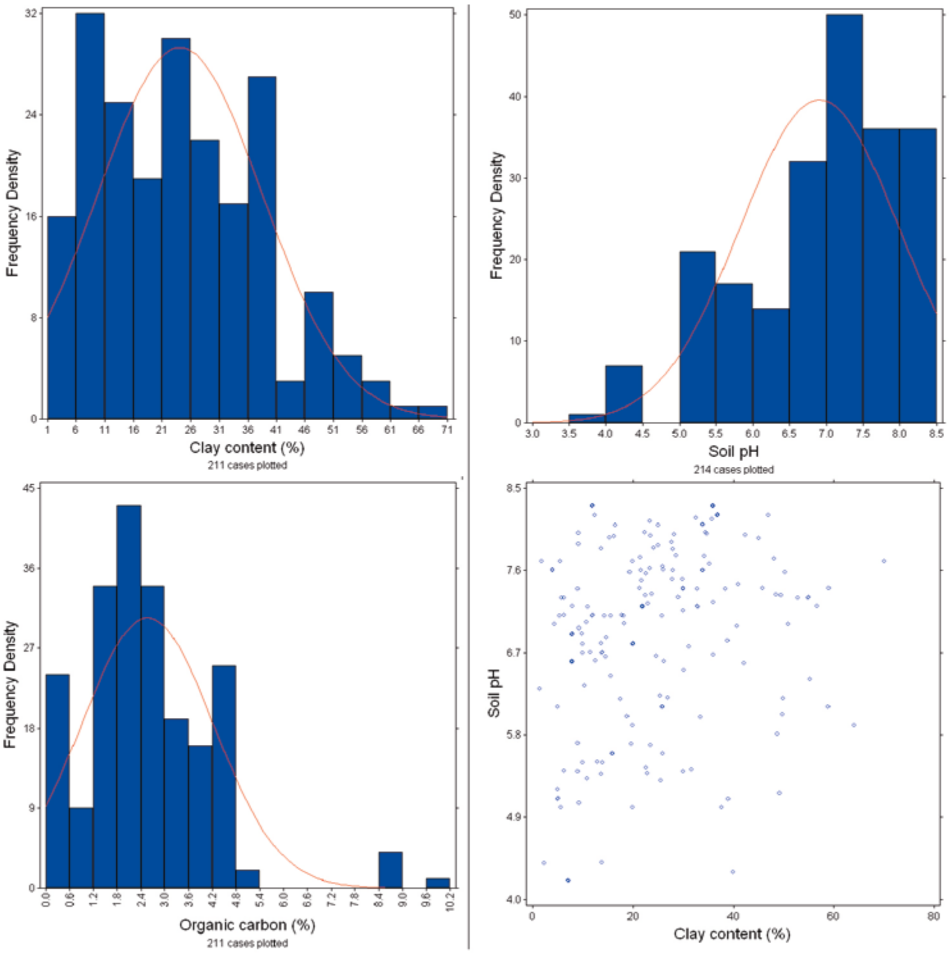

Figure 5-1. Frequency histograms for clay content, soil pH and organic carbon content for the NTE soils. Also shown is the scatter plot of pH versus clay content.

5.2.5 Newly compiled literature

In addition to the established data sets, a review was done of the recent literature. This was targeted to be of varying intensity. For well reported 'nuclear' elements such as Cs, Th and elements where recent reviews were already available (the OPG data sets and others), it was considered not necessary to do detailed further searches. For these elements, only particularly important recent papers were acquired. For most of the metals relevant to other industries, such as Cr, Ni and Pb, there is a lot of data and the search emphasis was on papers that dealt with a series of soils, or that presented predictive equations. For some elements, notably Ce, La, Nb, Nd, Sm and Yb, there is scant literature and it was necessary to be as exhaustive in the search as possible.

5.2.6 EMRAS data set

The report /IAEA 1994/ is undergoing revision, under the EMRAS program. The detailed data set compiled by EMRAS had been made available to date (October, 2008), and the best estimates and ranges were just recently made available for comparison only. Almost all values have been revised from the /IAEA 1994/ report (Figure 5-2). Comparison to the equations presented in this report was done by computing estimates for soils as described by EMRAS. The following properties were assumed to represent the EMRAS soils: All soils – clay 15%, pH 6.5, organic carbon 2%; Sand soils – clay 5%, pH 5.5, organic carbon 0.5%; Loam soils – clay 15%, pH 6.5, organic carbon 2%; Clay soils – clay 30%, pH 6.5, organic carbon 2%; Organic soils – clay 15%, pH 5.5, organic carbon 30%.

Note that there is in general a tenfold variation about the 1:1 line in Figure 5-2. The median GSD for the EMRAS data was 4, implying at least a 16-fold difference is needed to declare values significantly different. Obviously, half of the elements were even more variable than this.

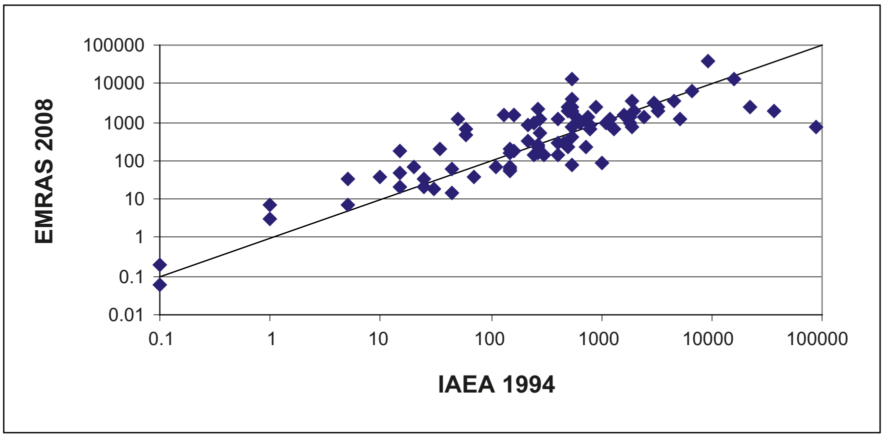

Figure 5-2. *Comparison of EMRAS values and /IAEA 1994/. Each point is a different element and soil type. Note that /IAEA 1994/ was based on the /Thibault et al. 1990/ used in this report, along with interpolation.*

5.3 Results

5.3.1 Presentation

Each element is discussed separately in the following sections, ordered alphabetically by the element symbol. For each, there is a table showing, where possible, the Kd values for samples of the 7 SKB sites. The predictions for these 7 sites using regression equations developed from the major data sets are shown, as a range 1 GSD above and below the actual estimate. Finally, in some cases predictions from other literature equations are shown.

Coloured fonts for the Kd values for the SKB sites denote the comparison to the data sets: pink indicates the observed SKB data were higher than 1 GSD above all the data set estimates, and blue indicates the observed SKB data were lower than 1 GSD below the data set estimates. These values are also in a box. Any values not marked in this manner were within 1 GSD of the predicted values.

Under the tables is a discussion, as for almost every element there were features that needed explanation or further development. Finally, based on the discussion, a proposed final equation, which does not include the results of the SKB sites, is presented.

5.3.2 Ce – Cerium

The Kd values for Ce were measured in the 7 SKB soils and in 209 Canadian soils in the NTE program (Table 5-1). The stepwise regression of the NTE soils included soil pH, sand content, silt content and the clay*pH interaction, with a residual GSD of 3.5.

Table 5-1. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Ce.

SKB soil	Measured Kd	NTE predicted Kd ± 1GSD
A Forsmark clay till	39,000	17,000–210,000
B Forsmark deep peat	4,100	980–12,000
C Simpevarp sandy till	1,700	3,100–37,000
D Simpevarp clay gyttja	5,900	2,800–34,000
E Simpevarp peat	3,800	1,200–14,000
F Simpevarp clay gyttja	4,400	3,200–38,000
G Forsmark clay till	88,000	5,600–67,000

To contrast with the above desorption Kd values, there have been some measures of sorption Kd. /Schimmack et al. 1987/ measured Kd every 3 m along a 150-m transect, using 20-d contact times with ^{139}Ce in a soil suspension. The median Kd was 310 L kg^{-1}, a range of 170–740 L kg^{-1}, all lower than the desorption Kd values above but within the 95th percentile confidence bounds based on a GSD of 3.5. Although these authors reported that equilibrium had been established, one could argue that it was not in equilibrium in the pedogenic time scales of millennia, and larger Kd values would apply for long-term assessment. /EMRAS 2008/ found 5 reports giving 11 Kd values with a GM of 1,200 L kg^{-1}, range of 120–20,000 L kg^{-1}, and GSD of 5.

However, the longest lived isotope, ^{144}Ce, is generated from fission reactions, and has a half life of 285 d. Thus, it is probably not a long-term hazard even from nuclear fuel waste, and so the value may not be critical to an assessment. Note that stable Ce is relatively abundant.

Soils from SKB sites C and G were outside the ±1 GSD range of the regression estimates (see above), but are both within the ±2 GSD range, thus within the 95th percentile confidence bounds. This includes soil B whose properties are well beyond the range in the NTE soils. Because the values are large, assessment results may not be strongly dependent on them, and so either the values measured in the SKB soils or predictions from the equations should be suitable for assessment purposes. The equation from the NTE data set is:

$$\text{Log(Kd)} = 3.51 + 0.54400 \cdot (\text{pH}) - 0.022 \cdot (\text{sand}) - 0.022 \cdot (\text{silt}) - 0.00471 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 3.5 and $r^2 = 0.27$. This gives values for the SKB soils of 59,000, 3,400, 11,000, 1,000, 4,200, 11,000 and 19,000 L kg^{-1}, respectively.

5.3.3 Cl – Chlorine

The Kd values for Cl were measured in the 7 SKB soils both using desorption of indigenous Cl and sorption of spiked stable Cl (Table 5-2). Additionally, there was an extensive literature search (OPG, /Sheppard et al. 2006/ and references therein), but this only yielded 7 other Kd values. These values were in either of two groups: mineral soil or organic soil.

The desorption Kd values were all higher than the corresponding sorption Kd values, consistent with the concept that less soluble forms may become more prominent with time. Some of this may be because of biological incorporation by soil microbes or by the plants of Cl into recalcitrant organic compounds, which then persist in the soil /Bastviken et al. 2007/ and related papers). The overall lack of data makes it necessary to combine all the SKB and literature desorption Kd values available (ignoring the sorption Kd values above), and the resulting GM values[2] are:

1.4 L kg^{-1} for mineral soils (< 20% organic carbon, Kd based on 11 soils) and,

150 L kg^{-1} for organic soils (> 20% organic carbon, Kd based on 3 soils).

Table 5-2. Measured (SKB) and predicted (using the OPG data set) Kd values (L kg^{-1}) for Cl.

SKB soil	Measured desorption Kd	Measured sorption Kd	OPG GM Kd (±1 GSD where GSD = 5)
A Forsmark clay till	4.4	0.0017	0.06–1.4
B Forsmark deep peat	37	3.5	440–11,000
C Simpevarp sandy till	7.1	0.76	0.06–1.4
D Simpevarp clay gyttja	0.036	1.1	see footnote[1]
E Simpevarp peat	41	2	440–11,000
F Simpevarp clay gyttja	5.2	2	see footnote[1]
G Forsmark clay till	12	0.24	0.06–1.4

[1] Sites A, C and G are clearly mineral soils and so the mineral-soil Kd is shown, sites B and E are clearly organic soils and the organic-soil Kd is shown. Sites D and F have just high enough organic carbon contents to be considered organic, but it is not clear which Kd values apply.

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[2] Note that because the Kd value for mineral soils approaches zero, and the data set included 3 zero values, the GM was computed from:

$$GM = \left\{10^{average[\log(Kd+1)]}\right\} - 1$$

/EMRAS 2008/ found 4 reports giving 22 Kd values with a GM of 0.3 L kg^{-1}, range of 0.04–0.9 L kg^{-1}, and GSD of 3.

Because of the very large uncertainty, more effort may be required to measure Kd for Cl, and a larger GSD is appropriate, perhaps about 10 (approximately the highest GSD generally found for Kd values).

5.3.4 Co – Cobalt

The Kd values for Co were measured in the 7 SKB soils and in 208 Canadian soils in the NTE program (Table 5-3). The stepwise regression of the NTE desorption Kd data included soil pH, sand content, silt content and the clay*pH interaction, with a residual GSD of 4.0. The stepwise regression of the /Thibault et al. 1990/ sorption Kd data included soil pH, organic carbon and clay, with a residual GSD of 7.2, see also Figure 5-3.

/EMRAS 2008/ found 8 reports giving a total of 118 Kd values, GM values were sand: 260 L kg^{-1}, loam: 810 L kg^{-1}, clay: 3,800 L kg^{-1} and organic: 87 L kg^{-1}. EMRAS also noted a low Kd for soils with $pH \leq 5$ of 12 L kg^{-1}.

In general, the Kd results from desorption of indigenous Co (SKB, NTE, /Gooddy et al. 1995/ and /Watmough et al. 2005/) agreed with those for sorption of radiocobalt /Thibault et al. 1990/, see Figure 5-3. The largest discrepancies were for SKB sites A, C and G which were the mineral soils, interesting because both the desorption and sorption data sets were strong in numbers of mineral soils and weak in numbers of organic soils. This suggests a more distinct aging effect in mineral soils than in organic soils. However, the 95th percentiles (2 GSD above and below the GM, note that Figure 5-3 shows 1 GSD above and below the GM) overlap. Thus, combining the NTE and Thibault et al. data sets is reasonable (n = 342) and by stepwise regression this gives the equation:

$$\mathrm{Log(Kd)} = 0.898 + 0.339 \cdot (\mathrm{pH}) + 0.0267 \cdot (\mathrm{organic\ carbon})$$

with a residual GSD of 8.4. This gives values for the SKB soils of 1,300, 3,800, 440, 1,400, 1,000, 1,300 and 650 L kg^{-1}, respectively.

5.3.5 Cr – Chromium

The Kd values for Cr were measured in the 7 SKB soils and in 112 Canadian soils in the NTE program (Table 5-4). The stepwise regression of the NTE soil included clay content and the clay*pH interaction, with a residual GSD of 3.1.

There is a marked difference between the desorption Kd values from the NTE study and the sorption Kd values reflected in the /Thibault et al. 1990/ and recently acquired literature data sets /Covelo et al. 2004, 2007ab, De Groot et al. 1998, Fulekar and Dave 1992, Hellerich and Nikolaidis 2005, Sheppard and Hawkins 1991, Vega et al. 2006/. The sorption Kd values may be lower because the recently applied Cr has not reached full equilibrium with the indigenous Cr (and may not for a very long time). Given that the longest-lived Cr isotope (^{51}Cr) has a half-life of 27.7 d, long-term equilibrium in soil is not relevant. Thus, the sorption data sets are the most relevant. /EMRAS 2008/ found 6 reports and 31 values, GM values were sand: 8.4 L kg^{-1}, loam: 45 L kg^{-1}, clay: 14 L kg^{-1} and organic: 160 L kg^{-1}, and these are also more consistent with the sorption data sets.

Table 5-3. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Co.

SKB soil	Measured desorption Kd	NTE predicted Kd ± 1GSD	Thibault et al. predicted Kd ± 1GSD
A Forsmark clay till	3,800	960–15,000	17–850
B Forsmark deep peat	1,000	260–4,200	5,600–290,000
C Simpevarp sandy till	1,100	270–4,300	3.7–190
D Simpevarp clay gyttja	1,700	420–6,700	99–5,100
E Simpevarp peat	1,100	260 –4,200	100–5,100
F Simpevarp clay gyttja	1,900	470–7,500	84–4,300
G Forsmark clay till	1,800	440–7,100	6.9–350

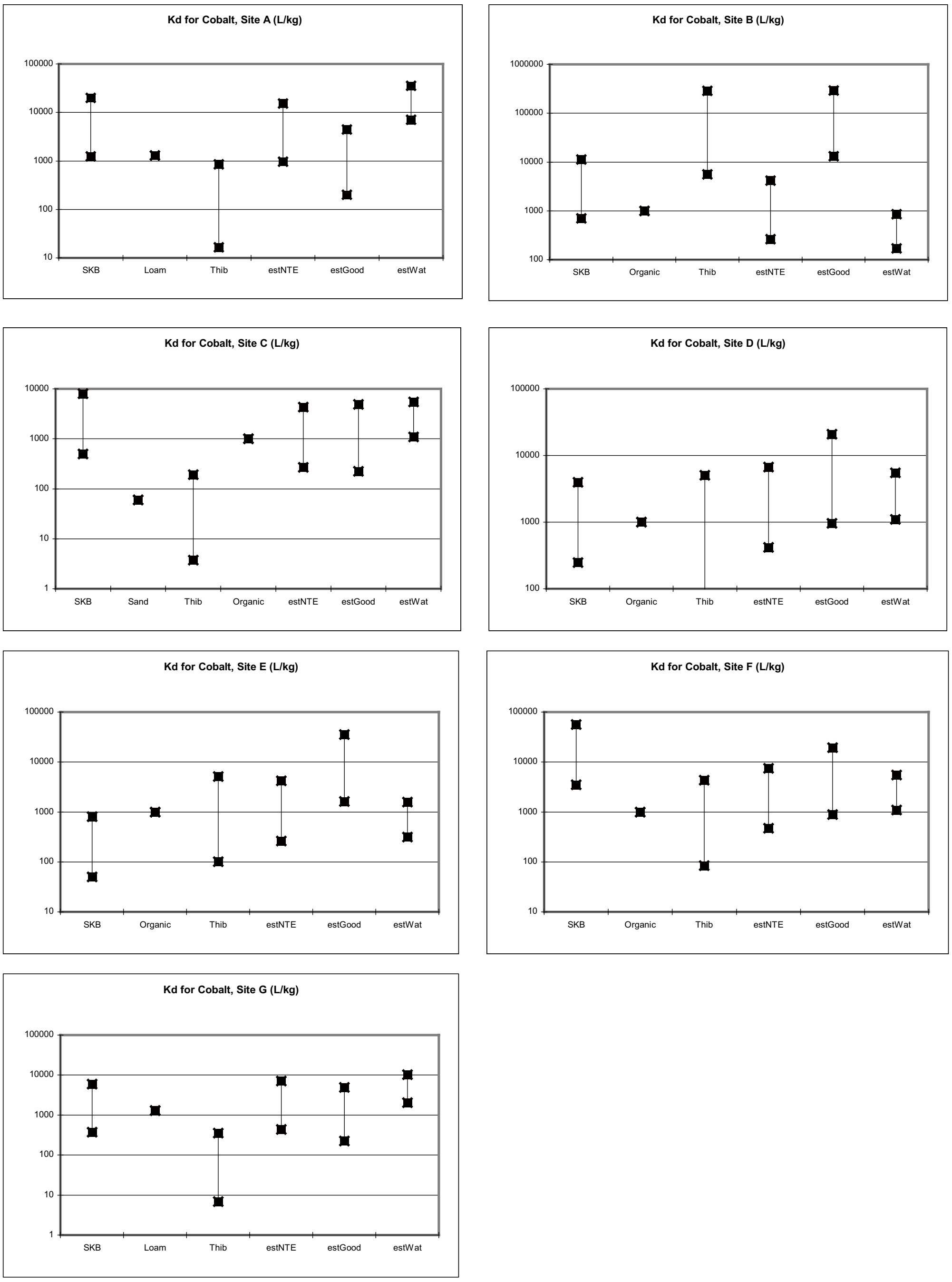

Figure 5-3. *Comparison of Kd from the 7 SKB soils (±1 GSD where GSD = 4.0) versus corresponding estimates, where sand, loam and organic are compendia recommendations from /Sheppard and Thibault 1990/, Thib is regression estimates from the /Thibault et al. 1990/ data set, estNTE is regression estimation from the Canadian NTE soils, estGood is regression estimation from the data of /Gooddy et al. 1995/, and estWat is regression estimation from /Watmough et al. 2005/. Variation on regression estimates correspond to the residual GSD.*

Table 5-4. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Cr.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. and literature sorption predicted Kd ± 1GSD
A Forsmark clay till	140	1,400–13,000	20–8,900
B Forsmark deep peat	900	67,000–630,000	98–43,000
C Simpevarp sandy till	250	1,500–14,000	14–5,900
D Simpevarp clay gyttja	430	3,500–33,000	28–12,000
E Simpevarp peat	370	8,800–82,000	28–12,000
F Simpevarp clay gyttja	480	4,200–40,000	27–12,000
G Forsmark clay till	270	1,600–15,000	15–6,700

/Thibault et al. 1990/ had relatively few (22) values for Cr. The 19 values for Cr(III) had a GM of 160 L kg^{-1} (GSD = 9.0), and the 3 values for Cr(VI) had a GM of 5.9 L kg^{-1} (GSD = 6.6). These were combined with more recent literature sources resulting in 135 values. Among these, there was an apparent difference in relationship to pH and clay content for low and high Kd values (shown for pH in Figure 5-4). Because speciation of Cr between relatively immobile Cr(III) and the mobile and ephemeral Cr(VI) is difficult to determine or predict in soil, there is a good possibility that the two apparent populations of data reflect the two species of Cr. Assuming this, the stepwise regression of the Kd values above 100 L kg^{-1} (83 values) gave the equation:

$$\text{Log(Kd)} = 1.79 + 0.287 \cdot (\text{pH})$$

with a residual GSD of 5.2 and $r^2 = 0.21$. This gives values for the SKB soils of 4,500, 2,000, 280, 590, 590, 560 and 320 L kg^{-1}, respectively, almost all above the observed values. A linear regression of the remaining 51 Kd values gave the equation:

$$\text{Log(Kd)} = 0.973$$

with a GSD of 3.6 (no independent variables were significant). This gives a value of 9.3 L kg^{-1}.

Because speciation of Cr is not easily predicted, it may be prudent in an assessment to use the functions that are the most conservative in dose estimation (gives the highest dose).

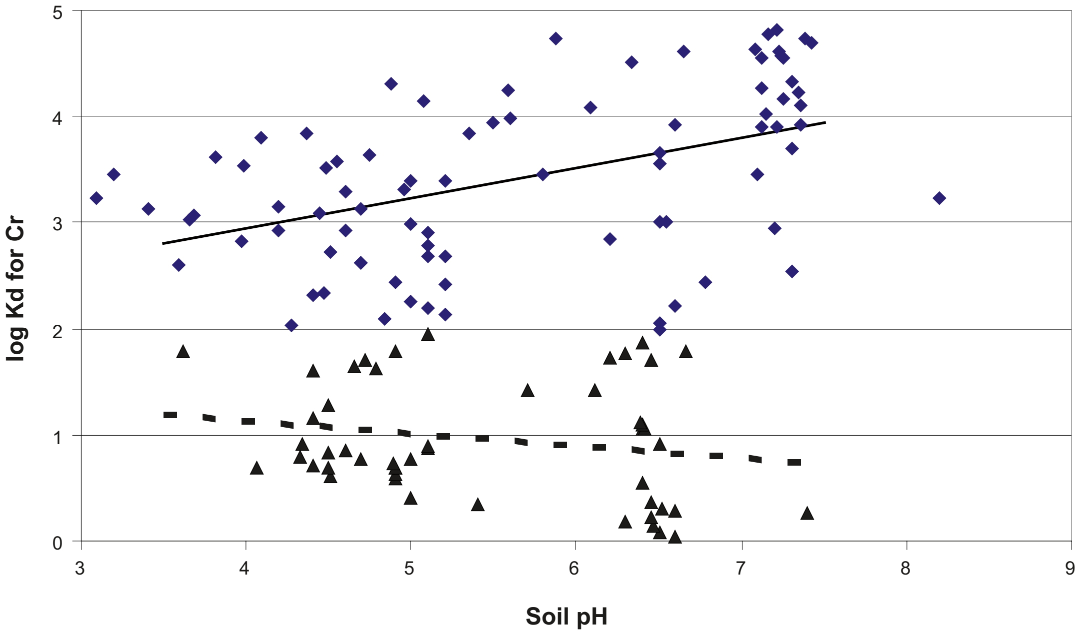

Figure 5-4. Scatter plot of Kd for Cr versus pH, showing the apparent separation of values above and below log (Kd) = 2 (100 L kg^{-1}).

5.3.6 Cs – Cesium

The Kd values for Cs were measured in the 7 SKB soils and in 171 Canadian soils in the NTE program (Table 5-5). The stepwise regression of the NTE soil included only the clay*pH interaction, with a residual GSD of 4.0. The stepwise regression of the /Thibault et al. 1990/ data set (246 values) included pH and the clay*pH interaction, with a residual GSD of 9.8. The stepwise regression of the /Sanchez et al. 2002/ data set (53 values, most very acidic and some highly organic) included pH and clay content, with a residual GSD of 5.0.

There is a notable difference between Kd based on desorption of indigenous Cs and Kd based on sorption of radiocesium (^{134}Cs or ^{137}Cs), which constitute most of the values summarised by /Thibault et al. 1990/. Cesium sorption reactions in soil are known to be slow, often referred to as aging and extending over several years or even decades. In part, this is because Cs slowly becomes entrapped in the interlayer spaces of clay mineral platelets. In contrast, indigenous Kd values account for much of the interlayer bound Cs. From the perspective of waste disposal, Kd values representing near steady state are probably most appropriate, so the values might be aligned with the upper values of sorption studies, but perhaps not as high as Kd values for desorption of indigenous Cs.

/EMRAS 2008/ found 32 reports and 469 values, GM values were sand: 530 L kg^{-1}, loam: 3,500 L kg^{-1}, clay: 5,500 L kg^{-1} and organic: 270 L kg^{-1}, and these are also more consistent with the sorption data sets rather than the long-term equilibration reflected in the desorption data sets.

Combining the data of /Thibault et al. 1990/ and /Sanchez et al. 2002/, a total of 299 values, the stepwise regression gave the equation:

$$\text{Log(Kd)} = 2.26 + 0.0112 \cdot (\text{pH}) + 0.127 \cdot (\text{clay})$$

for sorption Kd values, with a residual GSD of 9.5 and an $r^2 = 0.07$. This would be an appropriate equation for assessments dealing with ^{134}Cs (half-life = 2.06 d) and shorter lived isotopes, and perhaps ^{137}Cs (half-life = 30.2 a), because decay may prevent complete chemical equilibrium.

The NTE data, a total of 171 values, gave by stepwise regression the equation:

$$\text{Log(Kd)} = 4.31 + 0.00183 \cdot (\text{clay}) \cdot (\text{pH})$$

for desorption Kd values, with a residual GSD of 4.0 and an $r^2 = 0.09$. This would be an appropriate equation for assessments dealing with ^{135}Cs (half-life = $2 \cdot 10^6$ a), and perhaps ^{137}Cs, because complete chemical equilibrium may be reached.

The proposed equation was developed by stepwise regression from the combined desorption and sorption data (470 values), with a dummy variable to differentiate these (0 for desorption, 1 for sorption). Assuming an average of desorption and sorption is one approach for long-term assessment, the dummy variable is assigned the value 0.5, giving the equation:

$$\log(\text{Kd}) = 3.19 + 0.0798 \cdot (\text{pH}) + 0.00154 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 7.4 and an $r^2 = 0.40$. From this equation, the estimates for the SKB soils are 6,300, 6,000, 4,000, 6,300, 4,800, 6,000 and 4,700 L kg^{-1}, respectively. The scatter plot (Figure 5-5),

Table 5-5. Measured (SKB) and predicted (using the NTE, Thibault et al. and Sanchez et al. data sets) Kd values (L kg^{-1}) for Cs.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. sorption predicted Kd ± 1GSD	Sanchez et al. sorption predicted Kd ± 1GSD
A Forsmark clay till	28,000	6,500–110,000	140–14,000	2,200–54,000
B Forsmark deep peat	> 10,000nd	12,000–190,000	96–9,200	6,900–170,000
C Simpevarp sandy till	21,000	5,200–85,000	82–7,800	250–6,200
D Simpevarp clay gyttja	25,000	9,000–150,000	120–11,000	2,300–57,000
E Simpevarp peat	57,000	8,000–130,000	85–8,100	1,000–25,000
F Simpevarp clay gyttja	31,000	8,400–140,000	110–11,000	2,100–52,000
G Forsmark clay till	120,000	5,700–94,000	100–9,500	570–14,000

[nd] pore water concentration non-detectable, this value computed using detection limit.

differentiating the sorption from the desorption data shows the large variation in the data. /Sanchez et al. 2002/ and others point out that accounting for K behaviour in soil will markedly improve predictions of Kd for Cs, but this was not included here as the required information is not commonly available.

5.3.7 Fe – Iron

The Kd values for Fe were measured in the 7 SKB soils and in 127 Canadian soils in the NTE program (Table 5-6). The stepwise regression of the NTE soil included only soil pH, with a residual GSD of 4.0. The stepwise regression of the /Thibault et al. 1990/ data set (44 values) included only the clay*pH interaction, with a residual GSD of 5.2.

There is a notable difference between Kd based on desorption of indigenous Fe (the SKB and NTE soils, above) and Kd based on sorption of ^{59}Fe, which constitute most of the values summarised by /Thibault et al. 1990/. Iron sorption reactions in soil are confounded by Fe^{+++} precipitation as oxides on particle surfaces. These precipitates are not readily exchangeable with the Fe in soil pore water, except that there is redox variation in most soils over time that may cause some chemical reduction of precipitated Fe^{+++} to mobile Fe^{++}, which may then exchange with pore water Fe. As such, indigenous Fe will give high Kd values because these Kd values account for much of the Fe oxide coatings.

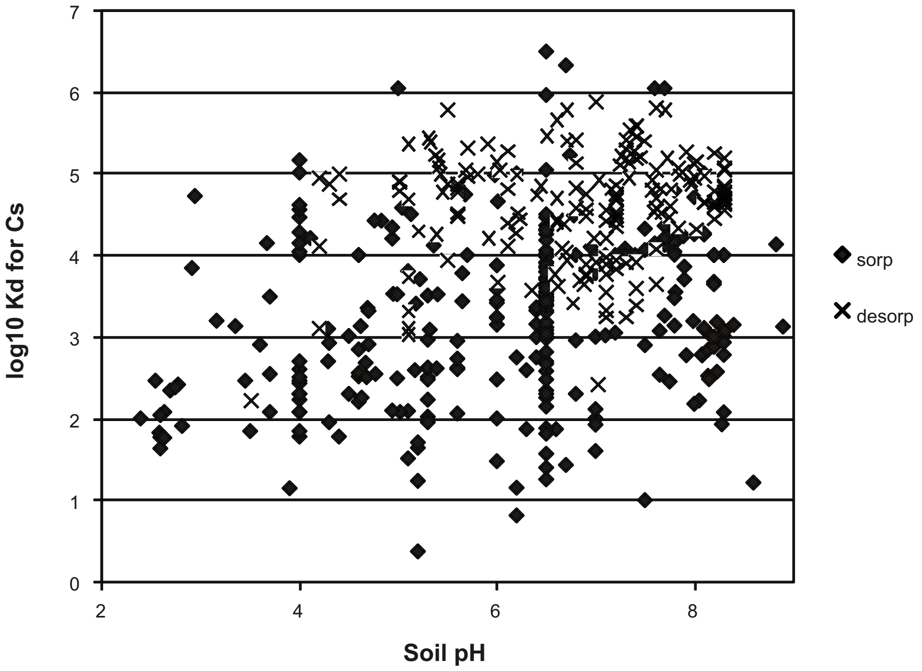

Figure 5-5. *Scatter plot of 470 values of Kd for Cs, differentiating those from sorption versus desorption methods.*

Table 5-6. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Fe.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. sorption predicted Kd ± 1GSD
A Forsmark clay till	25,000	7,300–120,000	36–960
B Forsmark deep peat	12,000	880–14,000	150–4,000
C Simpevarp sandy till	4,900	2,500–40,000	21–560
D Simpevarp clay gyttja	34,000	2,500–40,000	79–2,100
E Simpevarp peat	13,000	1,300–20,000	310–6,100
F Simpevarp clay gyttja	59,000	2,500–40,000	67–1,800
G Forsmark clay till	160,000	3,600–57,000	26–700

/EMRAS 2008/ found 2 reports and 23 values, GM values were sand: 320 L kg^{-1}, loam: 890 L kg^{-1}, clay: 1,600 L kg^{-1} and organic: 1,400 L kg^{-1}, and these are also more consistent with the sorption data sets rather than the long-term equilibration reflected in the desorption data sets.

Sorption of a spike of ^{59}Fe will initially be a surface sorption process, and this is what is most likely represented in the relatively low Kd values in /Thibault et al. 1990/. With time, it is anticipated that the ^{59}Fe will become more incorporated into the Fe precipitates. However, ^{55}Fe and ^{59}Fe are the only radioactive isotopes, and with half lives of 986 d and 45 d, neither represents a long-term radioactive hazard. Because Fe is so plentiful in the environment, small increments also do not present a chemical toxicity hazard.

Given the short half lives, the equation for sorption Kd values from Thibault et al. (results shown above) is proposed for use:

$$\log(Kd) = 2.01 + 0.00442 \cdot (clay) \cdot (pH)$$

with a residual GSD of 3.2 and an $r^2 = 0.22$. This gives values for the SKB soils of 190, 780, 110, 410, 310, 350 and 140 L kg^{-1}, respectively.

5.3.8 Ho – Holmium

The Kd values for Ho were measured in the 7 SKB soils and in 161 Canadian soils in the NTE program (Table 5-7). The stepwise regression of the NTE soil included soil pH and the clay*pH interaction, with a residual GSD of 3.1.

Interpolation of the data from /Thibault et al. 1990/ using plant/soil concentration ratio data was the methodology behind the Kd values in /IAEA 1994/. The Kd values for Ho (sand: 250, loam: 800, clay: 1,300 and organic: 3,000 L kg^{-1}) were not based on any measurements. /EMRAS 2008/ apparently found only the values from /IAEA 1994/ and recommended 930 L kg^{-1}. No other Kd data were found for Ho. It is proposed that the equation from the NTE data be used:

$$\mathrm{Log}(Kd) = 2.15 + 0.338 \cdot (pH) - 0.00094 \cdot (clay) \cdot (pH)$$

with a residual GSD of 3.1 and an $r^2 = 0.30$. This gives values for the SKB soils of 19,000, 1,400, 6,800, 5,100, 2,500, 5,300 and 9,600 L kg^{-1}, respectively.

5.3.9 I – Iodine

The Kd values for I were measured in the 7 SKB soils both using desorption of indigenous I and sorption of spiked stable I (Table 5-8). Additionally, there was an extensive literature search (OPG, /Sheppard et al. 2006/ and references therein) that documents Kd for 114 soils. These included, where appropriate, the data summarised by /Thibault et al. 1990/. The stepwise regression of the literature values included soil carbon content and the clay*pH interaction, with a residual GSD of 9.3.

The measured Kd values using indigenous I were almost all higher than the literature values plus 1 GSD, and in contrast the measured Kd values using a spike of stable I were almost all lower than the literature values less 1 GSD. Many of these measured values were within the range of the literature ±2 GSD, which corresponds with the 95th percentile confidence bounds. However, this range is ~7,500-fold, even after accounting for the most significant soil properties (organic carbon, pH and clay content).

Table 5-7. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Ho.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	37,000	6,300–60,000
B Forsmark deep peat	——[nd]	450–4,300
C Simpevarp sandy till	1,800	2,200–21,000
D Simpevarp clay gyttja	4,200	1,700–16,000
E Simpevarp peat	2,900	810–7,700
F Simpevarp clay gyttja	6,700	1,700–16,000
G Forsmark clay till	37,000	3,100–29,000

[nd] Not detectable in either solids or pore water.

Table 5-8. Measured (SKB) and predicted (using the OPG data set) Kd values (L kg^{-1}) for I.

SKB soil	Measured desorption Kd	Measured sorption Kd	OPG predicted Kd ±1 GSD	OPG predicted Kd ±2 GSD
A Forsmark clay till	> 90	0.0041	0.71–62	0.08–580
B Forsmark deep peat	< 140	2.9	58–5,000	6.2–47,000
C Simpevarp sandy till	60	0.28	0.63–55	0.067–510
D Simpevarp clay gyttja	3,900	8.8	4.0–350	0.43–3,300
E Simpevarp peat	1,600	1.7	5.8–510	0.62–4,700
F Simpevarp clay gyttja	> 8,300	2.9	3.5–310	0.38–2,900
G Forsmark clay till	> 210	0.069	0.69–60	0.074–560

Most of the Kd values from the literature search were based on contact times of at least several days, an essential feature because Kd for I will continue to increase for some time. This is probably part of the explanation for the large differences between the desorption and sorption Kd values measured for the SKB sites, where sorption was for 7 d. In addition, some of the indigenous I in these soils, which is included in the desorption Kd values, may have been biologically incorporated by plants and subsequently incorporated as strongly bound forms in recalcitrant organic matter. Including these more ecological processes would extend the equilibration time to many years. The desorption Kd values may better represent a very long-term steady state, and although it is not clear, these are probably the more appropriate data for a long-term assessment calculation.

The frequency distributions for log Kd and the independent variables in the literature data set from /Sheppard et al. 2006/ were relatively broad (Figure 5-6). Although included, there were few soils below pH 5 and above 20% organic carbon, and so the domain relevant to some of the SKB soils

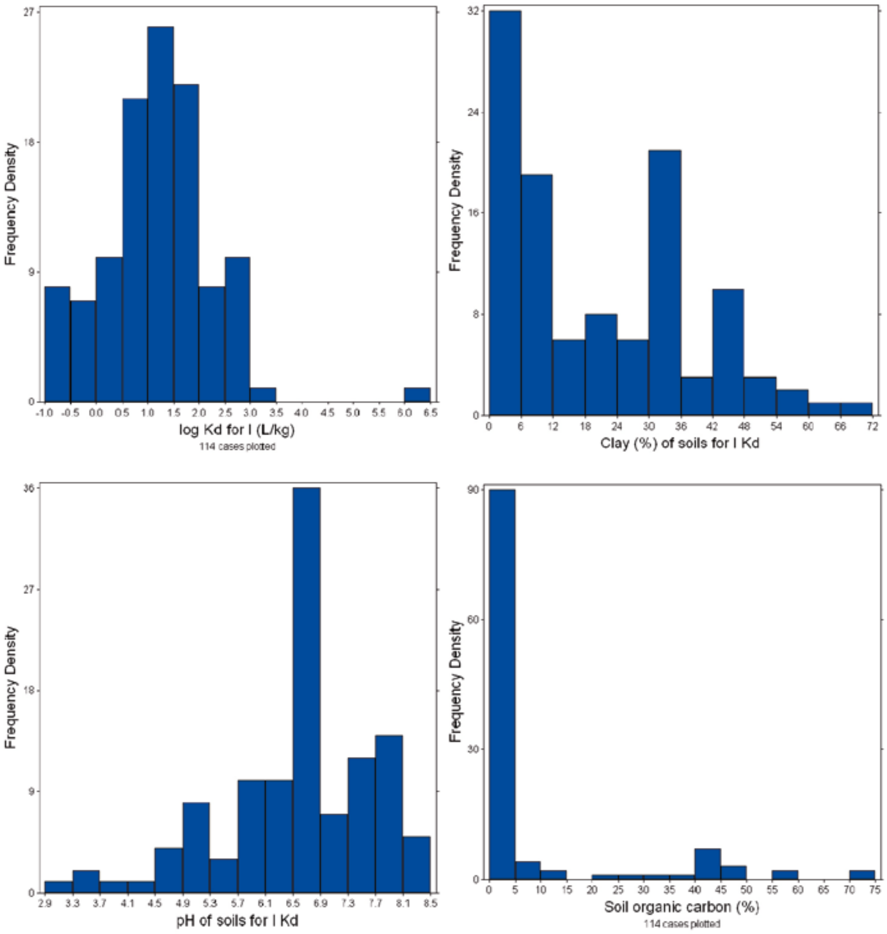

Figure 5-6. *Frequency histograms of log (Kd) and the various soil properties of the soils where Kd was measured in the literature.*

is not well represented. However, the results for the SKB soils that are outside the ±2 GSD range (above and Figure 5-7) from the literature are not just the exceptional soils in terms of pH and organic carbon.

/EMRAS 2008/ found 9 reports and 250 values, and they separated these into studies those with I^- versus IO_3^- and with soils with a range of soil organic matter contents, but none of the Kd values would be statistically different between these categories. Among soil textures, GM values were sand: 4.1 L kg^{-1}, loam: 8.0 L kg^{-1}, clay: 11 L kg^{-1} and organic: 32 L kg^{-1}. These values are more like the short-term sorption values.

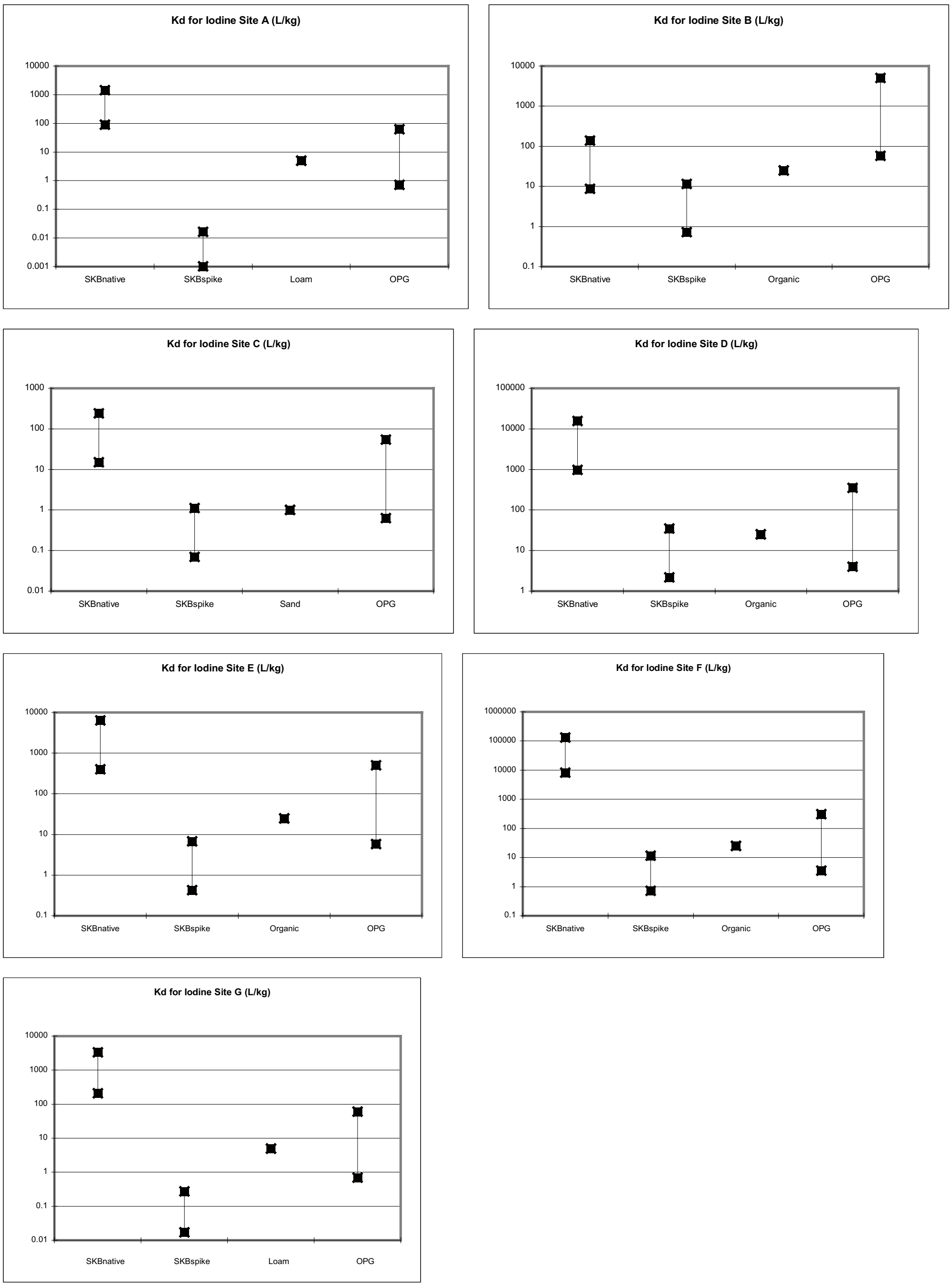

Figure 5-7. *Comparison of desorption (Kdnative) and sorption (Kdspike) Kd from the 7 SKB soils (±1 GSD where GSD = 4.0) versus corresponding estimates, where sand, loam and organic are compendia recommendations from /Sheppard and Thibault 1990/, OPG is regression estimates from the /Sheppard et al. 2006/ data set. Variation on regression estimates correspond to the residual GSD.*

The Kd values from the indigenous I at the SKB sites are 26-fold higher than the best estimate from the regression equation. It is proposed to adjust the predictions by this amount, so the equation becomes:

$$\text{Log(Kd)} = 2.13 + 0.00297 \cdot \text{(organic carbon)} + 0.00181 \cdot \text{(clay)} \cdot \text{(pH)}$$

5.3.10 La – Lanthanum

The Kd values for La were measured in the 7 SKB soils and in 209 Canadian soils in the NTE program (Table 5-9). The stepwise regression of the NTE soil included pH, clay, sand and silt contents, with a residual GSD of 3.0.

/Gooddy et al. 1995/ measured Kd in 18 acidic sandy soils, obtaining a GM of 14,000 L kg^{-1}, generally consistent with the values from the Canadian and SKB soils, albeit /Gooddy et al. 1995/ used similar methods. Five other values were found /Shan et al. 2002, Cao et al. 2001/, and they had a GM of 380 L kg^{-1}. EMRAS found one value for a sand of 5,300 L kg^{-1}. If one combines the above, the following regression equation can be formed:

$$\text{Log(Kd)} = 3.22 - 0.0506 \cdot \text{(clay)} + 0.230 \cdot \text{(pH)} + 0.00617 \cdot \text{(clay)} \cdot \text{(pH)}$$

with a residual GSD of 3.9, an $r^2 = 0.34$ and estimates for the SKB soils of 42,000, 240, 22,000, 6,900, 2,200, 6,200 and 25,000 L kg^{-1}, respectively.

5.3.11 Mn – Manganese

The Kd values for Mn were measured in the 7 SKB soils and in 205 Canadian soils in the NTE program (Table 5-10). The stepwise regression of the NTE soil included pH and the clay*pH interaction, with a residual GSD of 10. The stepwise regression of the /Thibault et al. 1990/ data set (113 values) included only soil pH, with a residual GSD of 7.0.

Table 5-9. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for La.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	44,000	14,000–130,000
B Forsmark deep peat	1,800	290–2,600
C Simpevarp sandy till	1,800	3,600–32,000
D Simpevarp clay gyttja	5,900	2,600–23,000
E Simpevarp peat	4,000	980–8,700
F Simpevarp clay gyttja	2,700	2,700–24,000
G Forsmark clay till	80,000	5,700–51,000

Table 5-10. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Mn.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. predicted Kd ± 1GSD
A Forsmark clay till	18,000	340–36,000	12–600
B Forsmark deep peat	1,200	10–1,100	0.088–4.3
C Simpevarp sandy till	3,600	100–11,000	1.1–51
D Simpevarp clay gyttja	730	57–6,100	1.1–51
E Simpevarp peat	230	24–2,600	0.20–9.7
F Simpevarp clay gyttja	130	61–6,500	1.1–51
G Forsmark clay till	22,000	150–16,000	2.3–120

The Kd values for the SKB and NTE soils represent desorption of indigenous Mn, whereas most of the studies in the summary by /Thibault et al. 1990/ represent sorption of ^{54}Mn. As with Fe, there are relatively stable Mn oxides in soils. These precipitates are not readily exchangeable with the Mn in soil pore water, except that there is some redox variation in most soils over time that may cause partial reduction to mobile Mn^{++} and then exchange with pore water Mn. Sorption of a spike of ^{54}Mn will initially be a surface sorption process, and this is what is most likely represented in the Kd values in /Thibault et al. 1990/. With time, it is anticipated that the ^{54}Mn will become more incorporated into the Mn precipitates. In contrast, indigenous Kd values account for much of the Mn oxide coatings.

/EMRAS 2008/ found 4 reports and 83 values, GM values were sand: 980 L kg^{-1}, loam: 1,100 L kg^{-1}, clay: 4,500 L kg^{-1} and organic: 160 L kg^{-1}.

The longer lived radioactive isotopes of Mn are ^{54}Mn (312 d) and ^{53}Mn ($3.8 \cdot 10^6$ a) so that, unlike Fe, it is not possible to limit the proposed Kd values to short-term equilibria. From the perspective of waste disposal, Kd values representing near steady state are probably most appropriate. So the values might be aligned with the upper values of sorption studies, but perhaps not as high as Kd values for desorption of indigenous Kd.

The proposed equation was developed by stepwise regression from the combined desorption and sorption data (318 values), along with 84 values from the more recent literature, using a dummy variable to differentiate these (0 for desorption, 1 for sorption). Assuming an average of desorption and sorption is appropriate for long-term assessment, the dummy variable is assigned the value 0.5, giving the equation:

$$\log (Kd) = -0.330 + 0.457 \cdot (pH)$$

with a residual GSD of 12, an $r^2 = 0.32$ and estimates for the SKB soils of 440, 19, 90, 90, 31, 90 and 150 L kg^{-1}, respectively.

5.3.12 Mo – Molybdenum

The Kd values for Mo were measured in the 7 SKB soils and in 185 Canadian soils in the NTE program (Table 5-11). The stepwise regression of the NTE soil included clay, silt and the clay*pH interaction, with a residual GSD of 2.5. The stepwise regression of the /Thibault et al. 1990/ data (24 values) included clay and soil pH, with a residual GSD of 3.2.

Table 5-11. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Mo.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. predicted sorption Kd ± 1GSD
A Forsmark clay till	70	48–310	3.1–32
B Forsmark deep peat	1,200	2,600–17,000	81,000–830,000
C Simpevarp sandy till	410	30–190	4.2–43
D Simpevarp clay gyttja	1,200	220–1,400	210–2,100
E Simpevarp peat	6,800	490–3,100	750–7,700
F Simpevarp clay gyttja	3,300	270–1,700	180–1,800
G Forsmark clay till	240	44–280	4.7–48

/EMRAS 2008/ found 5 reports and 9 values with an overall GM value of 380 L kg^{-1}. /Gooddy et al. 1995/ measured Kd for Mo on 6 acidic sandy soils with a GM of 1,600 L kg^{-1}, consistent with the above values. The desorption Kd values from the NTE data set and from /Gooddy et al. 1995/ also agree with the data of /Thibault et al. 1990/, which were mostly sorption of ^{99}Mo. Stepwise regression after combining these data (215 values) gave the equation:

$$\text{Log (Kd)} = 1.70 + 0.0718 \cdot \text{(clay)} + 0.0325 \cdot \text{(organic carbon)} - 0.00873 \cdot \text{(clay)} \cdot \text{(pH)}$$

with a residual GSD of 3.3 and $r^2 = 0.19$. This gave best estimate values for the SKB soils of 70, 740,000, 61, 1,400, 5,600, 1,500 and 76 L kg^{-1}, respectively, which are reasonable except for site B. Clearly, more data for very acidic peat soils is needed.

5.3.13 Nb – Niobium

The Kd values for Nb were measured in the 7 SKB soils and in 92 Canadian soils in the NTE program (Table 5-12). The stepwise regression of the NTE soil included clay, pH and the clay*pH interaction, with a residual GSD of 4.7. /Thibault et al. 1990/ did not find Kd values for Nb, and the values given in /IAEA 1994/ of 160 to 2,000 L kg^{-1} were only extrapolations.

EMRAS found 3 studies reporting a total of 11 values, with a GM of 1,500 L kg^{-1} and GSD of 4. No other reports were found. The regression from the NTE data was:

$$\text{Log (Kd)} = 2.45 + 0.0960 \cdot \text{(clay)} + 0.348 \cdot \text{(pH)} - 0.0159 \cdot \text{(clay)} \cdot \text{(pH)}$$

with a residual GSD of 4.7 and $r^2 = 0.13$. This gave best estimate values for the SKB soils of 44,000, 940,000, 16,000, 44,000, 82,000, 61,000 and 26,000 L kg^{-1}, respectively.

Table 5-12. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Nb.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	> 20,000[nd]	9,600–210,000
B Forsmark deep peat	> 8,000	200,000–4400,000
C Simpevarp sandy till	1,700	3,500–76,000
D Simpevarp clay gyttja	12,000	9,400–200,000
E Simpevarp peat	14,000	18,000–380,000
F Simpevarp clay gyttja	> 60,000	13,000–280,000
G Forsmark clay till	36,000	5,500–120,000

[nd] pore water concentration non-detectable, this value computed using detection limit.

Table 5-13. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Nd.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	46,000	13,000–130,000
B Forsmark deep peat	2,100	1,000–10,000
C Simpevarp sandy till	1,500	2,600–26,000
D Simpevarp clay gyttja	5,200	2,800–28,000
E Simpevarp peat	3,200	1,400–14,000
F Simpevarp clay gyttja	7,300	3,200–32,000
G Forsmark clay till	76,000	4,600–46,000

5.3.14 Nd – Neodymium

The Kd values for Nd were measured in the 7 SKB soils and in 208 Canadian soils in the NTE program (Table 5-13). The stepwise regression of the NTE soil included pH, sand, silt and the clay*pH interaction, with a residual GSD of 3.2.

/Gooddy et al. 1995/ measured Kd for Nd on 20 acidic sandy soils with a GM of 16,000 L kg^{-1}. Neither /IAEA 1994/ nor /EMRAS 2008/ have proposed values for Kd. There are no long-lived isotopes of Nd, so the Kd for indigenous Nd is probably appropriate. The equation from the NTE data set is:

$$\text{Log (Kd)} = 3.523 + 0.476 \cdot (\text{pH}) - 0.0200 \cdot (\text{sand}) - 0.0185 \cdot (\text{silt}) - 0.00435 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 3.2 and $r^2 = 0.25$. This gives values for the SKB soils of 40,000, 3,200, 8,300, 9,000, 4,300, 10,000 and 15,000 L kg^{-1}, respectively.

5.3.15 Ni – Nickel

The Kd values for Ni were measured in the 7 SKB soils and in 198 Canadian soils in the NTE program (Table 5-14). The stepwise regression of the NTE soil included pH, clay, sand and the clay*pH interaction, with a residual GSD of 2.4. The regressions of the data from /Thibault et al. 1990/ with 44 soils included only pH, and those of /Vandenhove et al. 2009/ with 143 soils, from /Sauvé et al. 2000/ with 139 soils and from /Watmough et al. 2005/ with 46 soils all included pH and log (organic carbon[1]).

The regressions of data from other literature (171 values all using stable Ni only, /Covelo et al. 2004, 2007ab, Fulekar and Dave 1992, De Groot et al. 1998, Echeverria et al. 1998, Gooddy et al. 1995, Jalali and Moharrami 2007, Lee 2006, Lin et al. 2004, Ponizovsky et al. 2008, Rabie Ahmed Usman 2008, Sauvé et al. 2003, Staunton 2004, Van den Hoop 1995, Veeresha et al. 2003, Vega et al. 2006, Watmough et al. 2005, Yin et al. 2002/) were done in three sets: sorption only, desorption of indigenous element only, and combined with a dummy variable to differentiate these (0 for desorption, 1 for sorption). Assuming an average of desorption and sorption was accomplished setting the dummy variable to 0.5.

SKB site B is different from the others with low pH and very high organic carbon content. The various estimates above differ most for this soil; the estimates from desorption data predict the highest Kds for site B, whereas the estimates from sorption data predict the lowest Kds. This implies that there are few surface sorption sites for Ni in this organic soil, but a substantial portion of the indigenous Ni is strongly held, perhaps non-exchangeably bonded to the organic matter.

/EMRAS 2008/ found 12 reports and 64 values; GM values were sand: 130 L kg^{-1}, loam: 180 L kg^{-1}, clay: 930 L kg^{-1} and organic: 1,100 L kg^{-1}. EMRAS also differentiated Kd based on pH, with $pH < 5$: 14 L kg^{-1}, $5 < pH < 6.5$: 58 L kg^{-1} and $pH > 6.5$: 820 L kg^{-1}.

Several Ni isotopes have long half lives; so in the context of long-term assessment of waste management, the desorption Kd values are of value. The relationship of sorption and desorption Kd values to pH, clay and organic carbon content were similar, with sorption Kd values effectively displaced about 60-fold lower. By using a dummy variable, both sorption and desorption data could be combined. This was done for all the data available (NTE, Thibault et al. and other literature), resulting in 410 values. The stepwise regression, substituting 0.5 for the dummy variable, was:

$$\text{Log (Kd)} = 0.718 + 0.233 \cdot (\text{pH}) + 0.0148 \cdot (\text{organic carbon})$$

with a residual GSD of 4.2 and $r^2 = 0.55$. The predictions for the SKB soils are 170, 230, 81, 160, 120, 150 and 110 L kg^{-1}, respectively.

5.3.16 Np – Neptunium

No Kd values for Np were measured in the SKB soils. There was an extensive literature search (OPG, /Sheppard et al. 2006/ and references therein) that summarised 82 Kd values (Table 5-15). For all of these soils, the stepwise regression included organic carbon and the clay*pH interaction with a residual GSD of 9.5. Of these there were 5 organic soils and these were best represented by a simple geometric mean (GSD = 4.0). The stepwise regression from the data of /Thibault et al. 1990/ included pH, organic carbon and the clay*pH interaction, with a residual GSD of 3.9.

Table 5-14. Measured (SKB) and predicted (using several literature data sets) Kd values (L kg^{-1}) for Ni.

SKB soil	Measured desorption Kd	NTE desorp. predicted Kd ± 1GSD	Thibault et al. predicted Kd ±1GSD	Vandenhove predicted Kd	Gil-Garcia et al. predicted Kd	Sauvé et al. predicted Kd	Watmough predicted Kd	Literature predicted Kd sorption – desorption – average
A Forsmark clay till	3,800	590–3,500	180–1,800	200	150	850	9,000	28–440–130
B Forsmark deep peat	> 3,000[nd]	2,000–12,000	5.6–55	11	0.22	5.9	920	0.95–16,000–250
C Simpevarp sandy till	530	230–1,400	31–310	46	11	71	2,900	5.1–340–57
D Simpevarp clay gyttja	460	890–5,400	31–310	49	1.8	71	2,900	5.1–1,500–130
E Simpevarp peat	710	990–5,900	9.9–98	18	0.58	13	1,300	1.7–2,100–99
F Simpevarp clay gyttja	450	1,100–6,300	31–310	49	1.9	71	2,900	5.2–1,400–130
G Forsmark clay till	3,000	370–2,200	56–550	77	16	160	4,200	9.0–390–76

[nd] pore water concentration non-detectable, this value computed using detection limit.

Table 5-15. Predicted (using the OPG and Thibault et al. data sets) Kd values (L kg^{-1}) for Np.

SKB soil	OPG predicted Kd all soils ±1 GSD	OPG GM Kd organic soils ±1 GSD	Thibault et al. predicted Kd ±1 GSD
A Forsmark clay till	0.45–40	––––^{nr}	4.3–67
B Forsmark deep peat	––––^{nr}	130–2,100	260–4,100
C Simpevarp sandy till	0.14–13	––––	0.78–12
D Simpevarp clay gyttja	18–1,600	––––	18–270
E Simpevarp peat	18–1,600	––––	11–170
F Simpevarp clay gyttja	11–1,000	––––	14–210
G Forsmark clay till	0.25–22	––––	1.5–24

^{nr} Not relevant.

The two sets of predicted values agree quite well, although there is the potential for some overlap in the original sources (it appears that none of the same papers were used, but some of the same authors appear in both data sets). Nonetheless, the two data sets were combined and the equation was:

$$\text{Log (Kd)} = -1.42 + 0.307 \cdot (\text{pH}) + 0.0429 \cdot (\text{organic carbon}) + 0.00638 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 6.8 and $r^2 = 0.43$. The estimates for the SKB soils are 9.2, 2,100, 1.7, 76, 50, 55 and 3.4 L kg^{-1}, respectively.

/EMRAS 2008/ found 3 reports and 26 values, GM values were sand: 14 L kg^{-1}, loam: 23 L kg^{-1} and organic: 81 L kg^{-1}. There was no estimate for clay but for all soils it was 36 L kg^{-1}.

5.3.17 Pa – Protactinium

The /IAEA 1994/ had interpolated values of 550, 1,800, 2,700 and 6,600 L kg^{-1} for sand, loam, clay and organic soils, respectively. There was no actual Kd data to support this; the interpolation was based on plant/soil concentration ratios, which may have themselves been extrapolated. /EMRAS 2008/ reported values of 2,000 L kg^{-1} for all soils, 1,380 L kg^{-1} for mineral soils and 6,600 L kg^{-1} for organic soils, based on 4 measurements. These values are recommended for SKB.

5.3.18 Pb – Lead

The Kd values for Pb were measured in the 7 SKB soils and in 197 Canadian soils in the NTE program (Table 5-16). The stepwise regression of the NTE soil included pH and sand content, with a residual GSD of 3.0. /Thibault et al. 1990/ only had 14 Kd values, so they were combined with recent literatures values /Covelo et al. 2004, 2007ab, Chen et al. 1995, De Groot et al. 1998, Echeverria et al. 1998, Fulekar and Dave 1992, Gooddy et al. 1995, Lee 2006, Lee et al. 1998, MacDonald and Hendershot 2006, Rabie Ahmed Usman 2008, Sastre et al. 2006, Van den Hoop 1995, Veeresha et al. 2003, Vega et al. 2006, Watmough et al. 2005/. The resulting stepwise regression included clay content, organic carbon and the clay*pH interaction, with a residual GSD of 10. The equation from /Carlon et al. 2004/ was based on 247 values from 5 authors, all of whom generated independent regression equations.

Table 5-16. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Pb.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. and literature sorption predicted Kd ± 1GSD	Carlon et al. predicted Kd ± 1GSD
A Forsmark clay till	6,000	4,600–42,000	170–18,000	5,200–520,000
B Forsmark deep peat	3,100	2,300–21,000	23–2,600	290–29,000
C Simpevarp sandy till	2,000	1,900–17,000	240–26,000	1,200–120,000
D Simpevarp clay	11,000	4,300–39,000	140–16,000	1,200–120,000
E Simpevarp peat	17,000	3,100–28,000	81–8,900	470–47,000
F Simpevarp clay	44,000	4,600–42,000	110–12,000	1,200–120,000
G Forsmark clay till	25,000	2,800–26,000	190–21,000	2,000–200,000

Note that the equation presented by /Watmough et al. 2005/ has coefficients almost identical to those of /Carlon et al. 2004/ and so is not computed separately here.

As with Fe and Mn, there is a large pool of stable element Pb in soil, and there is interest in both the Kd for radioactive isotopes and for stable Pb. Relatively few studies reporting Kd for Pb use ^{210}Pb (or other radioactive isotopes). From the perspective of nuclear fuel waste management, ^{210}Pb (half-life 22.3 a) will not reach the surface because of migration of ^{210}Pb, but rather the migration and decay of its nuclear parents. Some of this ^{210}Pb will be from decay of gaseous Rn that may decay in the soil or in the atmosphere over the soil. Thus, the entry of ^{210}Pb into the soil has unique aspects compared to all the other isotopes and elements of interest. Our assumption is that Kd values measured for stable indigenous Pb in soil are appropriate for ^{210}Pb.

/EMRAS 2008/ found 5 reports and 23 values, GM values were sand: 220 L kg^{-1}, loam: 10,000 L kg^{-1} and organic: 2,500 L kg^{-1}. It was not specified whether these were based on ^{210}Pb or stable Pb, the latter would be at much higher molar concentrations than the former, and this may affect Kd. The values from the NTE and /Thibault et al. 1990/ data sets and the recent literature generally overlap. Thus the NTE and Thibault et al. data were combined, giving the regression:

$$\text{Log (Kd)} = 2.41 - 0.043 \cdot (\text{clay}) + 0.216 \cdot (\text{pH}) + 0.0214 \cdot (\text{organic carbon}) + 0.00601 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 7.7 and $r^2 = 0.20$ and estimates for the SKB soils of 6,100, 1,300, 3,300, 3,900, 1,900, 3,300 and 3,900 L kg^{-1}, respectively.

5.3.19 Pu – Plutonium

No Kd values for Pu were measured in the SKB soils nor in Canadian soils in the NTE program. Stepwise regression of the /Thibault et al. 1990/ data set combined with the data of /Skipperud et al. 2000, Smith et al. 2006, Roussel-Debet 2005/ (176 values) included soil pH and organic carbon, with a residual GSD of 6.6 (Table 5-17). These are the data that were used to predict the Kd values for the SKB soils below.

/EMRAS 2008/ found 7 reports and 62 values, GM values were sand: 400 L kg^{-1}, loam: 950 L kg^{-1}, clay: 1,800 L kg^{-1} and organic: 760 L kg^{-1}.

The values for sites B, D, E and F, the organic soils, differ as a group from the remaining mineral soils. Clearly, organic carbon content seems to be important. Figure 5-8 shows the relationship, which appears to be heavily dependent on the few observations with high organic carbon content. The overall regression equation was:

$$\text{Log (Kd)} = 2.00 + 0.150 \cdot (\text{pH}) + 0.0318 \cdot (\text{organic carbon})$$

with a residual GSD of 6.6 and $r^2 = 0.11$. This gives values for the SKB soils of 960, 20,000, 630, 2,600, 3,100, 2,400 and 760 L kg^{-1}, respectively.

Table 5-17. Predicted (using literature data from several sources) Kd values (L kg^{-1}) for Pu.

SKB soil	Predicted Kd	± 1GSD
A Forsmark clay till	960	150–6,300
B Forsmark deep peat	20,000	3,000–130,000
C Simpevarp sandy till	630	96–4,200
D Simpevarp clay gyttja	2,600	390–17,000
E Simpevarp peat	3,100	470–20,000
F Simpevarp clay gyttja	2,400	370–16,000
G Forsmark clay till	760	120–5,000

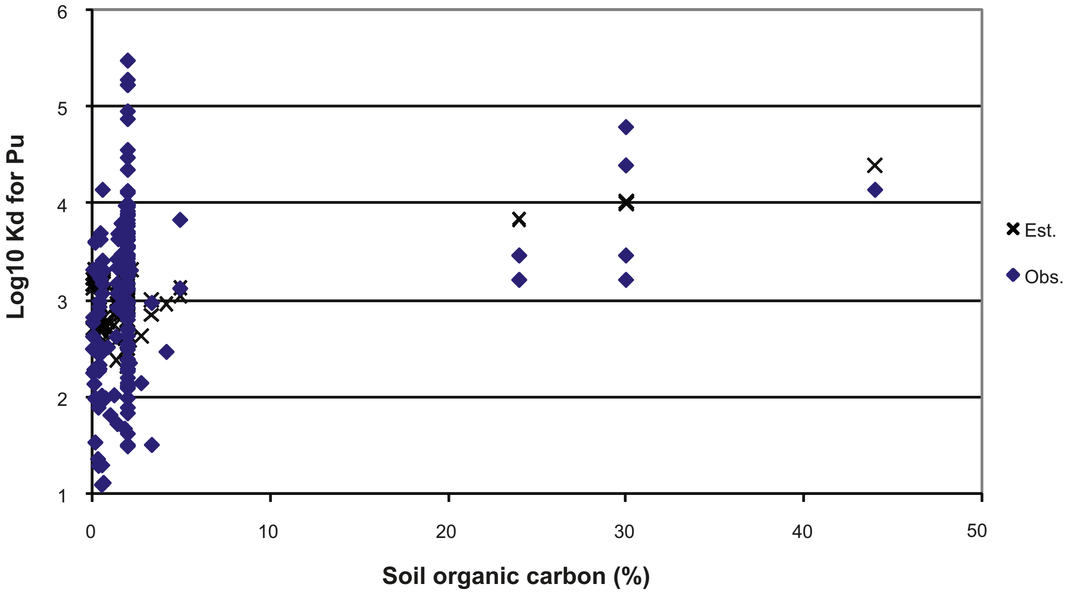

***Figure 5-8.** Observed and estimated Log_{10} Kd for Pu as a function of soil organic carbon content.*

5.3.20 Ra – Radium

No Kd values for Ra were measured in the SKB soils nor in Canadian soils in the NTE program. Stepwise regression of the data in a recent literature review (OPG, /Sheppard et al. 2006/ and references therein) data set (38 values) included only soil pH, with a residual GSD of 16 (Table 5-18). Stepwise regression of the /Thibault et al. 1990/ data set (19 values) included only clay content, with a residual GSD of 28. All of these soils had organic carbon contents < 20%, and so SKB site B was well outside the range, and sites D, E and F were just beyond the range.

/Vandenhove and van Hees 2007/ reported 9 Kd values with a GM of 12,000 L kg^{-1}. /EMRAS 2008/ found 8 reports and 51 values. GM values were sand: 3,100 L kg^{-1}, loam: 1,100 L kg^{-1} and clay: 38,000 L kg^{-1}.

Combining the data from /Sheppard et al. 2006, Thibault et al. 1990, Vandenhove and Van Hees 2007/ gave a total of 67 values, resulted in the equation:

$$\text{Log (Kd)} = 1.56 + 0.0599 \cdot (\text{clay})$$

with a residual GSD of 36 and $r^2 = 0.13$. This equation does not apply to soils much above 20% organic carbon. For the SKB sites, the estimates are 130, (95,000), 42, 1,500, 2,000, 1,300 and 73 L kg^{-1}, respectively. The value for site B is in brackets because it is an extrapolation in terms of organic carbon content; although the equation does not have organic carbon as an independent variable.

Table 5-18. Predicted (using the OPG and Thibault et al. data sets) Kd values (L kg^{-1}) for Ra.

SKB soil	OPG predicted Kd ± 1GSD	Thibault et al. predicted Kd ± 1GSD
A Forsmark clay till	3.5–910	27–20,000
B Forsmark deep peat	0.033–8.6	16,000–1.2E7
C Simpevarp sandy till	0.34–88	9.2–7,000
D Simpevarp clay gyttja	0.34–88	300–220,000
E Simpevarp peat	0.071–19	390–290,000
F Simpevarp clay gyttja	0.34–88	260–200,000
G Forsmark clay till	0.73–190	16–12,000

5.3.21 Sb – Antimony

The Kd values for Sb were measured in the 7 SKB soils and in 197 Canadian soils in the NTE program (Table 5-19). The stepwise regression of the NTE soil included pH and the clay*pH interaction, with a residual GSD of 2.2.

/EMRAS 2008/ found 5 reports and 152 values, GM values were sand: 17 L kg^{-1}, loam: 61 L kg^{-1}, clay: 140 L kg^{-1} and organic: 75 L kg^{-1}. /Gooddy et al. 1995/ measured desorption Kd for indigenous Sb in 13 acidic sandy soil with a GM of 580 L kg^{-1}. /Thibault et al. 1990/ found sorption Kd values for ^{125}Sb in 6 soils, with a GM of 2,400 L kg^{-1} (GSD 3.9). These few data agree with the predictions from the NTE data, and so the NTE equation is proposed for use:

$$\text{Log (Kd)} = 3.74 - 0.175 \cdot (\text{pH}) + 0.00083 \cdot (\text{clay}) \cdot (\text{pH})$$

with, as noted above, a residual GSD of 2.2 and $r^2 = 0.16$ and estimates for the SKB soils of 450, 2,000, 740, 940, 1,300, 920 and 630 L kg^{-1}, respectively.

5.3.22 Se – Selenium

The Kd values for Se were measured in the 7 SKB soils and in 82 Canadian soils in the NTE program (Table 5-20). The stepwise regression of the NTE soil included clay content and pH, with a residual GSD of 3.3. The stepwise regression of the /Thibault et al. 1990/ data included only soil pH, with a residual GSD of 1.7.

/EMRAS 2008/ found 10 reports and 172 values. GM values were sand: 56 L kg^{-1}, loam: 220 L kg^{-1} and clay: 240 L kg^{-1}. /Dhillon and Dhillon 1999, Wang and Liu 2005, Ashworth et al. 2008/ collectively reported 19 Kd values with an overall GM of 110 L kg^{-1}. Combining these 19 values with the NTE and Thibault et al. data sets (123 values) gave the regression equation:

$$\text{Log (Kd)} = 1.79 + 0.133 \cdot (\text{pH}) - 0.00163 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 3.4 and $r^2 = 0.13$ and estimates for the SKB soils of 360, 85, 280, 170, 140, 180 and 300 L kg^{-1}, respectively.

Table 5-19. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Sb.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	290	200–1,000
B Forsmark deep peat	180	870–4,400
C Simpevarp sandy till	260	330–1,700
D Simpevarp clay gyttja	1,200	420–2,100
E Simpevarp peat	1,300	600–3,000
F Simpevarp clay gyttja	1,100	410–2,100
G Forsmark clay till	440	280–1,400

Table 5-20. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Se.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. sorption predicted Kd ± 1GSD
A Forsmark clay till	10	150–1,600	41–120
B Forsmark deep peat	11	7.4–79	130–360
C Simpevarp sandy till	41	110–1,200	72–200
D Simpevarp clay gyttja	56	42–450	72–200
E Simpevarp peat	130	26–270	110–300
F Simpevarp clay gyttja	140	43–470	72–200
G Forsmark clay till	14	120–1,200	59–170

5.3.23 Sm – Samarium

The Kd values for Sm were measured in the 7 SKB soils and in 82 Canadian soils in the NTE program (Table 5-21). The stepwise regression of the NTE soil included soil pH, sand and silt content and the clay*pH interaction, with a residual GSD of 3.1.

/Gooddy et al. 1995/ reported Kd for 14 soils with a GM of 10,000 and GSD of 3.7. /EMRAS 2008/ reported only the values previously reported in /IAEA 1994/, which were interpolated and not based on measurement of Kd. Combining the Gooddy et al. data with the NTE data gave the regression:

$$\text{Log (Kd)} = 2.73 - 0.00862 \cdot (\text{clay}) + 0.290 \cdot (\text{pH})$$

with a residual GSD of 3.3 and $r^2 = 0.31$ and estimates for the SKB soils of 36,000, 1,800, 15,000, 8,900, 4,400, 9,000 and 19,000 L kg^{-1}, respectively.

5.4.24 Sn – Tin

The Kd values for Sn were measured in the 7 SKB soils and in 28 Canadian soils in the NTE program (Table 5-22). The stepwise regression of the NTE soil included clay content and the clay*pH interaction, with a residual GSD of 1.9.

/Nakamaru and Uchida 2008/ reported 4 Kd values with a GM of 9,400 L kg^{-1}. /EMRAS 2008/ found 4 studies and 12 Kd values, with an overall GM of 1,600 L kg^{-1}. Combining the /Nakamaru and Uchida 2008/ data with the NTE data gave the regression:

$$\text{Log (Kd)} = 3.60 - 0.0983 \cdot (\text{organic carbon})$$

with a residual GSD of 2.7 and $r^2 = 0.21$ and estimates for the SKB soils of 3,800, 0.013, 2,700, 35, 7.1, 43 and 2,700 L kg^{-1}, respectively.

Table 5-21. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Sm.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	49,000	9,600–92,000
B Forsmark deep peat	1,700	1,100–10,000
C Simpevarp sandy till	1,300	2,100–20,000
D Simpevarp clay gyttja	5,000	2,600–25,000
E Simpevarp peat	3,000	1,300–13,000
F Simpevarp clay gyttja	9,000	2,900–28,000
G Forsmark clay till	63,000	3,700–35,000

Table 5-22. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Sn.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	> 120[nd]	700–2,500
B Forsmark deep peat	> 340	0.0–0.0001
C Simpevarp sandy till	> 1,100	910–3,400
D Simpevarp clay gyttja	> 80	10–35
E Simpevarp peat	> 200	0.31–1.1
F Simpevarp clay gyttja	> 1,100	5.0–18
G Forsmark clay till	> 110	560–2,000

[nd] pore water concentrations non-detectable, these values computed using detection limit.

5.3.25 Sr – Strontium

The Kd values for Sr were measured in the 7 SKB soils and in 197 Canadian soils in the NTE program (Table 5-23). The stepwise regression of the NTE soil included pH and organic carbon content, with a residual GSD of 2.9. The stepwise regression of the /Thibault et al. 1990/ data (257 values) included only clay content, with a residual GSD of 5.7.

As with Cs, there is an expectation that Kd values for sorption of Sr, as is often considered with radiostrontium, will be lower than the Kd values for indigenous stable Sr. However, the NTE desorption and /Thibault et al. 1990/ sorption values do not differ substantially. Strontium does not undergo the interlayer fixation as does Cs, so more rapid equilibrium for Sr might be expected.

/EMRAS 2008/ found 28 reports and 255 values; GM values were sand: 22 L kg^{-1}, loam: 57 L kg^{-1}, clay: 95 L kg^{-1} and organic: 110 L kg^{-1}. EMRAS also categorised the Kd for soil cation exchange capacity, but this was not a continuous progression.

/Twining et al. 2004, Smith et al. 2006, Wang and Staunton 2005, Fernandez et al. 2006/ collectively reported another 7 values with an overall GM of 62 L kg^{-1}. Combining these 7 values with the NTE and Thibault et al. data sets (481 values) gives the equation:

$\text{Log (Kd)} = 1.55 + 0.0160 \cdot (\text{clay})$

with residual GSD of 7.4 and $r^2 = 0.07$ and estimates for the SKB soils of 49, 290, 37, 95, 100, 91 and 42 L kg^{-1}, respectively.

5.3.26 Tc – Technetium

No Kd values for Tc were measured in the SKB soils nor in the Canadian soils in the NTE program. The values from /Sheppard et al. 1990/ allow development of a regression for aerated values (soil pH was the only independent variable) and a single GM for anaerobic values (Table 5-24). These soils were predominantly organic. The residual GSD for aerated soils was 1.7 and the GSD for anaerobic soils was 4.4.

Table 5-23. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Sr.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. sorption predicted Kd ± 1GSD
A Forsmark clay till	340	180–1,500	3.0–97
B Forsmark deep peat	310	0.085–0.71	19–630
C Simpevarp sandy till	1,300	1,100–8,900	2.2–71
D Simpevarp clay gyttja	59	14–110	6.1–200
E Simpevarp peat	68	7.9–66	6.6–210
F Simpevarp clay gyttja	30	16–140	5.8–190
G Forsmark clay till	190	270–2,300	2.6–83

Table 5-24. Predicted (using the Sheppard et al. data set) Kd values (L kg^{-1}) for Tc.

SKB soil	Aerated predicted Kd ± 1GSD	Anaerobic Kd ± 1GSD
A Forsmark clay till	0.0[1]–0.48	15–280
B Forsmark deep peat	0.0–0.48	15–280
C Simpevarp sandy till	0.0–1.2	15–280
D Simpevarp clay gyttja	0.0–1.2	15–280
E Simpevarp peat	0.0–1.9	15–280
F Simpevarp clay gyttja	0.0–1.2	15–280
G Forsmark clay till	0.0–0.93	15–280

[1] The statistical range included negative Kd values as the lower value: although statistically correct, a negative Kd is not physically possible, so zero is reported here.

/EMRAS 2008/ found 4 reports with 33 values of Kd, GM values were sand: 0.04 L kg^{-1}, loam: 0.07 L kg^{-1}, clay: 0.09 L kg^{-1} and organic: 3.1 L kg^{-1}. /Echevarria et al. 2003/ reported Kd values ranging from 0.013 to 0.039 L kg^{-1} for 5 aerobic soils.

The Kd values reported by /Thibault et al. 1990/, excluding those of /Sheppard et al. 1990/, did not vary significantly with the independent variables, so the GM from those values of 3.3 is the appropriate summarization. In general, redox is the most important determinant of Kd for Tc, under aerated conditions the value is probably as low as can be measured regardless of soil type. Using all the data of /Thibault et al. 1990/ including the data for aerated soils from /Sheppard et al. 1990/, there was no statistically significant relationship to soil properties. It is proposed to use a constant:

the overall GM for aerated soils was 3.1 L kg^{-1} with a GSD of 5.8.

From /Sheppard et al. 1990/:

the overall GM for anaerobic soils was 65 L kg^{-1} with a GSD of 4.4.

5.3.27 Th – Thorium

The Kd values for Th were measured in the 7 SKB soils and in 198 Canadian soils in the NTE program (Table 5-25). The stepwise regression of the NTE soil included only pH, with a residual GSD of 3.4. The stepwise regression of the /Thibault et al. 1990/ data (23 values) included only pH, with a residual GSD of 15.

/EMRAS 2008/ found 8 reports with 46 values of Kd, GM values were sand: 700 L kg^{-1}, loam: 18,000 L kg^{-1}, clay: 4,500 L kg^{-1} and organic: 730 L kg^{-1}. /Gooddy et al. 1995/ reported 12 Kd values based on indigenous Th in acidic sandy soils, with a GM of 3,700 L kg^{-1}.

Combining the NTE and Thibault et al. data sets, along with data from /Sheppard and Hawkins 1991/ provided 39 values and the equation:

$\text{Log (Kd)} = 1.90 + 0.346 \cdot (\text{pH})$

with a residual GSD of 11 and $r^2 = 0.16$ and estimates for the SKB soils of 14,000, 1,300, 4,300, 4,300, 1,900, 4,300 and 6,400 L kg^{-1}, respectively.

5.3.28 Tm – Thulium

The Kd values for Tm were measured in the 7 SKB soils and in 87 Canadian soils in the NTE program (Table 5-26). The stepwise regression of the NTE soil included pH, sand, silt and the clay*pH interaction, with a residual GSD of 3.6. /IAEA 1994/ did not estimate a Kd for Tm, and /EMRAS 2008/ proposed the value 330 L kg^{-1} based on one measurement.

Table 5-25. Measured (SKB) and predicted (using the NTE and Thibault et al. data sets) Kd values (L kg^{-1}) for Th.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD	Thibault et al. sorption predicted Kd ± 1GSD
A Forsmark clay till	31,000	8,800–100,000	1,400–270,000
B Forsmark deep peat	34,000	1,300–15,000	18–3,500
C Simpevarp sandy till	5,500	3,400–39,000	160–30,000
D Simpevarp clay gyttja	14,000	3,400–39,000	160–30,000
E Simpevarp peat	9,300	1,800–21,000	37–7,100
F Simpevarp clay gyttja	34,000	3,400–39,000	160–30,000
G Forsmark clay till	25,000	4,700–54,000	330–62,000

Table 5-26. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Tm.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	> 2,000nd	4,700–61,000
B Forsmark deep peat	––nd	1,500–20,000
C Simpevarp sandy till	2,600	1,100–12,000
D Simpevarp clay gyttja	4,300	970–13,000
E Simpevarp peat	2,500	600–7,800
F Simpevarp clay gyttja	7,600	1,100–14,000
G Forsmark clay till	> 20,000	1,800–23,000

nd pore water concentration non-detectable for Sites A and G, these values computed using the pore water detection limit. Not detectable in both solids and pore water for Site B, so no limit value can be estimated.

/EMRAS 2008/ reported 1 Kd value of 330 L kg^{-1}. Because of the scarcity of data, the equation from the NTE data set is proposed:

$$\text{Log (Kd)} = 5.656 + 0.524 \cdot (\text{pH}) - 0.0464 \cdot (\text{sand}) - 0.0496 \cdot (\text{silt}) - 0.00841 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 3.6 and $r^2 = 0.29$. This gives values for the SKB soils of 17,000, 5,400, 4,000, 3,500, 2,200, 4,000 and 6,400 L kg^{-1}, respectively.

5.3.29 U – Uranium

The Kd values for U were measured in the 7 SKB soils and in 209 Canadian soils in the NTE program (Table 5-27). As noted in the review by /Sheppard et al. 2006/, Kd for U showed two different populations of data that responded differently to soil properties, divided by pH at about 5.5. Above a pH of 5.5, U tends to be solubilised by carbonate complexes, and so Kd decreases with pH above 5.5. Two stepwise regressions were developed. In addition, there were no NTE soils with pH > 5.5 and organic carbon > 20%. Simple stepwise regression included organic carbon as an independent variable, but because of a skew in this variable at high pH the predictions were spurious. Thus organic carbon was purposefully eliminated from the regression.

The stepwise regression of the NTE soil pH > 5.5 (182 soils) included pH, and clay, with a residual GSD of 4.2. Below pH 5.5 (30 soils), there was no significant equation and the GM was 3,200 L kg^{-1}, GSD 3.2. The data from the review by /Sheppard et al. 2006/ and references therein, which includes almost all the data from /Thibault et al. 1990/, was also handled in two regressions. For soils pH > 5.5 (123 soils), pH, organic carbon and the clay*pH interaction were included, with a residual GSD of 9.8. For soils pH < 5.5 (24 soils), only soil pH was included with a residual GSD of 11. /Vandenhove et al. 2007/ gave Kd values for 18 quite different mineral soils (24 soils, all low organic carbon), and the stepwise regression included pH and organic carbon, with a residual GSD of 2.3. From /Thibault et al. 1990/, with 46 values, there was no relationship to soil properties and the GM was 120 with a GSD of 33.

/EMRAS 2008/ found 22 reports with 178 values of Kd. GM values were sand: 110 L kg^{-1}, loam: 310 L kg^{-1}, clay: 28 L kg^{-1} and organic: 1,200 L kg^{-1}. EMRAS did note that Kd varied with pH and estimated Kd values as pH < 3: 71 L kg^{-1}, 5 < pH < 7: 740 L kg^{-1} and pH > 7: 65 L kg^{-1}.

Because there did not seem to be a consistent difference among the NTE, OPG and other data sets, they were combined. The resulting regression for soils pH > 5.5 (318 soils) and excluding organic carbon was:

$$\text{Log (Kd)} = 9.05 - 0.989 \cdot (\text{pH}) + 0.00290 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 6.6 and $r^2 = 0.44$. This gave predictions for the SKB sites A, C, D, F and G of 610, 2,000, 31,000, 28,000 and 4,900 L kg^{-1}, respectively.

The regression for soils pH < 5.5 (32 soils) was:

$$\text{log (Kd)} = 1.79 + 0.0152 \cdot (\text{clay}) \cdot (\text{pH})$$

Table 5-27. Measured (SKB) and predicted (using literature data from several sources) Kd values (L kg^{-1}) for U.

SKB soil	Measured desorption Kd	NTE desorption predicted pH < 5.5 ± 1GSD	NTE desorption predicted pH > 5.5 ± 1GSD	OPG sorption predicted pH < 5.5	OPG sorption predicted pH > 5.5	Vandenhove et al. 2007
A Forsmark clay till	610	(10^3–10^4)[1]	170–2,900	(120)	270	600
B Forsmark deep peat	1,500	10^3–10^4	——[1]	70	——	——[2]
C Simpevarp sandy till	1,800	10^3–10^4	(1,800–31,000)	92	(7,100)	3,200
D Simpevarp clay gyttja	5,700	10^3–10^4	(3,400–59,000)	92	(85,000)	——
E Simpevarp peat	12,000	10^3–10^4	——	77	——	——
F Simpevarp clay gyttja	44,000	10^3–10^4	(3,300–58,000)	92	(66,000)	——
G Forsmark clay till	3,300	10^3–10^4	840–15,000	100	2,600	2,300

[1] Because the regressions were specific to pH, estimates in brackets are for SKB sites where pH is within 1 unit of the range of the regression, and estimates are not shown if the pH of the site is more than 1 unit outside the range of the regression.

[2] The data of /Vandenhove et al. 2007/ had few high organic carbon soils, the extrapolated values seemed spurious (> 10^6 L kg^{-1}) and so are not shown.

with a residual GSD of 9.4 and $r^2 = 0.22$. This gave predictions for the SKB sites A through G of 470, 65,000, 73, 6,800, 2,700, 4,000 and 160 L kg^{-1}, respectively. These values do not agree well with those observed for the SKB soils. Clearly Kd for acidic organic soils is not well known. The combined data and the corresponding regression lines (clay = 15%) versus pH are shown in Figure 5-9. The combined data and the corresponding regression lines (pH = 5.5 and pH = 8.5) versus clay content are shown in Figure 5-10.

5.3.30 Yb – Ytterbium

The Kd values for Yb were measured in the 7 SKB soils and in 203 Canadian soils in the NTE program (Table 5-28). The stepwise regression of the NTE soil included pH, clay, sand and silt contents, with a residual GSD of 3.2. Neither /IAEA 1994/ nor /EMRAS 2008/ proposed Kd values for Yb.

Based on the NTE data set, the regression is:

$$\text{Log (Kd)} = 4.29 + 0.320 \cdot (\text{pH}) - 0.017 \cdot (\text{sand}) - 0.0194 \cdot (\text{silt}) - 0.0041 \cdot (\text{clay}) \cdot (\text{pH})$$

with a residual GSD of 3.2 and $r^2 = 0.065$. This gives values for the SKB soils of 31,000, 6,200, 14,000, 8,600, 5,700, 9,200 and 18,000 L kg^{-1}, respectively.

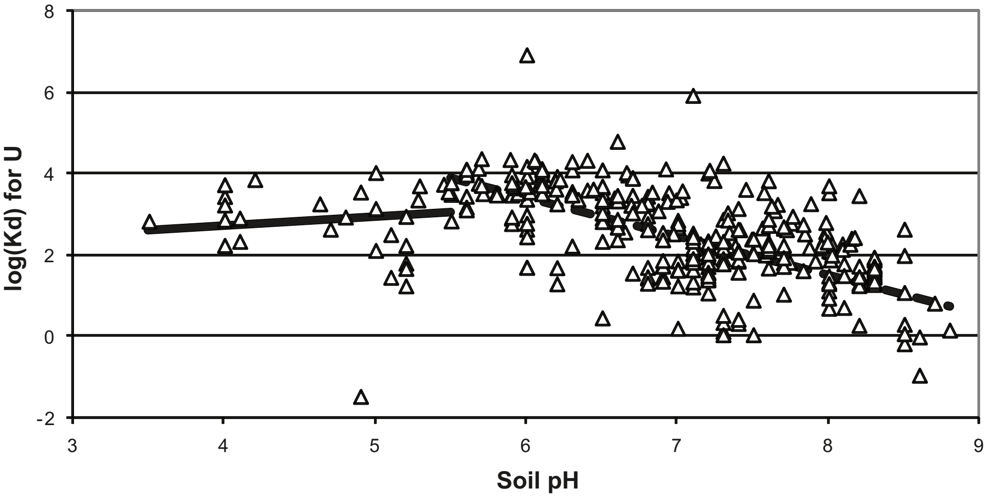

Figure 5-9. Scatter plot of combined Kd data for U versus pH with the lines showing the regression estimates for soils clay = 15% and pH < 5.5 or pH > 5.5.

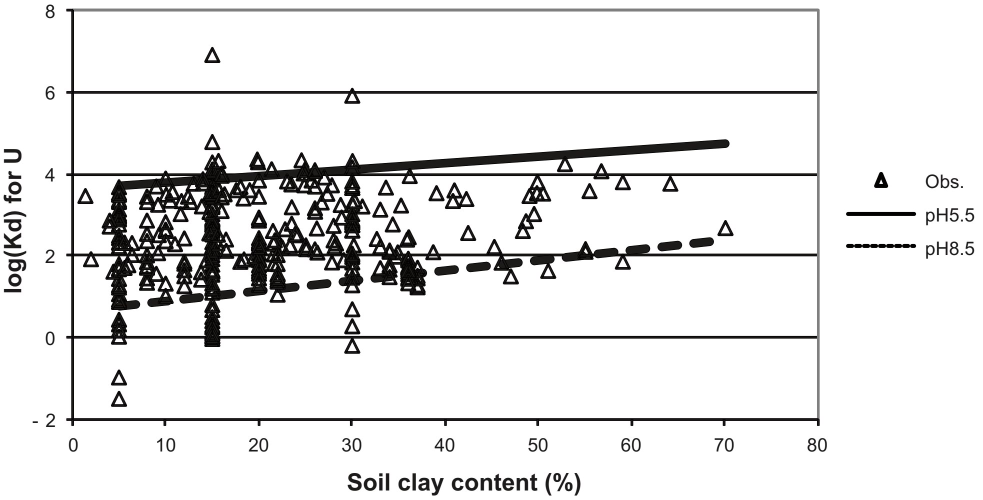

Figure 5-10. Scatter plot of combined Kd data for U versus clay content with the lines showing the regression estimates for soils pH = 5.5 and pH = 8.5.

Table 5-28. Measured (SKB) and predicted (using the NTE data set) Kd values (L kg^{-1}) for Yb.

SKB soil	Measured desorption Kd	NTE desorption predicted Kd ± 1GSD
A Forsmark clay till	18,000	9,700–98,000
B Forsmark deep peat	3,300	2,000–20,000
C Simpevarp sandy till	1,900	4,500–45,000
D Simpevarp clay gyttja	2,800	2,700–27,000
E Simpevarp peat	1,800	1,800–18,000
F Simpevarp clay gyttja	7,400	2,900–29,000
G Forsmark clay till	28,000	5,600–56,000

5.4 Summary and overview of soil Kd

Values of Kd have substantial inherent variation, a typical GSD is about 3 to 5, which implies a 95th percentile confidence range of about 80-fold to 600-fold. These ranges apply to situations where some attributes of the soil are known: variation on generic values is usually larger. As such, many efforts to obtain site-specific values are potentially misleading; differences in Kd of less than 10-fold are seldom meaningful. Nonetheless, developing statistically valid relationships to predict Kd values for specific soils is important, as long as the overall level of uncertainty is not forgotten.

In this report, we developed regression equations to predict Kd based on soil pH, clay content and organic carbon content – reporting only equations and independent variables that were statistically significant. Simultaneous to this work, the EMRAS organization has proposed revised soil Kd values that will be published as an IAEA Technical Document. These values are plotted (Figure 5-11), contrasting the estimates from our regressions versus EMRAS estimates to correspond to a specific acidic sandy soil at one of the SKB sites. Each point is a different element. Deviation of more than 10-fold is evident, and in general the estimates here are higher than those from EMRAS.

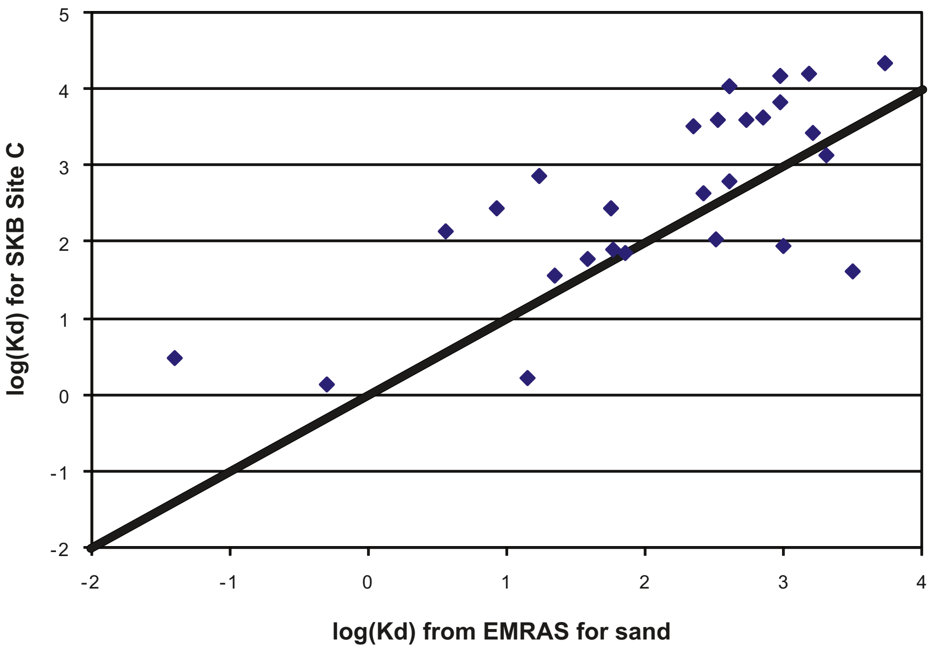

Figure 5-11. *Comparison of Kd estimates (each point is a different element) for the acidic sandy soil SKB-Site C from the regressions in this report versus those for sandy soils from /EMRAS 2008/.*

The independent variable most often found to significantly affect Kd was soil pH. The effect of pH on Kd for all the elements considered is shown in Figure 5-12. For most elements, the effect of a change in pH from 4 to 7 has less than a 20-fold effect, with higher Kd at higher pH. This level of effect is comparable to the random error in Kd values, and in many site-specific assessments there would not be such a broad range in soil pH. Several elements were not affected by pH, and Kd values for Mo, Sb, U and Tc decreased at higher pH. Note that for U, there are separate relationships for Kd above and below pH 5.5, and as developed they do not converge to the same Kd at pH 5.5 (this is considered a statistical feature, not a mechanistic discontinuity).

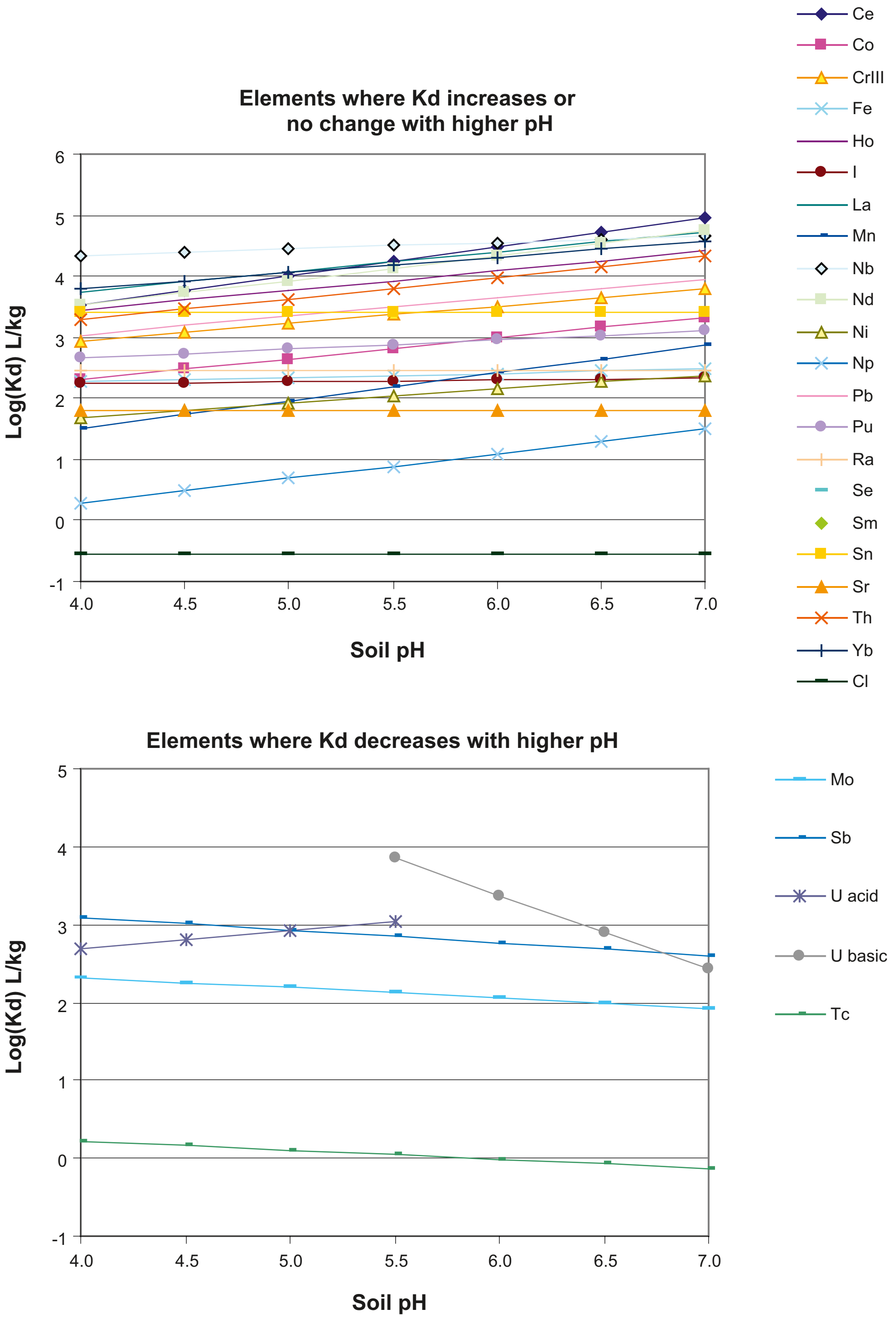

Figure 5-12. *Predicted log (Kd) versus soil pH for the elements considered, differentiating those where Kd increases (or no change) with pH (upper) versus those where Kd decreases with pH (lower).*

An interesting implication of the general effect of pH is that the environmental fates of most of these elements will co-vary with changes in pH. For example, in acidic soils, many of these elements will be more readily leached and probably more readily absorbed by plants. /Sheppard and Sheppard 1988/ explored the effect of soil acidification (acid rain) on dose with emphasis on Ra, in fact there would be an effect on dose from many of these elements. The change in combined dose with change in soil pH would be interesting.

5.5 Regression equations to predict soil Kd

Table 5-29 is a summary of regression equations for Kd (L kg^{-1}), showing the equation estimates for the acidic sandy till of Site C and the corresponding estimate from the /EMRAS 2008/ review.

Table 5-29. Summary of regression equations and comparison between prediction for Site C and corresponding estimate from the /EMRAS 2008/ review (Kd, L kg^{-1}).

Elem.	Regression equation	Est.	EMRAS
Ce	$\text{Log (Kd)} = 3.51 + 0.54400 \cdot \text{(pH)} - 0.022 \cdot \text{(sand)} - 0.022 \cdot \text{(silt)} - 0.00471 \cdot \text{(clay)} \cdot \text{(pH)}$	11,000	400
Cl	1.4 L kg^{-1} for mineral soils (< 20% organic carbon), 150 L kg^{-1} for organic soils	1.4	0.5
Co	$\text{Log (Kd)} = 0.898 + 0.339 \cdot \text{(pH)} + 0.0267 \cdot \text{(organic carbon)}$	440	260
Cr	$\text{Log (Kd)} = 1.79 + 0.287 \cdot \text{(pH)}$ where trivalent Cr is expected $\text{Log (Kd)} = 0.973$ where hexavalent Cr is expected	280	8.4
Cs	$\text{log (Kd)} = 3.19 + 0.0798 \cdot \text{(pH)} + 0.00154 \cdot \text{(clay)} \cdot \text{(pH)}$ for long-lived Cs	4,000	530
Fe	$\text{log (Kd)} = 2.01 + 0.00442 \cdot \text{(clay)} \cdot \text{(pH)}$	110	320
Ho	$\text{Log (Kd)} = 2.15 + 0.338 \cdot \text{(pH)} - 0.00094 \cdot \text{(clay)} \cdot \text{(pH)}$	6,800	930
I	$\text{Log (Kd)} = 2.13 + 0.00297 \cdot \text{(organic carbon)} + 0.00181 \cdot \text{(clay)} \cdot \text{(pH)}$	140	3.6
La	$\text{Log (Kd)} = 3.22 - 0.0506 \cdot \text{(clay)} + 0.230 \cdot \text{(pH)} + 0.00617 \cdot \text{(clay)} \cdot \text{(pH)}$	22,000	5,300
Mn	$\text{log (Kd)} = -0.330 + 0.457 \cdot \text{(pH)}$	90	980
Mo	$\text{Log (Kd)} = 1.70 + 0.0718 \cdot \text{(clay)} + 0.0325 \cdot \text{(organic carbon)} - 0.00873 \cdot \text{(clay)} \cdot \text{(pH)}$	61	38
Nb	$\text{Log (Kd)} = 2.45 + 0.0960 \cdot \text{(clay)} + 0.348 \cdot \text{(pH)} - 0.0159 \cdot \text{(clay)} \cdot \text{(pH)}$	16,000	1,500
Nd	$\text{Log (Kd)} = 3.523 + 0.476 \cdot \text{(pH)} - 0.0200 \cdot \text{(sand)} - 0.0185 \cdot \text{(silt)} - 0.00435 \cdot \text{(clay)} \cdot \text{(pH)}$	8,300	——[na]
Ni	$\text{Log (Kd)} = 0.718 + 0.233 \cdot \text{(pH)} + 0.0148 \cdot \text{(organic carbon)}$	81	58
Np	$\text{log (Kd)} = -1.42 + 0.307 \cdot \text{(pH)} + 0.0429 \cdot \text{(organic carbon)} + 0.00638 \cdot \text{(clay)} \cdot \text{(pH)}$	1.7	14
Pa	1380 L kg^{-1} for mineral soils, 6600 L kg^{-1} for organic soils	1,380	2,000
Pb	$\text{Log (Kd)} = 2.41 - 0.043 \cdot \text{(clay)} + 0.216 \cdot \text{(pH)} + 0.0214 \cdot \text{(organic carbon)} + 0.00601 \cdot \text{(clay)} \cdot \text{(pH)}$	3,300	220
Pu	$\text{Log (Kd)} = 2.00 + 0.150 \cdot \text{(pH)} + 0.0318 \cdot \text{(organic carbon)}$	630	400
Ra	$\text{Log (Kd)} = 1.56 + 0.0599 \cdot \text{(clay)}$	42	3,100
Sb	$\text{Log (Kd)} = 3.74 - 0.175 \cdot \text{(pH)} + 0.00083 \cdot \text{(clay)} \cdot \text{(pH)}$	740	17
Se	$\text{Log (Kd)} = 1.79 + 0.133 \cdot \text{(pH)} - 0.00163 \cdot \text{(clay)} \cdot \text{(pH)}$	280	56
Sm	$\text{Log (Kd)} = 2.73 - 0.00862 \cdot \text{(clay)} + 0.290 \cdot \text{(pH)}$	15,000	930
Sn	$\text{Log (Kd)} = 3.60 - 0.0983 \cdot \text{(organic carbon)}$	2,700	1,600
Sr	$\text{Log (Kd)} = 1.55 + 0.0160 \cdot \text{(clay)}$	37	22
Tc	3.1 L kg^{-1} for aerobic soils, 65 L kg^{-1} for anaerobic soils	3.1	0.04
Th	$\text{Log (Kd)} = 1.90 + 0.346 \cdot \text{(pH)}$	4,300	700
Tm	$\text{Log (Kd)} = 5.656 + 0.524 \cdot \text{(pH)} - 0.0464 \cdot \text{(sand)} - 0.0496 \cdot \text{(silt)} - 0.00841 \cdot \text{(clay)} \cdot \text{(pH)}$	4,000	330
U	if $\text{pH} > 5.5$ then $\text{Log (Kd)} = 9.05 - 0.989 \cdot \text{(pH)} + 0.00290 \cdot \text{(clay)} \cdot \text{(pH)}$ otherwise $\text{log (Kd)} = 1.79 + 0.0152 \cdot \text{(clay)} \cdot \text{(pH)}$	73	71
Yb	$\text{Log (Kd)} = 4.29 + 0.320 \cdot \text{(pH)} - 0.017 \cdot \text{(sand)} - 0.0194 \cdot \text{(silt)} - 0.0041 \cdot \text{(clay)} \cdot \text{(pH)}$	14,000	——[na]

[na] Not available.

6 Review and comparison of measured and literature aquatic Kd values

6.1 Introduction

Where soil Kd values differed to some extent among soil types, aquatic Kd values potentially differ between freshwater and marine, and between bottom sediment and suspended sediment. As with soil Kd, there may be differences in Kd depending on measurement method, which includes potential differences among data for stable elements versus their radioactive isotopes.

6.1.1 General methods

We conducted a meta-analysis to determine the Kd values of the 28 elements of interest to SKB, specifically within aquatic environments. We examined > 130 peer-reviewed articles, of which only a few were reviews or compendia of other works. We also made use of industry documents, as appropriate.

Our literature review covered as wide a range of source material as possible. Essentially, we conducted a stratified search of the literature where the strata were target element and type of aquatic environment. Overall, the literature revealed that research has been conducted in three main aquatic environments: marine, freshwater (usually lotic), and mixed – especially estuarine – systems. We obtained a range of publications representative of the field and experimental conditions under which Kd values have been reported. As such, this review has produced a solid representation of global Kd values for most of the target elements. Global Kd values were calculated from this pool of values and are presented along with values from /IAEA 1985, 2004/ and /EMRAS 2008/ and Kd values derived from information from the coastal area of eastern Sweden as measured by SKB.

Sediment Kd values are neither consistently defined nor reported within the literature. However, the most common definition and formula for Kd can be found in /Turner et al. 1992/, and is consistent with our usage throughout this report. We report Kd values in the most common units of L kg^{-1}, but confusion arises because many papers report Kd as unitless, with the implied units of (mg kg^{-1} dry sediment)/(mg kg^{-1} water). Given the variation in Kd, the assumption of 1 kg water = 1 L water is sufficiently accurate even for saline water, and we used this assumption for such data.

6.1.2 Determining literature-based Kd values

In most cases, Kd values were taken as reported in the literature. In a few cases, it was possible to interpolate or cautiously extrapolate values from literature that did not directly report Kd. Geometric means (GM) were used to summarize data within each study across replicates and across other factors that were not important to our overall objective. For example, time series samples at a given site would be considered one observation for this review, and a single GM would be recorded.

In some cases, both the minimum and maximum values from within a single literature source were entered into our master data set. In these circumstances, each value was used as a discrete data point from which the GM was later calculated. This approach maintains some measure of the variability found in the study. Another issue that arose was where authors reported only median values. With the assumption that Kd values are lognormally distributed (supported by the Central Limit Theorem and many empirical evaluations of the distribution of Kd), then median and GM should be similar and the medians were entered into our data set.

It was evident that some Kd values were based on many observations while others were based on relatively few. As a result, we decided to include as much information about number of observations as possible. Although it was not always possible to distinguish number of independent observations from the literature publications, we extracted > 220 estimates of Kd representing > 3,000 observations.

The number of observations for each Kd value, N (shown in Table 6-1) is reported at two levels: 1) the number of literature sources (publications) used to obtain the Kd data; and 2) the sum of the numbers of underlying observations within the literature sources. The second usually were comparatively large numbers and some of these were uncertain because of lack of clarity in the publications. In some cases, it appeared that results within a study were based on multiple observations, but some of these multiple observations may not have been truly independent observations. In other circumstances, extensive cross referencing revealed that the same data were used in more than one publication. However, we provide this number of apparent underlying observations as a gauge of the robustness of the data.

Although Kd values have been reported for various elements in a variety of aqueous environments, including estuaries, oceans, thermal vents and wetlands, we reduced the categorisation of our data into two broad groups of water: fresh and saline. The latter includes both estuaries and oceanic environments. While estuaries are generally less saline than oceans, they are usually only moderately so. The out-flowing riverine fresh water will generally lead to only limited dilution of the receiving salt water in the energetic tidal bays. It appears that estuaries were often sampled because they represented end-points for fluvial systems and therefore they offer the comparatively high concentrations of trace elements necessary for conducting the research.

Table 6-1. Sediment Kd values (L kg^{-1}) obtained from the literature reviewed in this study (indicated by 'Lit.') and from /EMRAS 2008/ and /IAEA 1985, 2004/. Where possible, EMRAS categorised Kd values and here the values from in situ studies (the most preferred values) or from desorption or adsorption studies that last at least 120 hours are reported. Note these EMRAS and IAEA values are for suspended sediment. The study types for the literature values include F = field in situ; E = experimental; L = lab-based and M = model.

	Fresh-water Kd	Source	N refs.	N obs.	Study type	Marine-water Kd	Source	N refs.	N obs.	Study type
Cl	98	Lit.	1	> 20	F	1	IAEA 04			
Co	390,000	EMRAS	2	14	des +120 hrs	23,000	Lit.	12	159	F
	150,000	EMRAS	11	101	ads +120 hrs	5 x 10^7	IAEA 04			
	44,000	EMRAS	5	29	in situ					
	60,000	Lit.	9	125	F					
Cr	42,000	Lit.	9	174	F	30,000	Lit.	2	10	F
						40,000	IAEA 04			
Cs	51,000	EMRAS	2	23	des +120 hrs	540	Lit.	11	57	F, M
	29,000	EMRAS	17	219	in situ	2,000	IAEA 85, 04			
	17,000	Lit.	9	> 69	F					
Fe	160,000	Lit.	11	181	F	2,900,000	Lit.	12	498	F
	5,000	EMRAS				2 x 10^8	IAEA 04			
Ho	230,000	Lit.	1	12	F					
I	220	EMRAS	10	158	ads	30	Lit.	1	28	F
	29	Lit.	3	39	F	200	IAEA 85, 04			
La	300,000	Lit.	3	> 60	F					
Mn	660,000	EMRAS	3	20	des 2–24 hrs	48,000	Lit.	18	> 494	F
	540,000	EMRAS	6	20	ads +120 hrs	2 x 10^8	IAEA 85, 04			
	78,000	EMRAS	4	20	in situ					
	28,000	Lit.	16	215	F					
Mo	1,100	Lit.	3	79	F					
Nb	10,000	IAEA 85				300,000	IAEA 04			
Nd	870,000	Lit.	2	30	F					
Ni	1,500	Lit.	15	247	F, M	17,000	Lit.	21	> 717	F, M
						300,000	IAEA 04			

Table 6-1 Continued.

	Fresh-water Kd	Source	N refs.	N obs.	Study type	Marine-water Kd	Source	N refs.	N obs.	Study type
Np	10	EMRAS			F	5,000	Lit.	1	4	F
						1,000	IAEA 04			
Pa						1,100,000	Lit.	4	> 95	F
						5,000,000	IAEA 04			
Pb	130,000	Lit.	13	333	F	180,000	Lit.	19	> 715	F
						1×10^7	IAEA 04			
Pu	300,000	EMRAS	4	41	des	1,100,000	Lit.	15	> 541	F & M
	240,000	EMRAS	6	79	in situ	100,000	IAEA 85, 04			
	79,000	EMRAS	3	37	ads					
Ra	7,400	EMRAS	7	75		30	Lit.	1	12	F
						4,000	IAEA 04			
Sb	5,000	EMRAS	5	23		1,100	Lit.	1	3	F
	4,100	Lit.	4	> 88	F	4,000	IAEA 04			
Se	470	Lit.	2	61	F	260	Lit.	1	3	F
						1,000	IAEA 85, 04			
Sm	430,000	Lit.	3	> 50	F	500,000	IAEA 04			
Sn						300,000	IAEA 04			
Sr	1,200	EMRAS	3	13	in situ	5	Lit.	4	> 6	F, E
	620	EMRAS	3	34	des	200	IAEA 85, 04			
	540	EMRAS	2	13	ads+120 hrs					
	480	Lit.	8	> 102	F					
Tc	0	Lit.	1	9	F	1,400	Lit.	1	12	F
	5	EMRAS				100	IAEA 85, 04			
Th	180,000	EMRAS	7	63		4,500,000	Lit.	6	> 125	F, M
	87,000	Lit.	2	> 40	F	5,000,000	IAEA 85, 04			
Tm	220,000	Lit.	2	23	F	200,000	IAEA 04			
U	15,000	Lit.	3	> 58	F	860	Lit.	3	31	F
						500	IAEA 85, 04			
Yb	350,000	Lit.	1	12	F	200,000	IAEA 04			

The Kd values were also reported for a variety of isotopes and chemical species. Information about isotope and chemical species was retained in the master data set. However, at present Kd values of all species and isotopes of an element were treated as those of the element. We anticipate that future work could be done to seek finer resolution in Kd values by examining Kd values for each of the chemical species of each element under a full suite of environmental conditions.

6.1.3 IAEA and EMRAS Kd values

We offer for comparative purposes, the marine pelagic and coastal Kd values reported by /IAEA 1985, 2004/ and freshwater suspended sediment values in the draft report obtained from the EMRAS program /EMRAS 2008/. /EMRAS 2008/ will in future be assigned an IAEA Technical Report Series number.

6.1.4 Determining SKB Kd values

In theory, Kd values could be considered site specific because they are empirical and depend upon many environmental (and methodological) variables. For example /Comber et al. 1995/ reported differences in Kd by an order of magnitude from two proximate English rivers – the Mersey which drains the northwest and the Humber which drains the northeast. Thus, site-specific Kd values may be quite different from the global values for any element.

We calculated site-specific Kd values for the Swedish east coastal area, based on data produced by SKB (the appendices in /Engdahl et al. 2008/ and from /Kumblad and Bradshaw 2008/. The Kd values for 23 of the 27 target elements were calculated for freshwater and marine benthic (bottom) sediments for both the Forsmark and Oskarshamn (Simpevarp) locations. The Kd values were also determined for suspended particulate matter (SPM) for both marine and freshwater systems, again for both Forsmark and Oskarshamn.

Benthic sediment Kd values were computed from /Engdahl et al. 2008/ for Forsmark as the concentration of filtered sediment (dry-weight basis, their Appendix 9) divided by the corresponding pore water concentrations (their Appendix 11), with appropriate unit conversions. For the Oskarsham site it was concentration of filtered sediment (dry-weight basis, their Appendix 12) divided by pore water concentrations (their Appendix 14).

Values of Kd for suspended sediment (suspended particulate matter – SPM) for Forsmark used the total elemental content of the sediment and filter (their Appendix 1) less the blank for this type of filter along with any chemi-sorbed element (their Appendix 2), all divided by the corresponding water concentrations (their Appendix 4). The corresponding values for Oskarsham involved their Appendices 5, 6 and 8. The Kd values from the study by /Kumblad and Bradshaw 2008/ were tabulated by the authors, and were converted to units of L kg^{-1} for this report.

All of the SKB Kd values were combined into one data set, and log-transformed for statistical analysis. Each study had different numbers of sampling locations and other factors, resulting in an imbalanced design for statistical interpretation. The objective of the statistical interpretation was to provide a systematic means to determine which factors could be averaged and which required specific consideration. This was done with analysis of variance (ANOVA) of each element separately, following a stepwise analysis plan to deal with the imbalance. The first series of ANOVA were completely randomized designs with only one factor specified in each ANOVA. The factors, considered in separate ANOVA for each factor and each element, were water type (fresh versus marine), sediment type (benthic versus suspended), study (/Engdahl et al. 2008/ versus /Kumblad and Bradshaw 2008/), site (Forsmark versus Oskarsham) or sediment depth (3 depths were given by /Kumblad and Bradshaw 2008/).

The factors study, site and sediment depth were statistically significant for only 1 or 2 elements, whereas sediment type was statistically significant for almost all elements. The effects of water type were intermediate. It was decided that this was sufficient justification to attribute differences between sites, studies and sediment depth as statistical error, where the statistical objective was to characterize Kd for sites along the eastern coast. This improved the balance of the statistical design, so the next series of ANOVA were two-way factorials of water and sediment types along with the two-way interaction of these factors.

For several elements, the two-way interaction was significant, implying that to properly represent the data, it was necessary to derive values for both water types and both sediment types. As a result, the tables of values show four Kd values for each element. Clearly, the four values are not statistically different for all elements, but they are for some elements. The residual variance (the error term) from the ANOVA was used to compute a residual GSD. The interpretation of this GSD is the unexplained variance after accounting for the effects of water type and sediment type.

6.2 Results

The Kd values obtained from the literature and from /EMRAS 2008/ and /IAEA 1985, 2004/ are shown in Table 6-1, and references from the literature are shown in Tables 6-2 and 6-3. More detailed comparison of these values and estimates of variation are given in later tables, the most revealing information from Table 6-1 is the numbers of observations supporting each value and the differences between field in situ values and values obtained from sorption and desorption experiments. In general, most of these values are supported by substantial numbers of both reports and individual observations. Among the /EMRAS 2008/ data, the in situ values were lower than the experimental values except for Pu and Sr.

Table 6-2. Literature used to determine Kd values for freshwater sediment, both suspended and benthic.

	Literature Kd	References
Cl	98	/Vesely et al. 2001/
Co	60,000	/Ciffroy et al. 2003, Marcussen et al. 2007, Neal et al. 1997, Nguyen et al. 2005, Queirazza et al. 1987, Roychoudhury and Starke 2006, Tipping et al. 1998, Vesely et al. 2001, Zhang et al. 1994, Zhang 1995/
Cr	42,000	/Davide et al. 2003, Marcussen et al. 2007, Pertsemli and Voutsa 2007, Queirazza et al. 1987, Roychoudhury and Starke 2006, Vesely et al. 2001, Routh and Ikramuddin 1996, Neal et al. 1997, Turner 1996/
Cs	17,000	/Ciffroy et al. 2003, Cochran et al. 2000, Oughton et al. 1997, Queirazza et al. 1987, Stephens et al. 1998, Strand et al. 1999, Turner 1996, Turner et al. 1993, Vesely et al. 2001/
Fe	160,000	/Ciffroy et al. 2003, Drndarski et al. 1993, Marcussen et al. 2007, Neal et al. 1997, Pertsemli and Voutsa 2007, Queirazza et al. 1987, Routh and Ikramuddin 1996, Roychoudhury and Starke 2006, Vesely et al. 2001, Zhang et al. 1994, Zhang 1995/
Ho	230,000	/Weltje et al. 2002/
I	29	/Bird and Schwartz 1996, Cochran et al. 2000, Drndarski et al. 1993/
La	300,000	/Tosiani et al. 2004, Vesely et al. 2001, Weltje et al. 2002/
Mn	28,000	/Ciffroy et al. 2003, Dassenakis et al. 1997, Davide et al. 2003, Davis et al. 1991, Drndarski et al. 1993, Marcussen et al. 2007, Neal et al. 1997, Pertsemli and Voutsa 2007, Queirazza et al. 1987, Routh and Ikramuddin 1996, Roychoudhury and Starke 2006, Standring et al. 2002, Turner et al. 1993, Vesely et al. 2001, Zhang et al. 1994, Zhang 1995/
Mo	1,100	/Routh and Ikramuddin 1996, Roychoudhury and Starke 2006, Marcussen et al. 2007/
Nd	870,000	/Marcussen et al. 2007, Weltje et al. 2002/
Ni	1,500	/Christensen et al. 1996, Dassenakis et al. 1997, Davide et al. 2003, Drndarski et al. 1993, Marcussen et al. 2007, Neal et al. 1997, Nguyen et al. 2005, Pertsemli and Voutsa 2007, Routh and Ikramuddin 1996, Roychoudhury and Starke 2006, Tipping et al. 1998, Turner 2007, Young and Harvey 1992, Zhang 1995, Zhang et al. 1994/
Pb	130,000	/Dassenakis et al. 1997, Davide et al. 2003, Drndarski et al. 1993, Marcussen et al. 2007, Neal et al. 1997, Nguyen et al. 2005, Pertsemli and Voutsa 2007, Routh and Ikramuddin 1996, Roychoudhury and Starke 2006, Tipping et al. 1998, Vesely et al. 2001, Zhang 1995, Zhang et al. 1994/
Sb	4,100	/Drndarski et al. 1993, Marcussen et al. 2007, Routh and Ikramuddin 1996, Vesely et al. 2001/
Se	470	/Routh and Ikramuddin 1996, Roychoudhury and Starke 2006/
Sm	430,000	/Weltje et al. 2002, Vesely et al. 2001, Marcussen et al. 2007/
Sr	480	/Drndarski et al. 1993, Oughton et al. 1997, Queirazza et al. 1987, Routh and Ikramuddin 1996, Roychoudhury and Starke 2006, Stephens et al. 1998, Strand et al. 1999, Vesely et al. 2001/
Tc	0	/Krupka and Serne 2002/
Th	87,000	/Vesely et al. 2001, Roychoudhury and Starke 2006/
Tm	220,000	/Marcussen et al. 2007, Weltje et al. 2002/
U	15,000	/Marcussen et al. 2007, Roychoudhury and Starke 2006, Vesely et al. 2001/
Yb	350,000	/Weltje et al. 2002/

The comparison of /EMRAS 2008/ values to the literature is best illustrated by Figure 6-1. In general, the two sources agreed within a factor of 10, with the exception of Fe. The Kd for Fe may be responsive to redox conditions, and this may account for the difference between the sources. The grey lines in Figure 6-1 are 2 GSD above and below the 1:1 line, using the median GSD across elements to give an approximation of 95th percentile confidence bounds. Thus, if there are sufficient numbers of observations, only elements outside the grey lines would be cases where the EMRAS and literature data were statistically different. The results indicate that the two sources of data were not different, which is expected because both may have drawn on some of the same primary literature.

The freshwater Kd values for the SKB sites are shown in Table 6-4 along with the corresponding /EMRAS 2008/ and literature values. Note that the residual GSD for the SKB sites is almost always lower than the GSD from the literature, and this makes sense because the residual GSD reflects variation among sampling locations that are relatively close together and where consistent analytical methods were applied. The GSD from the literature includes variation among globally dispersed sites and among laboratories and measurement methods. The extreme GSD values (above ~12) from the literature probably should not be used for stochastic assessment, as it is not clear what portion of that variance is the result of methodological issues or unusual site characteristics.

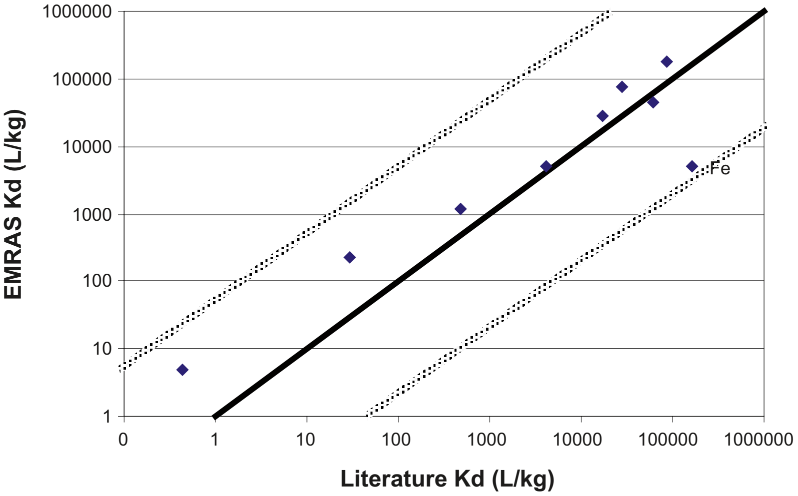

Figure 6-1. Comparison of /EMRAS 2008/ values (the in situ values when these were specified) versus values obtained from the literature for freshwater. Each point is a different element (data from Table 6-1), and Fe is labelled as the element furthest from the 1:1 line. The Kd for Fe is redox sensitive and this may account for the differences in values. The grey lines are 2 GSD above and below the 1:1 line, approximating the 95th percentile confidence bounds using the median GSD across elements for literature data.

The comparison of SKB values to the literature is most easily seen in Figure 6-2. Although some values deviate from the 1:1 line by more than a factor of 10, they generally are symmetrical about the 1:1 line and within the approximate 95th percentile confidence bounds indicated by the grey lines. This suggests that the Kd values for SKB sites belong in the same data population as the literature, suggesting that literature values are applicable to the SKB sites. Values of Kd above 10,000 L kg^{-1} indicate strong particle reactivity, and accuracy on values this large is probably not very important in risk assessment. Accuracy is important for lower Kd values, and in this case these largest discrepancies for low-Kd elements are for I and Cl, and possibly for Se, Mn and Ni.

The comparison of Kd values for benthic sediment versus suspended particulate matter is illustrated in Figure 6-3. The ANOVA described earlier indicated that for many elements there was a significant difference between these sets of values, with benthic Kd values generally lower than suspended sediment Kd values. This was especially the case for I and Mn. The suspended particulate matter includes living and recently living organisms, whereas the benthic sediment will typically have more mineral matter and more aged organic matter.

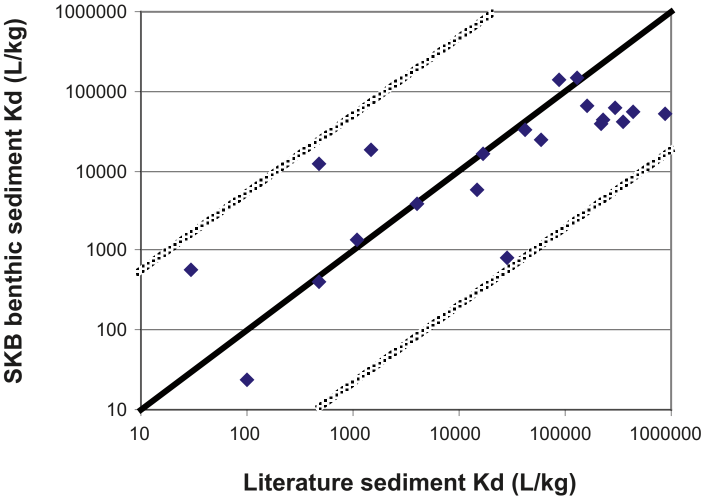

Figure 6-2. Freshwater sediment Kd values, comparing benthic Kd values from the SKB sites with literature values (which are not exclusively for benthic sediment). Each point is a different element, data the same as in Table 6-4. The grey lines are 2 GSD above and below the 1:1 line, approximating the 95th percentile confidence bounds using the median GSD across elements for literature data.

Table 6-3. Literature used to determine Kd values for marine sediment, both suspended and benthic.

	Literature Kd	References
Co	23,000	/Aston and Duursma 1973, Balistrieri and Murray 1986, Chiffoleau et al. 1994, Duursma and Eisma 1973, Ewais et al. 2000, Nyffler et al. 1984, Sirinawin et al. 1998/
Cr	30,000	/Turner 1996/
Cs	540	/Aston and Duursma 1973, Balistrieri and Murray 1986, Vives i Batlle et al. 2008, Cochran et al. 2000, Duursma and Eisma 1973, Ewais et al. 2000, Guscó et al. 2002, Nyffler et al. 1984, Oughton et al. 1997, Turner et al. 1992a, 1993/
Fe	2,900,000	/Balistrieri and Murray 1986, Benoit et al. 1994, Brugman et al. 1992, Duursma and Eisma 1973, Gavriil and Angelidis 2006, Hall et al. 1996, Koshikawa et al. 2007, Martin et al. 1994, Munksgaard and Parry 2001, Nyffler et al. 1984, Turner et al. 1992b, Zhou et al. 2003/
I	30	/Cochran et al. 2000/
Mn	48,000	/Balistrieri and Murray 1986, Balls et al. 1994, Billon et al. 2002, Dassenakis et al. 1997, Duursma and Eisma 1973, Gavriil and Angelidis 2006, Hall et al. 1996, Hatje et al. 2003, Hydes and Kremking 1993, Koshikawa et al. 2007, McCubbin and Leonard 1996, Nyffler et al. 1984, Owens et al. 1997, Paucot and Wollast 1997, Turner et al. 1993, Turner et al. 1992a, b, Zhou et al. 2003/
Ni	17,000	/Balistrieri and Murray 1986, Balls 1989, Balls et al. 1994, Brugman et al. 1992, Chiffoleau et al. 1994, Comber et al. 1995, Dassenakis et al. 1997, Gavriil and Angelidis 2006, Gee and Bruland 2002, Hall et al. 1996, Hatje et al. 2003, Jiann et al. 2005, Martin et al. 1994, Martino et al. 2004, Munksgaard and Parry 2001, Owens et al. 1997, Paucot and Wollast 1997, Turner 2007, Turner et al. 1992b, Turner et al. 1998, Yee et al. 2007, Zhou et al. 2003/
Np	5,000	/Pentreath 1987/
Pa	1,100,000	/Chase et al. 2002, Geibert and Usbeck 2004, Nyffler et al. 1984, Scholten et al. 2005/
Pb	180,000	/Balistrieri and Murray 1986, Balls 1989, Balls et al. 1994, Baskaran and Santschi 1993, Baskaran et al. 1997, Benoit et al. 1994, Brugman et al. 1992, Chiffoleau et al. 1994, Dassenakis et al. 1997, Hall et al. 1996, Helmers 1996, Jiann et al. 2005, Martin et al. 1994, Munksgaard and Parry 2001, Owens et al. 1997, Ravichandran et al. 1995, Rivera-Duarte and Flegal 1994, Sirinawin et al. 1998, Turner et al. 1992b/
Pu	1,200,000	/Balistrieri and Murray 1986, Baskaran et al. 1997, Vives i Batlle et al. 2008, Guscó et al. 2002, Hamilton-Taylor et al. 1993, Kershaw and Young 1998, Kershaw et al. 1999, Lindahl et al. 2005, Malcolm et al. 1990, McCubbin and Leonard 1996, McDonald et al. 2001, McKay and Pattenden 1993, Mitchell et al. 1999, Pentreath 1987, Walker and McKay 1991/
Ra	30	/Rama and Moore 1996/
Sb	1,100	/Nyffler et al. 1984/
Se	260	/Nyffler et al. 1984/
Sn	33,000	/Balistrieri and Murray 1986, Langston and Pope 1995, Nyffler et al. 1984/
Sr	5	/Billon et al. 2002, Duursma and Eisma 1973, Nyffler et al. 1984, Oughton et al. 1997/
Tc	1,400	/McCubbin et al. 2006/
Th	4,500,000	/Vives i Batlle et al. 2008, Chase et al. 2002, Geibert and Usbeck 2004, Kershaw and Young 1998, Nyffler et al. 1984, Scholten et al. 2005/
U	860	/Balistrieri and Murray 1986, Kershaw and Young 1998/

The suspended material, if more organic, will have a lower particle density (mass per unit volume), and may, because it has remained suspended, be of smaller particle size. Thus it will have a higher specific surface area (surface area when hydrated per unit mass when dry). This will tend to result in higher Kd values. Iodine may be especially affected because algae will accumulate I. Another attribute of the suspended sediment versus the benthic sediment is redox: the benthic sediment may have lower redox potential, and this may explain the large difference seen for Mn. Manganese is relatively insoluble (high apparent[3] Kd) in aerated environments and relatively soluble (low apparent Kd) is reducing environments.

The marine Kd values for the SKB sites are shown in Table 6-5 along with the corresponding /IAEA 1985/ and literature values. Note that the residual GSD for the SKB sites is again often lower than the GSD from the literature. The comparison of SKB values to the literature is most easily seen in Figure 6-4. Again, some values deviate from the 1:1 line by more than a factor of 10, but generally they are symmetrical about the 1:1 line.

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[3] The word ‘apparent’ is used because Kd in theory implies an exchange reaction, for Mn and many other elements, chemical precipitation or occlusion may be the actual retention mechanism involved.

Table 6-4. Freshwater sediment Kd values (L kg^{-1}) as measured in benthic and suspended particulate matter at the SKB sites and from the /EMRAS 2008/ and literature sources. The residual geometric standard deviation (GSD) is the residual following ANOVA (see text), and so reflects variation among the SKB sampling locations. The literature GSD is among all entries obtained from the literature, and so reflects global site-to-site and study-to-study variation.

	SKB benthic Kd	SKB susp. Kd	Residual GSD[a]	EMRAS susp. Kd (in situ)	Literature Kd	Literature GSD[a]
Cl	24		1.9		98	
Co	25,000	88,000	3.1	44,000	60,000	7.8
Cr	32,000	110,000	2.4		42,000	10
Cs	16,000	100,000	2.4	29,000	17,000	5.0
Fe	67,000	320,000	7.9	5,000	160,000	11
Ho	43,000	75,000	2.6		230,000	1.6
I	580	16,000	2.4	220	29	9.8
La	61,000	180,000	2.0		300,000	4.3
Mn	820	210,000	3.6	78,000	28,000	32
Mo	1,400	12,000	3.3		1,100	18
Nb	130,000	270,000	2.8	10,000		
Nd	52,000	110,000	2.4		870,000	4
Ni	18,000	28,000	2.2		1,500	36
Np				10		
Pa						
Pb	150,000	540,000	2.4		130,000	14
Pu				240,000		
Ra				7,400		
Sb	3,900	24,000	2.3	5,000	4,100	18
Se	12,000	8,100	3.2		470	
Sm	54,000	91,000	2.5		430,000	3.0
Sn	6,600		3.2			
Sr	410	1,300	1.9	1,200	480	3.1
Tc				5	0	4.9
Th	140,000	190,000	2.7	180,000	87,000	2.2
Tm	39,000	72,000	3.4		220,000	2.1
U	5,900	8,400	3.4		15,000	39
Yb	42,000	50,000	2.5		350,000	2.7

[a] The median GSD of SKB data was 2.5 and for literature data was 6.4.

Table 6-5. Marine sediment Kd values (L kg^{-1}) as measured in benthic and suspended particulate matter at the SKB sites and from the /IAEA 1985/ and literature sources. The residual geometric standard deviation (GSD) is the residual following ANOVA (see text), and so reflects variation among the SKB sampling locations. The literature GSD is among all entries obtained from the literature, and so reflects global site-to-site and study-to-study variation.

	SKB benthic Kd	SKB susp. Kd	Residual GSD[a]	IAEA pelagic Kd	IAEA coastal Kd	Literature Kd	Literature GSD[a]
Cl	3	0.5	1.9	1	0.03		
Co	25,000	120,000	3.1	5 x 10^7	300,000	23,000	13
Cr	37,000	240,000	2.4	40,000	50,000	30,000	1.0
Cs	31,000	40,000	2.4	2,000	4,000	540	5.1
Fe	71,000	3,100,000	7.9	2 x 10^8	3 x 10^8	2,900,000	11
Ho	17,000	160,000	2.6				
I	130	6,100	2.4	200	70	30	
La	82,000	1,200,000	2				
Mn	1,700	210,000	3.6	2 x 10^8	2,000,000	48,000	34
Mo	440	1,100	3.3				
Nb	75,000	650,000	2.8	300,000	800,000		
Nd	45,000	880,000	2.4				
Ni	12,000	20,000	2.2	300,000	20,000	17,000	11
Np				1,000	1,000	5,000	
Pa				5,000,000	5,000,000	1,100,000	8.6
Pb	150,000	480,000	2.4	1 x 10^7	100,000	180,000	9.3
Pu				100,000	100,000	1,200,000	25
Ra				4,000	2,000	30	2.4
Sb	2,400	6,100	2.3	4,000	2,000	1,100	13
Se	1,900	18,000	3.2	1,000	3,000	260	2.8
Sm	33,000	620,000	2.5	500,000	3,000,000		
Sn	13,000		3.2	300,000	4,000,000	33,000	5.3
Sr	77	130	1.9	200	8	5	19
Tc				100	100	1,400	4
Th	91,000	990,000	2.7	5,000,000	3,000,000	4,500,000	3.8
Tm	13,000	150,000	3.4	200,000	1,000,000		
U	2,500	1,900	3.4	500	1,000	860	3.1
Yb	20,000	190,000	2.5	200,000	1,000,000		

[a] The median GSD of SKB data was 2.5 and for literature data was 8.6.

There are several points about 2 GSD from the 1:1 line, but these had GSD higher than the median GSD used to draw the grey lines in Figure 6-4. Thus even for these elements, one can argue the Kd values for SKB sites belong in the same data population as the literature, suggesting that literature values are applicable to the SKB sites. Fortunately, among the low Kd values that are important to risk assessment, there is better agreement. As with the freshwater Kd values, the marine benthic Kd values tend to be lower than the corresponding suspended sediment values (Figure 6-5).

6.3 Selection of values for use in risk assessment

In general, given the inherent variability in sediment Kd values, the differences among the sources of data were not statistically meaningful. Data specific to the SKB sites were not different, overall, from the global literature. One could justifiably draw Kd values for use in risk assessment from any one of these sources, or from them all collectively. For a few elements, notably Cl, the number of observations from the SKB sites is comparable to the numbers of observations supporting the values derived from the global literature. However, for most elements there are many more observations in the global literature than from the SKB sites, which suggests the global literature should be used.

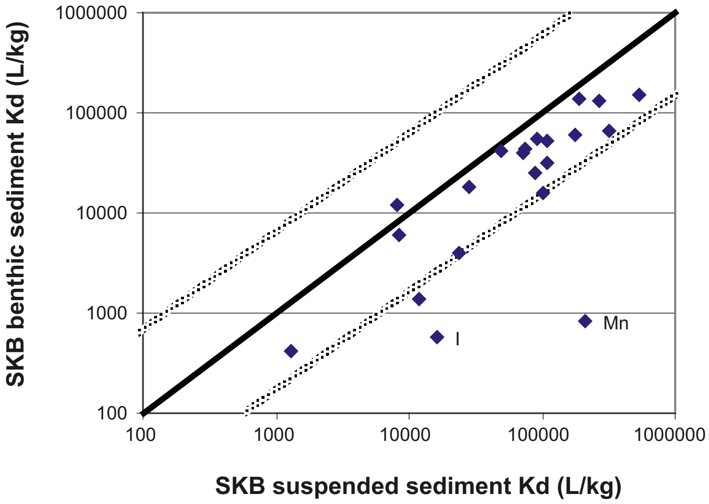

Figure 6-3. *Freshwater sediment Kd values, comparing benthic Kd values from the SKB sites with suspended sediment Kd values from the SKB sites. Each point is a different element, data the same as in Table 6-2. The points for I and Mn are labelled as they are furthest from the 1:1 line. The grey lines are 2 GSD above and below the 1:1 line, approximating the 95th percentile confidence bounds using the median GSD across elements for SKB data.*

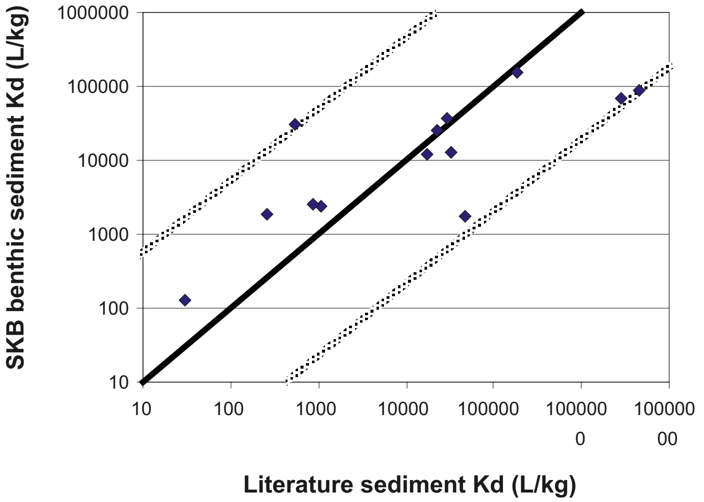

Figure 6-4. *Marine sediment Kd values, comparing benthic Kd values from the SKB sites with literature values (which are not exclusively for benthic sediment). Each point is a different element, data the same as in Table 6-5. The grey lines are 2 GSD above and below the 1:1 line, approximating the 95th percentile confidence bounds using the median GSD across elements for literature data.*

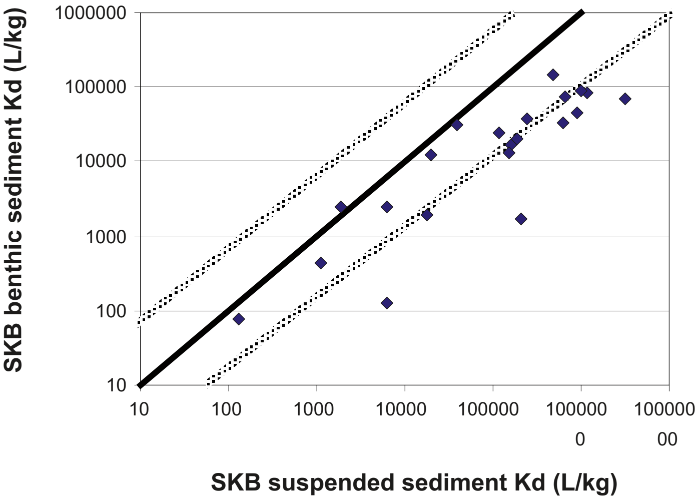

Figure 6-5. *Marine sediment Kd values, comparing benthic Kd values from the SKB sites with suspended sediment Kd values from the SKB sites. Each point is a different element, data the same as in Table 6-5. The grey lines are 2 GSD above and below the 1:1 line, approximating the 95th percentile confidence bounds using the median GSD across elements for SKB data.*

In addition to numbers of samples, the global literature represents a broader range of conditions, not only spatially but in terms of water movement (tides, currents and fluvial inputs), water chemistry and water-column flora and fauna. The present situation for these conditions along the eastern coast of Sweden may change with time, so there is merit in considering the global literature as a sampling of possible future states. This argument is relevant for the GM values, but even more importantly for the GSD that would be used in a stochastic risk assessment. As described above, the GSD for the SKB sites may be too small to encompass the full spatial variation intended, and very likely too small to represent any temporal effects. In contrast, the literature GSD values may encompass too many uncertainties, such as measurement methods.

Key advantages of the SKB data are the consistent measurement methods and the clear differentiation in values between Kd for benthic sediments and suspended particulate matter. It is also important to note that the SKB freshwater Kd values for I and Cl, and perhaps also for Se, Mn and Ni, were in the value range where Kd is important to risk assessment and deviated to some extent from the corresponding global literature. These elements may need special attention, and perhaps more detailed study.

Given these arguments, it may be appropriate to use the SKB data for risk assessment of sites on the east coast of Sweden, when available. For elements where freshwater SKB data are not available, the /EMRAS 2008/ values for freshwater have the advantage of promulgation by the IAEA, and would therefore be the next best choice. For elements where marine SKB data are not available, the literature reviewed here may be a better choice than /IAEA 1985, 2004/, where Kd were not necessarily derived from paired water and sediment samples.

7 Overall conclusions

It is possible to select values of Kd for soils and sediments that are the best estimates from the data available, and to show that these values would differ in a statistically significant manner as site conditions differed. For example, the soil Kd values for U varied notably with pH, and so if soil pH varied from site to site, or with time because of natural processes or human activities, then it is possible to assign revised best estimate Kd values for the changed soil pH. However, it is important to bear in mind the overall variability of Kd data.

Among the regression equations to predict soil Kd values, the median residual GSD was 4.3, which implies 95th percentile confidence bounds 18-fold above and below the recommended values. The 5th percentile and 95th percentile values are more than 300-fold different from each other. This is clearly a large enough variance to move some elements from what might be deemed an 'immobile' category into a category that might be deemed 'very mobile', or vice versa. In many respects, the best that can be said of Kd is that it can differentiate the extremely mobile elements such as Cl from the extremely immobile elements such as Pb and Th. However, this does not imply a fault with Kd as a model of sorption, but rather that many variables, both known and unknown, have a strong influence.

For the sediment Kd values, the median GSD for data measured at the SKB sites was only 2.5, but from the literature the median was 6.4 or 8.6 (Tables 6.1 and 6.4). This implies 95th percentile confidence bounds for marine Kd as high as 74-fold above and below the recommended values (for freshwater Kd they are 41-fold). As discussed by /Sheppard 2005b/, there is an asymptotic minimum variation in Kd, further specification of environmental conditions will not substantially lessen this variation. Therefore, it is important to account for variation in an assessment application.

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