

# SKBF TECHNICAL KBS REPORT

**83-63**

## Sorption of radionuclides in geologic systems

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

A list of other reports published in this series during 1983 is attached at the end of this report. Information on KBS technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26), 1981 (TR 81-17) and 1982 (TR 82-28) is available through SKBF/KBS.

SORPTION OF RADIONUCLIDES  
IN GEOLOGIC SYSTEMS

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## SUMMARY

The sorption of radionuclides (cesium, strontium, iodide, technetium and carbon-14) has been studied as a function of pH, nuclide concentration, ionic strength and contact time on geologic material in distribution experiments (batch-wise equilibrium studies and transport studies in columns).

Important parameters governing the interaction of radionuclides with geologic materials are pH, the ionic strength and the amount of complex-forming agents in the aqueous phase, and the mineralogy of the geologic media, the cation exchange capacity of the corresponding minerals, the surface to mass ratio and the amount of complex-forming agents in the solid phase.

Three main sorption mechanisms have been exemplified: Electrostatic interactions (ion exchange processes), which depend on the exchange capacity of the solid, the pH and ionic strength of the water and concentration of the sorbing species; chemisorption processes, where complex-forming agents, either in solution or on the solid phase interact with the radionuclide; physical adsorption phenomena, where cationic and neutral complexes are sorbed on solid surfaces by non-specific forces.

The sorption mechanism for cesium and strontium is mainly ion exchange and for iodide, technetium and carbon (as carbonate) chemisorption under oxic conditions. Technetium in the reduced form (as technetium(IV)hydroxide/oxide) sorbs like the actinides mainly by physical adsorption.

Radionuclides sorbing by ion exchange mechanisms are found to migrate in rock through microfissures and high-capacity minerals. A similar migration was not found for radionuclides sorbing mainly by chemisorption or physical adsorption processes.

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## 1. INTRODUCTION

The release of radionuclides into the biosphere from a deep underground repository for high-level radioactive waste or spent fuel will be prevented by both engineered barriers (sparingly soluble waste-form, over-pack, encapsulation, possibly a back-fill with certain additives) as well as natural barriers (low groundwater flow, retention of the nuclides due to the interaction with geologic media). A thorough knowledge of the chemical properties of the various long-lived radionuclides in spent fuel as well as of the interactions with components of the bedrock would be required and a necessary basis for predictions of nuclide mobilities and transport times.

The actinides with daughter products as well as a few long-lived fission products (e.g.  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{135}\text{Cs}$ ; in the short time-perspective  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ) dominate the biological hazards of spent fuel. A few additional activation products would be present in low- and medium-level waste (e.g.  $^3\text{H}$ ,  $^{13}\text{C}$ ,  $^{59}\text{Ni}$ ,  $^{95}\text{Nb}$ ).

Some qualitative and quantitative observations of sorption phenomena involving some of the important fission products (primarily Sr, Cs and I; also some data for Tc and carbonate) are discussed in this report, which is largely a summary of previously published or presented material (1-14), representing studies over the period 1978-1982 at the Dept. of Nuclear Chemistry, Chalmers University of Technology, Sweden. Similar summaries of actinide sorption studies have previously been presented (15-16). Comparisons with literature data are given elsewhere (17).

## 2. THE GROUNDWATER-ROCK SYSTEM

Spent nuclear fuel or high-level reprocessing waste will be stored in crystalline rock, probably of granitic or granodioritic composition, at great depth in the present Swedish concept for radioactive waste deposition. The variety in chemical environments encountered in the proposed bedrock-groundwater systems must be considered in any systematic study of radionuclide behaviour and mobility.

### 2.1. Mineralogic composition of igneous rocks

Igneous rocks like granite are composed of a small number of major rock-forming minerals like quartz, feldspars (orthoclase, microcline, plagi-

clase), micas (biotite), amphiboles (hornblende), pyroxenes and olivine. Moreover, there are accessory minerals like fluorite, calcite, apatite, magnetite etc. that would have some influence on the interactions with trace elements in the groundwater, especially if these minerals have chemisorbing capacities or particularly high exchange capacities.

Of special interest are the fracture filling products which usually form a coating on water-bearing fractures (18,19). Thus, the dissolved trace components in the groundwater will primarily interact with these minerals rather than with the underlying bulk rock components.

## 2.2. Groundwater composition in igneous rocks

The most important chemical system in many shallow or deep groundwaters is the  $H_2O-CO_2$ -system, which largely controls pH (through the protolysis of  $H_2CO_3$ ). The total carbonate concentration determines the maximum calcium concentration (precipitation of calcite,  $CaCO_3(s)$ ), which in turn sets an upper limit for the fluoride concentration due to the low solubility of fluorite ( $CaF_2(s)$ ). Some interrelations exist between the calcium, magnesium, sodium and potassium concentrations through ion exchange reactions with weathering products (mostly clay minerals). Thus, the concentrations of  $HCO_3^-$ ,  $Ca^{2+}$ ,  $Na^+$ ,  $Mg^{2+}$ ,  $K^+$ ,  $F^-$  (as well as  $SiO_2$ ,  $HPO_4^{2-}$  and occasionally  $SO_4^{2-}$ ) are interdependent and related to the various solid phases present. Another important interdependent system is Fe(II)-Fe(III)- $O_2$ , which largely governs the redox conditions.

Saline waters with total salt concentrations (mainly  $Na^+$ ,  $Ca^{2+}$  and  $Cl^-$ ) of up to several thousand mg/l (normally less than 300 mg/l) are not uncommon in deep groundwaters. (Further aspects on groundwater chemistry is given in ref. 20).

Concentration ranges of major species in deep groundwaters are summarized in Table 1, as well as maximum concentrations in saline waters and the composition of an artificial groundwater previously used in sorption studies (7,15).

Table 1 Composition of deep groundwaters (mg/l)

	Normal concentration range	Max. concentration <sup>b</sup> in saline waters	Artificial groundwater
HCO <sub>3</sub> <sup>-</sup>	90-275	(400)	123
SO <sub>4</sub> <sup>2-</sup>	0.5-15	40(370)	9.6
HPO <sub>4</sub> <sup>2-</sup>	0.01-0.2		
F <sup>-</sup>	0.5-5	5(7.5)	
Cl <sup>-</sup>	5-15	350(6300)	70
Ca <sup>2+</sup>	10-40	90(2100)	18
Mg <sup>2+</sup>	1-10	20(210)	4.3
Na <sup>+</sup>	10-100	300(1660)	65
K <sup>+</sup>	1-5	10(37)	3.9
Fe(II)	0.02-5		
SiO <sub>2</sub> (tot)	3-14	20	11
TOC <sup>a</sup>	1-8		
pH	7-9		8.2

<sup>a</sup> Total organic carbon

<sup>b</sup> Common upper concentration levels are given; values within parentheses represent the highest values encountered in any location.

### 2.3. Selection of solid and aqueous phases for sorption studies

The general principle in the selection of minerals for the experimental studies has been to include representatives of the most common rock-forming components as well as minerals that for various reasons would be expected to interact strongly with the radionuclide (high exchange capacity, chemisorption). The following groups of rocks and minerals are represented:

- o Major rock-forming minerals
- o Common accessory minerals, including some fracture filling products and weathering products
- o High-capacity minerals (high anion- or cation exchange capacity; see ref. 21)
- o Chemisorbing minerals (for I<sup>-</sup> and H<sup>14</sup>CO<sub>3</sub><sup>-</sup>)
- o Hydroxides, representing corrosion products and fresh metal precipitates with large surface/volume ratios



- o Igneous rocks; granite, gneiss and diabase from Finnsjön, Stripa, Studsvik and Blekinge.

The selected mineral systems are listed in Table 2.

The artificial groundwater, Table 1, representative of deep non-saline groundwaters, was normally used as aqueous phase, and 4 M NaCl was representing very saline waters or brine. The transport studies simulating the field measurements in Studsvik were performed with an artificial groundwater with a composition based on analyses from drilling holes in the area (12,14).

Table 2 Minerals and rocks selected for sorption studies

Class	Mineral <sup>a</sup>	Trace element	
Sulfides	Galena	I	
	Chalcopyrite	I	
	Cinnabar	I	
	Pyrite	Sr, Tc, Cs	
Oxides, Hydroxides	Corundum	Cs	
	Hematite	Sr, Cs	
	Magnetite	Sr, Tc, Cs	
	Limonite	I	
	Fe(OH) <sub>3</sub>	Sr, I, Cs	
	Al(OH) <sub>3</sub>	Sr, I, Cs	
	Pb(OH) <sub>2</sub>	Sr, I, Cs	
	Cu(OH) <sub>2</sub>	Sr, Cs	
Halides, Carbonates, Sulfates, Phosphates	Fluorite	Sr, Cs	
	Calcite	C, Sr, Cs	
	Dolomite	Sr, Cs	
	Gypsum	Sr, Cs	
	Apatite	Sr, Cs	
	AgCl	Sr, I, Cs	
Silicates	Nesosilicates	Olivine	I
		Sorosilicates	Epidote
	Prehnite		Sr, Cs
	Inosilicates	Hornblende	Sr, Tc, Cs
		Attapulgite	I
	Phyllosilicates	Kaolinite	Sr, Cs
		Serpentine	Sr, I, Cs
		Halloysite	I
		Illite	Sr, Cs
		Montmorillonite	C, Sr, Cs
		Muscovite	Sr, Cs
		Biotite	Sr, Tc, Cs
		Chlorite	Sr, Tc, Cs
	Tectosilicates	Quartz	Sr, Cs
		Microcline	Sr, Cs
		Orthoclase	Sr, Cs
		Plagioclase	Sr, Cs
		Laumontite	Sr, Cs
		Stilbite	Sr, Cs
Igneous rocks	Granite, Finnsjön (Fi)	Sr, Cs	
	Granite, Stripa (Str)	C, Sr, Tc, Cs	
	Gneiss/Granite, Studsvik (Stu)	Sr, Cs	
	Gneiss/Granite, Blekinge (G-B1)	Sr, Cs	
	Diabase, Blekinge (D-B1)	Sr, Cs	

<sup>a</sup> See e.g. ref. 22.

### 3. EXPERIMENTAL

#### 3.1. Preparation and characterization of sorbents

Pure, crystalline products from natural sources were used. Mineral grains that appeared to be reasonably pure were selected from a crushed or coarsely ground material and then further ground and sieved. Usually the size fraction 0.045-0.063 or 0.090-0.125 mm was used. Large samples of homogeneous rocks were crushed and sieved, and only the larger size fraction 0.090-0.125 mm was used. The purity was checked by X-ray diffraction, and for some of the minerals also by chemical analysis. Anion exchange capacities (AEC) and cation exchange capacities (CEC) as well as surface/mass ratios were determined (21). (Detailed data concerning mineral composition and purity, sources etc. are available but not included in this report.) The mineralogic composition of the rock samples is given in Table 3.

Table 3 Composition of rock samples

Mineral	Composition, wt-% <sup>a</sup>		
	Finnsjön	Stripa	Studsvik
Quartz	39	37	40-70
Plagioclase	26	29	10-40
Microcline	17	26	
Biotite	6	2	b
Muscovite	-	4	
Hornblende	8	-	b
Epidote	3	-	
Accessories	1	2	b

<sup>a</sup> From ref. 12, 18 and 19

<sup>b</sup> Hornblende, biotite and accessories all together 20 weight %

#### 3.2. Sorption studies

##### 3.2.1. Radionuclides

The radionuclides used in the sorption and migration studies are listed in

Table 4. Acidic stock solutions (pH 1-2) with a total concentration of  $10^{-5}$  -  $10^{-6}$  M were generally prepared.

### 3.2.2. Batch measurements

The crushed (in agate mortar) and sieved material was prewashed and equilibrated with aqueous phase for one week. The solid was allowed to settle by gravity or by a light centrifugation after a short vigorous shaking of the sample. The aqueous phase was removed and discharged and new aqueous phase was added. The system was allowed to equilibrate for another week before the acidic active nuclide solution was added (typically 0.1 ml). After the addition of the spike solution pH was adjusted to roughly the desired level (between 5 and 10) by the use of 0.1 or 0.01 M NaOH or HCl.

The activity remaining in the system was determined using a Ge(Li)-detector (after phase separation by centrifugation), as well as pH, after various contact times. The distribution coefficient  $K_d$  ( $m^3/kg$ ) was calculated according to

$$K_d = (c_o - c)V/cm \quad (1)$$

where  $c_o$  = initial concentration (M),  
 $c$  = measured concentration after a certain contact time (M)  
 $V$  = volume of aqueous phase ( $m^3$ )  
 $m$  = mass of solid phase (kg).

The sorption on the walls of the vials was measured and found to be negligible in the presence of crushed material with a large accessible surface.

The composition of the artificial groundwater was usually not significantly altered under the present experimental conditions except for pH (15).

All experiments were performed in aerated systems, except for some batch measurements, where the technetium sorption under reducing conditions was studied.

The experimental conditions are given in Table 5.

Table 4 Radionuclides used in the sorption studies.

Nuclide	Half-life
$^{14}\text{C}$ a	5730 y
$^{85}\text{Sr}$	64.8 d
$^{99\text{m}}\text{Tc}$ b	6.0 h
$^{131}\text{I}$	8.0 d
$^{134}\text{Cs}$	2.06 y

a As  $\text{H}^{14}\text{CO}_3^-$

b From  $^{99}\text{Mo}$ ,  $t_{1/2} = 66$  h

Table 5 Conditions for batch experiments

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Solid phase:	Minerals according to Table 2	
Grain size		0.045-0.063 mm 0.090-0.125 mm
Aqueous phase:	Artificial groundwater according to Table 1 ( <u>standard</u> )	
	Various salt solutions	
Solid/liquid ratio		ca 0.5-1g per 45 ml <sup>a</sup> (10-20 g/l)
Initial element concentration		usually $10^{-7}$ - $10^{-8}$ M <sup>a</sup> ( <u>standard</u> )
Other parameters		
Temperature		22±2°C
Contact time		1d, 1w ( <u>standard</u> ) or longer
Phase separation		Centrifugation; 1h at 4000g (27000g for some clay systems)
Equipment		Polypropylene, glass

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<sup>a</sup> Exact amounts of solids and aqueous phases are known for each of the ca 1500 measurements given in the subsequent figures and table but not included in this report.

### 3.2.3. Column measurements

A column, 0.1 m high and Ø 4 mm, was packed with crushed rock or mineral (0.090-0.125 mm), and a pulse of radionuclide solution (usually 0.05 ml) was added to the column. Artificial groundwater was pumped through the column by means of a HPLC-pump and was collected by an automatic fraction collector.

The flow rate through the column was varied between 0.01 and 0.08 ml/min.

The retention of a certain radionuclide in the column was obtained from comparison with a non-sorbing tracer. The distribution coefficient was estimated from:

$$R = u_w/u_n = 1 + K_d \rho_p (1-\epsilon)/\epsilon \quad (2)$$

where R = retention factor

$u_n$  = velocity of nuclide (m/s)

$u_w$  = velocity of water (= velocity of non-sorbing tracer) (m/s)

$\rho_p$  = density of solid ( $m^3/kg$ )

$\epsilon$  = porosity of column filling.

#### 3.2.4. In situ measurements

Boreholes in the bedrock at the Studsvik laboratory area were used for retention studies in situ. (12,14) The mineralogy of a drilling core from the area was determined and fracture filling minerals identified. Two boreholes with an interconnecting fracture at about 100 m depth were chosen for the migration experiments. A pump was installed in one of the holes to remove and sample water coming from the fracture. The composition of this water was determined at regular intervals during the preparations and subsequent experiments. The hydraulic properties of the flow paths were determined by adding a pulse of  $^{131}I$  to the injection hole followed by  $\gamma$ -activity measurements of the water coming from the pump-hole. The result in terms of water flow conditions was interpreted as a combination of several flowpaths where one with a residence time of 288 h was dominating. After this preliminary test-run a simultaneous injection of  $^3H$ ,  $^{85}Sr$  and  $^{131}I$  was performed and the activity in the pumphole water was recorded vs. time.

## 4. RESULTS AND DISCUSSION

### 4.1. Strontium and cesium

#### 4.1.1. Composition of the solid

The sorption, expressed as the distribution coefficient, of cesium and strontium on minerals and rocks is given in Figure 1 and 2, respectively and in Table 6a-c (7,10,11).

Cesium does not form complexes with species normally found in groundwaters, and interacts with geologic materials as the non-complexed monovalent cation  $\text{Cs}^+$ . The sorption is mainly a cation exchange process dependent both on the pH of the water and on the properties of the solid. As expected for cation exchange processes the sorption of cesium increases with increasing pH of the water (Figure 1). Furthermore, minerals with high CEC, like biotite and hornblende exhibit high cesium sorption, while minerals with low CEC (e.g. quartz) sorb cesium poorly (cf. Figure 3).

Cesium was found to sorb on natural fracture surfaces, especially on fracture fillings with high CEC with a consecutive diffusion through the fracture filling into the bulk rock. Cesium migrated millimeters into the rock at an exposure time of a couple of months of the fracture surface to a spiked groundwater. The major transport pathways into the rock were found to be microfissures and high-capacity mineral veins (13).

The sorption of strontium is, similar to cesium, largely a cation exchange process; that is, highly related to the pH of the water and the CEC of the solid (cf. Figure 2 and 3). Strontium acts predominantly as the divalent cation  $\text{Sr}^{2+}$  in most environmental waters. At high pH (>9) and fairly high carbonate concentration, a formation of hydrolysis products or carbonate complexes is possible leading to an increased sorption due to a physical adsorption of neutral or near neutral complexes on the solid.

High sorption of strontium was found in phosphate or fluoride rich waters, probably due to a formation of sparingly soluble strontium phosphate and fluoride complexes.



Table 6a Distribution coefficients for cesium and strontium on minerals from batch experiments. (Table 5, groundwater systems)

Solid	pH	$K_d, m^3/kg$	
		Sr	Cs
Pyrite	8-8.5	0.003	0.003
Corundum	7-8		0.000
Hematite	4-6	0.003	0.002
	6-8	0.004	0.005
	8-9	0.007	0.006
Magnetite	4-6	0.002	
	6-8	0.002	0.007
	8-10	0.002	0.007
Fluorite	4-6	0.003	0.000
	6-8	0.003	0.000
	8-9	0.002	0.000
Calcite	6-8	0.002	
	8-9	0.003	0.001
a	8-8.5	0.004	0.006
Dolomite	8-8.5	0.003	0.005
Gypsum	8-8.5	0.003	0.006
Apatite	4-6	0.003	0.006
	6-8	0.005	0.000
	8-9	0.008	0.003
Epidote	8-8.5	0.010	0.019
Prehnite	8-8.5	0.010	0.013
	b	8-8.5	0.016
Hornblende	4-6	0.002	
	6-8	0.005	0.156
	8-9	0.018	0.213
Biotite	4-6	0.004	0.925
	6-8	0.016	0.610
	8-9	0.024	0.624
Muscovite	4-6	0.014	0.684
	6-8	0.071	0.591
	8-9	0.114	0.765
Serpentine	8-9	0.010	0.012
Chlorite	8-8.5	0.050	0.128
Kaolinite	8-8.5	0.029	0.059
Illite	8-8.5	0.335	> 5
Bentonite <sup>c</sup>	8-9	3.5	1.1
Quartz	4-6	0.000	
	6-8	0.000	
	8-9	0.001	0.001
Orthoclase	4-6	0.001	0.007
	6-8	0.002	0.013
	8-9	0.005	0.011
Plagioclase	8-8.5	0.004	0.011
Microcline	8-8.5	0.012	0.177
Laumontite	8-8.5	0.029	0.117
Stilbite	8-8.5	0.188	0.950

a Fracture filling in drill core at 309 m depth, Finnsjön

b Fracture filling in drill hole at 519 m depth, Finnsjön

c Commercial Wyoming bentonite, MX-80, mainly Na-montmorillonite

Table 6b Distribution coefficient for cesium and strontium on rocks  
from batch experiments. (Table 5, groundwater systems)

Solid	pH	$K_d, m^3/kg$	
		Sr	Cs
Stripa, granite	3-7		0.089
	7-9	0.006	0.152
	9-11	0.057	0.152
Finnsjön, granite	3-7	0.007	0.264
	7-9	0.020	0.490
	9-11	0.259	0.639
Studsvik, gneiss/granit	6-7	0.008	1.215
	7-8	0.012	0.106
	8-9	0.021	0.948
a	7-8	0.004	
a	8-9	0.006	
a	> 9	0.030	
b	7-8	0.043	1.214
Blekinge, gneiss	3-7	0.001	0.100
	7-9	0.009	0.107
	9-11	0.103	0.083
Blekinge, diabase	3-7	0.006	0.116
	7-9	0.010	0.368
	9-11	0.163	

a In natural water from Studsvik borehole

b Artificially weathered, autoclave 150°C, 1 week

Table 6c Distribution coefficients for cesium and strontium on hydroxides (corrosion products) from batch experiments. (Table 5, ground-water systems)

Solid	pH	$K_d, m^3/kg$	
		Sr	Cs
$Fe(OH)_3$	8-8.5	1.20	0.010
$Pb(OH)_2$	8-8.5	0.000	0.000
$Cu(OH)_2$	8-8.5	0.001	0.000

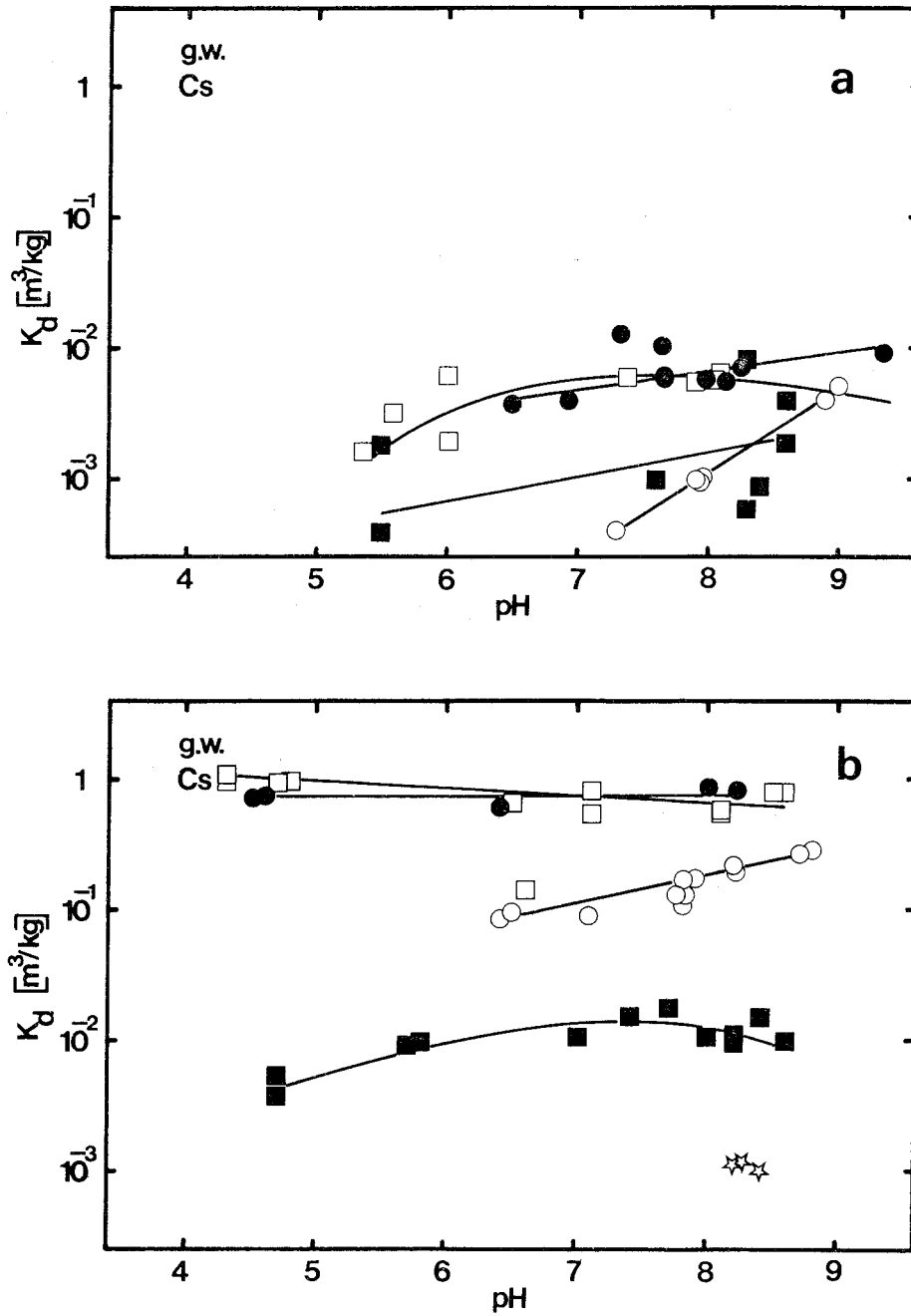


Figure 1 Distribution coefficients for cesium from batch experiments (Table 5, groundwater systems); Effect of pH

a. Non-silicates

● Magnetite, □ Hematite, ○ Calcite,  
■ Apatite

b. Silicates

○ Hornblende, ● Muscovite, □ Biotite,  
■ Orthoclase, ☆ Quartz

c. Rocks

● Granite-Fi, ○ Granite-Str, □ Gneiss-Stu

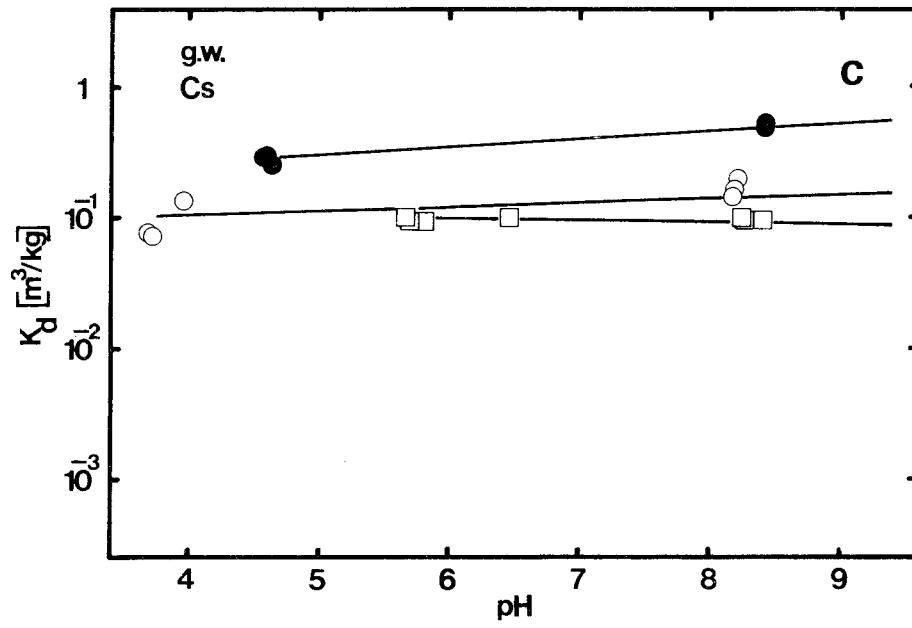


Figure 1 Distribution coefficients for cesium from batch experiments;  
Effect of pH (continued)

c. Rocks

● Granite-Fi, ○ Granite-Str, □ Gneiss-Stu

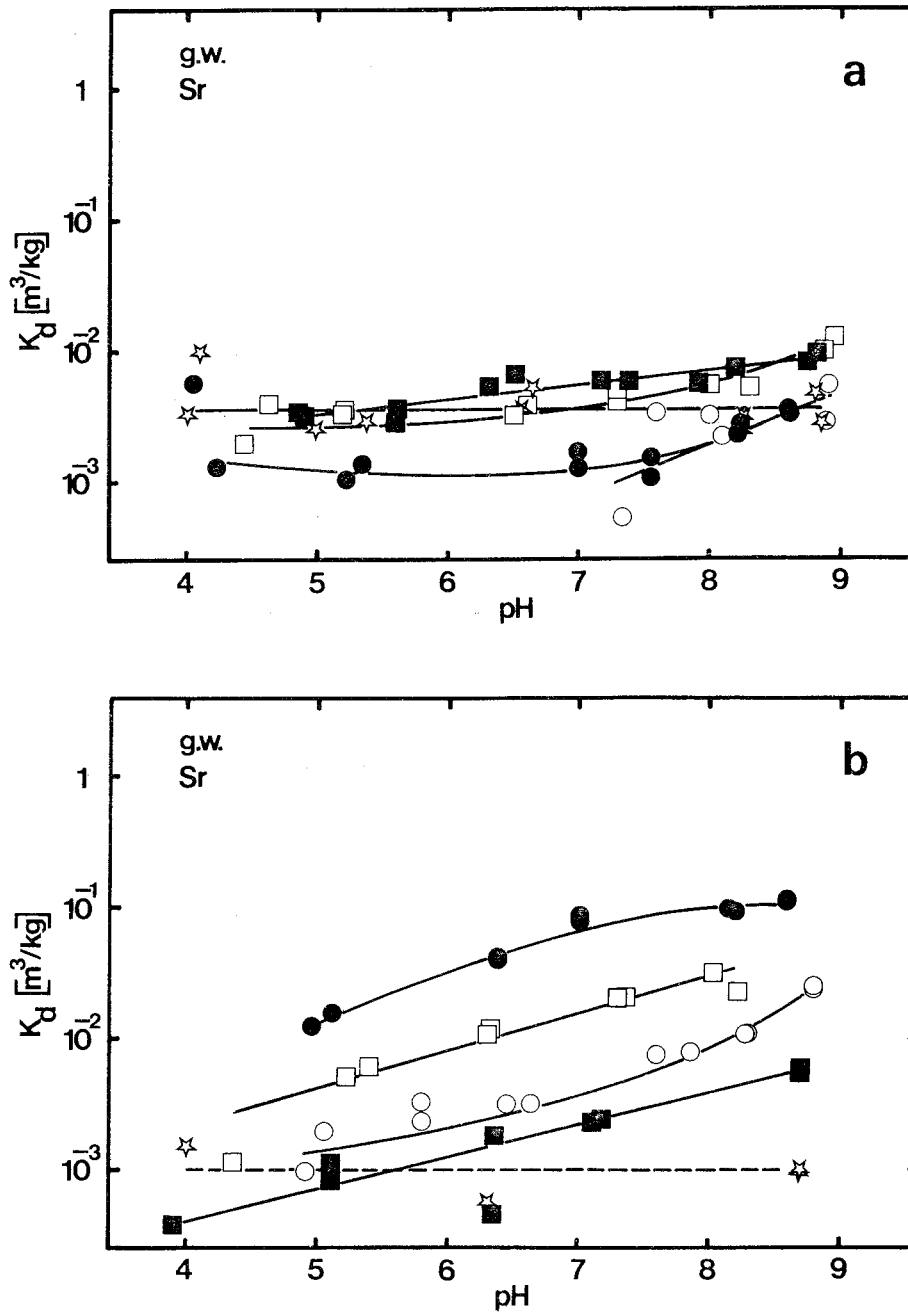


Figure 2 Distribution coefficients for strontium from batch experiments (Table 5, groundwater systems); Effects of pH

a. Non-silicates

- Magnetite, □ Hematite, ○ Calcite,
- Apatite, ☆ Fluorite

b. Silicates

- Hornblende, ● Muscovite, □ Biotite,
- Orthoclase, ☆ Quartz

c. Rocks

- Granite-Fi, ○ Granite-Str, □ Gneiss-Stu

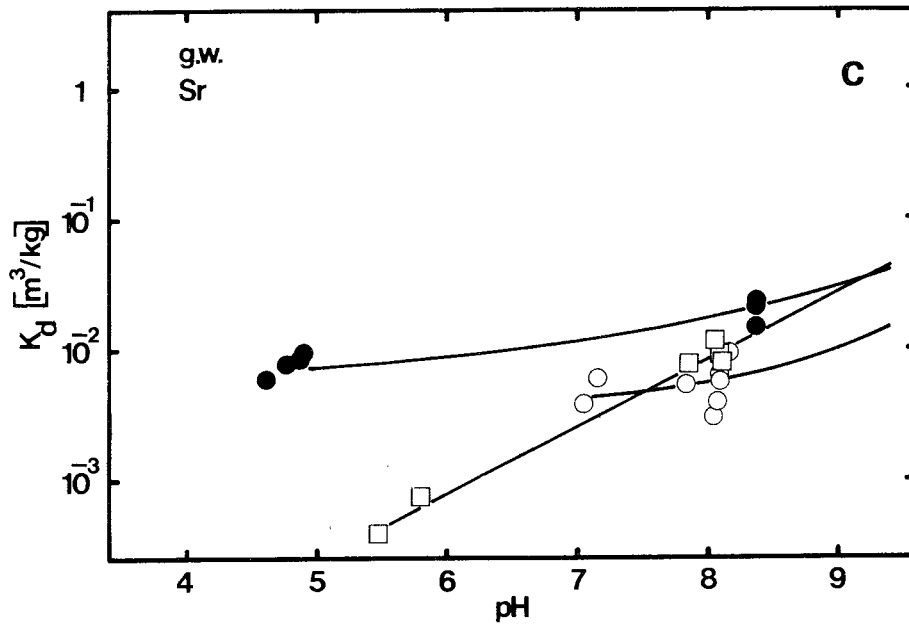


Figure 2 Distribution coefficients for strontium from batch experiments;  
Effect of pH (continued)

c. Rocks

● Granite-Fi, ○ Granite-Str, □ Gneiss-Stu

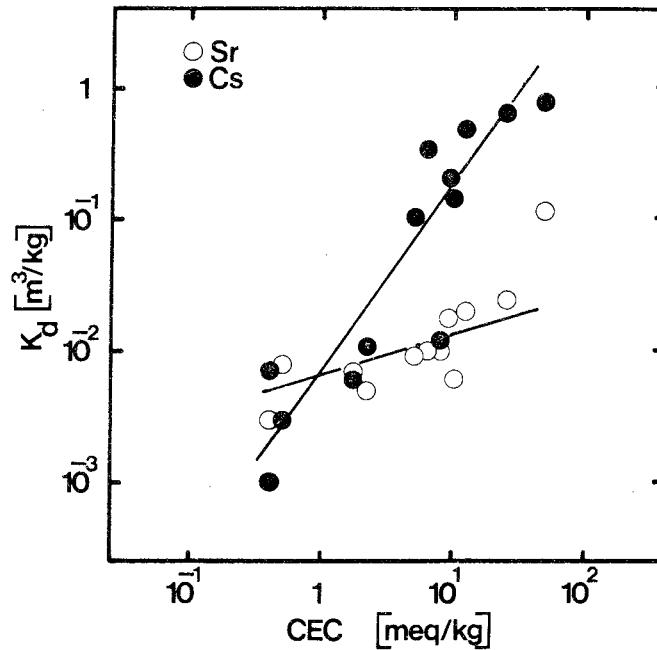


Figure 3 Distribution coefficients for cesium and strontium at pH 8; Effect of cation exchange capacity (CEC) of the solid Minerals and rocks in order of increasing CEC: Calcite, magnetite apatite, hematite, orthoclase, gneiss-Stu, diabase, serpentine, hornblende, granite-Str, granite-Fi, biotite, muscovite.

#### 4.1.2. Nuclide concentration and ionic strength

The ionic strength is important for ion exchange processes, there is a stronger competition for the sorption sites on the solid in a concentrated ionic media than in a dilute. In Table 7 the distribution coefficients are given for cesium and strontium in three ionic medias; groundwater, 0.1 M NaCl and 4 M NaCl. As expected the distribution coefficients are much lower in the 4 M salt solution than in groundwater and 0.1 M NaCl (cf. Figure 4) (10). For low nuclide concentrations ( $<10^{-7}$  M) on solids with high CEC (e.g. granite, hornblende, montmorillonite, etc.) linear sorption isotherms have been found. In these cases the sorption follows a Freundlich type of sorption isotherm, expressed as

$$q = Kc^n$$

where  $q$  = concentration in solid (mol/kg),  $c$  = concentration in liquid (mol/m<sup>3</sup>) and  $K$  and  $n$  are system constants ( $K$  is the distribution coefficient)



ent  $K_d$ ). For bentonite  $n$  is found to be 0.94 and 0.91 for cesium and strontium, respectively. Different exponents have been calculated for the granites, e.g. for cesium 0.78 on Studsvik granite and 0.57 on Stripa granite.

Table 7 Influence of ionic strength on the distribution of cesium and strontium

Solid	Ionic medium	$K_d, m^3/kg$	
		Cs	Sr
Fi	Groundwater	0.331	0.014
	0.1 M NaCl	0.135	0.016
	4 M NaCl	0.005	0
Str	Groundwater	0.043	0.006
	0.1 M NaCl	0.160	0.007
	4 M NaCl	0.015	0
G-B1	Groundwater	0.138	0.003
	0.1 M NaCl	0.065	0.004
	4 M NaCl	0.002	0
D-Be	Groundwater	0.220	0.005
	0.1 M NaCl	0.100	0.008
	4 M NaCl	0.004	0

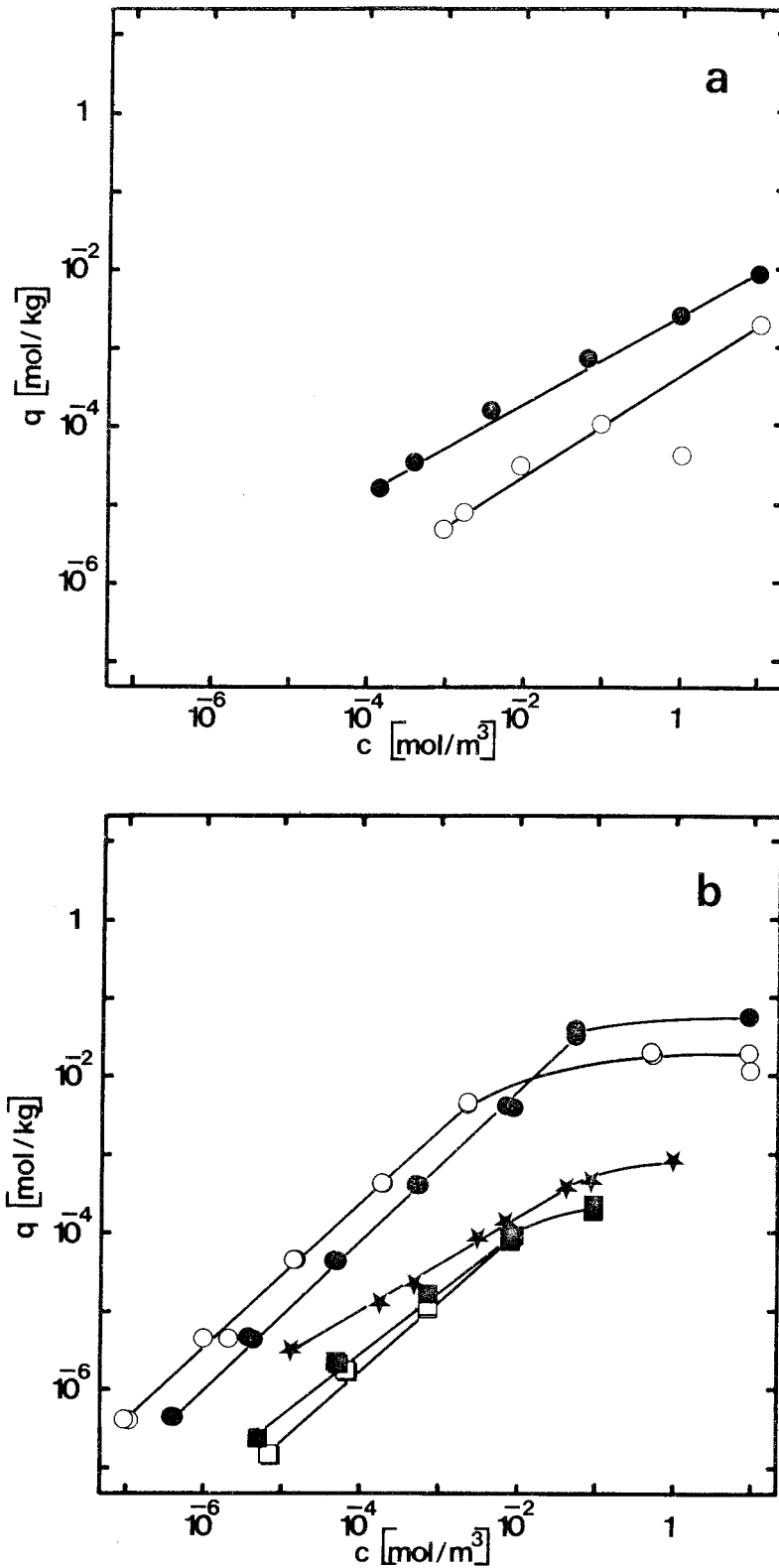


Figure 4 Sorption isotherms for cesium and strontium.

$q$  = concentration in solid,  $c$  = concentration in solution

a. cesium

● Hornblende, ○ Orthoclase

b. cesium (filled symbols) and strontium (open symbols)

○ Montmorillonite (bentonite), □ Stu, ☆ Str

#### 4.1.3. Contact time

The distribution coefficients for cesium and strontium vs. contact time are illustrated in Figure 5. Two different sorption mechanisms are found for both cesium and strontium; first an initial fast (minutes or hours) surface dependent reaction followed by a slower (up to years) process, probably related to a diffusion into the solid matrix. After long contact time (more than 6 months) the weathering and alteration of the solid could lead to either an increased sorption or a desorption of the nuclides (10).

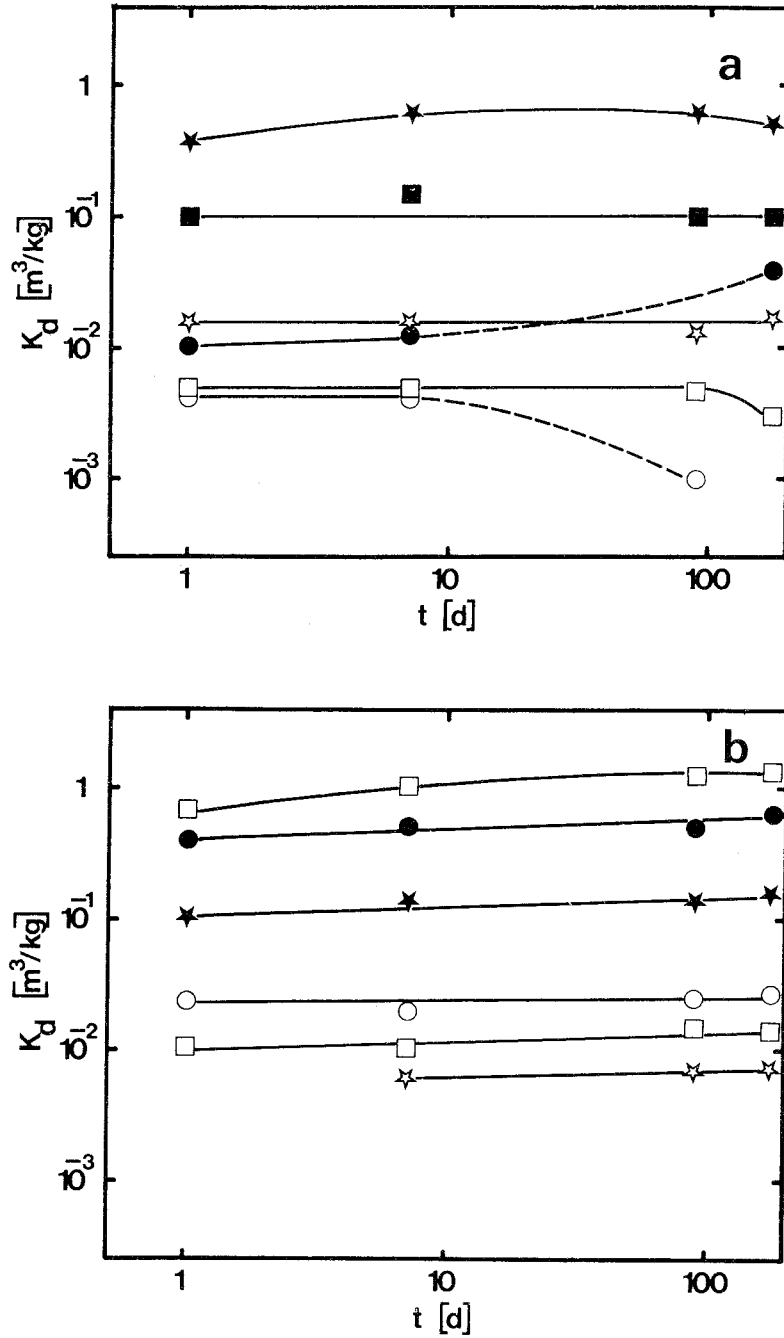


Figure 5 Distribution coefficients for cesium and strontium; Effect of contact time

a. Silicates

□ Hornblende, ○ Orthoclase, ☆ Biotite

b. Rocks

○ Fi, □ Stu, ☆ Str

cesium (filled symbols) and strontium (open symbols)

#### 4.2. Iodide

Iodine in groundwater will be present mainly as the iodide ion,  $I^-$ , but other species may be formed under influence of light or bacteria (23,24). Since most minerals have a negative surface charge in groundwater environment the anion exchange capacity (AEC) is low compared to CEC.

The sorption of iodide on minerals is given in Table 8 (23) and Figures 6 and 7. Iodides of low solubility are formed by a number of metals (Hg, Ag, Bi, Cu, Pb, etc), thus minerals containing these could sorb iodide by chemisorption. In Table 9 the solubility products of some metal iodides are given and when comparing these with the data in Table 9, it is worth noting that cinnabar contains Hg, chalcopyrite Cu and galena Pb, i.e.  $K_d$  increases with decreasing solubility of the iodides. At high ionic strength the sorption decreases even for these minerals (cf. Figure 7). There are some other minerals that are sorbing iodide, probably due to large surface areas. The highest sorption is obtained at low pH as expected for an anion exchange process.

For Stripa granite no sorption has been detected while a low but significant  $K_d$  ( $0.004 \text{ m}^3/\text{kg}$ ) was observed for Studsvik granite at 1 week contact time.

Table 8 Distribution coefficients for iodide from batch experiments  
(Table 5)

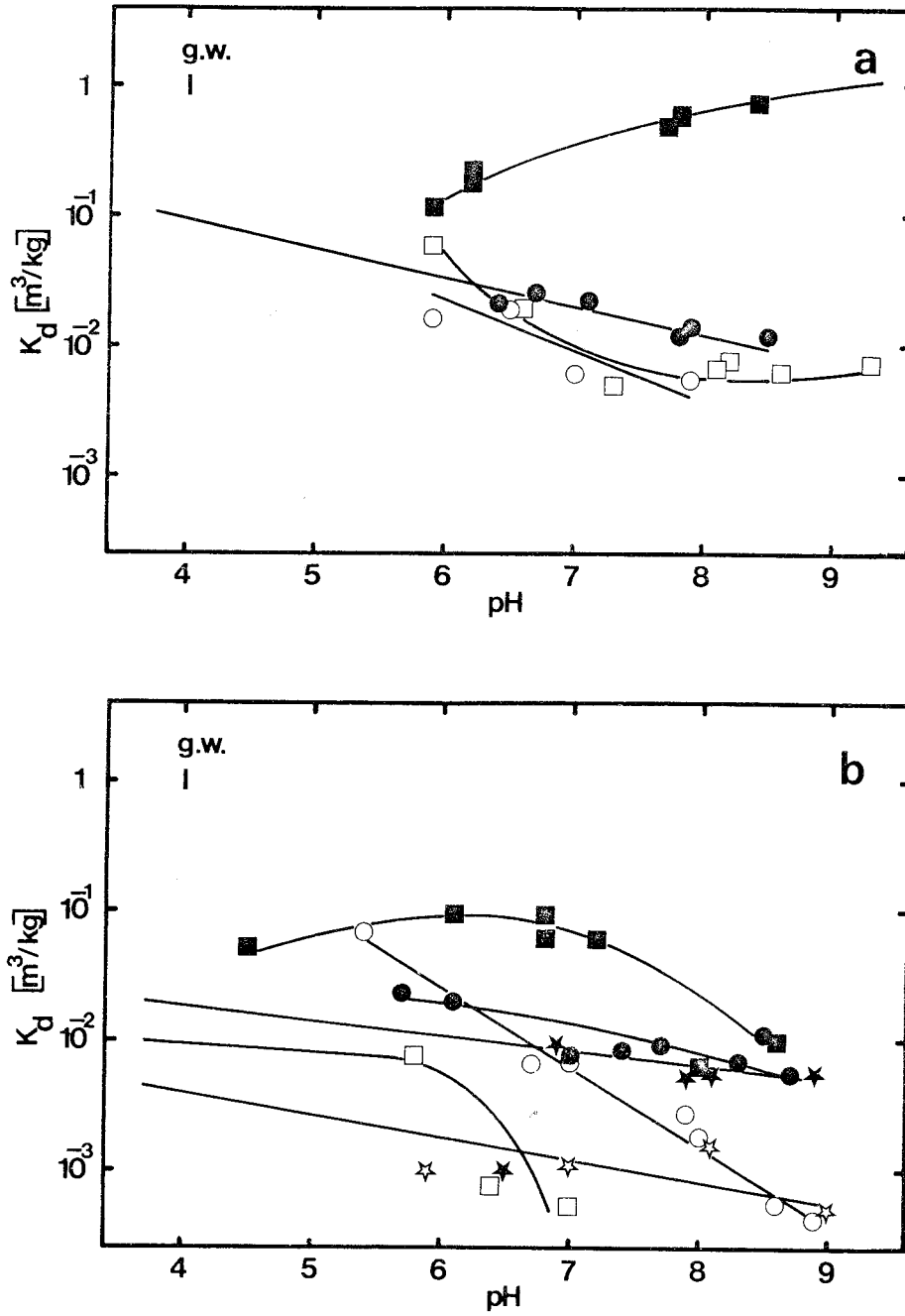
Solid	pH	$K_d, m^3/kg$	
		Artificial groundwater	4 M NaCl
Chalcopyrite	4-6	0.125	0.005
	6-8	0.025	0.003
	8-10	0.005	0.001
Galena	2-4	0.040	0.002
	4-6	0.012	0.002
	6-8	0.006	0.001
	8-10	0.004	0.002
Cinnabar	2-4	1.836	0.030
	4-6	-	0.020
	6-8	0.187	0.025
	8-10	0.043	0.011
Limonite	4-6	0.019	0.037
	6-8	0.010	0.008
	8-10	0.000	0.003
Olivine	4-6	0.066	0.012
	6-8	0.005	0.006
	8-10	0.001	0.003
Attapulgate	4-6	0.000	0.005
	6-8	0.048	0.012
	8-10	0.072	0.008
Serpentine	4-6	0.022	0.084
	6-8	0.011	0.008
	8-10	0.008	0.003
Montmorillonite	2-4	0.010	0.151
	4-6	0.007	0.071
	6-8	0.000	0.050
	8-10	0.000	0.005
Halloysite	2-4	0.020	-
	4-6	-	0.054
	6-8	0.005	0.015
	8-10	0.005	0.015
Quartz	2-4	0.008	0.005
	4-6	0.001	0.000
	6-8	0.001	0.000
	8-10	0.000	0.000

Table 9 Solubility products for metal iodides (25)

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Metal	$\log K_s$
$\text{Cu}^+$	-12.0
$\text{Ag}^+$	-16.08
$\text{Hg}^{2+}$	-28.33
$\text{Pb}^{2+}$	- 8.10

---



**Figure 6** Distribution coefficients for iodide from batch experiments  
(Table 5, groundwater)

a. Hydroxides

- Limonite, ● Fe(III)-hydroxide
- Al-hydroxide, ■ Pb(II)-hydroxide

b. Silicates

- Olivine, ■ Attapulgite, ● Serpentine,
- ★ Halloysite, □ Montmorillonite, ☆ Quartz

c. Others

- Chalcopyrite, ● Galena, □ Cinnabar, ■ AgCl



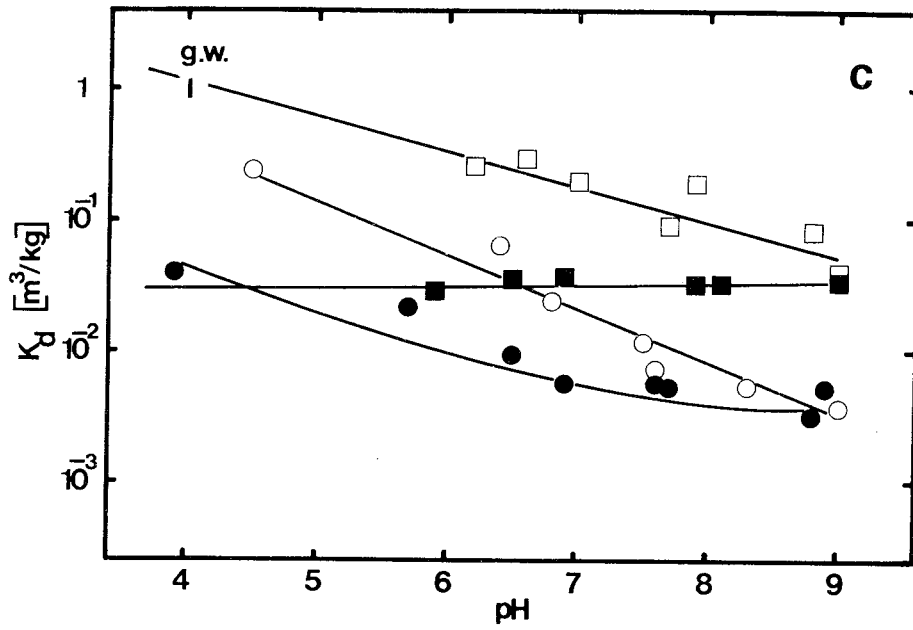


Figure 6 Distribution coefficients for iodide from batch experiments (continued)

c. Others

○ Chalcopyrite, ● Galena, □ Cinnabar, ■ AgCl

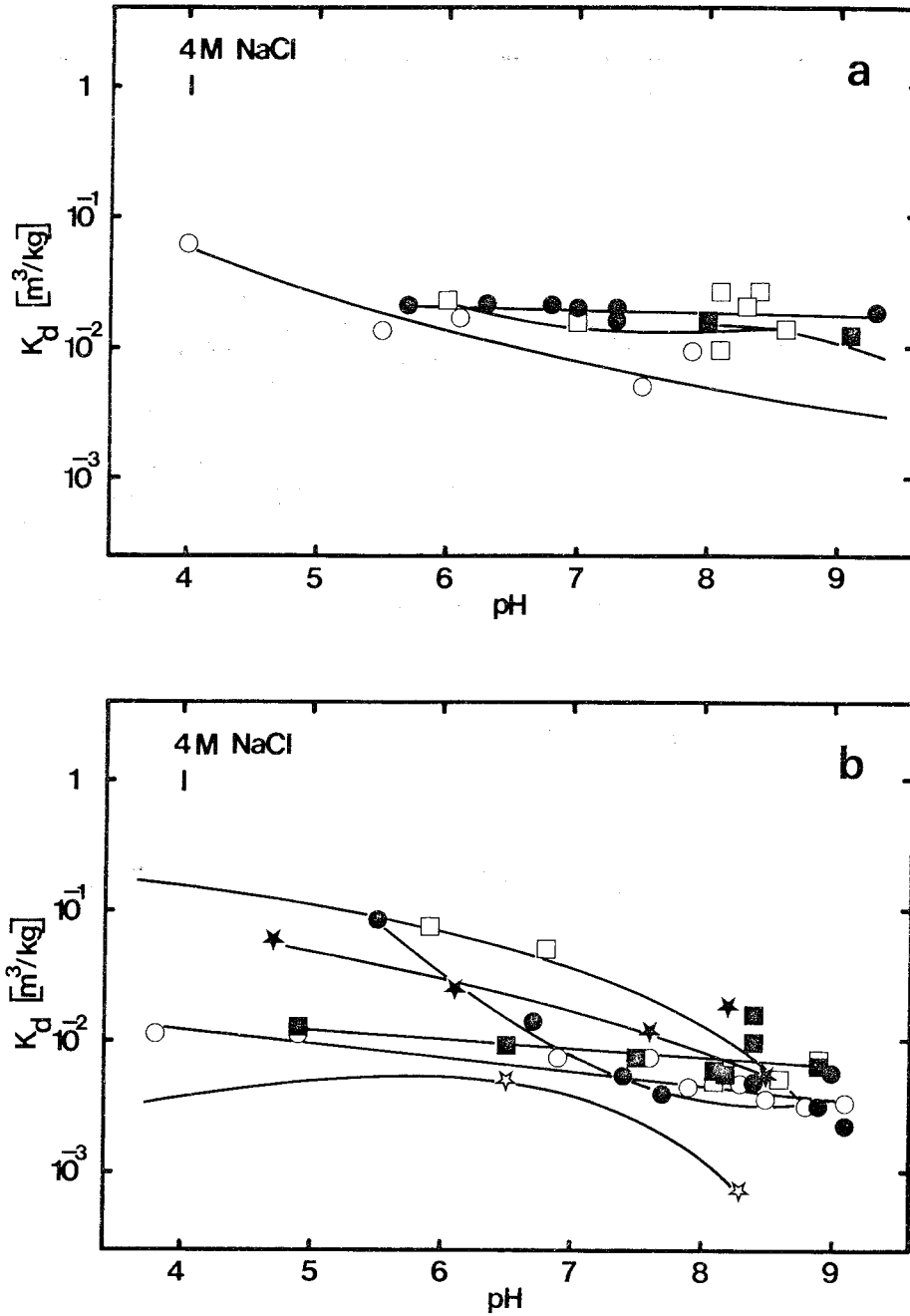


Figure 7 Distribution coefficients for iodide from batch experiments  
(Table 5, 4 M NaCl)

a. Hydroxides

- Limonite, ● Fe(III)-hydroxide
- Al-hydroxide, ■ Pb(II)-hydroxide

b. Silicates

- Olivine, ■ Attapulgite, ● Serpentine,
- ★ Halloysite, □ Montmorillonite, ☆ Quartz

c. Others

- Chalcopyrite, ● Galena, □ Cinnabar, ■ AgCl

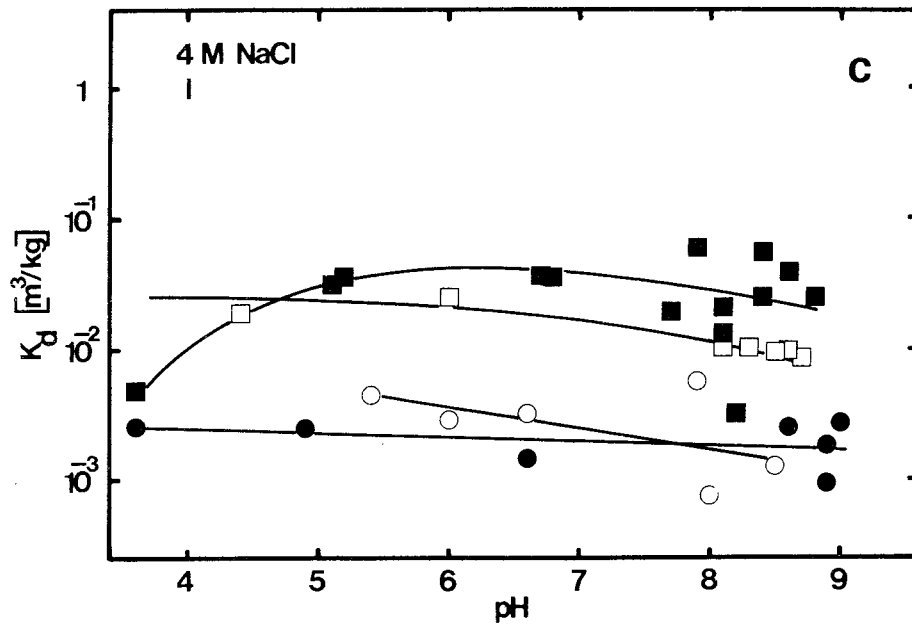


Figure 7 Distribution coefficients for iodide from batch experiments  
(continued)

c. Others

○ Chalcopyrite, ● Galena, □ Cinnabar, ■ AgCl

#### 4.3. Technetium

Technetium exists predominantly as pertechnetate,  $\text{TcO}_4^-$ , in aqueous solution under oxidizing conditions, and in the tetravalent state, either as a hydroxide or as an oxide, in a reducing environment (6).

The sorption of pertechnetate is poor on geologic materials (cf. Table 10), but in the presence of a reducing agent (Fe(II)) there is a small but significant sorption, probably an adsorption or precipitation of technetium (IV)-hydroxide/oxide (3).

Table 10 Distribution coefficients for technetium ( $\text{TcO}_4^-$ ) from batch experiments (Table 5, groundwater system, 1d contact time)

Solid	Aqueous phase	$K_d, \text{m}^3/\text{kg}$
Granite	Aerated	< 0.001
	Deaerated	< 0.001
	Deaerated, 20 ppm Fe(II)	0.050
Biotite	Deaerated	0.001-0.003
Chlorite		
Hornblende		
Pyrite		
Magnetite	Deaerated	0.006

#### 4.4. Carbonate

Groundwater contains a fairly high concentration of carbonate/hydrogen carbonate (in the mM-range), partly from exchange and contact with the biosphere and the atmosphere.

Carbon-14 released into the groundwater will participate in the continuous precipitation/dissolution of calcium carbonate (e.g. the formation and precipitation of the fracture mineral calcite) and will thus be retained compared to the groundwater transport.

The sorption of  $^{14}\text{C}$ , as  $\text{H}^{14}\text{CO}_3^-$ , on calcium and/or carbonate containing minerals, granite and concrete has been determined in batch experiments (cf. Table 11) (5,8). A significant sorption is found for clayish moraine,

calcite and concrete. The high sorption on concrete is probably because of a formation and precipitation of  $\text{CaCO}_3(\text{s})$ .

Table 11 Distribution coefficients for  $\text{HCO}_3^-$  from batch experiments  
(Table 5, groundwater)

Solid	3d	$K_d \times 10^3, \text{m}^3/\text{kg}$		
		1w	5w	6 months
Stripa granite	0	0	0	0
Na-montmorillonite	0	0	0	0
Bentonite/quartz	2.8	8.6	7.6	7.8
Sandy moraine	0.8	1.1	2.6	2.2
Clayish moraine	1.3	2.0	3.0	2.3
Calcite	3.7	3.5	9.0	83
Concrete	5.3	1600	$>10^4$	

#### 4.5. Comparison of batch and column measurements

Retention factors (water velocity/nuclide velocity) and distribution coefficients obtained from column measurements are given in Table 12. When comparing these  $K_d$ -values with  $K_d$ -values from batch measurements it is obvious that at least for Cs, batch data seems to be higher than column data. The only measurement for strontium on granite indicates the opposite, but more measurements would be required to determine this. A deviation towards lower  $K_d$ -values in column measurements would, however, be expected when sorption kinetics are slow, as the contact times in the column are much shorter than in batch measurements.

In a natural fissure the water velocity will be 5-6 orders of magnitude lower than in the laboratory column experiments and the sorption kinetics will probably have less influence on the sorption.

Table 12 Comparison of distribution coefficients ( $K_d$ ) in batch and column experiments

Solid	Tracer	Retention factor	$K_d \times 10^3, m^3/kg$	
			Column	Batch
Calcite	Cs	3	0.5	2
Quartz	Cs	4	0.8	1
Orthoclase	Cs	11	3	10
Hematite	Cs	>22	>3	6
Magnetite	Cs	>100	>12	6
Hornblende	Cs	>250	>50	190
Granite, Stu	Sr	89	20	12

#### 4.6. In situ measurements

A retention factor of the same order of magnitude as for laboratory column tests for crushed bulk rock has been obtained in the field test ( $R = 15-17$  in field and 30-35 at lab) (12). The difference may be explained by the fact that the column study was performed on drill cuttings from the borehole (i.e. representative for the average rock composition) whereas in the field test the radionuclide interact with the fracture surfaces, which may have slightly different sorption properties. As distribution coefficients for Sr do not differ by more than one order of magnitude for most rocks and minerals, the expected differences between field and laboratory measurements due to different mineral composition are of the same order of magnitude as that measured.

In the experiment a small fraction of strontium was transported with the velocity of the water. This may be due to some transport of strontium sorbed on particles in the water (i.e.  $FeOOH(s)$  which may precipitate when the water is areated during the injection), an effect that has been observed also in column studies of radionuclides in clay suspensions (pseudocolloids).

#### 4.7. Predictions of distribution coefficients

There are many different parameters determining the distribution coefficient. One parameter that will vary in nature is the mineralogic composition

of the rock, and it would be of great interest to be able to predict the effect of a change in mineralogy without performing laboratory measurements for each rock. A comparison of predicted  $K_d$ -values for granites, using data for the pure minerals and measured  $K_d$ -values has been performed for cesium, strontium, americium and neptunium, cf. Table 13. For cesium and strontium the predicted values are too low, while the difference for the actinides is smaller. Not even the probable presence of small amounts of weathering products with high  $K_d$ -values can wholly explain the deviations for cesium and strontium. For actinides the mineral composition would be expected to be of minor importance as  $K_d$ -values usually are within one order of magnitude at the same pH.

Table 13 Comparison of predicted and measured distribution coefficients

Rock	Predicted $K_d$ , m <sup>3</sup> /kg				Measured $K_d$ , m <sup>3</sup> /kg			
	Cs	Sr	Am	Np	Cs	Sr	Am	Np
Granite, Str	0.056	0.002	-	-	0.15	0.015	-	-
Granite, Fi	0.062	0.002	-	-	0.49	0.03	-	-
Climax stock granite <sup>a</sup>	-	-	8	0.050	-	-	20	0.100
Westerly granite <sup>a</sup>	-	-	6.3	0.050	-	-	2.5	0.032

<sup>a</sup> See ref 15

## 5. CONCLUSIONS

The sorption of radionuclides at trace concentrations in a geologic groundwater environment is related to both physical and chemical parameters.

Physical parameters of importance are

- o the groundwater velocity (determining the contact time)
- o the surface/mass ratio of the solid
- o the surface texture and frequency of microfissures in the solid.

The most important chemical parameters of the aqueous phase are

- o pH
- o the redox potential (determining the oxidation state)
- o presence of complex-forming agents,

and of the solid phase

- o the mineralogy and degree of weathering
- o the CEC (cation exchange capacity)
- o presence of reducing agents (e.g. divalent iron).

Three main categories of sorption mechanisms influences the over-all transport rate of radionuclides. These processes are: Electrostatic interactions (ion exchange processes) related to the exchange capacity of the solid, the pH and ionic strength of the water and the concentration of the sorbing species; chemisorption processes, where complex-forming agents, either in solution or on the solid phase, interact with the radionuclide; physical adsorption phenomena, where cationic or neutral species of mainly oxides/hydroxides interact with solid surfaces by non-specific adsorption forces.

Cesium and strontium which are non-hydrolyzed metals in environmental waters, participate mainly in ion exchange reactions in geologic systems. Hydrolyzed species, like the actinides and tetravalent technetium are expected to interact through adsorption processes and to some extent by chemisorption/mineralization with geologic media. The negatively charged species, iodide and carbonate, have a sorption largely related to the presence of metals forming sparingly soluble iodide and carbonate complexes, thus interacting through chemisorption processes.

The sorption of redox-sensitive nuclides, like technetium, uranium, neptunium and plutonium, is strongly related to the oxidation state, determined by the redox conditions of the system.

Radionuclides participating in ion exchange reactions, like cesium, are expected to sorb strongly on high-capacity minerals on the fracture surface and be transported into the bulk rock in microfissures and high-capacity minerals. A similar migration into the rock is not expected for nuclides sorbing mainly by chemisorption or physical adsorption.

## 6. ACKNOWLEDGEMENTS

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## SELECTION OF DISTRIBUTION COEFFICIENTS FOR A GRANITE

A list of recommended distribution coefficients for a pilot case used in migration modeling has been compiled, cf. Table A1. The groundwater and rock compositions are typical of the Finnsjön area north of Stockholm and the recommended  $K_d$ -values are to be regarded as conservative (i.e. they may be higher but not lower).

Some comments to the choice of constants:

Cobalt and nickel may be expected to have a similar behaviour. There are very few measurements of distribution coefficients for these metals on granite, but a value of  $0.4 \text{ m}^3/\text{kg}$  at trace concentration of Ni and pH 8 has been measured at CTH. A recommended value for Co and Ni of  $0.2 \text{ m}^3/\text{kg}$  seems feasible, considering a possible higher ionic strength in Finnsjön water than in the synthetic groundwater used in the measurement.

Strontium is sorbed mainly by an ion exchange mechanism and is thus sensitive to ionic strength and pH. The proposed  $0.004 \text{ m}^3/\text{kg}$  is reduced due to the risk of high ionic strengths at Finnsjön. In groundwater of lower ionic strength a higher  $K_d$  - at least  $0.010 \text{ m}^3/\text{kg}$  would be expected for a granite of similar composition. For fracture filling minerals from Finnsjön lower  $K_d$ -values than that for the pure granite have been obtained.

Zirconium and niobium have been found to have very high  $K_d$ -values at pH 8-9 and here  $4 \text{ m}^3/\text{kg}$  may be a conservative value. At higher pH  $K_d$  decreases.

Technetium is redox sensitive but even under oxidizing conditions a  $K_d \neq 0$ , although probably  $<0.001 \text{ m}^3/\text{kg}$  may be expected. At short contact times ( $\sim 1\text{d}$ ) distribution coefficients of  $0.050 \text{ m}^3/\text{kg}$  have been found under reducing conditions, and this may be regarded as a lower limit for longer contact times.

Iodide may be sorbed by some minerals, but there is no reason to use  $K_d > 0$  for a safety analysis as granites usually have  $K_d \approx 0$ .

Cesium sorption is dependent on pH, ionic strength and nuclide concentration. In the case of high ionic strength a quite low value has been chosen -  $0.050 \text{ m}^3/\text{kg}$ . Much higher values have, however, been encountered for granites. These values are for trace concentrations ( $\sim 10^{-8} \text{ M}$ ) of cesium. At higher concentrations lower  $K_d$  may be expected.

Appendix A2

Cerium, neodymium and europium may be expected to behave very similarly to Th, U and Am and thus undergo hydrolysis. The distribution coefficients will be high;  $>5 \text{ m}^3/\text{kg}$  may be expected (cf. ref. 15 where data for all the actinides are discussed).

Table A1 Distribution coefficients for pilot case "Finnsjön"

Nuclide		Granite	Fracture fillings	Comments
Co		0.2		
Ni		0.2		
Sr		0.004	> 0.001	
Zr		4		
Nb		4		
Tc	ox	0.0002		
	red	0.05		
I		0		
Cs		0.05	> 0.001	
Ce, Nd, Eu		$\geq 5$		
Ra		0.1		
Th		$\geq 5$		
Pa		$\geq 5$		
U	ox	0.01		U(VI)
	red	$\geq 5$		U(IV)
Np	ox	0.01		Np(V)
	red	$\geq 5$		Np(IV)
Pu	ox	3		Pu(IV)+Pu(V)
	red	$\geq 5$		Pu(III)
Am		$\geq 5$		

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