

**Concentrations of particulate matter  
and humic substances in deep  
groundwaters and estimated effects  
on the adsorption and transport of  
radionuclides**

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CONCENTRATIONS OF PARTICULATE MATTER AND HUMIC  
SUBSTANCES IN DEEP GROUNDWATERS AND ESTIMATED EFFECTS  
ON THE ADSORPTION AND TRANSPORT OF RADIONUCLIDES

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**ABSTRACT**

The concentration of particulate matter such as colloids and microbes in deep Swedish groundwaters has been measured and has been found to be low in all waters. The results are summarized in this paper. The sorption capacity of relevant radionuclides on the particulate matter has been assessed based on many direct measurements and on comparisons with measurements on similar systems. The maximum transport capacity of nuclides by the particulate matter has been estimated for reversible as well as irreversible sorption of nuclides to particles.

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## SUMMARY

Groundwater contains colloids and microbial matter on which radionuclides may attach themselves. The nuclides attached to the particles may travel with the velocity of water and might not be retarded in the same way as the fraction of the nuclides which is in true solution in the water. The latter may be retarded due to sorption on the rock surfaces.

The concentration of colloidal matter was found to vary between 0.1 and 400 mg/m<sup>3</sup> and that of microbes between 0.1 and 50 mg/m<sup>3</sup>. Humic and fulvic substances which also may influence nuclide transport are present in concentration between 0.1 and 500 mg/m<sup>3</sup>. The particulate matter may be filtered out or may attach itself to the rock surfaces by sorption but it has not been able to model this quantitatively in a reliable way. In this paper we have chosen to assume for illustrative purposes the case where all particulate matter stays in the water.

Nuclides will sorb on the particulate matter but the mechanisms are not so well understood that reliable predictive modelling can be made at present. Two limiting cases were studied. In one case reversible sorption was assumed and it was found that in this case the impact of the particles is negligible. In another case the sorption was assumed to be irreversible. Then the particles may transport the sorbed fraction of the nuclides with the velocity of the water and they may not have the same travel time and opportunity to decay as the dissolved fraction. Sample calculations indicate, however, that the sorbed fraction does not seem to be able give very high doses even in a case where all the leaking nuclides go to one and the same well.

## 1 BACKGROUND AND INTRODUCTION

The transport of radionuclides from a repository for radioactive waste would be influenced by the interactions between dissolved species and exposed solid surfaces of geologic materials etc. Radionuclide sorption would take place on stationary macro surfaces (fracture and fissure surfaces, etc) as well as on mobile solid particulate matter (inorganic colloids, microorganisms) present in the groundwater. Thus, sorption on stationary surfaces would reduce the transport rate, while sorption on a particulate mobile phase might enhance the transport rate, the particles serving as carriers.

Most important hydrochemical parameters are (at a given redox potential) pH of the system (determining the degree of hydrolysis of many of the relevant radionuclides as well as the charge properties of the solid systems), the presence of complexing agents (notably organic humic substances) and to some extent the total salinity. The importance of colloidal matter in deep groundwaters (inorganic geologic materials as well as microorganisms) on the mobility of radionuclides (under given pH, salinity and redox conditions) are discussed in this note.

## 2 CONCENTRATIONS OF COLLOIDS IN DEEP GROUNDWATERS

Inorganic colloidal particles are found in groundwaters at all depths. Analyses of groundwater samples taken as deep as 800 m show that content and composition of the colloids vary with depth /AR 90-37/. Calcite particles dominate in the deep samples. Less abundant but also represented are iron hydroxide, gibbsite, iron sulphides and quartz.

Analyses of particle contents in groundwater tend to be conservative. Drilling operation generates particles of e.g. quartz and clay. Pumping can mobilise e.g. clay particles. Degassing or uptake of carbon dioxide or oxygen can cause precipitation of particles composed of calcite or iron hydroxide. Mixing of groundwater from different levels during sampling can cause precipitation of e.g. iron sulphide. The reason for that is the relatively high content of iron(II) ions at intermediate depths and sulphide which is more abundant at depth.

The ease by which colloids and particles are created and their relative instability have been demonstrated by an in situ experiment in Fjällveden (Wikberg, 1987). One cubic meter of aerated oxidizing water was pumped into a deep aquifer. The aquifer had been penetrated by a borehole and the borehole section containing the conductive zone had been isolated with packer sleeves. The groundwater was sampled at regular intervals after the injection. It was found that iron and sulphur containing samples were generated in abundance by the mixing of oxidizing water and natural reducing deep groundwater. It was also found that the concentration of particles dropped back to ambient levels after a few days.

The reference AR 90-37 reports a median value of  $1.56 \cdot 10^{-4}$  kg/m<sup>3</sup> of particles and states that concentrations are expected to be lower than  $4 \cdot 10^{-4}$  kg/m<sup>3</sup>. Based on these results a central value of  $10^{-4}$  kg/m<sup>3</sup> is chosen, as well as a maximum value of  $4 \cdot 10^{-4}$  kg/m<sup>3</sup> and a minimum value of  $10^{-5}$  kg/m<sup>3</sup> (see Table 1). The sorption properties expected are those of calcite and quartz particles rather than clay which is not abundant at depth in these waters.

It should be noted that the deep Swedish granitic groundwater is characterized by a relatively high ionic strength and abundance of divalent ions such as calcium. This will act to destabilize colloids which is reflected in the relatively low concentrations of particles. A low salinity groundwater with monovalent cations like e.g. sodium instead of divalent ions would promote colloid stability. Especially clay particles may become released from fracture minerals.



## 2.1 Concentration of microbes

Microbes have been sampled in deep groundwaters by means of a down-hole probe. The sampling device was, during the sampling operation, isolated between rubber packers in water conducting sections of boreholes. Sampling were performed at depths ranging from 100 to 900 m in Ävrö, Äspö, Laxemar and Stripa /SKB TR 90-05/.

Concentrations of bacteria are typically around 100 000 bacteria per ml. This has been chosen as the central case. A maximum value was obtained from the variation of analytical data. The selected value is 500 000 bacteria per ml.

Spherical bacteria with a radius of  $0.2 \cdot 10^{-6}$  m would have a weight of approximately  $10^{-17}$  kg for one cell. Assuming that part of the bacteria are larger such as rod shaped cells, a ten times higher value of  $10^{-16}$  kg per cell has been used. This gives the concentration of bacteria in the central case of  $10^{-5}$  kg/m<sup>3</sup> with a maximum value of  $5 \cdot 10^{-5}$  kg/m<sup>3</sup> (see Table 1).

## 2.2 Concentration of humic and fulvic acids

Measurements of TOC (total organic carbon) are frequently in the range of one to a few mg/l. However, careful analyses of organic content in deep groundwater show that only a small part consists of complex forming humic and fulvic acids. The rest is partly contaminants e.g. organics bleeding from plastic materials and partly natural but simple organic compounds. According to experiences from isolation of humic substances from deep groundwater samples concentrations are generally well below 0.1 mg/l. This value,  $10^{-4}$  kg/m<sup>3</sup> is chosen as a central case. A maximum value of  $5 \cdot 10^{-4}$  kg/m<sup>3</sup> has been selected.

## 2.3 Reference data

The assessed concentration levels of colloids, microorganisms and humic substances (humic and fulvic acids) are given in Table 1 (central case, as well as maximum concentrations).

*Table 1* Assessed concentrations of colloids and microorganisms as well as humic substances in groundwaters

Concentration, (kg/m <sup>3</sup> )·10 <sup>6</sup>	Central value	Max value
Inorganic colloids	100	400
Humic substances (HA, FA)	100	500
Microorganisms	10	50

### 3 RADIONUCLIDE ADSORPTION ON PARTICULATE MATTER

The groundwater composition expected in a repository is shown in appendix 1. The following conditions, representative of a deep repository in crystalline granitic bedrock, are discussed:

- \* Water with either high salinity (HIS; 0.1 to 0.5 M) or low salinity (LIS; <0.05 M)
- \* High or low pH (8 and 6, respectively)
- \* Presence (central case, Table 1) or absence of dissolved humic substances

The low pH (6) is not representative of deep groundwaters. The presence of humic substances has to be considered in all natural systems. Thus, the most realistic case is:

- \* LIS (or HIS, representing salt water intrusion) at pH 7 to 8 with humics present and reducing conditions.

All waters would contain a particulate fraction (c.f. Table 1). A realistic system is assumed to be:

- \* HIS, pH 7, humics present and reducing conditions; colloids and microorganisms corresponding to the central case present.

The sorption is discussed in terms of over-all distribution coefficients,  $K_d$ ; (mol/kg solid phase)/(mol/m<sup>3</sup> aqueous phase) under the specific conditions of the reference cases. Distribution coefficients for relevant radionuclides under the above conditions (at low concentrations, well below total adsorption capacities) are discussed on the basis of chemical properties of individual or groups of radionuclides.

Recent experimental investigations of sorption on inorganic particles /SKB AR 90-20/ and on microbes /SKB TR 90-26/ have been used as a base for the evaluation.

The uptake and transport of radionuclides by means of colloidal particles, have been investigated in studies of natural analogues to radioactive waste disposal. In the Poços de Caldas project the dissolution and transport of thorium and lanthanides from natural ore concentrations were investigated as one of the subprojects (Chapman et al, 1990). Two sites near the town of Poços de Caldas in Brazil were chosen: a thorium, rare earth element mineralization at Morro do Ferro and a uranium mine at Osamu Utsumi. Sampling and analysis of groundwater at the sites reveal the strong tendency of tri- and tetravalent ions of lanthanides and thorium to get sorbed on natural occurring particles. However, there are very low concentrations downstream the mineralizations and consequently no evidence of mobility due to particle transport. The same conclusions were reported from observations of thorium in groundwater at the Broubster analogue site in Caithness, UK (Longworth et al, 1989).

### 3.1 Monovalent elements

#### Cesium

Cesium does not hydrolyze or form significant complexes with humic substances. The predominant adsorption mechanism is electrostatic interaction, determined by the properties of the solid (mineralogic composition) as well as the hydrochemistry (pH, ionic strength). In a heterogeneous crystalline rock the silicates (feldspars, micas, amphiboles, pyroxenes, as well as their weathering products, clay minerals), have large ion exchange capacities, and a substantial Cs adsorption has been reported in the literature for such systems. For LIS and pH 8 a value of 0.25 m<sup>3</sup>/kg was selected. An increased ionic strength would reduce the adsorption. A reduction by a factor of 5, when going from LIS to HIS, has been assumed. A pH-decrease would reduce the negative surface charge, and thereby the electrostatic adsorption. A similar reduction of the distribution coefficient when going from pH 8 to 6 (by a factor of 5) has been assumed. Both the salinity effect (from LIS to HIS) and pH-effect (from 8 to 6) are overestimated.

The presence of humics would not effect the adsorption of Cs.

Natural colloids (quartz, calcite, pyrite, goethite) generally have low cation exchange capacities. A reduction of  $K_d$  by a factor of 5, compared to the rock system (HIS, pH 8), is assumed and also indicated by reported literature data (in the absence of colloidal clay minerals, which are not normally encountered in deep groundwaters). Also for bacteria the same value is assumed, since no enhanced electrostatic interaction, or specific uptake, is expected or reported.

### 3.2 Divalent elements

#### Strontium and radium

Strontium would not hydrolyze at pH below 8. Complexation with humics could possibly be significant although far from dominating. Just as for Cs electrostatic interaction would be the dominating adsorption mechanism. Reported distribution coefficients (e.g corresponding to the LIS pH 8 case) are, however, significantly lower than for Cs. Values for Sr corresponding to the Cs-value, reduced by a factor of 5, are assessed for LIS/HIS at pH 6/pH 8, respectively.

For the humic system, a reduction by a factor of 2 is assumed, to account for the possible effect of a formation of low-sorbing humic complexes at pH above the point of zero charge (PZC) of the solid surface (which is below pH 8 in the rock system).

Identical sorption on bacteria and rock (HIS, pH 8) is assumed. No specific sorption effects are reported (or expected).

The sorption of Ra is generally at least one order of magnitude higher than for Sr under otherwise identical conditions. Distribution coefficients a factor of 10 higher than the corresponding Sr-values are assessed.

A certain affinity for the colloidal fraction must be considered for both Sr and Ra, considering that most groundwaters are saturated with respect to calcite. Thus, a precipitation/coprecipitation of Sr (as well as Ra) is feasible. Distribution coefficients of 1 and 10, respectively, are assessed.

#### Nickel

Nickel would resemble Sr at low pH, i.e. hydrolysis is insignificant, and electrostatic interaction would be the dominating adsorption mechanism. At high pH, however, the hydrolysis may enhance the adsorption significantly, and values a factor of 2 higher than for Sr are assumed.

For the uptake in the presence of humics and on bacteria values are selected in analogy with the Sr-systems.

Since a coprecipitation with Fe can not be excluded a high distribution coefficient has to be assumed for the colloid system (a value of 1 is assessed).

#### Lead

Lead differs significantly from both Sr and Ni in terms of much more pronounced hydrolysis. Thus, Pb is essentially quantitatively hydrolyzed already at pH 6, leading to high adsorption on oxide- silicate surfaces. This is e.g. the case in most surface water systems, where Pb is immobilized at pH above 5-6. A  $K_d$ -value of 1 is assessed, to include the hydrolysis effect, although lowered by a factor of 2 when going from pH 8 to pH 6, to include any pH-related reduction. No significant ionic strength effects are expected at the low salinity. However, at pH 6, a reduction by a factor of 2 is assumed when increasing the salinity from LIS to HIS to account for a possible ion exchange contribution

A reduction by a factor of 2 is assumed for the humic acid system.

A certain sorption would be expected on bacteria surfaces, although these have a hydrophobic character. However, if some strong specific organic surface complex would be formed, this would probably leave the surface at the high pH (e.g. at pH 8; above PZC), as is observed for the trivalent elements (see below). The same  $K_d$ -value as for Ni is assumed.

Lead is predominantly associated with colloidal particulate matter (oxides/hydroxides) in surface water systems. A similar reaction could be possible in groundwaters, and a high  $K_d$ -value is suggested for the colloid case.

Palladium, tin

Both Pd and Sn (although divalent) exist predominantly in anionic forms. For Pd a formation of strong chloride complexes is expected, while anionic hydroxides could be formed for Sn at high pH. A significant, but low, adsorption is assumed for both elements in all the cases (c.f. discussion on see below). Justified distinctions between various ionic strengths and pH etc can not be given.

### 3.3 Trivalent elements

Lanthanides, actinides

All the relevant lanthanides (Ce, Sm) and trivalent actinides (Pu, Am, Cm) tend to hydrolyse at a fairly low pH and form very strong complexes with humic substances. Thus, at high pH, regardless of the ionic strength, a high  $K_d$ -value (3) is suggested, and justified considering several reported studies. A reduction (by a factor of 3) is assumed when going from pH 8 to 6, as well as by a factor of 2 when increasing the salinity from LIS to HIS (in analogy with Pb).

The complexes with humics are strong, and a significant fraction of trivalent elements could be complexed by e.g. aquatic fulvic acids. A reduction of the distribution coefficient by a factor of 10 is assumed, corresponding to the formation of low-sorbing complexes at pH above PZC. Even larger reductions in  $K_d$  are reported in the literature, however at much higher total concentrations of humic materials.

A high sorption of trivalent elements on bacteria has been observed at low pH. However, at pH 8 (above PZC), the observed distribution coefficients are drastically reduced, which can indicate the release of a complexing exudate at high pH. Thus, this effect would be similar to an increasing concentration of strong complexing agents with humic-like properties, however at a concentration level far below the assumed total concentration of humics (c.f. Table 1).

A very high sorption (coprecipitation ?) of trivalent lanthanides/actinides on representative colloidal matter has been observed, both in laboratory systems and in situ. A  $K_d$ -value of 30, corresponding to an increase by a factor of 10, is suggested.

### 3.4 Tetravalent elements

#### Actinides

The actinides (Th, U and Np under reducing conditions; possibly a minor fraction of Pu at high pH) hydrolyse already at pH far below environmental levels, and the adsorption would be high and independent of the salinity at pH 8. A distribution coefficient of 5 is assessed for pH 8 and reduced to 3 for pH 6, which over-estimates the effect of a reduction of pH.

Some reduction in the adsorption due to complexation with humics is feasible, but much less than for the trivalent state (a reduction of the distribution coefficient from 5 to 3 suggested).

A certain sorption to bacterial surfaces can not be excluded, although a specific uptake, or the release of a strongly complexing exudate, would not be expected, considering the dominating hydrolysis. Just as for the trivalent state, the tetravalent state is strongly associated with the colloidal fraction, and the same  $K_d$ -value as for the trivalent elements is assumed.

#### Technetium, zirconium, niobium, protactinium

Both Tc and Zr resemble the larger tetravalent elements with respect to the very strong hydrolysis. However, the higher hydrolysis would extend to a dehydration, and divalent species of "metalyI"-type can be formed. However, cationic or neutral hydrolysis species would still be expected, and anionic carbonates are not formed (unlike the uranyl carbonates for U(VI) under oxidizing conditions).

Distribution coefficient that are a factor of 2-3 lower than for the tetravalent actinides are selected except for the colloid case, where identical interaction is assumed.

The elements Pa and Nb (pentavalent) are not entirely analogous to either the tetravalent actinides Th, U and Np or the smaller tetravalent Tc and Zr. However, hydrolysis is dominating and interaction with solid surfaces is generally pronounced. The same distribution coefficients as for Tc and Zr are assessed.

### 3.5 Others

#### Carbon, selenium

Both C and Se exist as anions (carbonate, selenite). Selenite adsorbs strongly on e.g. oxide surfaces at low pH, just as arsenite and arsenate. However, at pH 8 the adsorption is minor.

Carbonate would interact to some extent with solid phases at pH 8, considering the fact that the systems generally are saturated with respect to calcite. Substitution reactions with solid calcium carbonate on fissure surfaces and in the colloidal fraction is feasible with time.

The same  $K_d$ -value (0.001) is suggested for Se and C as for the anionic Pd and Sn, except for the uptake of C by microorganism. Here a distribution coefficient corresponding to an even distribution between inorganic carbonate in solution (at 100 mg/l-level) and organic carbon in the microorganisms (50 % C) is assumed as the limiting case, giving a  $K_d$ -value of 25.

### Halogens

The halogens Cl and I are both anionic under reducing conditions. Distribution coefficients of 0 are assumed.

The suggested distribution coefficient corresponding to the various cases are summarized in Table 2.



Table 2 Selected distribution coefficients ( $K_d$ ,  $m^3/kg$ ) for granitic rock under reducing conditions neglecting the effects of humic substances. LIS = low ionic strength (<0.05 M), HIS = high ionic strength (0.1 to 0.5 M) /SKB AR 91-07/.

Ox.State	Element	$K_d(\text{rock})^a$					
		pH8		pH7		pH6	
		LIS	HIS	LIS	HIS	LIS	HIS
M(I)	Cs	0.25	0.05	0.1	0.02	0.05	0.01
M(II)	Sr	0.05	0.01	0.02	0.004	0.01	0.002
	Ra	0.5	0.1	0.2	0.04	0.1	0.02
	Ni	0.1	0.02	0.02	0.004	0.01	0.002
	Pb	1	1	0.7	0.7	0.5	0.25
	Pd,Sn	0.001	0.001	0.001	0.001	0.001	0.001
M(III)	Ln,An <sup>b</sup>	3	3	1.7	1.7	1	0.5
M(IV)	An <sup>c</sup>	5	5	3.9	3.9	3	3
	Tc,Zr, Nb <sup>d</sup> ,Pa	2	2	1.4	1.4	1	1
Others	C	0.001	0.001	0.001	0.001	0.001	0.001
	X <sup>e</sup>	0	0	0	0	0	0
	Se	0.001	0.001	0.001	0.001	0.001	0.001

<sup>a</sup> No humics present

<sup>b</sup> Lanthanides; Ce, Sm; actinides: Pu, Am

<sup>c</sup> Actinides; Th, U, Np

<sup>d</sup> Nb is pentavalent

<sup>e</sup> Halogens; Cl, I

*Table 3* Selected distribution coefficients ( $K_d$ ,  $m^3/kg$ ) for reference case (HIS, neutral pH, reducing conditions, presence of humic substances, low radionuclide concentrations).  $K_{part}$  denotes sorption coefficients for particulate matter (colloids and bacteria, respectively).

Ox.State	Element	$K_d(\text{rock})$	$K_{part}$ Colloids	$K_{part}$ Bacteria
M(I)	Cs	0.03 <sup>f</sup>	0.01	0.01
M(II)	Sr	0.003 <sup>f</sup>	1	0.01
	Ra	0.03 <sup>f</sup>	10	0.1
	Ni	0.006 <sup>f</sup>	1	0.02
	Pb	0.3	10	0.02
	Pd,Sn	0.001	0.001	0.001
M(III)	Ln,An <sup>b</sup>	0.2	30	0.01
M(IV)	An <sup>c</sup>	2	30	0.5
	Tc,Zr,	1	30	0.2
	Nb <sup>d</sup> ,Pa	1	30	0.2
Others	C	0.001	0.001	25
	X <sup>e</sup>	0	0	0
	Se	0.001	0.001	0.001

<sup>b</sup> Lanthanides; Ce, Sm; actinides: Pu, Am

<sup>c</sup> Actinides; Th, U, Np

<sup>d</sup> Nb is pentavalent

<sup>e</sup> Halogens; Cl, I

<sup>f</sup> For LIS these values are increased by a factor of 5

## 4 TRANSPORT BY PARTICULATE MATTER

There are many mechanisms by which dissolved species are attached onto particulate matter. They have been discussed extensively in the literature. A recent thorough literature review has been made by Grauer (1990). We use the word sorbed for all the attachment mechanisms in this paper. Grauer concludes that there are still many unknown factors which makes it difficult to predict the fate of colloids and the attached matter. It is not even possible to predict if the sorption is reversible or irreversible. We will not discuss the mechanisms in this paper but instead investigate the consequences for the transport of the two extreme cases: fully reversible linear sorption and irreversible sorption.

### 4.1 Reversible sorption case

We consider a system where there is a solid phase on (in) which the particle can sorb reversibly by linear sorption. The distribution coefficient is denoted by  $K_d$ . The nuclide can also sorb on the particulate matter with a sorption coefficient  $K_{part}$ . The nuclide will divide itself between the three phases; water, solid and particles according to the equilibrium relations.

The "effective" sorption capacity  $K_{eff}$  of the nuclide on the rock is the ratio of the concentration on the solid,  $K_d \cdot C$  to the total concentration of the nuclide in the liquid phase

$$K_{eff} = K_d / (1 + S \cdot K_{part}) \quad (1)$$

This means that the amount of the nuclide which attaches itself to the solid phase is decreased by a factor  $1/(1 + S \cdot K_{part})$  compared to a system where there is no particulate matter. The fraction of nuclides in the water which are attached to the particulate matter is

$$S \cdot K_{part} / (1 + S \cdot K_{part}) \quad (2)$$

The effective sorption coefficient  $K_{eff}$  can be used instead of  $K_d$  in transport models based on retardation due to volume or surface sorption and also in models where matrix diffusion and sorption in the internal rock matrix take place.

For porous media flow and retardation the effective distribution coefficient can be used to modify the retardation factor  $R_d$ . This becomes

$$R_{d,eff} = 1 + \frac{\rho(1-\epsilon)K_{eff}}{\epsilon} = 1 + \frac{\rho(1-\epsilon)}{\epsilon} \frac{K_d}{(1+S \cdot K_{part})} \quad (3)$$

For systems with small porosities as in fractured rock the presence of the particulates will decrease the retardation by a factor  $1/(1 + S \cdot K_{part})$  for the porous flow case. For the matrix diffusion case the relations are more complex.

When there is more than one kind of particulate matter in the water, colloids, microbes etc, the effective sorption coefficient becomes

$$K_{eff} = K_d / (1 + \Sigma S \cdot K_{part}) \quad (4)$$

where the the summation is made over all the particulate species. If the nuclide exists in more than on species which can sorb, e.g. several complexes with different sorption coefficients, the above relations are not entirely correct. Because, however, the  $K_d$ 's in practice do not make this distinction and are used to describe the sum effects the errors are acceptable.

The figures in tables 1 and 2 indicate that even a combination of the highest particle concentration  $S=500 \cdot 10^{-6} \text{ kg/m}^3$  and the highest  $K_{part}$  value which is 30 will only influence the  $K_d$  value by 1%. This is entirely negligible in relation to the error bounds on  $K_d$ 's. When there is reversible sorption the impact of the presence of particulate matter will be negligible.

## 4.2 Irreversible sorption case

Nuclides may sorb irreversibly on the particle by a number mechanisms. By irreversible sorption in this paper is meant that during the time of interest no significant release of the nuclide takes place. This may mean that the release rate is slow in comparison to the time it takes for the particles to travel to the biosphere. These times may vary from a few years to many thousands of years and for our purposes it is sufficient that most of the sorbed nuclide will still be attached after these times in order for it to be "irreversibly" sorbed. There is a variety of mechanisms by which "irreversible" sorption could take place. Surface precipitation on colloid surfaces, covalent bonding to surfaces and in microbes, formation of solid solutions in the particle phase, slow desorption kinetics and several others. As it has not been possible to definitely exclude the possibility of "irreversible" sorption we have felt it necessary to explore the potential importance of this effect on the transport of the nuclides.

It is assumed that the nuclide will distribute itself between the particle phase and the water in a given ratio given as it leaves the near field. As the sorption mechanisms are not known it is not possible to determine the capacity of the colloids to sorb on the nuclides. It may be argued that the particles will not sorb more of the nuclides irreversibly than they would sorb in experiments performed to determine sorption coefficients. If it is assumed that the higher  $K_d$  values observed in laboratory experiments may have been caused by irreversible sorption the mass of nuclides attached to the particulate phase can be determined. The concentration in the water of nuclide attached to the particles  $C_s$  then is given by equation (5)

$$C_s = C \cdot S \cdot K_{part} \quad (5)$$

That part of the nuclides which resides on the particles will travel to the biosphere with the water velocity and will not further interact with the rock. For most cases it can be assumed that the whole rock is short circuited and that all the nuclide released from the near field which was attached to the particles arrives directly to the biosphere.

## 5 DISCUSSION AND CONCLUSIONS

Particulate matter suspended in water can attach nuclides by various mechanisms. The particles with the nuclides may move with a velocity different from that of the flowing water. It does not seem possible with the present understanding of the behaviour of particulate matter to make reliable quantitative predictions of either the sorption effects or of the particulate transport processes. There seems to be little doubt that particles are filtered out to a large extent during their movement but as the processes cannot be quantified, especially in fractured rocks, firm predictions are not possible.

In this paper we have chosen to look at some bounding cases which we consider to be the worst from the nuclide transport point of view. We have assumed that all particles which are present in the water when it passes the repository also are present in the water as it reaches the biosphere. With this basic assumption two further cases are considered. When the nuclides are reversibly sorbed this will have a negligible influence on the transport because the amount of the nuclide on the colloid is much less than 1 % of that in the water and as it is reversibly sorbed it will influence the retardation due to sorption on the rock by the same negligible fraction. Thus if the nuclide would decay a significant fraction during the transport this would not be influenced.

A totally different situation occurs if the nuclide is attached irreversibly. Then the particle with its attached nuclide will travel with the velocity of water (in our assumptions) and will not be desorbed. The nuclide on the particles may thus have a much shorter residence time than that dissolved in the water. The latter may be retarded by sorption on the rock. Even if the fraction on the particles is small, the effect can be considerable in some cases where the fraction in the water decays much and that on the particles does not. The sample calculations in appendix 3 indicate that even in the case of irreversible sorption the carrying capacity of the particles is very small and the potential dose from the nuclides on the particles is small.

## 6 NOTATION

C	Concentration of nuclide	kg/m <sup>3</sup>
C <sub>s</sub>	Concentration of nuclide attached to particles in water	kg/m <sup>3</sup>
K <sub>d</sub>	Sorption coefficient on rock	m <sup>3</sup> /kg
K <sub>eff</sub>	Effective sorption coefficient on rock	m <sup>3</sup> /kg
K <sub>part</sub>	Sorption coefficient on particles	m <sup>3</sup> /kg
R <sub>d,eff</sub>	Effective retardation factor	-
S	Concentration of particulate matter	kg/m <sup>3</sup>
ε	Porosity	-
ρ	Density of rock	kg/m <sup>3</sup>

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## Appendix 1: Groundwater composition

The groundwaters studied in SKB 91 are two typical Finnsjö-waters (Ahlbom et al 1989), one saline, and one high in carbonate. The concentrations are given in g/m<sup>3</sup>.

	Fresh	Saline
Ca	76	1600
Na	23	1700
Mg	6.3	120
K	3.2	13
Fe	9.0	0.016
Si	6.0	6.0
Cl	61	5500
Br	0.3	29
F	0.6	1.2
NO <sub>3</sub>	0.5	
SO <sub>4</sub>	8.9	380
HCO <sub>3</sub>	220	48
pH	6.9	7.0
Eh(mV) (Hydrogen scale)	-200/650	-200/650

Only these two waters are considered in the safety analysis. Chemical effects on the groundwater caused by bentonite reactions are neglected.

Groundwater below 100 m in Finnsjön is reducing in character with the redox potential Eh in the range -200 to -300 mV. This has been measured with Eh electrodes (Ahlbom et al 1989). The measured redox potential is controlled by the iron system Fe<sup>2+</sup>-Fe(OH)<sub>3</sub>(s) (Grenthe 1991). Uranium solubility under those conditions is expected to be controlled by uraninite UO<sub>2</sub>. This is also indicated by low total concentrations of uranium, in the order of a few micrograms per litre and relatively high activity ratios of the uranium isotopes <sup>234</sup>U to <sup>238</sup>U. Sorption of redox sensitive elements like uranium, neptunium and technetium at repository depths will correspond to natural reducing conditions as demonstrated by laboratory experiments (Eriksen 1988).

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## Appendix 2: Dose conversion factors for some nuclides

Table A2 Dose conversion factors Sv/Bq, Well case, the only exposure path is by drinking the water. Halflives and sorption coefficients on particular matter

	Ingestion Sv/Bq	T <sub>half</sub> Years	K <sub>part</sub> m <sup>3</sup> /kg Colloids	K <sub>part</sub> m <sup>3</sup> /kg Bacteria	
C-14	5.6·10 <sup>-10</sup>	5730	0.001	25	(ICRP56)
Se-79	2.3·10 <sup>-9</sup>	6.4·10 <sup>4</sup>	0.001	0.001	(ICRP30)
Tc-99	3.4·10 <sup>-10</sup>	2.1·10 <sup>5</sup>	30	0.2	(ICRP30)
Sn-126	4.7·10 <sup>-9</sup>	1.0·10 <sup>5</sup>	0.001	0.001	(ICRP30)
I-129	6.4·10 <sup>-8</sup>	1.6·10 <sup>7</sup>	0	0.001	(ICRP56)
Cs-135	1.9·10 <sup>-9</sup>	2.3·10 <sup>6</sup>	0.01	0.01	(ICRP30)
Pb-210	1.4·10 <sup>-6</sup>	22.3	30 <sup>a</sup>	0.5 <sup>a</sup>	(ICRP30)
Ra-225	3.1·10 <sup>-7</sup>	14.8	30 <sup>a</sup>	0.5 <sup>a</sup>	(ICRP30)
Ra-226	3.1·10 <sup>-7</sup>	1602	10	0.1	(ICRP30)
Ac-227	3.8·10 <sup>-6</sup>	2.18	30 <sup>a</sup>	0.5 <sup>a</sup>	(ICRP30)
Th-229	9.4·10 <sup>-7</sup>	7340	30	0.5	(ICRP30)
Th-230	1.6·10 <sup>-7</sup>	7.7·10 <sup>4</sup>	30	0.5	(Joh 82)
Pa-231	2.2·10 <sup>-5</sup>	3.2·10 <sup>4</sup>	30	0.2	(Joh 82)
U-233	3.1·10 <sup>-7</sup>	1.6·10 <sup>5</sup>	30	0.5	(Joh 82)*
U-234	3.0·10 <sup>-7</sup>	2.5·10 <sup>4</sup>	30	0.5	(Joh 82)*
U-235	2.8·10 <sup>-7</sup>	2.0·10 <sup>8</sup>	30	0.5	(Joh 84)
U-236	2.9·10 <sup>-7</sup>	2.3·10 <sup>7</sup>	30	0.5	(Joh 82)*
U-238	2.7·10 <sup>-7</sup>	4.5·10 <sup>9</sup>	30	0.5	(Joh 84)
Np-237	4.5·10 <sup>-7</sup>	2.1·10 <sup>6</sup>	30	0.5	(ICRP56)
Pu-239	9.7·10 <sup>-7</sup>	2.4·10 <sup>4</sup>	30	0.01	(ICRP56)

<sup>a</sup>As for precursor with largest K<sub>part</sub>

ICRP56, International Commission on Radiologic Protection. Age dependent doses to members of the public from intake of radionuclides. Annals of the ICRP, ICRP Publication no 56, Part 1, 1990

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\*Based on values for U-235 and U-238 given by Johansson (1984).

### Appendix 3: Some sample dose calculations of the effects of irreversible sorption of nuclides

In the examples below it is assumed that a person drinks 600 l/year of the water in a well. The total flowrate of water to the well is 4000 m<sup>3</sup>/a. All the nuclides which are attached to particulate matter are assumed to end up in this well. The repository contains 6000 canisters each containing 1 tonne of uranium. After 1000 years 6 canisters are breached and leach with a rate of 10<sup>-7</sup> fractions per year. The fraction of the released nuclides which attach themselves to the particulate matter is  $S \cdot K_{\text{part}} / (1 + S \cdot K_{\text{part}})$  from equation (2). The data used in the calculations are summarized in table A3.1 below.

Table A3.1 Basic data used in sample dose calculations

Uranium in repository tonnes	6000
1 canister in 1000 is releasing gives fraction leaching	0.001
Release rate. Fraction per year	1·10 <sup>-7</sup>
Fraction of released nuclides to one well with 4000 m <sup>3</sup> /a water	1
Man drinks 0.6 m <sup>3</sup> /a, ie 0.6/4000 parts of what ends up in the well	0.00015
Fraction of release that is particle bound is $S \cdot K_{\text{part}} / (1 + S \cdot K_{\text{part}})$	
Example for actinide $S = 1 \cdot 10^{-4}$ kg/m <sup>3</sup> , $K_d = 30$	0.003
Tonnes per year that ends up in one man is prod of column above	2.7·10 <sup>-13</sup>
Multiply this by Bq/tonne Uranium to get Bq/a to our drinking man	
For other fractions on particulate matter use these figures	

Table A3.2 shows the calculated results using the data in table A3.1.

It may be noted that the right hand column is based upon the unlikely assumption that all nuclides released from the repository go directly to the well. This column is included to illustrate the upper bound of possible doses.

*Table A3.2* Doses from some selected nuclides. See Table A3.1 and the description of the conditions under which these dose estimates were made.

Nuclide	Dose covers. factor Sv/Bq	Inventory* GBq tonne U at 1000 years	Fract on particles	Dose to man by particles Sv/a	Dose to man if all goes to well Sv/a
Se-79	2.3E-09	17	0.001	3.9E-16	3.5E-09
Tc-99	3.4E-10	560	3.00E-03	5.1E-11	1.7E-08
Sn-126	4.7E-09	34	1.10E-07	1.6E-15	1.4E-08
I-129	6.4E-08	1.3	1.00E-08	7.5E-17	7.5E-09
Cs-135	1.9E-09	14	1.10E-06	2.6E-15	2.4E-09
Pb-210	1.4E-06	0.13	3.01E-03	4.9E-11	1.6E-08
Ra-225	3.1E-07	0.006	3.01E-03	5.0E-13	1.7E-10
Ra-226	3.1E-07	0.13	1.00E-03	3.6E-12	3.6E-09
Ac-227	3.8E-06	0.011	3.01E-03	1.1E-11	3.8E-09
Th-229	9.4E-07	0.006	3.01E-03	1.5E-12	5.1E-10
Th-230	1.6E-07	0.74	3.01E-03	3.2E-11	1.1E-08
Pa-231	2.2E-05	0.011	3.00E-03	6.5E-11	2.2E-08
U-233	3.1E-07	0.15	3.01E-03	1.3E-11	4.2E-09
U-234	3.0E-07	89	3.01E-03	7.2E-09	2.4E-06
U-235	2.8E-07	0.52	3.01E-03	3.9E-11	1.3E-08
U-236	2.9E-07	10	3.01E-03	7.8E-10	2.6E-07
U-238	2.7E-07	12	3.01E-03	8.8E-10	2.9E-07
Np-237	4.5E-07	48	3.01E-03	5.8E-09	1.9E-06
Pu-239	9.7E-07	11000	3.00E-03	2.9E-06	9.6E-04

\* Data from KBS-3, 1983.

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<sup>1</sup>Environmental Services, Studsvik Nuclear,  
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<sup>2</sup>MBT Tecnologia Ambiental, CENT, Cerdanyola,  
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<sup>2</sup>Sveriges Geologiska AB  
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<sup>1</sup>Vattenfall HydroPower AB  
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<sup>2</sup>Chalmers Institute of Technology  
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<sup>2</sup>SKB AB  
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<sup>2</sup>Chalmers Institute of Technology

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<sup>1</sup>Conterra AB  
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**Description of transport pathways in a KBS-3 type repository**  
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