Division of Safety and Radiation Protection (S)

Partition Coefficients (K_d) for the Modelling of Transport Processes of Radionuclides in Groundwater

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Table of Contents

List of Figures and Tables

Figures

Tables

1 Introduction

1.1 EURISOL Design Study and SAFERIB

The EURISOL (European Isotope Separation On-Line Radioactive Ion Beam Facility) project is aimed at the design - and eventual construction - of the "next-generation" European ISOL radioactive ion beam (RIB) facility.

The EURISOL Design Study (EURISOL DS) is a project funded by the EUROPEAN COMMUNITY (chap. 8) within the 6th Framework Programme as a Research Infrastructures Action under the "Structuring the European Research Area Specific Programme". The Project started officially on 1st February 2005, for the duration of 4 years. The EURISOL design study produced detailed engineering-oriented studies and technical prototyping work for the next-generation ISOL Radioactive Ion Beam (RIB) facility in Europe.

Twenty institutes and laboratories within Europe had offered to take part in the design study as full participants, with an additional 20 institutions – in Europe, North America and Asia – collaborating as contributors.

The work of EURISOL DS was divided into 12 tasks:

- Task 1: Management
- Task 2: Multi-MW target station
- Task 3: Direct (100-kW) target
- Task 4: Fission target
- Task 5: Safety & radioprotection
- Task 6: Heavy-ion accelerator
- Task 7: Proton accelerator
- Task 8: Superconducting cavity development
- Task 9: Beam Preparation
- Task 10: Physics and instrumentation
- Task 11: Beam intensity calculations
- Task 12: Beta-beam aspects

Task 5 of the project was "Safety and Radioprotection". Within this task work on activity generation and transport, shielding, decommissioning, and legislation were performed. Here, activation of soil and groundwater and activity transport in ground wa-

ter was studied, in close cooperation with the project SAFERIB (safety of radioactive ion beam facilities) which was also funded by EU's Framework Programme.

1.2   Accelerators/ion beam facilities and groundwater

Accelerators/ion beam facilities are widely used instruments using particles such as protons which are "fired" on a specific target after passing the accelerator.

Before operation and even before starting the construction of a high-energy accelerator a complex licensing procedure has to be performed which addresses also safety issues. Part of the licensing documents is usually a safety analysis report (SAR). Within this report possible impacts on humans and the environment have to be estimated.

Depending on their energy, particles like protons and neutrons can activate substances in the soil, shielding and groundwater such as organic carbon, chemical compounds and ions which are present in the subsurface and the soil/groundwater close to the sources of radiation, e.g. an accelerator or a target station (Figure 1-1).
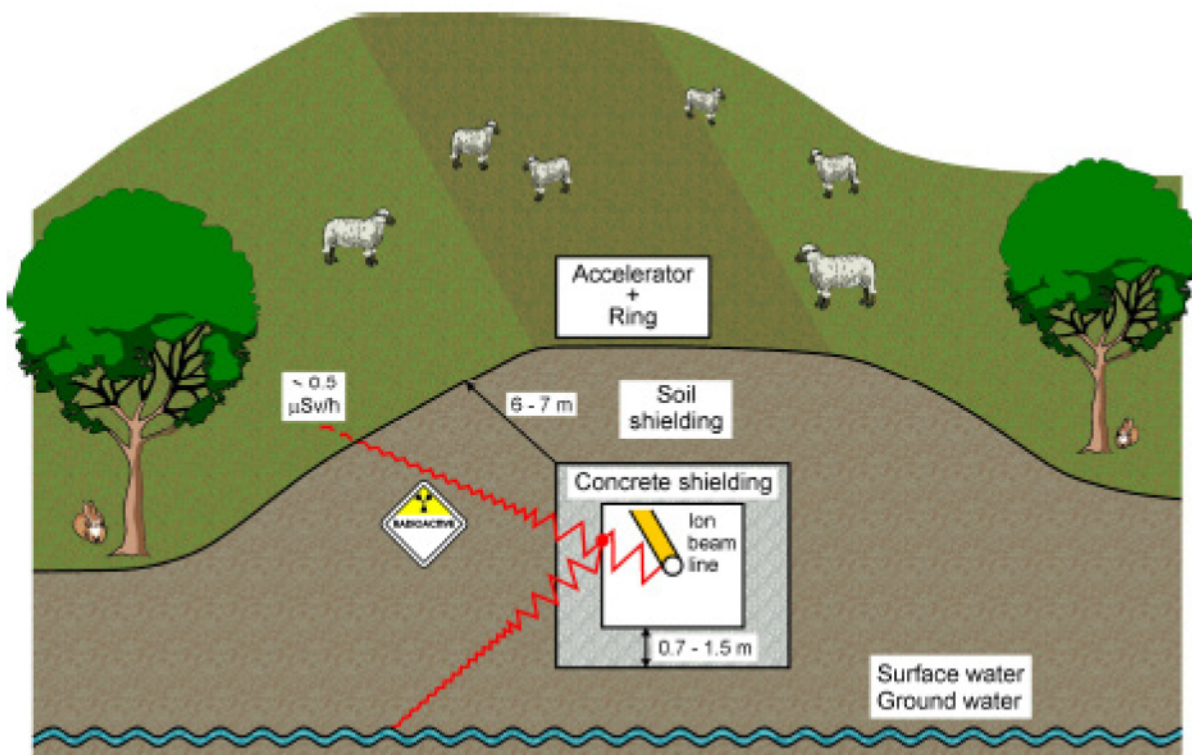
Figure 1-1:   Schematic drawing of the shielding of an accelerator (Moormann et al., 2003)

As part of the licensing procedure it has to be shown that any radiation coming from an accelerator does not exceed the limits given by the relevant regulations. Direct

radiation from e. g. a beam or a target station can be routinely modeled resulting in dose rates. Figure 1-2 shows calculated dose rates for a planned linac.

For the transfer of radionuclides in the groundwater this proof can be demonstrated by the application of a computer based transport model (Heuel-Fabianek et al., 2003). This modeling is relevant to all sites where radionuclides can reach the groundwater by both direct activation within an aquifer (saturated flow conditions) and transport by infiltrating surface waters (unsaturated flow conditions).

The understanding of the transport and fate of radionuclides in the subsurface is of major importance for the setup of groundwater / transport model and the interpretation of the related results (output). For this purpose the partition (or distribution) coefficient K_d estimates the migration potential of a specific contaminant in aqueous solution in contact to solid phases.

Position	A	B	C	D
Dose rate [µSv/h]	37	0.8	2.74	0.92
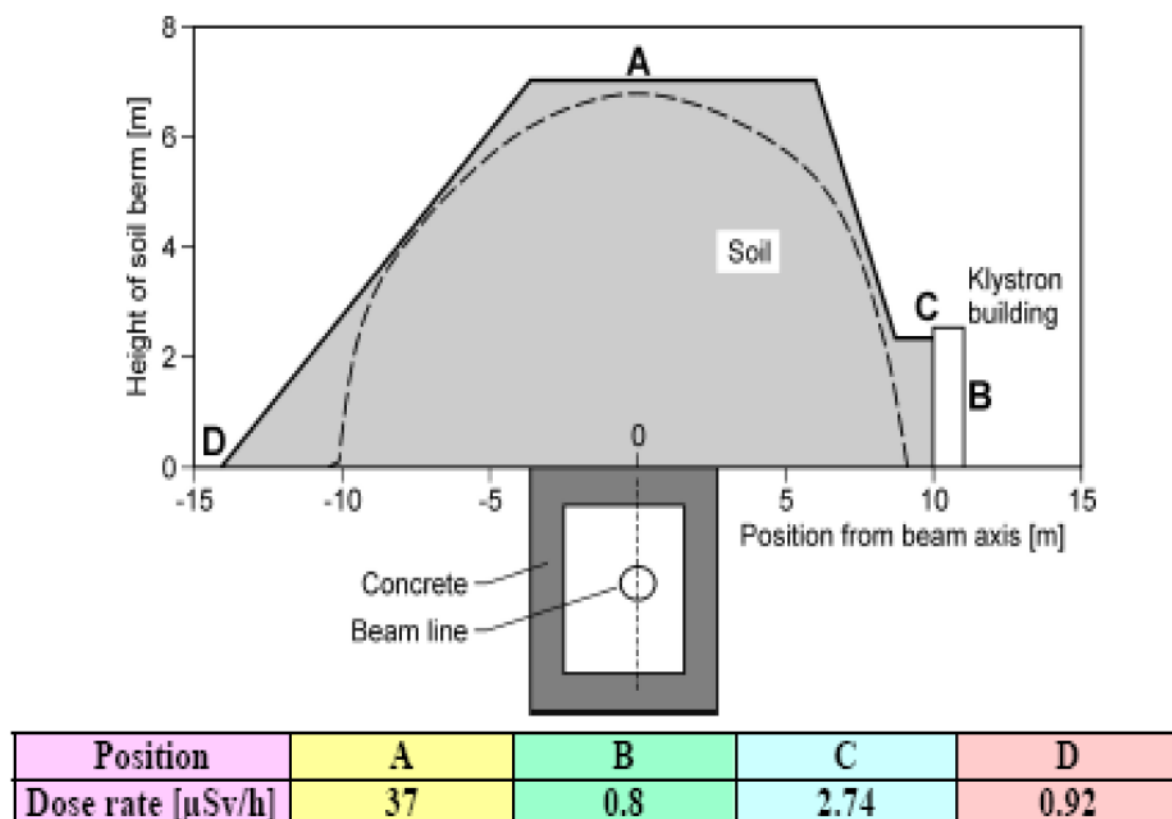
Figure 1-2: Minimum soil profile (broken line) and real soil profile (solid curve) for the high-energy part of a planned linac and calculated dose rates (Moormann et al., 2003)

This approach can be used not only for radionuclides entering the groundwater "regularly" as part of a permitted use of ionizing radiation but also if radionuclides enter the environment by other processes, e. g. uranium mining activities, nuclear accidents or even assaults with radiological dispersal devices (RDD).

Due to a great variety of parameters influencing the migration of contaminants/radionuclide (Figure 1-3), e.g. kind of clay minerals, suspended solids, interaction between contaminants, sorption/desorption processes, bacterial activity, physico-chemical properties of groundwater, decay of contaminants the use of K_d values in transport modeling is always a simplifying estimation. Szermerski (2010) clearly showed advantages and disadvantages of using K_d in models.
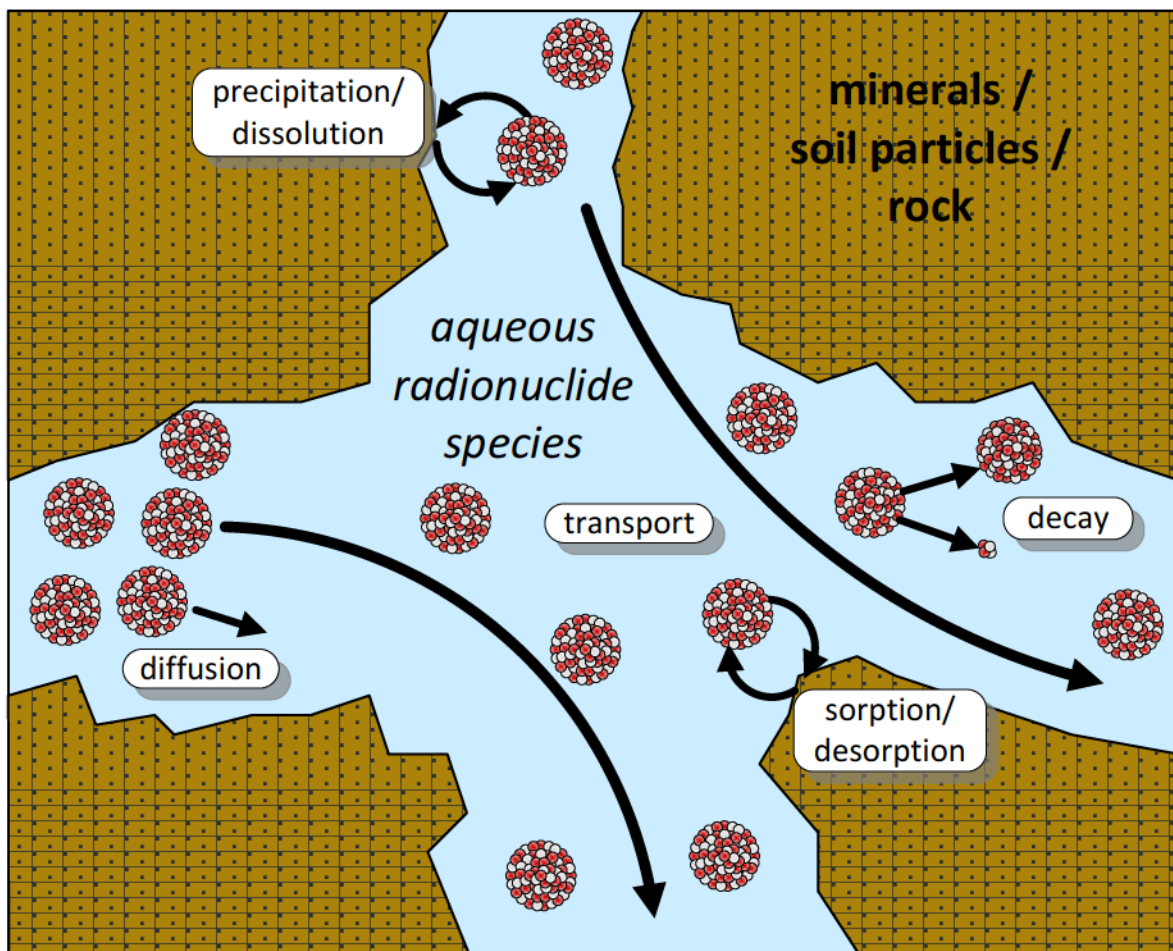
Figure 1-3:   Transport and fate of radionuclides in groundwater

For a sophisticated modeling it is preferable to determine K_d values for the specific soil and the contaminants of concern by laboratory methods (e.g. batch methods,

flow-through methods) or, even better, in the field. Unfortunately these activities are very time and resources consuming.

Often the values of the above mentioned parameters greatly vary in the area of concern. In this case results from laboratory and field methods characterize just spots in a three-dimensional aquifer and do not represent the total volume.

A typical result of modeling the transfer of radionuclides in the groundwater at a specific site is shown in Figure 1-4.
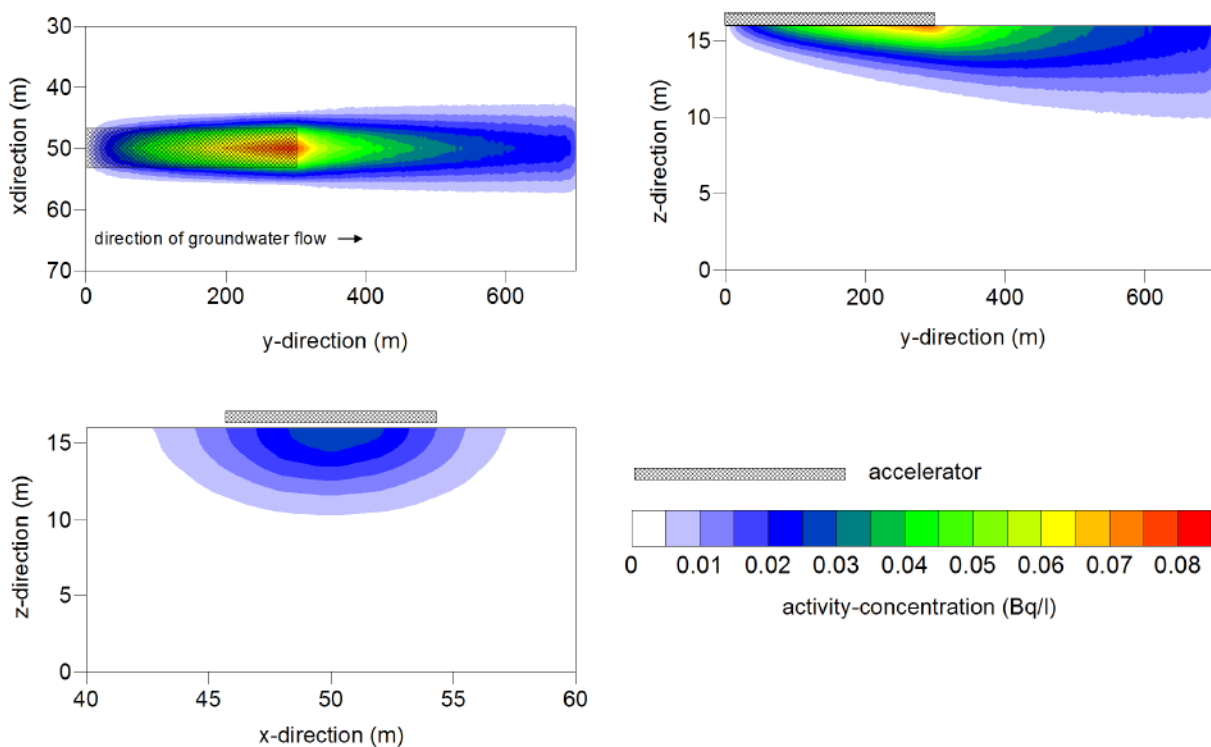
Figure 1-4:   Resultant activity concentration plume of ^{3}H under an accelerator with continuous contamination after steady-state conditions occur (Prolingheuer et al. 2009)

For reasonable results during a screening process it is often more effective to use K_d values from the literature and estimate critically their applicability for the specific task. Some reports are citing K_d values collected from many sources including estimated values found in numerous publications (chapter 1.6) to give an overview about K_d values relevant for specific modeling purposes, e.g. IAEA (1994), Thibault, D.H. et al. (1990), U.S. EPA (1999b, 2004, 2005).

This report is also a collection of K$_d$ values for radionuclides. The radionuclides were selected with respect to transport process in groundwater at (potential) accelerators sites.

K$_d$-data and information regarding geochemical behavior of radionuclides relevant in other processes such as nuclear accidents or assaults can be found in the cited literature (chap. 7), e. g. United States Environmental Protection Agency (1999b; cadmium, cesium, plutonium, radon, uranium etc.) and Pacific Northwest National Laboratory (2003; americium, cesium, iodine, uranium, plutonium etc.).

A "conservative" approach during the process of using "collected" K$_d$ values, which results in a potential worst-case concentration, will increase the acceptance of the results especially if regulatory limits for the protection of humans or the environment have to be reached.

2 Relevant isotopes

For the derivation of the K_d values it is necessary to analyze the chemical constituents of the groundwater which can be activated if passing an area where the groundwater is exposed to thermal neutrons and protons. In addition, activated elements can enter an aquifer after "production" in the shield (concrete, soil etc.) or surrounding soil of an accelerator if these elements can be mobilized e.g. by infiltrating surface water or by direct contact of shield and groundwater. Additionally, water itself should be taken into account before starting activation calculations (Schlögl et al., 2007).

Diffusion of activated elements from soil particles or mineral phases into the ground water within the aquifer or the soil moisture in the vadose zone can, in most cases, be ignored because the water is usually in equilibrium with the local soil. Therefore, natural occurring radioactive constituents of the soil or soil particles are not relevant because they can be estimated as immobile or fixed (Brock, 1978).

Table 2-1:   Constituents of the groundwater of the shallow aquifer upstream a proposed site southeast of Forschungszentrum Jülich, Germany

parameter	concentration[#]	unit	parameter	concentration[#]	unit
Oxygen °	14,1	mg/l	K [#]	2,67	mg/l
Nitrite-N °	0,01	mg/l	Na [#]	17,10	mg/l
Nitrate-N °	61,2	mg/l	Ca [#]	137,0	mg/l
Chloride °	87,2	mg/l	Mn [#]	20,6	mg/l
Sulfate °	151,0	mg/l	Fe [#]	417,0	mg/l
			As [#]	0,35	µg/l
			Ba [#]	16,70	µg/l
			Co [#]	0,17	µg/l
			Cr [#]	3,70	µg/l
			Cu [#]	5,62	µg/l
			Mo [#]	0,022	µg/l
			Ni [#]	5,38	µg/l
			Pb [#]	0,41	µg/l
[#] average values from 2 analysis of wells I and II, February 2006			Rb [#]	0,18	µg/l
			Tl [#]	0,0015	µg/l
° values from analysis of well I, November 2005			U [#]	1,29	µg/l
			Zn [#]	25,30	µg/l

An example for the constituents of groundwater beneath a potential accelerator site is shown in Table 2-1. The data of Table 2-1 are from a sample taken at the shallow wells I and II (total depth: 20 m) of the Forschungszentrum Jülich, Germany. These wells are upstream a potential accelerator site (Mank et al., 2003).

Table 2-2 shows an example for the radionuclides which has been evaluated as important as input for a specific application of a transport model.

The radionuclides of Table 2-2 were selected in accordance with a safety report for a licensing procedure for an accelerator (COSY) at the Research Center Jülich (Probst, 1992).

Table 2-2:   Relevant radionuclides for groundwater modeling for the COSY facility at Forschungszentrum Jülich (Probst, 1992)

radionuclide	radionuclide
^{3}H	^{48}Sc
^{7}Be	^{32}Si
^{22}Na	^{32}P
^{35}S	^{33}P
^{37}Ar	^{44}Ti
^{39}Ar	^{48}V
^{42}Ar	^{49}V
^{45}Ca	^{51}Cr
^{47}Ca	^{52}Mn
^{44m}Sc	^{54}Mn
^{46}Sc	^{55}Fe
^{47}Sc	^{59}Fe

Additionally these and other radionuclides "produced" by activation of soil have been described as radiological relevant or significant at existing or planned facilities/sites, e.g.:

- ^{3}H, ^{22}Na; Superconducting Super Collider Laboratory SSCL, Waxahachie/Texas, USA (Romero, V. et a., 1994, Baker, S. et al.,1994)

- ^{3}H, ^{7}Be, ^{22}Na, ^{24}Na, ^{45}Ca, ^{54}Mn, ^{55}Fe; Next Linear Collider NLC, USA (Rokni, S., 2000)

- ^{3}H, ^{22}Na; Linac Coherent Light Source LCLS at Stanford Linear Accelerator Center SLAC, USA (Mao, X.S. et al., 2006)

- ^{3}H, ^{22}Na, ^{54}Mn, ^{60}Co, ^{152}Eu; High Energy Accelerator Research Organization KEK, Japan (Miura, T. et al., 2005)

- ^{7}Be, ^{22}Na, ^{24}Na, ^{35}S, ^{37}Ar, ^{45}Ca, ^{51}Cr, ^{52}Mn, ^{54}Mn, ^{56}Mn, ^{55}Fe, ^{60}Co, ^{152}Eu; Proton Accelerator Facility of PEFP, Korea (An, S.H. et al., 2007)

- ^{3}H, ^{7}Be, ^{14}C, ^{22}Na, ^{26}Al, ^{36}Cl, ^{40}K, ^{55}Fe; "large neutron sources", considering only those radionuclides with a half-life greater than 10 hours (Schlögl, B. et al., 2007)

Depending on their half-life and the partition coefficient Prolingheuer et al. (2006) set up a matrix of radionuclides relevant in soil typical at Forschungszentrum Jülich site (Figure 2-1). Most relevant for dose calculations are those radionuclides with a long half-life and a low K_d-value. Therefore, a main focus should be on ^{36}Cl, ^{3}H, ^{60}Co, and ^{14}C.

P-32	Ca-45	H-3	Cl-36
14.26 d	163 d	12.323 a	300000 a
5 cm³/g	5 cm³/g	0 cm³/g	0 cm³/g
5.97E-08 Bq/m³	1.00E-08 Bq/m³	3.43E-05 Bq/m³	1.30E-05 Bq/m³
Co-55	S-35	Co-60	C-14
17.54 h	87.5 d	5.272 a	5730 a
30 cm³/g	14 cm³/g	30 cm³/g	7 cm³/g
9.47E-13 Bq/m³	1.23E-07 Bq/m³	2.72E-12 Bq/m³	3.28E-06 Bq/m³
Na-24	Co-57	Mn-54	Si-32
14.96 h	271.79 d	312.2 d	172 a
76 cm³/g	30 cm³/g	50 cm³/g	35 cm³/g
7.08E-08 Bq/m³	3.55E-12 Bq/m³	2.40E-12 Bq/m³	1.41E-10 Bq/m³

Partition coefficient K_D

Half-life $T_{1/2}$

Figure 2-1:   Matrix of radionuclides relevant in Jülich soil (half-life, K_d-value, saturation concentration below 0.8 m concrete shielding for a loss of 1 W/m) (Prolingheuer, N. et al, 2006)

Miura et al. (2005) showed results of measurements of radionuclides under a 12 GeV proton accelerator facility at the High Energy Accelerator Research Organization (KEK) (Figure 2-2).

The measured radionuclides were induced by activation. The groundwater table was at a depth of 1 m. Thus, the radionuclides under the groundwater table could be directly transported depending on their geochemical behavior in the specific aquifer.
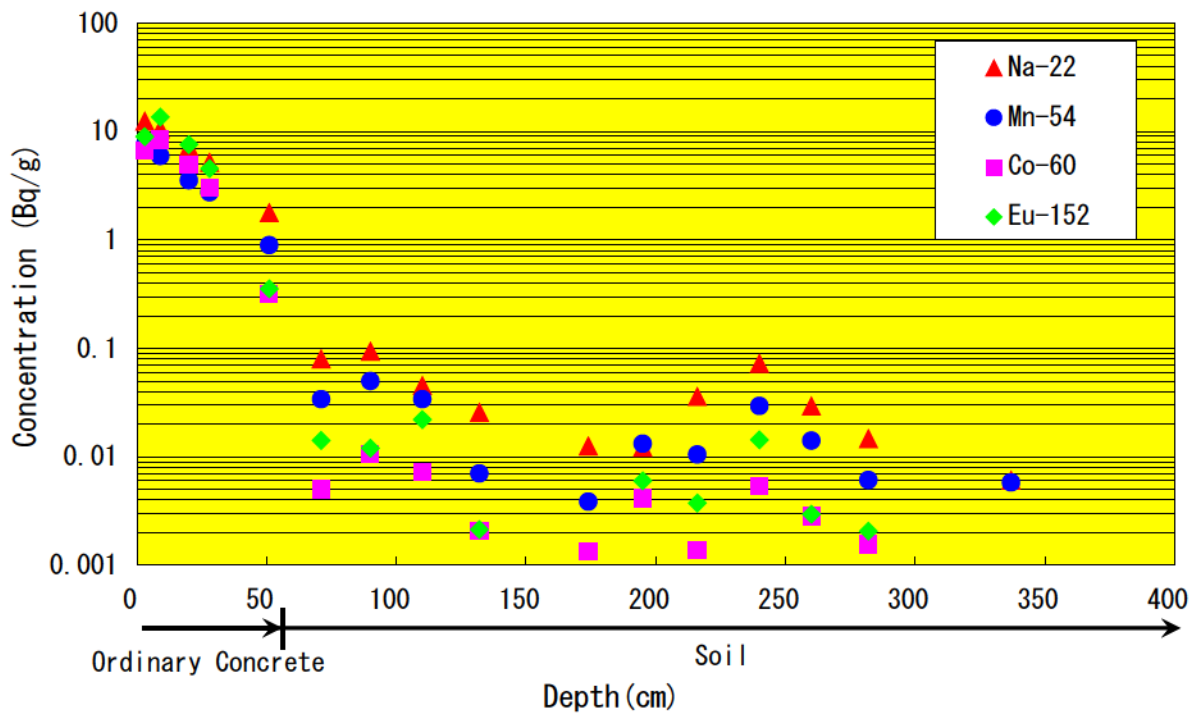
Figure 2-2:   Vertical profiles of radionuclides in the soil around a beam line at KEK, Japan (Miura et al., 2005)

3   Partition coefficient K_d - background

3.1   Definition

K_d was first introduced by Mayer and Tompkins (1947) based on activities in the United States related to the separation of elements which are products of uranium fission in ion exchange columns.

The Partition coefficient is defined as the concentration of solute in the adsorbed phase (mass of solute per unit mass of soil) divided by the concentration of the solute in the solution phase (mass of solute per unit volume of soil pore water). The units for K_d are usually given as mL/g (see Equation 3-1).

$$K_d = \left(\frac{C_0}{C} - 1 \right) \frac{V}{M} \qquad \text{Equation 3-1}$$

where C_0 is tracer concentration in the solution before adding the soil, C is the solute concentration in the liquid phase of a soil water suspension, V is the volume of water, and M is the mass of soil.

Thus, K_d is a factor related to the partitioning of a solute (or contaminant) between the solid and the aqueous phases.

3.2   Assumptions and limitations

The sorption from the solid to the aqueous phase can be defined as equilibrium-partitioning process with a linear isotherm at low solute concentrations (e.g. either $\leq 10^{-5}$ molar, or less than half the solubility, whichever is lower (U.S. EPA, 1999a)).

The derivation of K_d values from thermodynamic data is based on some assumptions:

- the adsorption of the solute is fully reversible (adsorption rate = desorption rate)

- the reaction is independent of the concentration of the solute

- pH and temperature are fixed

- there is only one type of dissolved species

- there is only one type of adsorption site

K_d values which can be used in modeling transport of contaminants can also be derived from field data and observations. These field empiricised K_d values should be

used, if available, with a critical review of the soil specific parameters which influence sorption processes. A method for estimating K$_d$ values is described in chapter 1.6.

Using K$_d$ values ignores the knowledge of the complex adsorption process itself. More advanced models of the surface complexation in a soil / solution system, such as the so-called double or triple layer models, are based on electrostatic attraction between the metal in solution and the particle surface. These theories have been applied successfully to ideal laboratory systems (Albrecht, 1998).

Crawford et al. (2006) summarizes uncertainties in the acquisition and use of K$_d$ data at different levels:

- Uncertainty in the recommended K$_d$ data itself
  This includes sources of random error, mineralogical variability of rock samples, subtle differences in water chemistry, methodological flaws in measurement and interpretation, documentation flaws, as well as sources of systematic bias ("frame shift" and "frame dilation").

- Uncertainty related to the use of generic data in site-specific safety assessment (SA)
  Application conditions may not exactly match the conditions under which the experimental data have been obtained. This could potentially include both differences in water chemistry as well as the use of generic data for rock types where site-specific data is unavailable.

- Uncertainty in the application conditions
  It is not possible to know the actual flow paths and rock types that a water package encounters while flowing through fractured rock and consequently, the material properties averaged over a representative volume (block) is uncertain. In addition, the future state of relevant geochemical parameters may not be accurately known owing to transient flow effects that have not been well characterized.

However, more complex models addressing these uncertainties are often not applicable due to the difficulties in getting reliable values for relevant parameters, e.g. surface complexation constants. Therefore the simplifying K$_d$ concept is still in use if the transport of inorganic contaminants has to be modeled.

3.3   Methods for measuring K$_d$

<u>Laboratory batch studies</u> are most common for determining K$_d$ values. The principle is adding a known volume of an aqueous contaminant solution at known concentration to a soil of known mass. After mixing and completed sorption the remaining concentration of the contaminant in the solution is measured.

In-situ K$_d$ values can be gained out of core samples from an aquifer of a specific site. The aqueous phase is separated from the solid phase and then analyzed for the solute concentration. The solid is also analyzed for the contaminant(s) of concern. This <u>in-situ batch method</u> (U.S. EPA, 1999a) results in more site specific K$_d$ values.

A column of packed soil can be infiltrated by a solution containing known concentration of the contaminant. This <u>laboratory column (flow-through) method</u> produces data of concentrations of a contaminant in the effluent of the column as a function of time. The analysis of the residence time of the contaminant followed by the calculation of a retardation factor results in a calculated soil specific K$_d$ value.

Site specific K$_d$ values can be derived by the use of transport models and existing groundwater monitoring data for the contaminant of concern (<u>field modeling method</u>).

3.4   Methods for estimating K$_d$

Even among soils with greatly different characteristics a significant relationship among retention parameters (e.g. K$_d$) and soil and element properties exist (Buchter et al., 1989). If this relationship can be quantified, an estimation of K$_d$ is possible.

$$\ln K_d = \left[4.62 + stex - 0.56(\ln CR)\right]$$ Equation 3-2

where,

if soil = sand (≥70 % sand-sized particles)	⇒ stex = -2.51
if soil = loam (even distribution of sand, silt, clay; ≤ 80 % silt sized particles)	⇒ stex = -1.26
if soil = clay (≥ 35 % clay-sized particles)	⇒ stex = -0.84
if soil = organic (≥ 30 organic matter)	⇒ stex = 0

Baes (1984) developed a method to predict K$_d$ values based on the bioavailability. The so-called *"soil-to-plant concentration ratio"* (CR, Table 3-1), which is an indicator for

the bioavailability, shows a strong negative correlation to K_d values (Equation 3-2, Thibault et al. (1990)).

Table 3-1:   Concentration ratio (CR) values (wet weight basis) used for estimation of K_d values

Element	CR	Element	CR
Ag	$1,0 * 10^{-1}$	Hg	$2,3 * 10^{-1}$
Au	$1,0 * 10^{-1}$	Mn	$6.3 * 10^{-2}$
Be	$2.5 * 10^{-4}$	Na	$1,9 * 10^{-2}$
C	$1.4 * 10^{-0}$	P	$8.7 * 10^{-1}$
Ca	$8.8 * 10^{-1}$	S	$3,8 * 10^{-1}$
Co	$5.0 * 10^{-3}$	Sc	$1,5 * 10^{-3}$
Cr	$1.9 * 10^{-3}$	Si	$8.8 * 10^{-2}$
Cu	$1,0 * 10^{-1}$	Sr	$6,3 * 10^{-1}$
Eu	$2,5 * 10^{-3}$	Ti	$1,4 * 10^{-3}$
Fe	$1.0 * 10^{-3}$	V	$1,4 * 10^{-3}$
H	$1.2 * 10^{-0}$		

CR values derived from Baes et al. (1984) divided by 4
to get CR on a wet weight basis (Thibault et al., 1990)

An estimation of K_d values can also be based on a study of the specific literature. If the K_d values found for a specific contaminant can be related to soil types they can be used for obtaining a bandwidth for K_d. Preferable are K_d values which are based on monitored or calculated migration in the field (see chapter 1.5).

3.5   Variations of K_d values

The partition coefficient K_d for a specific contaminant can extend over a very wide range even for a single type of soil. Therefore the determination of K_d values has major importance for modeling and has to take the soil mechanical and mineralogical parameters for a layer of a computer model into account.

Deriving K_d values from on-site data will result in a bandwidth of realistic values. These variations of K_d with respect to a specific contaminant are based on soil specific parameters, hydraulic conditions, physical and chemical properties of the groundwater and the applied monitoring procedures or calculations. During the study of such data from literature these issues have to be addressed and should be evaluated carefully before using the respective K_d for a planned modeling process.

Some reports are just citing K_d collected from many sources including estimated values found in numerous publications (chapter 1.6) to give an overview about K_d values relevant for specific modeling purposes, e. g. U.S EPA (1999b, 2004, 2005), IAEA (1994).

3.6 K_d in computer modeling

The solute migration in soil and unconsolidated geological material can be described and predicted by computer models. These models are widely used and are more or less complex depending on the purpose of the user and the hardware/software available.

The model must predict the time of arrival of a contaminant at the appropriate location and the magnitude of the concentration. The location has to be defined by the user of the model, whereas migration time and magnitude/concentration are influenced by the specific K_d values.

A partial differential equations for solute transport (Istok, 1989) used in computer models is shown in Equation 3-3.

$$\partial(\Theta C) = D_x \frac{\partial^2}{\partial x^2}(\Theta C) + D_y \frac{\partial^2}{\partial y^2}(\Theta C) + D_z \frac{\partial^2}{\partial z^2}(\Theta C)$$
$$- \frac{\partial}{\partial x}(v_x C) - \frac{\partial}{\partial t}(\rho_b K_d C) - \lambda(\Theta C + \rho_b K_d C)$$

Equation 3-3

where C is solute concentration, D_x, D_y, D_z are dispersion coefficients, Θ is the volumetric water content, v_x is apparent groundwater velocity in the x coordinate direction, ρ is bulk density, K_d is the equilibrium partition coefficient for a particular sorption/desorption reaction involving the solute and the porous media, and λ is the solute decay coefficient.

Diffusion processes have no significant influence even at very low hydraulic conductivities (k_f in the order of 10^{-9} to 10^{-10} m/s) at difference in concentration of 2.200 mg/l for common cations like Na^+, K^+, Ca^{2+} (Heuel, 1991). Moreover, the relevance of diffusion driven migration will decrease at lower differences in concentration and greater permeability.

In absence of K_d values based on site-specific observations K_d values from the literature can be selected for groundwater modeling. These values should come from observations or calculations under similar conditions as existing at the site of concern.

The influence of solid and aqueous phase components (e.g. organic matter, coating of soil particles, dissolved carbonate, clay concentrations, Eh) on the sorption of a specific contaminant should be estimated if these data are available before using K$_d$ values from the literature in transport models.

4 Partition coefficients K$_d$ of selected elements in groundwater

4.1 Background

The partition coefficient of a specific element varies e.g. with respect to the soil type, the pH of the groundwater and the concentrations of other elements.

The following subchapters are partly referring to a publication by Thibault et al. (1990) and Sheppard et al. (1990). This publication divided the different soil textures in 4 groups; a way to handle the great variability of soil textures, mineralogical composition and chemical constituents by reducing characterizing parameters to particle size distribution and organic content:

- sand soils: ≥ 70 % sand sized particles

- clay soils: ≥ 35 % clay-sized

- loam: ≤ 80 % silt-sized particle or even distribution of sand-, clay- and silt-sized particles

- organic soil: > 30 % organic matter

This grouping is the most practicable way of addressing soil specific parameters which influence the specific K$_d$ because many publications of empiricised K$_d$ values refer to this major soil types.

However, K$_d$ values depend on more parameters than just the grain size distribution. Thus, the following subchapters will also include a short description of some geochemical aspects of the behavior of the specific contaminant which can be used for a further interpretation of the K$_d$ values or for adjusting the K$_d$ values to specific site conditions.

Thibault et al (1990) and Sheppard et al. (1990) compiled their data from literature. Where no data exists in the literature they used the soil-to-plant concentration ratio to predict K$_d$.

4.2 Geochemical aspects

As described above radioisotopes can enter an aquifer after the process of activation in the shield (concrete, soil etc.) or surrounding soil/rock of an accelerator assuming

that these elements can be mobilized by infiltrating surface water or by direct contact of shield and groundwater.

To give at least a first impression of the geochemical behaviour of a specific element or its species after entering an aquifer some Eh-pH-diagrams are given in the following. Eh-pH-diagrams visualize the influence of the reduction potential (Eh) and the pH on the stability of a specific mineral, soluble species and gas in an aqueous electrochemical system such as groundwater. A predominant ion boundary is represented by a line. Most lines are straight and are functions of pH alone (vertical lines), Eh alone (horizontal lines) or both (sloped lines). Details about the background and the use of Eh-pH diagrams are given by Brookins (1988) and Takeno (2005).

4.3 Beryllium

Beryllium occurs in aquatic systems usually as Be^{2+} and solid BeO (Bromellite) strongly depending on pH, whereas BeO dissolves under acidic to near neutral condition to form Be^{2+} (Figure 4-1).
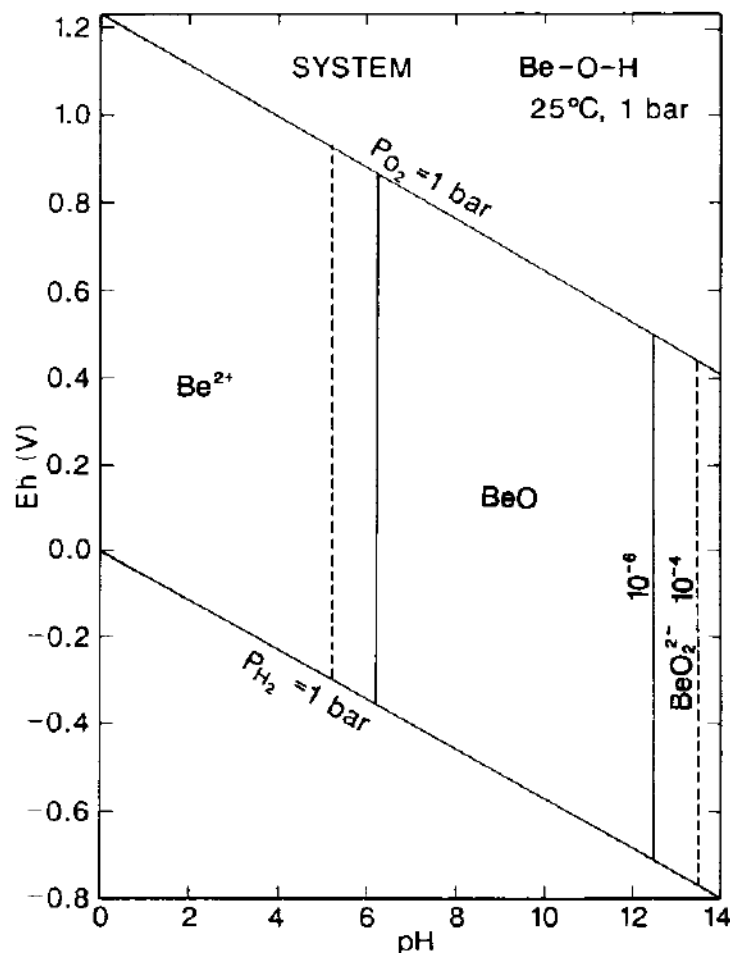
Figure 4-1:   Eh-pH diagrams of the system Be-O-H. Assumed activities for dissolved Be=10$^{-6, -4}$ (Brookins, 1988)

Yet, beryllium is known to be quite insoluble in nature (Brookins, 1988). Therefore Be is expected to have relatively high K$_d$ values.

Empiric K$_d$ values were not found in the cited literature (chapter 7). Thus an estimation is made (see chapter 1.6) based on the bioavailability of Be resulting in values given in Table 4-1.

Table 4-1:   Beryllium - K$_d$ values based on concentration ratio (CR) values derived from Baes et al. (1984)

soil texture	sand	silt	clay	organic
K$_d$ [L/kg]	250	800	1.300	3.000

derived from Baes et al. (1984) by using the soil characterization described above

4.4 Calcium

In general the stability fields of calcium species depend on pH and are not sensitive to the redox potential (Figure 4-2).
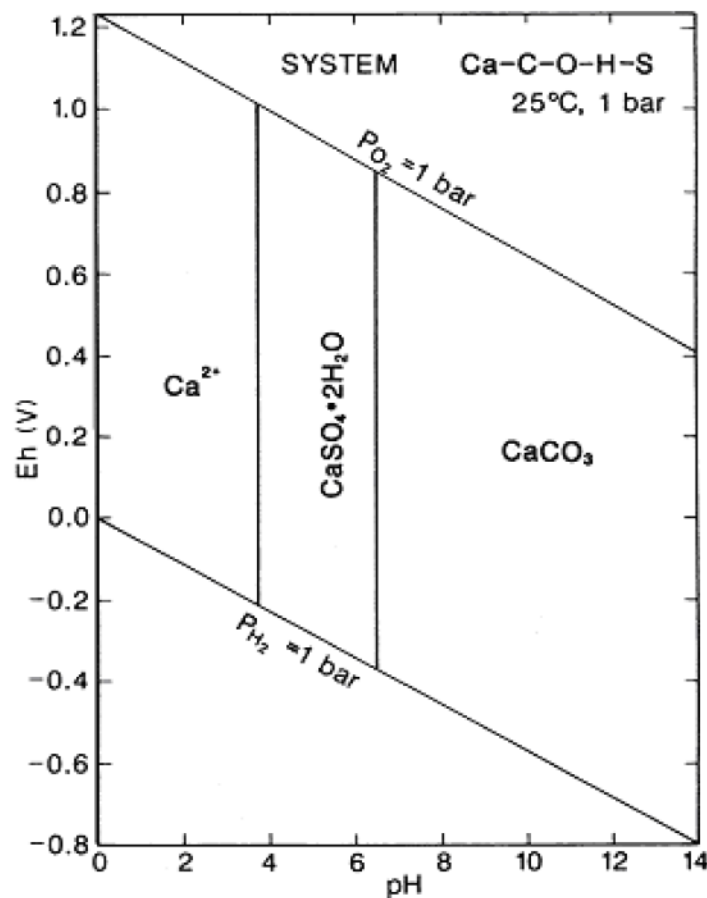
Figure 4-2:   Eh-pH diagrams of the system Ca-C-O-H-S. Assumed activities for dissolved species Ca=10$^{-2.5}$, S=10^{-3}, C=10^{-3} (Brookins, 1988)

Most common species are calcite ($CaCO_3$) and gypsum ($Ca\,SO_4 \cdot 2H_2O$) which can be formed due to water incorporation out of anhydrite ($CaSO_4$).

Anhydrite can only be stable with respect to gypsum under specific conditions (excess of sodium or potassium chloride). Usually it is altered to gypsum by absorption of water.

Table 4-2:   Calcium - K_d values based on concentration ratio (CR) values (Sheppard, M. I. and Thibault, D. H., 1990).

soil texture	sand	silt	clay	Organic
K_d [L/kg]	5	30	50	90

4.5 Carbon

In natural environments carbon exists in numerous species.
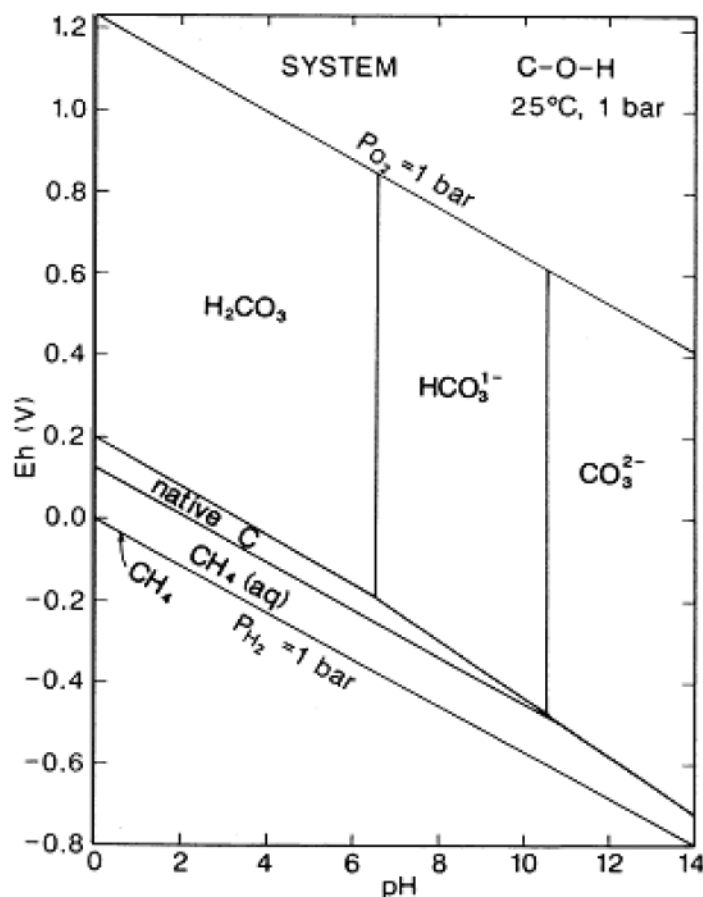
Figure 4-3:   Eh-pH diagrams of the system C-O-H. Assumed activities for dissolved $C = 10^{-3}$ (Brookins, 1988)

However, under oxidizing conditions H_2CO_3 (carbonic acid), HCO_3^{-1} and CO_3^{2-} (with increasing pH) are mostly the dominant species in common groundwater depending on the pH value as shown in an Eh-pH-diagram (Figure 4-3).

Sorption of C is generally low, but appeared to increase with increasing calcium contend of the solid (Allard et al., 1981).

Due to the lack of information with respect to K_d values of carbon a conservative retardation factor or a K_d of 0 L/kg is recommended by Sheppard et al. (1984). However, Thibault et al. (1990) showed K_d values based on concentration ratio (CR) values and from literature (Table 4-3).

Table 4-3:   Carbon - K_d values based on concentration ratio (CR) values for silt and clay and from literature for sand (Thibault et al., 1990).

soil texture	sand	silt	clay	Organic
K_d [L/kg]	5	20	1	70

4.6 Chlorine

Chlorides (e.g. halite (NaCL) or sylvite (KCl)) easily dissociates in groundwater to form chloride (Cl$^-$). Cl$^-$ is the stable species of chlorine in Eh-pH diagrams within the stability field of groundwater (Figure 4-4).

Chloride as anion has a negative charge. Most silicate surfaces onto which chloride could adsorb are negative. Due to their own negative charge, chloride ions do not adsorb onto these silicate surfaces and therefore move at approximately the same rate as the groundwater (Bentley, H.W. et al., 1986).

However, Sheppard et al. (2006) stated that even a very modest degree of sorption in soil could markedly change the dose estimates, e.g. a very low K_d of 0.1 L/kg could result in a near doubling of the steady-state soil ^{36}Cl concentrations. Therefore, a retardation of chloride ions in a groundwater system can be assumed at a very low level.

Sheppard et al. (1996) and Zach et al. (1996) derived K_d values for Cl from a regression across many elements, with plant/soil concentration ratios as the independent variable (Table 4-4).

Figure 4-4:   Eh-pH diagrams of the system Cl-O-H. Σ Cl = 10^{-10}, 298.15 K, 10^5 Pa
                (Takeno, 2005)

Table 4-4:   Chlorine - K$_d$ values (Sheppard et al. (1996), Zach et al. (1996)).

soil texture	sand	silt	clay	Organic
K$_d$ [L/kg]	0.8	0.25	4.4	11

4.7 Chromium

The adsorption behavior of chromium is influenced by a variety of chemical and phys-
ico-chemical factors (Brookins (1988), U.S. EPA (1999b)). These factors can be sum-
marized as follows:

- Concentrations of Cr(III) in soil solutions are typically controlled by dissolution/
  precipitation reactions; therefore, adsorption reactions are not significant in soil
  Cr(III) chemistry.

- Above pH 5 chromium is present in its stable form Cr_2O_3 (or incorporated into
  chromites and other chromian spinels).

- pH values above 13.5, where CrO_2^- is the dominant chromium species (Figure 4-5), is usually not relevant for common types of soil and groundwater.

- Increased pH decreases adsorption (decrease in K_d) of Cr(IV) on minerals and soils.

- E_h of the soil affects chromium adsorption. Cr(III) oxidizes to form Cr(VI) as $HCrO^{4-}$ and CrO_4^{2-} ions at high Eh. Ferrous iron associated with iron oxide/hydroxide minerals can reduce Cr(VI) which results in precipitation (higher K_d). Lower K_d values can be determined if Mn oxides are present resulting in an oxidation of Cr(III) into Cr(VI).

- The presence of competing anions (e.g. HPO_4^{2-}, $H_2PO_4^-$ >> SO_4^{2-}, CO_3^-, Cl^-, NO_3^-) reduce Cr(IV) adsorption.

- Cr(VI) is readily be reduced to Cr(III) by soil organic matter (Kephalopoulos et al, 1988).
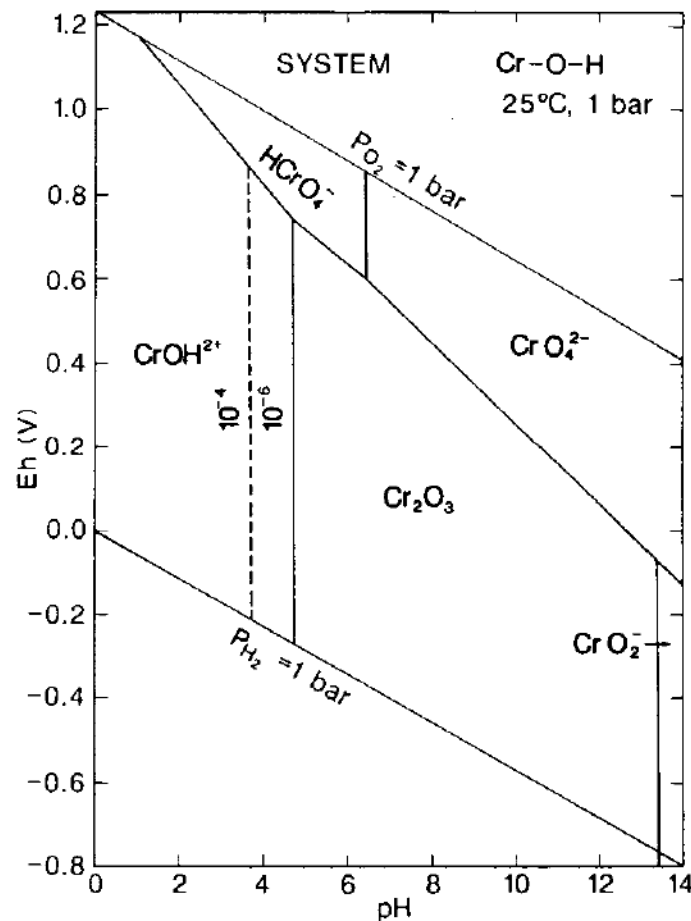
Figure 4-5: Eh-pH diagrams of the system Cr-O-H. Assumed activities of dissolved Cr $=10^{-6}$ (Brookins, 1988)

These data are corresponding to Kephalopoulos et al. (1988) with K_d values from column experiments for sand (17 L/kg), sandy soil (24 L/kg), sandy loam (123 L/kg).

Table 4-5:   Chromium(IV) - K_d values (Sheppard, M. I. and Thibault, D. H., 1990).

soil texture	sand	silt	clay	organic
K_d [L/kg]	70	30	1.500	270

4.8 Cobalt

The mobility of Co is mostly limited by adsorption and coprecipitation reactions with manganese and iron oxide minerals. Under normal geochemical, conditions Co(II) - not Co(III) - is the dominant co-oxidation state (see Eh-pH-diagram, Figure 4-6). Uncomplexed Co^{2+} dominates under oxidizing condition below pH 8 to 9. Because Co readily adsorbs to minerals and has a short half-life relative to those of other radionuclides of environmental concern, the mobility of Co should be limited in soil pore (Krupka et al., 2002).
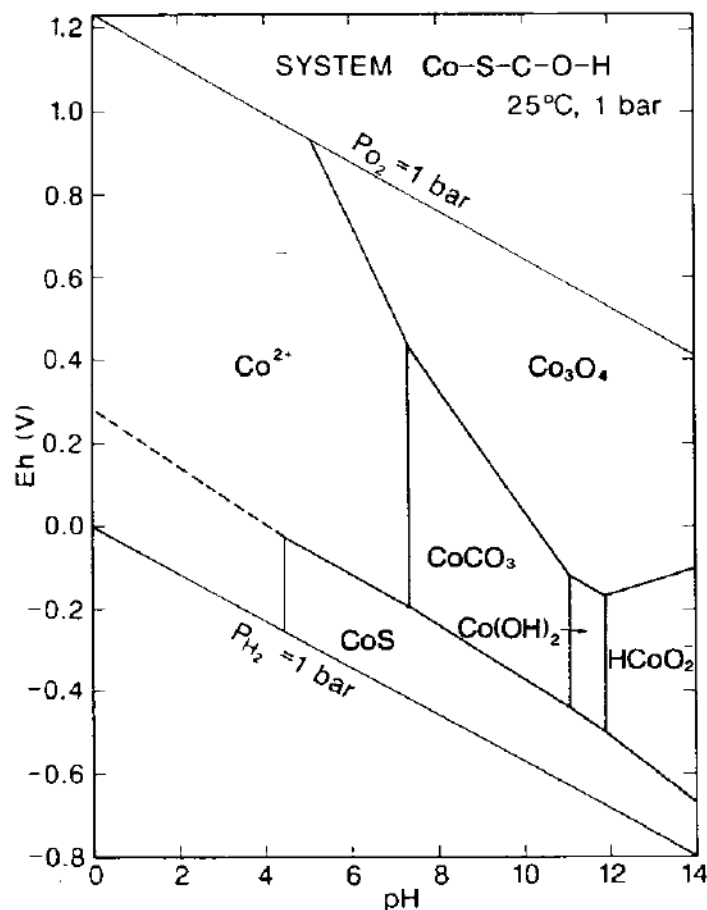
Figure 4-6:   Eh-pH diagrams for part of the system Co-S-O-H. Assumed activities for dissolved species are: Co=10^{-6}, C=10^{-3}, S=10^{-3} (Brookins, 1988)

Table 4-6: Cobald - K_d values (Sheppard, M. I. and Thibault, D. H., 1990).

soil texture	sand	silt	clay	Organic
K_d [L/kg]	60	1.300	550	1.000

4.9 Copper

In general, the mobility of copper increases with decreasing pH. Depending on the presence of other ions solid copper species are formed under reducing conditions and alkaline pH (e.g. native copper, CuO/Cu_2O, CuS/Cu_2S).

If C is present cupric carbonates, mostly important malachite ($Cu_2(OH)_2CO_3$), can be formed at medium and higher pH replacing CuO (Brookins, 1988).
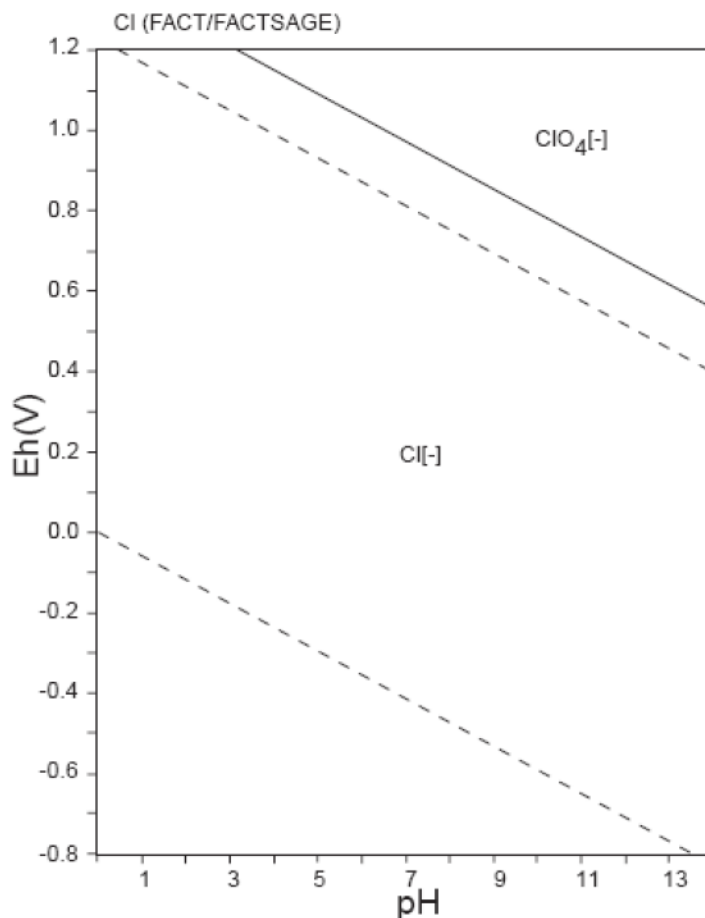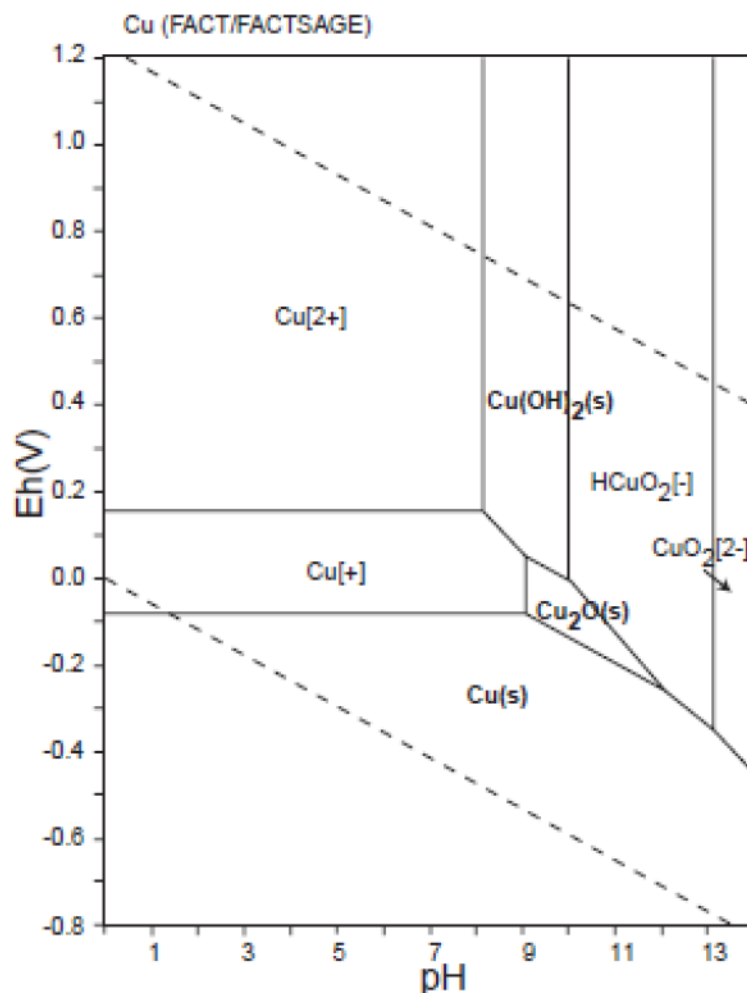

Figure 4-7: Eh-pH diagrams of the system Cu-O-H. Σ Cu = 10^{-10}, 298.15 K, 10^5 Pa (Takeno, 2005)

25

Table 4-7:　　Copper - K$_d$ values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K$_d$ [L/kg]	30	105	159	369

derived by Equation 3-2 using the soil characterization described above

4.10 Europium

Europium is one of the rare earth elements (REE). Eu^{3+} is the most stable oxidation state. The abundance of different Eu^{3+}-species strongly depends on the pH of an aquatic system. Eu^{3+} behaves similar to other rare earth elements (e.g. Ce, Y, Lu, Sm, Tm) with a 3+ oxidation state. The dominant species at acidic pH values in a ground-water system is the uncomplexed ion Eu^{3+}. At medium and alkaline pH Eu^{3+} tends to form stable complexes with carbonate, hydroxide, sulfate etc. (Krupka et al., 2002).
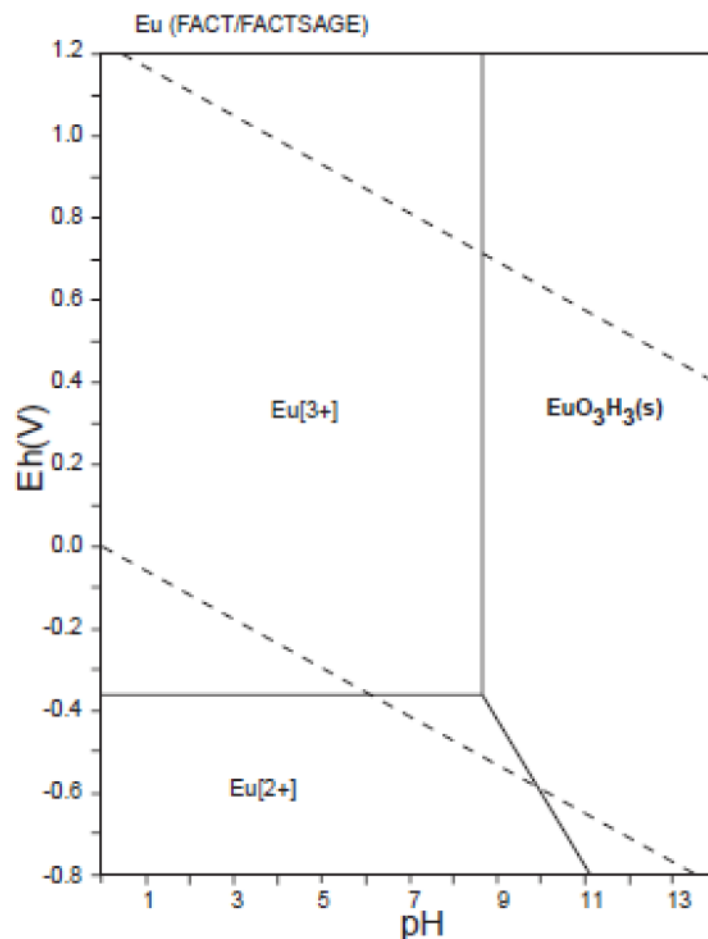
Figure 4-8:　　Eh-pH diagrams of the system Eu-O-H. Σ Eu = 10^{-10}, 298.15 K, 10^5 Pa (Takeno, 2005)

Trivalent elements are considered to be highly sorbed in sediments - i.e., exhibit high K_d values - and thus immobile in most environments (EPA, 2004). Eu(III) is considered very insoluble in environmental systems. Its low solubility may be a contributing factor to the large partition coefficient K_d.

The existence of anionic hydroxide and carbonate species at alkaline pH values should result in increased mobility of Eu(III) in the geochemical environment. Such mobility however is inconsistent with the observed sorption behaviour of europium (Krupka et al., 2002).

Unlike most other rare earth elements Europium can form an (II+) species at extremely reducing conditions and a medium pH (Brookins, 1988). The formed Eu^{2+} is mobile in contrast to trivalent rare earth elements.

Table 4-8:   Europium - K_d values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K_d [L/kg]	236	825	1.255	2.908

derived by Equation 3-2 using the soil characterization described above

4.11 Gold

Native Au is the dominant species under most conditions in natural groundwater (Figure 4-9). Only in very acidic and oxidizing environments Ag can be mobilized e.g. as $AuCl_2^-$.

Table 4-9:   Gold - K_d values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K_d [L/kg]	30	105	159	369

derived by Equation 3-2 using the soil characterization described above
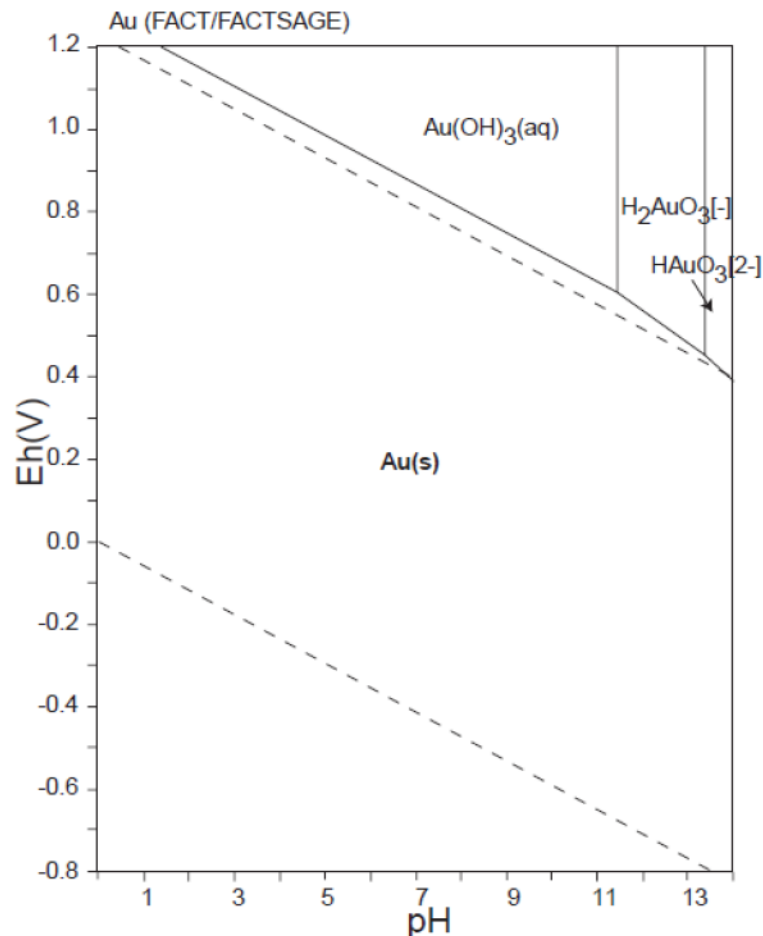
Figure 4-9:   Eh-pH diagrams of the system Au-O-H. Σ Au = 10^{-10}, 298.15 K, 10^5 Pa
(Takeno, 2005)

4.12 Hydrogen/Tritium

Tritium (^{3}H or T), being a radioactive isotope of hydrogen, usually migrates as part of the compound water at the same velocity as groundwater. Due to its half-life of 12.3 years tritium is used for age determination of near surface groundwater and for studies related to hydrological flow conditions.

Tritium behaves hydrologically like hydrogen (^{1}H) and therefore exists in ionic, gaseous and liquid forms. Because free tritium is oxidizing rapidly to form water (^{3}H$_2$O or ^{3}HHO) its migration and mobility is practically independent of any precipitation or sorption processes. For groundwater modeling the migration of tritium is just depending on the input concentration, the velocity of the groundwater flow and the half-life of tritium.

Due to this independence, K$_d$ values for tritium related to specific soil types are given only for comparison in this report. Using concentration ratio (CR) values derived by

Equation 3-2 K_d values for tritium can be calculated as shown in Table 4-10. If a specification is necessary for calculation or modeling, the K_d value for tritium is 0.

Table 4-10:   Hydrogen/Tritium - K_d values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K_d [L/kg]	7	26	40	92

derived by Equation 3-2 using the soil characterization described above

4.13 Iron

The dominant iron species in many natural groundwaters are Hematite (Fe_2O_3) and Fe^{2+} whereas Hematite is stable at higher pH and Eh and Fe^{2+} at lower pH and Eh (Figure 4-10). Depending on the concentration of (additional) elements/ions, such as C or Si additional iron species (e.g. $FeCO_3$ (C; medium to high pH), $Fe(OH)_3$ (absence of S), Fe_3O_4, $FeSiO_3$ (additional Si), $Fe·OH$) can be dominant under specific redox condition and pH.

Figure 4-10:   Eh-pH diagrams of the system Fe-O-H. Σ Fe = 10^{-10}, 298.15 K, 10^5 Pa (Takeno, 2005)

Table 4-11:   Iron - K$_d$ values (Sheppard, M. I. and Thibault, D. H., 1990).

soil texture	sand	silt	clay	organic
K$_d$ [L/kg]	220	800	165	600

4.14 Manganese

The dominant species of manganese at acidic to medium conditions up to a pH of 10 (depending on the redox-potential and e.g. the occurrence of S and C) is Mg^{2+}. Above this boundary manganese (hydro)oxides (MgO$_2$, MgO, Mn$_3$O$_4$, Mn(OH)$_2$ etc.) and also carbonate (MnCO$_3$) can occur.

Table 4-12:   Manganese - K$_d$ values (Sheppard, M. I. and Thibault, D. H., 1990).

soil texture	sand	silt	clay	organic
K$_d$ [L/kg]	50	750	180	150
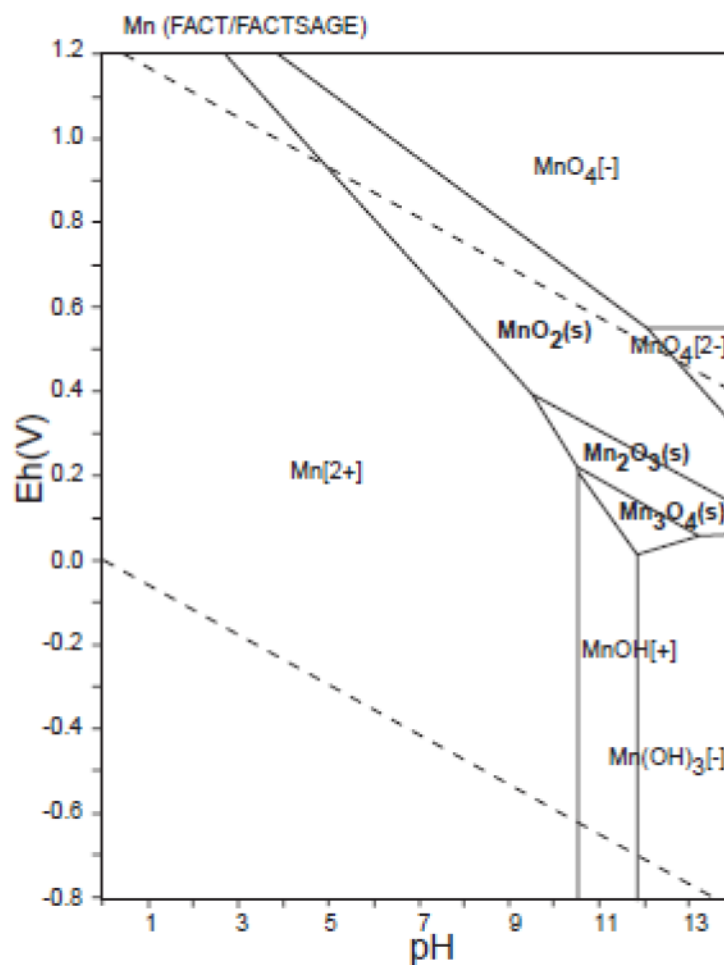
Figure 4-11:   Eh-pH diagrams of the system Mn-O-H. Σ Mn = 10^{-10}, 298.15 K, 10^5 Pa (Takeno, 2005)

4.15 Mercury

Under a broad range of pH- and redox-condition (neural to higher pH, medium redox potential) in groundwater Mercury is stable as native Hg. HgO forms between pH 5.4 and 10,8 at higher redox (Brookins, 1988). Between pH 5.5 and 2.5 K$_D$ values for Hg increases.

Chloride ions are playing an important role with respect to the formation of Hg species. In presence of Cl HgCl$^{2-}_4$ and Hg$_2$Cl$_2$ are the dominant species at lower pH and higher redox.

In general organic matter binding Hg plays the dominant role in controlling Hg sorption onto soil (Lee et al., 2001).

Figure 4-12: Eh-pH diagrams of the system Hg-O-H. Σ Hg = 10^{-10}, 298.15 K, 10^5 Pa (Takeno, 2005)

Table 4-13: Mercury - K_d values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K_d [L/kg]	19	66	101	234

derived by Equation 3-2 using the soil characterization described above

A K_d value of 52 L/kg is given by the Alaska Department of Environmental Conservation (1999) without further specifying soil type or particle size distribution. This value corresponds to the values in Table 4-13 for sand/silt sized particles.

4.16 Phosphorus

Phosphorus in groundwater exists mainly as phosphoric acid (H_3PO_4) at low pH and the related dissociation products ($H_2PO_4^-$ (medium low to medium pH), HPO_4^{2-} (medium to medium high pH), PO_4^{3-} (very high pH)). Metal phosphates and their ion species can occur commonly depending on the concentration of a specific metal cation and the general groundwater conditions (Eh, pH etc.).
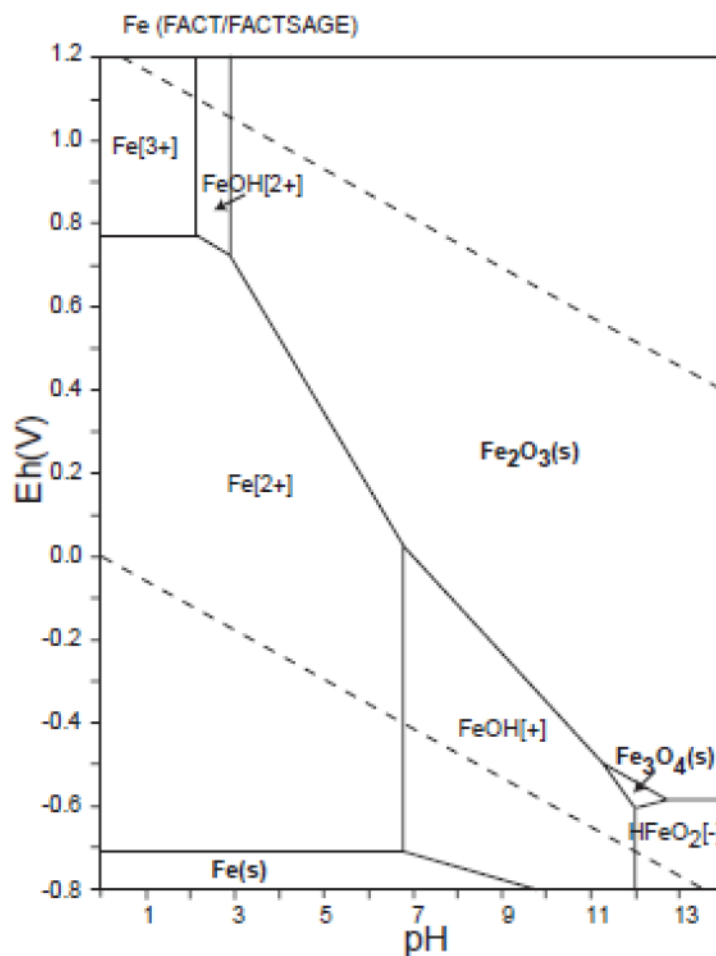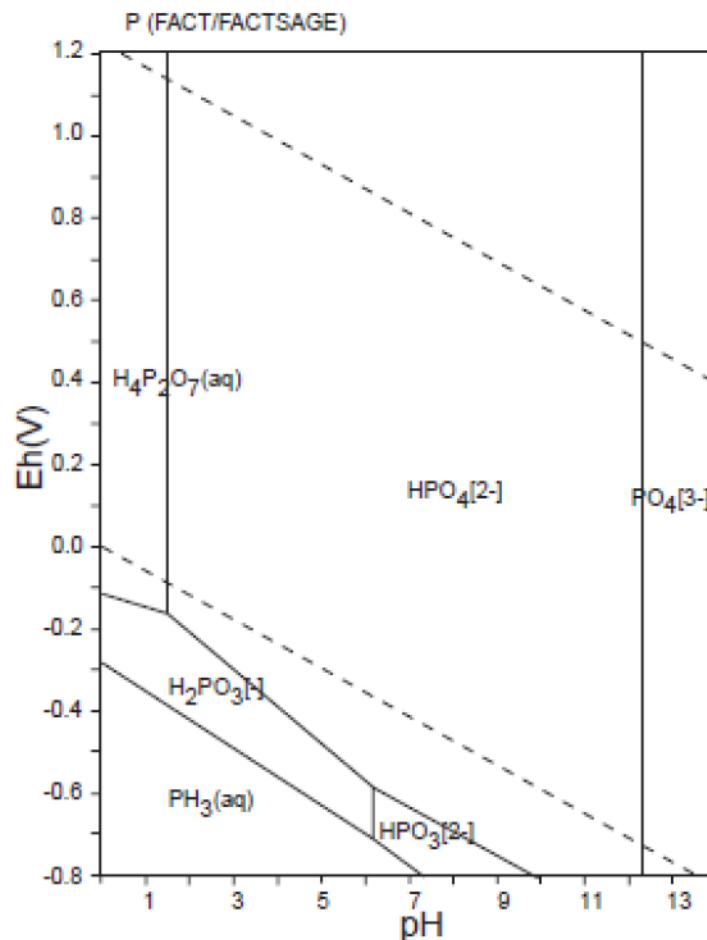

Figure 4-13: Eh-pH diagrams of the system P-O-H. $\Sigma\ P = 10^{-10}$, 298.15 K, 10^5 Pa (Takeno, 2005)

Table 4-14:   Phosphorus - K$_d$ values based on concentration ratio (CR) values (Sheppard, M. I. and Thibault, D. H., 1990).

soil texture	sand	Silt	clay	organic
K$_d$ [L/kg]	5	25	35	90

4.17 Scandium

Scandium belonging to the rare-earth metals often follows iron (Fe^{3+}) in natural occurring minerals such as hematite, pyroxene or muscovite. There is a different chemical behavior of scandium during oxidation-reduction processes where Mn is present resulting in Fe-Mn nodules. These nodules show typically a depletion of Sc relative to Fe (Aide, 2009).

The solubility of Sc depends on the pH of the groundwater; there is no Eh dependence. Between the pH range of approx. 4 to 10/11 ScOH^{2+} is the acidic species, whereas Sc(OH)$_3$ is the dominant species at higher pH.

Table 4-15:   Scandium - K$_d$ values based on concentration ratio (CR) values

soil texture	sand	Silt	clay	organic
K$_d$ [L/kg]	315	1.098	1.671	3.871

derived by Equation 3-2 using the soil characterization described above

Sc$_2$O$_3$ is stable like hematite (Fe$_2$O$_3$, see chap. 1.21) with respect to pH and redox potential. Sc forms no sulfites in natural groundwater.

4.18 Silicon

Only one common silicon species exists under natural conditions: silica (SiO$_2$) which is extremely insoluble. Only under high pH conditions (above pH 12) silica starts to dissolve to form H$_3$SiO$_4^-$. Therefore, K$_d$ values for Si in natural groundwater are higher than for more soluble compounds.

Table 4-16:   Silicon - K$_d$ values based on concentration ratio (CR) values (Sheppard, M. I. and Thibault, D. H., 1990).

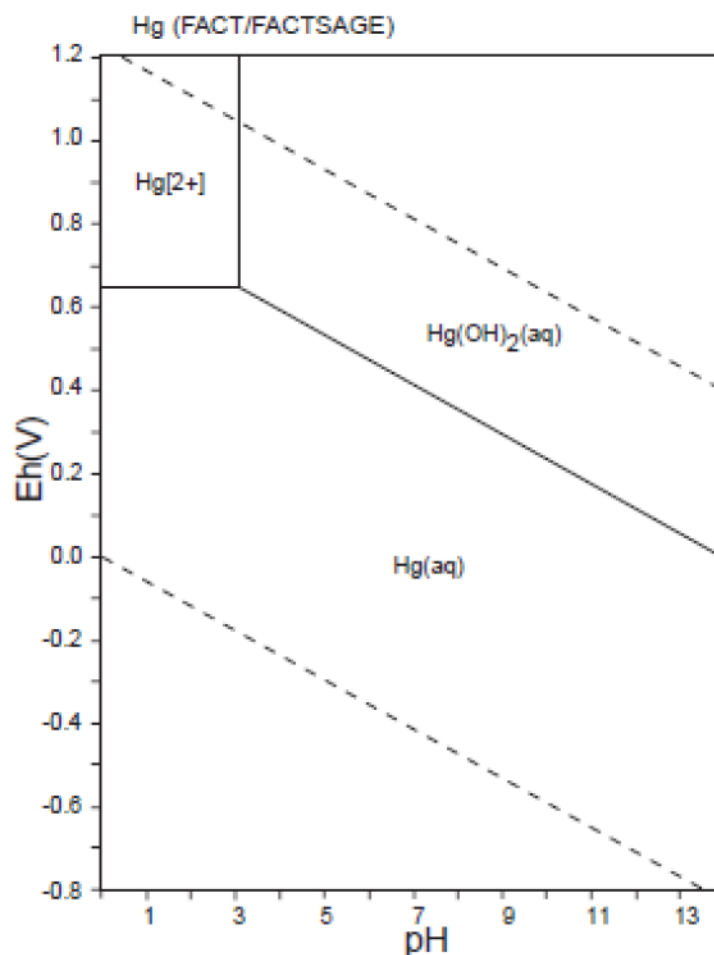| soil texture	sand	silt	clay	Organic
K$_d$ [L/kg]	35	110	180	400
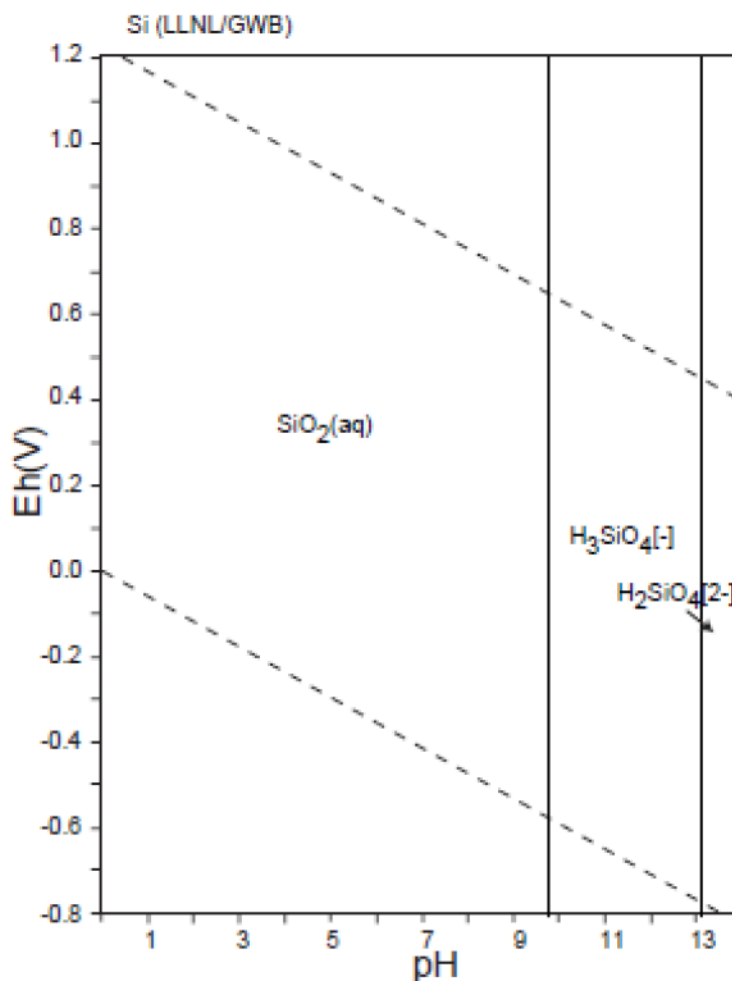
Figure 4-14:   Eh-pH diagrams of the system Si-O-H. Σ Si = 10^{-10}, 298.15 K, 10^5 Pa
(Takeno, 2005)

4.19 Silver

Under most natural conditions Ag occurs preferable as native Ag (Figure 4-15). If sulfur is present, Ag forms Ag_2S at low Eh (reducing conditions). Under strongly oxidizing conditions Ag^+ or $Ag(OH)_2^-$ is dominating or, if Cl is present, $AgCl_2^-$ replaces Ag^+. Thus, dissolved Cl plays a major part on the transport of Ag under oxidizing, acidic conditions (Brookins, 1988).

Table 4-17:   Silver - K_d values (Sheppard, M. I. and Thibault, D. H., 1990).

soil texture	sand	silt	clay	organic
K_d [L/kg]	90	120	180	15.000
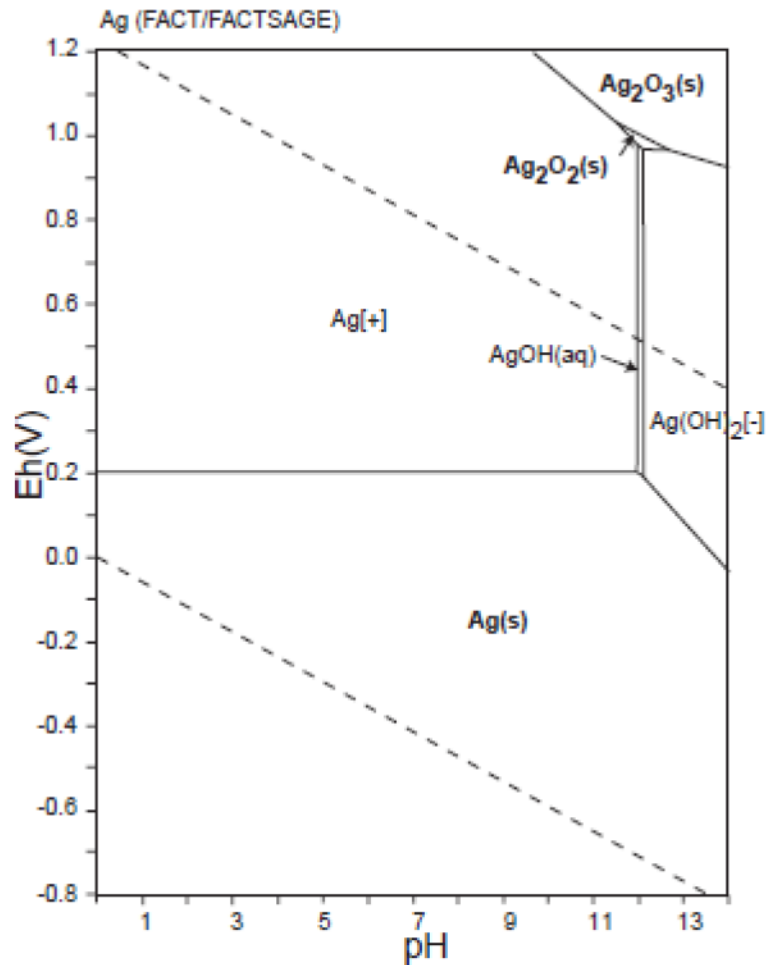
Figure 4-15: Eh-pH diagrams of the system Ag-O-H. Σ Ag = 10^{-10}, 298.15 K, 10^5 Pa (Takeno, 2005)

4.20 Sodium

Unlike calcium, magnesium and silica sodium is not an essential constituent of many common minerals. However, main source of sodium in groundwater is the weathering of sodium bearing minerals, i.e. plagioclase.

Table 4-18: Sodium - K$_d$ values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K$_d$ [L/kg]	76	265	403	934

derived by Equation 3-2 using the soil characterization described above

In general sodium salts are highly soluble and will not precipitate under common concentration in natural ground waters. The only common mechanism of sodium (Na$^+$) removal from groundwater is the ion exchange usually driven by the ion exchange capacity of the clay minerals in a specific aquifer.

4.21 Strontium

Under mildly acidic to basic pH conditions the dominant strontium species are insoluble $SrSO_4$ or $SrCO_3$ (basic pH). The stability of this strontium species is not depending on the Eh of the system as shown in an Eh-pH diagram (Figure 4-16). At intermediate to strongly acidic pH Sr^{2+} can be expected as the dominant species (Brookins, 1988).
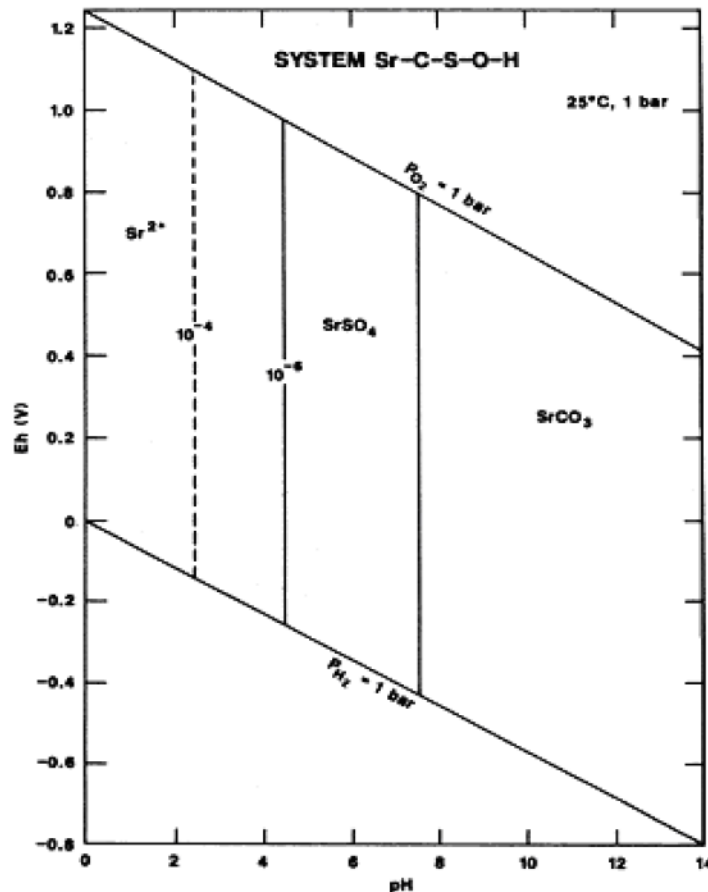
Figure 4-16: Eh-pH diagram for part of the system Sr-C-S-O-H. Assumed activities for dissolved species are: Sr=$10^{-6,-4}$, S=10^{-3}, C=10^{-3} (Brookins, 1988)

Strontium behaves chemically much like calcium. In the presence of calcium ions strontium commonly forms coprecipitates with common calcium minerals such as calcite and anhydrite at an increased ph. In the vadose zone or phreatic aquifer where CO_2 removal occurs (e.g. by weathering) precipitation of calcite is further enhanced (Mitchell et al., 2005). Thus, coprecipitation of strontium can be stimulated additionally.

Dissolved strontium is expected to partition to the solid phase at intermediate to acidic pH by cation exchange (U.S. EPA, 1999b) and not (co)precipitation. Therefore,

the cation exchange capacity (CEC) and also the related clay content of a soil is of major relevance for any modeling of the transport of Sr (see Table 4-19, Table 4-20) at these conditions.

Table 4-19: Strontium - K_d values for different soil textures (Sheppard, M. I., Thibault, D. H., 1990)

soil texture	sand	silt	clay	organic
K_d [L/kg]	15	20	110	150

Table 6-1 which is summarizing all K_d values from the subchapters uses the experienced data from Table 4-20. These values correspond with values of 44 to 86 L/kg for a sand shale used by Malkovsky (2009).

Table 4-20: Strontium - K_d values [L/kg] based on CEC, clay content and pH (U.S. EPA, 1999b)

	CEC [meq/100g] / clay content [wt.%]								
	3 / < 4			3 – 10 / 4 - 20			10 – 50 / 20 - 60		
	pH			pH			pH		
	< 5	5-8	8-10	< 5	5-8	8-10	< 5	5-8	8-10
Minimum	1	2	3	10	15	20	100	200	300
Maximum	40	60	120	150	200	300	1.500	1.600	1.700
mean	21	31	62	80	108	160	800	900	1.000

note: all values for natural soils, low ionic strength (< 0.1 M), low humic material concentration (< 5 mg/l), no organic chelates (e.g. EDTA), and oxidizing conditions

4.22 Sulfur

Most common species of sulfur in natural waters are sulfates. Sources of sulfates in groundwater are sulfate minerals and atmospheric input. Some sediments (e.g. organic shale) containing sulfites (e.g. pyrite) or other anionic sulfur can also contribute sulfate if oxidation processes occur.

A very important boundary in groundwater is the boundary between S(-II) and S(VI) (Figure 4-17). The oxidation of S(-II) species to S(VI) is very energetic with respect to thermodynamic behavior.

Table 4-21:   Sulfur - K_d values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K_d [L/kg]	14	49	75	174

derived by Equation 3-2 using the soil characterization described above
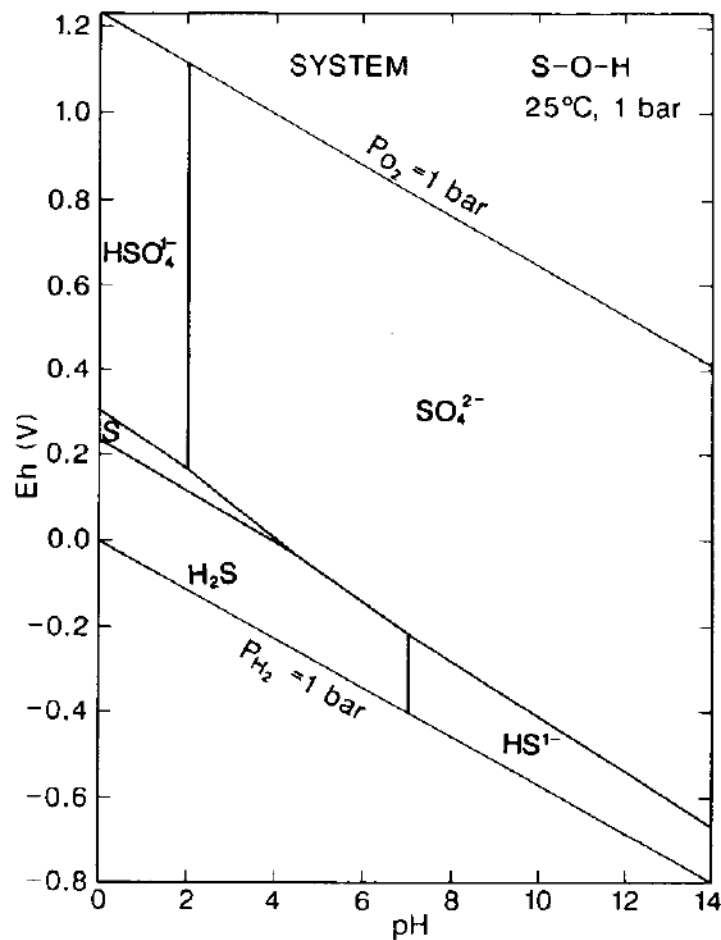
Figure 4-17:   Eh-pH diagram for part of the system S-O-H. Assumed activities for dissolved S=10^{-3} (Brookins, 1988)

4.23 Titanium

Titanium is highly insoluble in natural groundwater forming TiO_2 or $TiO(OH)_2$ (Brookins, 1988) resulting in high K_d values (Table 4-22).

Table 4-22:   Titanium - K_d values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K_d [L/kg]	327	1.141	1.737	4.024

derived by Equation 3-2 using the soil characterization described above

4.24 Vanadium

Most V species (e.g. VO^{2+}, $H_2VO_4^2$, H_2VO_4) are soluble under common groundwater conditions. Exceptions are V_2O_4 und V_2O_3, which are occurring at medium pH and low to medium redox potential. Nevertheless, V (III, IV) is often incorporated into clay minerals (octahedral site) instead of forming separate V-oxides (Brookins, 1988). V-ions are also strongly adsorbed by Al- and Ti-oxides (Bockting et al., 1992).

Table 4-23:   Vanadium - K$_d$ values based on concentration ratio (CR) values

soil texture	sand	silt	clay	organic
K$_d$ [L/kg]	327	1.141	1.737	4.024

derived by Equation 3-2 using the soil characterization described above
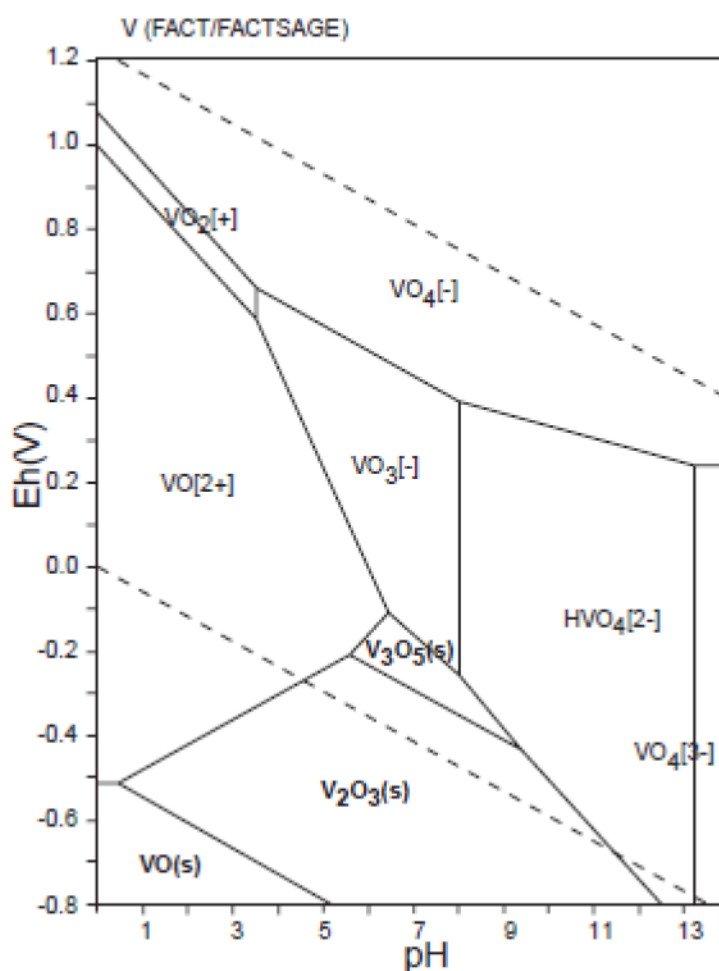
Figure 4-18: Eh-pH diagrams of the system V-O-H. Σ Mn = 10^{-10}, 298.15 K, 10^5 Pa
           (Takeno, 2005)

5   Half-life of relevant radionuclides

Any decay of a radionuclide (parent nuclide) produces a daughter nuclide. Some of these daughter nuclide may be stable where others may be unstable i.e. radioactive. Even though the decay is a random process on an atomic level the decay rate or half-life is predictable if a large number of atoms of a specific radionuclide are present.

By looking at the half-life of mobile radionuclides a second process besides retardation reduces the activity of a radionuclide released at a specific site further downstream. Depending on the half-life of a radionuclide its importance for modeling of the transfer in groundwater can vary strongly. Radionuclides with a half-life of some days or even less, e.g. ^{7}Be, ^{24}Na, ^{32}P/^{33}P and ^{55}Co (Table 5-1), are less important for the calculation of the dose of people in the public because the velocity of groundwater usually is within a range of some centimeters to a few meters a day.

Table 5-1:   Half-life of relevant radionuclides

radionuclide	half-life	radionuclide	half-life
^{3}H	12.32*10^3 d	^{45}Ca	163 d
^{7}Be	53,3 d	^{46}Sc	83.8 d
^{14}C	5.73*10^3 d	^{47}Ca	4.54 d
^{22}Na	949,7 d	^{47}Sc	3.35 d
^{24}Na	14.96 h	^{48}Sc	1.82 d
^{32}Si	3.69*10^4 d	^{48}V	16.0 d
^{32}P	14.3 d	^{49}V	330 d
^{33}P	25.3 d	^{51}Cr	27.7 d
^{35}S	87,5 d	^{52}Mn	5.6 d
^{36}Cl	301*10^3 d	^{54}Mn	312.2 d
^{37}Ar	35,0 d	^{55}Co	17.53 h
^{39}Ar	9.82*10^4 d	^{55}Fe	985.5 d
^{42}Ar	1.20*10^4 d	^{57}Co	271.8 d
^{44m}Sc	2.44 d	^{59}Fe	45.1 d
^{44}Ti	1.73*10^4 d	^{60}Co	5.27 d
		^{90}Sr	29.1 a

dark grey: radionuclide has stable daughter nuclide/s

light grey: radionuclide has radioactive daughter nuclide/s

Thus, there is enough time to significantly reduce the activity of short-living radionu-clides in groundwater by decay until the groundwater leaves the site of an accelera-tor or other radionuclide releasing facilities.

Radioactive daughter nuclides can have an influence on the activity concentration and further more on the effective dose at a specific location. Therefore, it is recom-mended to carefully investigate the possible daughter nuclides of migrating radionu-clides with respect to their K_d, their half-life and their radiotoxicity. Uncritical radio-nuclides with respect to daughter nuclides are e.g. tritium (^{3}H), which e. g. decays to form stable helium, carbon - ^{14}C is changed by beta decay into stable nitrogen (^{14}N) -, ^{36}Cl, which decays by producing mainly stable ^{36}Ar, and ^{45}Ca which daughter nuclide is stable Sc.

Other radionuclides can have instable daughter nuclides, e.g. ^{32}Si, which daughter ^{32}P decays with a half-life of 14,26 days to form stable ^{32}S, or ^{47}Ca which is changed by beta decay into its radioactive daughter ^{47}Sc, which itself decays with a half-life of 3,35 days to form stable ^{47}Ti.

Another example is ^{90}Sr (Table 5-1). ^{90}Sr is changed by beta decay into its radioactive daughter ^{90}Y (half-life 2.7 d) which decays by beta decay into stable ^{90}Zr.

Charts of the nuclides provide information about the kind of decay and the daughter nuclides of radionuclides. Sources for a chart of nuclides:

- Interactive Chart of Nuclides of Brookhaven National Laboratory: http://www.nndc.bnl.gov/chart

- Karlsruhe Nuclide Chart: http://www.nucleonica.net/nuclidechart.aspx

- Interactive Table of Nuclides from the Korea Atomic Energy Research Institute: http://atom.kaeri.re.kr/

- Chart of the Nuclides (2010) of the Japan Atomic Energy Agency: http://wwwndc.jaea.go.jp/CN10/index.html

6 Summary and conclusion

Transport and fate of radionuclide are essential factors for assessing the impact of an accelerator on the groundwater. For an assessment which can be used in licensing processes or as an argument in public discussion, it is necessary to quantify travel time and element concentration or activity in the aquifer. Computer modeling is the common tool for receiving this information.

The same approach has to be performed if radionuclides enter the environment by other processes, e. g. uranium mining activities, nuclear accidents or even assaults with radiological dispersal devices (RDD).

To address soil specific behavior of a solute, a parameter, which quantifies the interaction of a migrating element with the surrounding soil/rock, is introduced in many computer codes. This parameter, the partition coefficient K_d, describes the partitioning of a solute (or contaminant/radionuclide) between the solid and the aqueous phases.

One way to handle the great variability of soil textures, mineralogical composition and chemical constituents is the reduction of characterizing parameters to particle size distribution (sand, loam, clay) and organic content. If measured K_d values are not available in the literature or data bases, an established method to predict K_d values based on the bioavailability can be used. The *"soil-to-plant concentration ratio"* (CR), which is an indicator for the bioavailability, shows a strong negative correlation to K_d values.

Retardation by sorption and precipitation processes causes the mean velocity of a specific radionuclide to be slower than the groundwater velocity. From all reported radionuclides tritium (hydrogen) and chlorine are the most mobile elements – more or less unaffected by these processes - followed by carbon, phosphorus and calcium. Other elements such as beryllium, europium, iron, scandium, titanium and vanadium can be estimated as relatively immobile.

For the use of these values in computer modeling it is necessary to show values with respect to the specific soil type.

Table 6-1 gives an overview bandwidth of K_d values which were found in the cited An initial estimation with respect to the potential radioactive contamination of ground-

water downstream of an accelerator site can be made by looking at potential travel time of a mobile radionuclide and its half-life.

If mobile and sufficiently long living nuclides are identified e.g. by activation calculations these nuclides can be used for a first modeling of radionuclide transport in groundwater.literature. It distinguishes between four major types of soil (see chapter 3.4). If no experienced value from the literature could be derived predicted K$_d$ values were used which are based on the soil-to-plant concentration ration (chap. 3.4) mentioned above. More information about the K$_d$ values can be found in the element specific subchapters of chapter 4.

Table 6-1:   K$_d$ values [L/kg] for different soil types

element	sand[1]	silt[2]	clay[3]	organic[4]
Beryllium	*250*	*800*	*1.300*	*3.000*
Calcium	5	30	50	90
Carbon	5	20	1	70
Chlorine	0.8	0.25	4.4	11
Chromium(IV)	70	30	1.500	270
Cobald	60	1300	550	1.000
Copper	*30*	*105*	*159*	*369*
Europium	*236*	*825*	*1.255*	*2.908*
Gold	*30*	*105*	*159*	*369*
Hydrogen/Tritium	0	0	0	0
Iron	220	800	165	600
Manganese	50	750	180	150
Mercury	*19*	*66*	*101*	*234*
Phosphorus	5	25	35	90
Scandium	*315*	*1.098*	*1.671*	*3.871*
Silicon	*35*	*110*	*180*	*400*
Silver	90	120	180	15.000
Sodium	*76*	*265*	*403*	*934*
Strontium	31	108	900	1.540
Sulfur	*14*	*49*	*75*	*174*
Titanium	*327*	*1.141*	*1.737*	*4.024*
Vanadium	*327*	*1.141*	*1.737*	*4.024*

[1] sand soils: ≥ 70 % sand sized particles

[2] clay soils: ≥ 35 % clay-sized particles

[3] loam: ≤ 80 % silt-sized particle or even distribution of sand-, clay- and silt-sized particles

[4] organic soil: > 30 % organic matter

italic: predicted K$_d$ values using the "soil-to-plant concentration ration" (CR) from Baes et al. (1990)

normal: experienced K$_d$ values taken from the literature

By looking at the half-life of mobile radionuclides a second process reduces the activity further downstream of a radionuclide released at a specific site.

EU Council Directive 96/29/EURATOM (EURATOM, 1996) gives basic safety standards for all EU countries for the protection of the health of workers and the general public against the dangers arising from ionizing radiation. This directive limits the total annual effective dose of 1 mSv to any member of the public (assuming the continuous presence of a person at the worst possible position of a fence enclosing a facility).

Effective dose, by definition, is the sum of the weighed equivalent doses in all the tissues and organs of the body specified in Annex II (EU Council Directive 96/29/EURATOM) from internal and external irradiation.

Additionally, fulfillment of the ALARA principle (doses have to remain "As Low As Reasonably Achievable") is required.

Figure 6-1 shows the main protection levels. More information about licensing procedures for ion beam facilities can be found in a technical report of the ESS project (European Spallation Source / Europäische Spallations-Neutronenquelle, Moormann et al. (2003)).
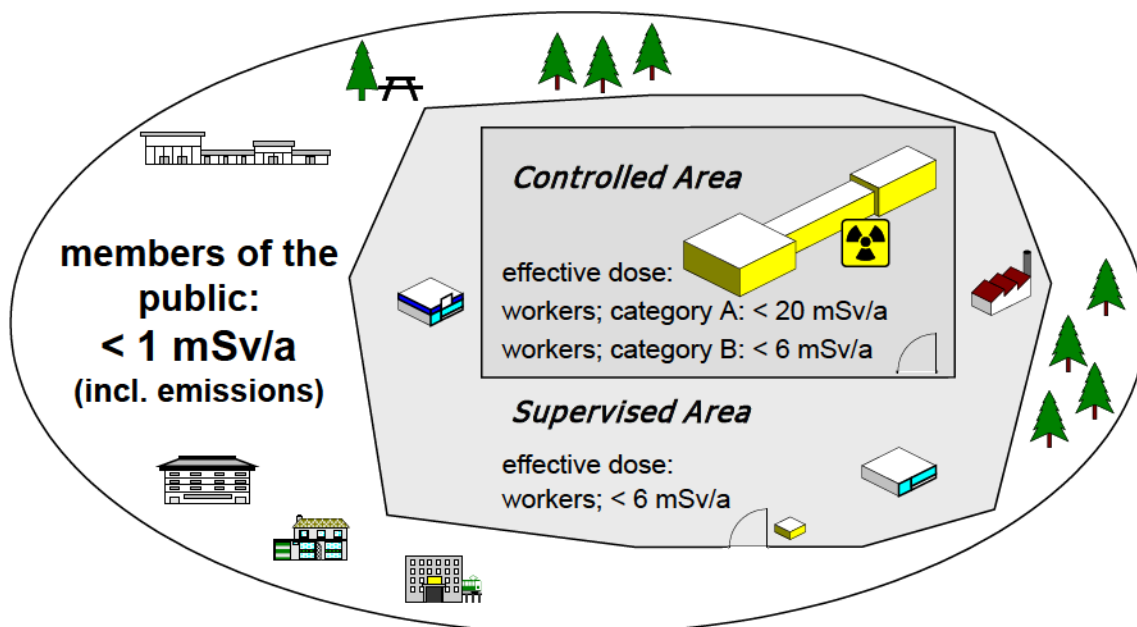
Figure 6-1:   "1 mSv effective dose concept" of Council Directive 96/29/EURATOM

Due to differences in sensitivity of tissues/organs the effective dose has to be calculated using weighing factors for radiation and for tissues. Therefore, after modeling the transport of specific radionuclides, calculation of their concentrations and activity

concentrations at a specific location, a final calculation has to be made to receive/define the effective dose. This dose is the most relevant factor for any discussion about effectiveness and optimisation of the shielding with respect to radiation protection of the public and the environment.

7 Literature

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