

Morsleben Salt Dome (Germany)

Description: Low and intermediate, non heat generating radioactive wastes have been disposed of in a repository constructed in the former Morsleben salt mine, in a salt dome in Northern Germany.

The repository ceased operation in 1998 and currently a safety assessment is being performed prior to closure. The host rock and associated technical features represent an important barrier to radionuclide migration. An additional barrier is provided by the sediments overlying the salt dome, where radionuclides are retarded by sorption. In safety assessment studies this radionuclide retention is modelled by means of distribution coefficients (K_d -values). This analogue study deals with the retention processes in the sediments overlying the salt dome at the Morsleben site.

Selected sediment samples from aquifers in the overburden at the Morsleben site have been analysed to determine their content of those naturally occurring elements of relevance in performance assessment. In-situ K_d -values have been determined from element concentrations in the sediment and in the corresponding groundwater. These values have been compared with K_d -values determined by laboratory batch experiments on similar sediments from the site.

In total, 15 sediments from 13 different boreholes at the Morsleben site have been investigated. The majority of the samples come from the so-called Alleringersleben sandy horizons, with one sample from the Buntsandstein horizon. In-situ K_d -values have been derived for strontium, uranium, thorium and rare earth elements, all of which are considered as homologues for trivalent actinides.

The use of in-situ K_d -values has been widely discussed in the literature. The K_d -value describes the ratio between the sorbed and dissolved concentration of an element, and assumes the existence of a reversible sorption process. Therefore, the first prerequisite for the determination of an in-situ K_d -value is that the natural system is in a state of equilibrium. Secondly, the water-sediment pair must be taken from the same location. Thirdly, only the surface sorbed fraction, which is accessible to groundwater, is considered in the natural sediment sample.

One commonly applied procedure for determination of the sorbed element fraction involves the use of selective leachants. In this procedure the surface sorbed fraction of the natural sediment sample is leached by a specific agent, leaving the bulk phase as far as possible undisturbed. Within the study presented here, the sorbed element fraction has been quantified by leaching with 1 M acetic acid at pH 2.4. However, it is important to note that acetic acid will not only leach the sorbed element fraction but will also dissolve carbonate minerals. This must be taken into account during interpretation of the results.

Figure 1 presents the results for uranium, by way of example. For most of the samples the agreement between in-situ and laboratory K_d -value is fairly close. There is no sediment-water system where the in-situ K_d -value is significantly lower than that measured in the laboratory. This is an important observation for building confidence in the laboratory K_d -values. In those cases where the sediments contain a high amount of calcite, the results are enclosed by blue boxes. In these cases the in-situ K_d -values are significantly higher than the values from laboratory experiments. This is due to the fact that part of the uranium is incorporated in the calcite mineral. As already discussed, in the selective extraction procedure, the 1 M acetic acid dissolves the calcite minerals and therefore an additional amount of uranium is leached, resulting in an increased value for the in-situ K_d .

The investigations have also shown that the leachable fractions of strontium and rare earth elements are even more affected by the content of calcite compared to uranium, i.e. about 100 % of Sr and 50 % of REE can be fixed in the calcite structure.

This fact also demonstrates another aspect of radionuclide retardation in natural systems. Calcite can be considered as a potential host phase for Sr and trivalent actinides. However, due to limited

knowledge, especially with regard to the time scale of this incorporation (co-precipitation) process, it has not been considered in performance assessments up to the present time.

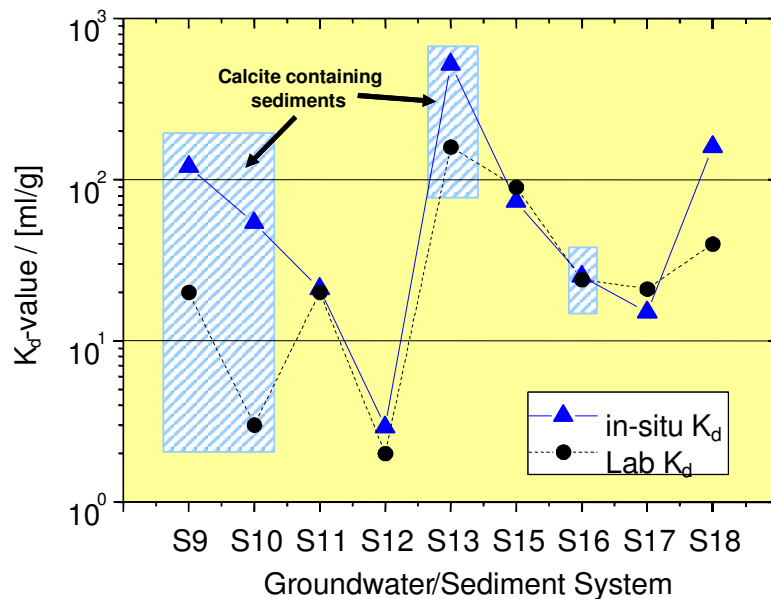


Figure 1: In-situ K_d -values versus lab K_d -values for uranium

Relevance: The overburden can be an important barrier for radionuclide transport out of the repository into the biosphere. The travel time of the respective radionuclide depends on the sorption properties of the overburden. In cases where the radionuclide travel time is greater than its half life, its concentration will be significantly reduced during transport through the overburden.

Position(s) in the matrix tables: The study illustrates chemical RN retardation at low temperatures in sandstones in the far-field matrix table.

Limitations: The application of in-situ K_d -values is limited due to the principal uncertainty in clearly identifying sorption mechanisms by such studies. The selective extraction procedures do not allow a more precise determination of the sorbed metal ion species. In-situ K_d values have to be compared with laboratory sorption data obtained under controlled conditions. It should be noted that laboratory K_d values do of course also suffer from uncertainties. Agreement between both types of values, can, however, increase confidence in sorption data.

Quantitative information: In-situ K_d -values for Sr, U, Th and REE are derived for selected sediment-water pairs in the overburden at the Morsleben site. Due to the limitations of the method, values will not be directly used in PA, but are of use in building confidence in laboratory measured K_d -values.

Uncertainties: The main uncertainties are due to the determination of the sorbed element fraction by the sequential extraction procedure and to difficulties in the comparability of samples. This is due to the heterogeneity of the overburden, which can also affect the laboratory K_d -values.

Time-scale: The time-scale addressed by the study is geological.

PA/safety case applications: The in-situ K_d -values have not been applied directly as parameters in performance assessment due to the limitations mentioned. They have however been used to strengthen confidence in the laboratory K_d -values. It can be shown that in-situ K_d -values agree rather well with laboratory K_d -values in similar sediment-water systems, where the minerals contained little or no trace amounts of calcite.

Communication applications: The licensing procedure for closure of the repository has not yet started. Therefore the results have not yet been used for communication purposes.

References:

Geckeis, H., Klenze, R., Kim, J.I. (1999) Solid water interface reactions of actinides and homologues: Sorption onto Mineral surfaces. *Radiochim. Acta* 87, 13-21.

Geckeis, H.; Vejmelka, P., Degering, D. Pohlmann, C., Gompper, K.; Hentschel, D., Kim, J.-I. (1999) In-situ distribution of trace metals in a natural aquifer overlying the nuclear repository site Morsleben, Germany. In: *Proceedings of the workshop on 'Mineral/water interactions close to equilibrium'*. FZKA 6291.

Geckeis, H., Ngo Manh, Th., Bouby, M., Kim, J.I. (2003) Aquatic colloids relevant to radionuclide migration characterization by size fractionation and ICP-mass spectrometric detection. *Colloids and Surface A* 217, 101-108.

Added value comments: A similar study has also been performed for sediment-water systems at the Gorleben site.

Potential follow-up work: not planned

Keywords: sorption, in-situ K_d , strontium, uranium, thorium, rare earth elements, sandy sediments

Reviewers and dates: Ulrich Noseck, GRS (April, 2004)