Heselbach (Germany)

Description: The intent of the natural analogue study at Heselbach in Bavaria, Germany is to understand and quantify the relevant geochemical and hydraulic processes and to identify the mechanisms leading to mobilisation/immobilisation of uranium, thorium and trace elements in argillaceous sediments with seams of lignite.

Since 2001 the Heselbach site has been part of an investigation program which includes an extensive drilling-campaign with sample collection and regular groundwater analysis including on-site measurements. After detailed chemical characterization of the site, a 2D-flow-model has been developed to simulate the groundwater flow. Further transport modelling will be undertaken to quantify the uranium migration and fixation in the sediments (Brasser et al., 2002; Noseck et al., 2002).

Heselbach is located 30 km N of Regensburg at the NE rim of the former mining district Wackersdorf. The uranium accumulation is located within a tertiary basin structure fromed during the Miocene age (18-20 Ma). Clayey sediments at the bottom of the tertiary basin structure represent a barrier to the underlying Keuper sandstone. Several lignite seams within the tertiary basin were formed and overlayed with a sandy-silty horizon (Figure 1). The actual uranium accumulation is limited to the rim of a small E-oriented side-bay extending over just a few tens of meters.

Uranium has been concentrated in lignite seams and clays along the uppermost margin in depth down to 0.5-8 m. The enrichment reaches concentrations up to 1100 ppm in the lignite horizon and 700 ppm in the underlying clay. Quartz, feldspar, muscovite, kaolinite and in some cases pyrite and gypsum have been identified as main minerals; organic components in the lignite horizon amount to max. 34 wt%.

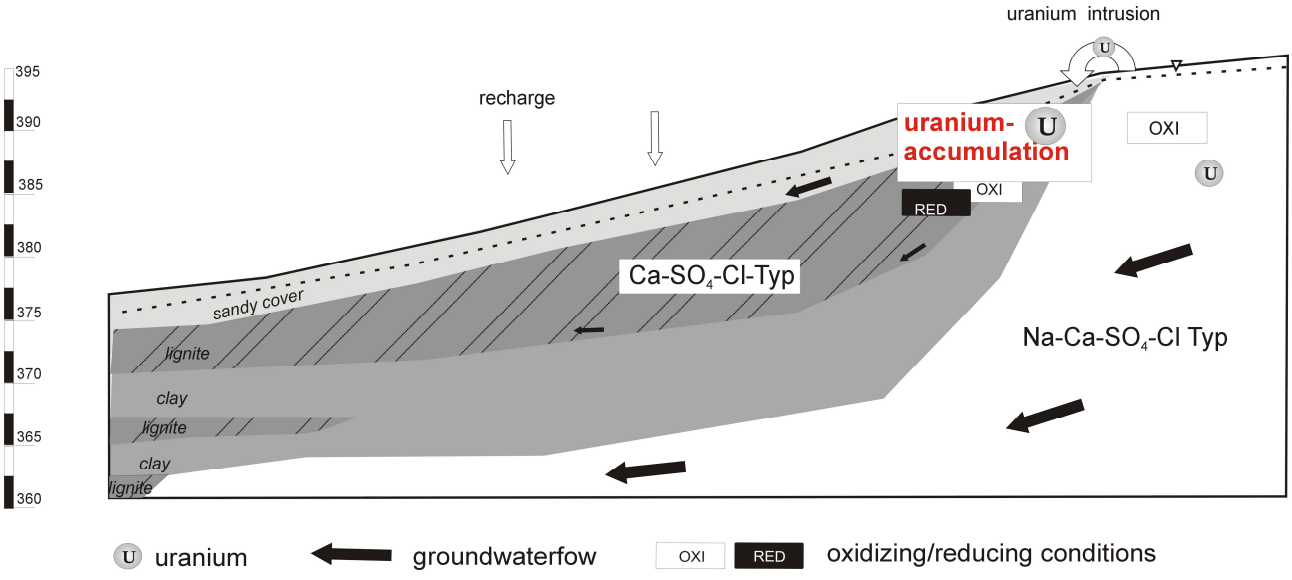

Figure 1. Conceptual model for the uranium migration into the tertiary basin at Heselbach site

Figure 1 demonstrates the geological situation at Heselbach site before coal-mining occurred. The supposed uranium source is the flanking Keuper sandstone. Fixed uranium from the Keuper sandstone was oxidized, remobilised and transported as uranyl (UO_2^{2+}) within the groundwater. An influx to the tertiary basin over the overlaying sandy layer could have been the consequence of Quaternary meltwater intrusions.

On-site-characterisation of groundwater chemistry shows slightly positive Eh- and low pH-values (5 to 6.5). Photometric determination of Fe^{2+}, Fe_{tot}, SO_4^{2-} and S^{2-} clearly demonstrates the dominance of SO_4^{2-} and Fe^{2+} ions. Generally groundwater from Heselbach site is of Ca-SO4-Cl-type. O_2- and

HCO_3^--concentrations at the rim of the tertiary basin are variable and lead locally to a recent uranium solution.

The uranium phases have been characterized by SEM-EDX, EXAFS and XANES-measurements, as well as sequential extraction techniques. Any evidence for the existence of uranium minerals could not been found. Uranium exists finely distributed within the sediment and is likely to be associated with the organic phase. Chemical separation techniques used to determine the oxidation state of uranium as well as XAFS measurements clearly indicate U (VI) as predominant phase. Measurements of the bond-length of uranium with oxygen identified uranylion as main species, so that, at the current stage, it is most likely that the uranium accumulation is composed of Uranyl-humat-complexes.

Activity-ratios of uranium series isotopes below unity (Figure 2) clearly indicate uranium enrichment processes for the lignite horizon within the last 300 000 years.

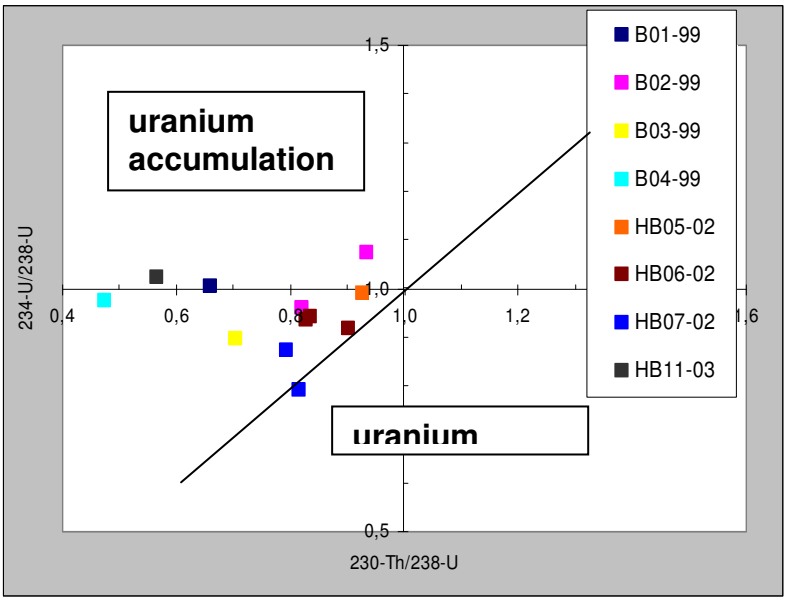

Figure 2. Activity-ratios of uranium-series-isotopes in the Thiel-diagram from lignite sediment samples of different boreholes

Relevance: The uranium transport at Heselbach site occurs in sandstones which are overlaid by tertiary argillaceous sediments and lignite horizons. This geological situation is similar to the tertiary and quaternary sediments making up the overburden of salt domes in northern Germany, which are foreseen as a potential host rock for repositories with radioactive waste.

This study is focussed on transport and fixation processes of uranium in tertiary sediments under natural conditions over long time scales, i.e. the interaction of U(VI) with lignite sediments and uranium diffusion in clay under natural conditions.

Position(s) in the matrix tables: RN migration in the far-field matrix table.

Limitations: At Heselbach site the hydrological situation and the formerly flow system have changed due to coal mining in the last two centuries. The uranium deposit is near surface and accordingly disturbed by recent Eh/pH-value changes. During the latest paving, parts of the site were covered by material containing hydrogen carbonates. Local rising of the pH-value and solution of uranium from the sediment are the consequence.

Quantitative information: Hydrological investigations lead to the verification of the conceptual hydrological model. Quantitative information will be received by uranium transport modelling (in

progress). The results could be used to test numerical models applied in PA for radionuclide transport under typical far-field conditions including heterogeneities.

Evaluation of diffusion profiles in clay will be accompanied by modelling in order to derive apparent diffusion coefficients for uranium in natural clays (cf. follow up work).

Uncertainties: One uncertainty is associated with the determination of the fine distributed immobile uranium source. Uncertainty for flow and transport modelling is caused by uncertainties in the knowledge of duration and time period of the uranium influx.

Time-scale: The time-scale addressed by this study is geological < 1 Ma. Potential older uranium accumulation processes have been displaced by young (< 300.000 a) events.

PA/safety case applications: None identified.

Communication applications: None identified.

References:

Brasser Th, Noseck U and Schönwiese D. 2002. Uranium Migration in Argillaceous Sediments as Analogue for Transport Processes in the Far Field of Repositories (Heselbach Site, Germany). in Uranium in the Aquatic Environment. B.J. Merkel, B. Planer-Friedrich, C. Wolkersdorfer (eds.) Springer-Verlag 2002.

Noseck U, Brasser Th and Pohl W. 2002. Tertiäre Sedimente als Barriere für die U- / Th-Migration im Fernfeld von Endlagern. Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, GRS-176, Braunschweig März 2002.

Added value comments: None identified

Potential follow-up work: The project is due to complete in 2005. Uranium transport calculations are currently in progress. Analyses of activity ratios are being undertaken in order to identify diffusion processes within the uranium clay enrichment and will be accompanied by modelling.

Keywords: uranium, thorium, organic material, argillaceous sediments, diffusion

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