Ruprechtov (Czech Republic)

Description: The Ruprechtov natural analogue study is focused on the investigation and quantification of relevant geochemical and hydraulic processes related to mobilisation and immobilisation of uranium and other trace elements in Tertiary argillaceous sediments. Such sediments are characteristic for the overburden of host rocks at several potential repository sites.

The Ruprechtov site is situated in the north-western part of the Czech Republic where several Tertiary basins with clay, sand and organic material (coal, lignite) are located. Granites form the base of the basin and its surroundings. The granites have been strongly kaolinized at their surface mainly during Miocene. These kaolinic horizons can reach thicknesses of up to some tens of meters (Figure 1).

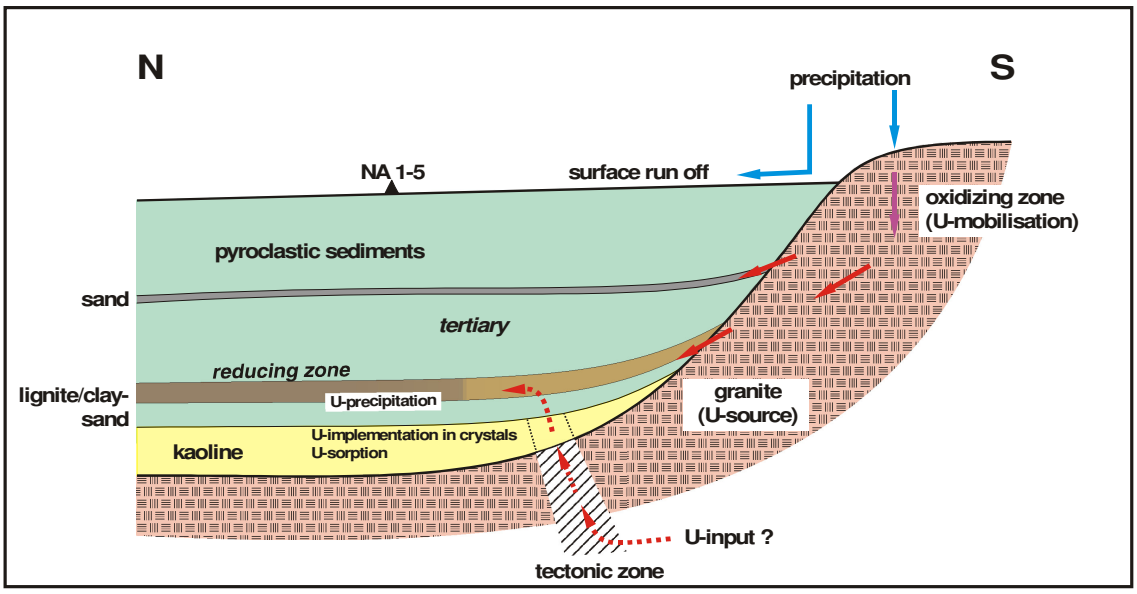

Figure 1: Conceptual model for the uranium migration at Ruprechtov site.

The tertiary sediments are mainly composed of quartz, kaolinite, muscovite/illite, montmorillonite, anatase and siderite; the occurrence of pyrite is less frequent. A characteristic feature of the site is the heterogeneity of sediments at small scale. Irregularly distributed uranium accumulations have been identified, mainly connected to sediments with high content of organic carbon, at the interface between the kaolinic basement and overlying sediments in the so-called lignite/clay-sand horizon. Adjacent to these uranium enrichments a permeable horizon with thickness of about 1-2 m and hydraulic conductivity in the range of $1x10^{-6}$ m/s exists. In contrast, the upper and lower clay layers are characterized by rather low permeability (10^{-11} m/s).

The uranium enrichments appear in very narrow sections extending only a few decimetres. The highest U-concentrations measured come to about 300 mg/kg. Uranium phases have been characterized by sequential extraction, SEM-EDX, XAFS and micro-x-ray fluorescence techniques. The existence of phosphates such as ningyoite ($CaU[PO_4]_2$) and traces of uraninite minerals in the zones of highest U-enrichment has been detected. Furthermore, uranium bearing monazites have been found indicating that part of uranium is of detrital origin. By using micro-XRF and XAFS it is has been confirmed that U(IV) represents the predominant oxidation state of uranium. Iron, probably pyrite nodules is adjoining Uranium accumulations, frequently accompanied by Arsenic. Activity ratios of members of the uranium decay show slight disequilibrium states, which indicate uranium mobilisation/immobilisation processes at least within the last 500,000 years.

Groundwater from the permeable lignite/clay-sand layer is Ca-Na-HCO_3. The pH-values range from 7 to 8, the Eh-values vary between 0 and -280 mV and uranium concentrations have been detected in the range of 10^{-8} to 10^{-9} mol/l. The extracted porewater is characterised by higher salinity, uranium and other trace elements concentrations compared to the granitic porewater

which is generally less mineralized and more oxidizing, with slightly lower pH-values of 6.2 to 6.8 and uranium concentrations up to 10^{-7} mol/l.

At current stage of the project it is supposed that mainly three processes are responsible for uranium migration and accumulation at the site. The primary uranium source has been identified to be the surrounding "Erzgebirge granite" with its generally high uranium content (20 – 40 ppm) and a U/Th ratio of >1. Mobilisation of uranium started from underlying granite followed by a) transport via diffusion as well as b) advective transport through fault zones in the kaolin up to permeable lignite/clay-sand horizon. A third process (c) is the mobilisation of uranium from outcropping granite and transport via permeable layers. The main accumulations are found close to the permeable clay/lignite-sand horizon. Due to the occurrence of detrital monazites an additional syn-sedimentary U-enrichment also has to be taken into account.

Relevance: Uranium and trace metal mobilisation and immobilisation processes at Ruprechtov site have taken place in a stratigraphic sequence typified by argillaceous sediments and lignite/clay-sand horizons with partly high content of organic matter. Similar geological features are found in the overburden of host rocks as the salt domes in northern Germany, which might be potential host rocks for deep geological repositories for radioactive wastes.

This study enables the investigation of the main processes leading to a retention of the transport of uranium, thorium and to some extent for other PA relevant elements in these argillaceous sediments enriched with organic material under natural (reducing) conditions over long time scales. In particular the study will contribute to the:

– testing of geochemical models and data for uranium and trace elements under typical far-field conditions (including impact of humic colloids); and
– testing/confirmation of conceptual models used in PA for radionuclide transport under typical far-field conditions in Tertiary sediments.

Position(s) in the matrix tables: The study illustrates the far-field process of RN-migration in mudrocks (clays) at low temperature - in particular impact of organics and colloids - and chemical RN-retardation in mudrocks at low temperature.

Limitations: The site is rather complex. Different processes of uranium mobilisation and immobilisation have occurred in the geological past, which might be difficult to differentiate. High heterogeneity on small spatial scale and complex hydrogeology of the site limits the application of transport models.

Quantitative information: Project is not yet finished and it is expected to output the following data:

– Groundwater data with concentrations of major and trace elements from different areas of the site will be available including also uranium disequilibrium series, natural isotopes (δD δ^{18}O in water, δ^{13}C and ^{14}C in various forms of carbon) and colloids.
– Well characterized sediment data with uranium concentrations, uranium series disequilibrium states, and characterized immobile uranium phases will be available. Analysis of the U(IV)/U(VI) distribution including ^{234}U/^{238}U activity ratios in both oxidation states is underway and will be available at the end of the project.
– Data will be used for application and testing of geochemical models.

Uncertainties:
In general, there are good quality groundwater and sediment data, including concentration of major and trace elements, uranium disequilibrium series disequilibria and natural isotopes. The main uncertainties are correlated with the limitations, i.e. the complexity of the system and the problem to differentiate between the distinct uranium mobilisation/immobilisation processes. Uncertainties are also connected to uncertainties in the exact time scales of the respective processes. This makes it difficult to apply transport models.

Time-scale: The time-scale addressed by the study is geological. Firstly uranium deposition stems from Lower Miocene. Due to disequilibrium states in the uranium decay chain it is clear that uranium mobilisation/immobilisation processes occurred within the last 500 000 years.

PA/safety case applications: None identified.

Communication applications: None identified.

References:

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Added value comments: None identified.

Potential follow-up work: The project is still running, at least until 2005. Characterisation of new drill cores and water from new wells are currently on the way.

Keywords: uranium, thorium, argillaceous sediments, sorption, precipitation, organics

Reviewers and dates: Ulrich Noseck, GRS; Ales Laciok, NRI (April, 2004)