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Migration lengths of uranium in the Opalinus Clay are determined by the pore water geochemistry and hydrogeological setting

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Any safety assessment of radioactive waste disposal sites is done based on the simulation of migration lengths of radionuclides through the host formation. This is done by the application of transport parameters obtained from experiments with homogeneous, e.g. geochemically and mineralogically constant, conditions. However, such an assumption of homogeneity is no longer applicable on the host rock scale (>200 m). Consequently, experimentally determined transport parameters might no longer represent the host rock on larger scales.

Uranium, as main component of spent fuel, is used here as an example to evaluate the impact of heterogeneous systems on the total migration lengths compared to homogeneous ones. For this, the hydrogeological system of the Swiss Opalinus Clay, a potential host rock, is modelled in one-dimensional diffusion simulations with PHREEQC. Since sorption and hence migration of uranium is primarily governed by pCO_2 > Ca^{2+} > pH > pe > clay mineral quantity [1], the focus is on the simulation of the geochemically heterogeneity, and thus on the hydrogeological system. Sorption is quantified with mechanistic surface complexation models and cation exchange. At Mont Terri, geochemical gradients established towards the embedding aquifers due to diffusive exchange over millions of years as a consequence of the Jura folding and associated erosion history [2].

First, measured pore water profiles were confirmed by the simulations. They served as starting profiles for the subsequent uranium migration that was quantified in a second step. By comparing migration lengths after a simulation time of one million years with results of homogeneous simulations, it has been shown that uranium migration is enhanced by up to several tens of meters depending on the pCO_2. Consequently, the entire hydrogeological system needs to be taken into account and the governing parameters can be prioritized as follows: pCO_2 > hydrogeology > mineralogy.

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