Mobility of long-lived U-series radionuclides at an interface between claystone hosting old reducing pore water and an aquifer carrying young meteoric water

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The Jurassic Opalinus Clay formation (OPA) in northern Switzerland is considered as a potential host rock for a deep underground nuclear waste repository. We are interested in the naturally occuring radionuclides which can provide valuable information regarding their long-term in-situ behaviour in this formation.

We studied the systematics of long-lived U-series radionuclides from Opalinus Clay (OPA) at its interface with an overlying limestone aquifer at the Mont Terri in the Jura Mts. This aquifer was activated ~2.5 Ma ago due to erosional incision that enabled inflow of young, meteoric water [1]. As a result reducing and stagnant pore water of OPA has been exposed to the oxidizing aquifer. This hydrogeological setting provides a unique opportunity to study the behaviour of long-lived U-series radionuclides at the interface of these two distinct environments and gives an insight into processes that govern their transport in OPA.

We investigated rock samples of OPA from 5 boreholes in close vicinity and at the interface with the aquifer. Two sampling approaches were employed: averaging of the core material to look at the record on a large scale (meters) and small-scale drilling within the cores in order to check for possible mobility on a centimeter scale. Samples were studied by sequential leaching with total digestion as a final step. ^{238}U, ^{234}U and ^{230}Th concentrations and ratios were determined using MC-ICP-MS.

Our data points to a very slow diffusive redistribution of U in the OPA. Results obtained indicate two distinct periods of geochemical evolution: one before and one after the infiltration of meteoric waters into the aquifer. During the former a centimeter scale diffusion of ^{234}U governed by the in-situ supply from rock operated. This resulted in development of bulk rock ($^{234}U/^{238}U$) disequilibria above and below one in the regions of smaller and larger U content respectively. The fresh water infiltration changed the local gradients of both ^{234}U and ^{238}U in the pore water of portions of OPA directly bordering on the aquifer. As a consequence the previous regime record was overprinted indicating the depth of influence of the overlying oxidizing aquifer.

References

[1] Bossart P. and Wermeille S. (1999) Rep. Swiss nat. hydrol. and geol. Surv. **23**, Bern, 5-14.