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Assessing long-term microbial impact on mineralogical trapping of uranium from deep groundwater at Äspö Hard Rock Laboratory, Sweden

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Natural uranium (U) in deep groundwater has been extensively studied in connection to the search for suitable locations for final disposal of spent nuclear fuel (SNF). The U removal process depends on environmental and geochemical conditions and is often associated with fractionation of the main 'stable' isotopes, ^{238}U and ^{235}U (denoted as $\delta^{238}\text{U}$), during reduction of hexavalent (U(VI)) to tetravalent (U(IV)) species. Thus, $\delta^{238}\text{U}$ serves as an important tracer for redox specific scenarios, in local to global temporal and spatial scales. In this contribution, one specific borehole setting built at 415 m depth into Paleoproterozoic granitoid rock at the Äspö Hard Rock Laboratory (HRL), Sweden, is investigated after 17-year experiment period (1995-2012). The HRL is constructed and operated by the Swedish Nuclear Fuel and Waste Management Co. and serves as a full-scale tunnel as a test-facility for the actual SNF repository to be built at Forsmark. We show how various analytical techniques, modelling, and isotope methods can be utilized to reveal U speciation and removal pathways, associated redox changes and related U isotope fractionations in the deep aquifer and during U mineralogical trapping. Synchrotron-based spectroscopic techniques reveal that calcite and Fe(II) sulfides precipitated on the borehole equipment contains intermittent highly elevated U, occurring as U(IV), and hence serves as a mineralogical sink for U. Thermodynamic modelling shows that aqueous Fe(II) is the main driving force for the reduction of U(VI) in the borehole water, alongside sulfides formed from bacterial sulfate reduction. The bacteria-driven degradation of technical polymer constituents present in the borehole equipment is central to processes forming the carbonates and sulfides that facilitate reduction of U(VI) and subsequent immobilisation of U(IV) into the calcite and Fe sulfides. We use $\delta^{238}\text{U}$ to show that U(VI) undergoes redox transformations in granitic rock aquifers, involving mineralogical and microbial pathways. The recorded $\delta^{238}\text{U}$ provides evidence for reductive removal of U(VI) from fracture water, presumably taking place along redox fronts in the fracture network. The obtained data on removal of U by calcite and Fe sulfides provide important insights for the assessment of the geochemical behaviour of U and other redox-sensitive species in deep anoxic aquifers, that is relevant for trace metal mobility and long-term storage of SNF and nuclear waste.

Primary authors: Dr PIDCHENKO, Ivan (Department of Biology and Environmental Science, Linnaeus University, Kalmar, Sweden); Dr DRAKE, Henrik (Department of Biology and Environmental Science, Linnaeus University, Kalmar, Sweden)

Co-authors: Dr CHRISTENSEN, John (Energy Geosciences Division, Lawrence Berkeley National Laboratory, Berkeley, USA); Dr IGNATYEV, Konstantin (Diamond Light Source, Harwell Science and Innovation Campus, Didcot, Oxon, UK); Dr PUIGDOMENECH, Ignasi (Swedish Nuclear Fuel and Waste Management Co., Solna, Sweden); Dr RASBURY, Troy (Department of Geosciences, FIRST, Stony Brook University, Stony Brook, USA)

Presenter: Dr PIDCHENKO, Ivan (Department of Biology and Environmental Science, Linnaeus University, Kalmar, Sweden)

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