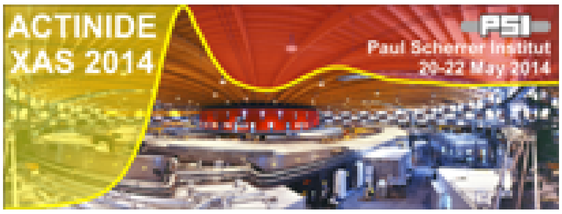

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Using XAS to probe radionuclide biogeochemistry in complex environmental systems

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Understanding the behaviour of radionuclides in natural and engineered environments is key to the management and control of radioactively contaminated materials. Recent work has demonstrated that biogeochemical cycling of radionuclides is critical in controlling radionuclide behaviour in the shallow sub-surface. More recently, we have been exploring the role of microbial processes in controlling radionuclide behaviour under conditions relevant to geological disposal of radioactive waste and to some radioactively contaminated land scenarios. Specifically we have explored both high pH conditions and the indirect effects of microbial reduction on silicate minerals and the resultant reactivity of the bioreduced mineral phases on radionuclide speciation. For the high pH geomicrobiology work, we took alkaline sediments from an old lime workings, and incubated the sediments with an electron donor and uranium-(VI), technetium-(VII) and neptunium-(V). In addition, we also added excess Fe(III) as ferrihydrite into an additional set of experiments to probe the impact of microbial Fe(III)-reduction on radionuclide behaviour. As the systems became reducing, we tracked the biogeochemical behaviour of the stable elements and tracked the solubility and speciation of U, Tc and Np using geochemical and spectroscopic techniques (XANES and EXAFS). For the experiments exploring the indirect impacts of microbially induced Fe(III)-reduction on radionuclide speciation, we exposed chlorite and biotite to the model Fe(III)-reducing microorganism Geobacter sulfurreducens in the presence of an electron donor. The resultant bioreduced minerals contained elevated levels of Fe(II) compared to the unreacted biotite and chlorite. After washing to remove cells, the bioreduced minerals were then spiked with radionuclides to explore the reactivity of the bioreduced minerals compared to the unreacted materials. U, Tc and Np solubility and speciation were again tracked using geochemical and, at selected time points, spectroscopic techniques (XANES and EXAFS). Here, we present the results of this work and highlight the significance of biogeochemistry in underpinning the understanding of radionuclide speciation and fate in the environment.

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