Neptunium Biogeochemistry Across the Nuclear Waste Legacy

GARETH LAW¹, ADAM WILLIAMSON², CLARE THORPE², PIETER BOTS², CHRISTOPHER BOOTHMAN², SAM SHAW², FRANCIS LIVENS¹, JORG ROTHE³, KATHY DARDENNE³, MELISSA DENECKE¹, JON LLOYD² AND KATH MORRIS²

¹School of Chemistry, The Univ. of Manchester, Manchester, M13 9PL, UK (Correspondence: gareth.law@man.ac.uk);

²School of Earth, Atmospheric and Environmental Sciences, The Univ. of Manchester, Manchester, M13 9PL, UK;

³KIT-INE, Eggenstein-Karlsruhe, D-76344, Germany.

Neptunium is a key risk-driving radionuclide in nuclear legacy management and it is becoming increasingly clear that microbially-mediated redox cycling is pertinant to Np mobility under both contaminated land and geodisposal conditions. Here, NpO_2^+ is predicted to be the most mobile transuranic species in the sub-surface; however, compared to other radionuclides, there is a distinct paucity of information concerning Np biogeochemistry.

Here we present an overview of Np environmental behaviour under both contaminated land (circumneutral pH) and geodisposal (pH 10-13) settings. Data presented encompasses experimentation conducted across a range of hetergeneous sediment and pure mineral systems, with XAS used throughout to monitor changes in Np speciation and local coordination. Importantly, these data highlight the co-cycling of Np with microbially-mediated Fe reduction between pH 6-10. Here, abiotic reduction of Np(V) to Np(IV) *via* reaction with biogenic Fe(II) limits Np solubility. Further, co-cycling with Mn appears relevant at circumneutral pH. Conversely, under conditions relevant to the geodisposal chemically disturbed zone (pH 10-13), Np(V) sorption and potential uptake into the lattice of neo-forming Fe(III) minerals (e.g. heamatite) appears important.