The Biogeochemistry of Pu and U: Distribution of Radionuclides Affected by Microorganisms and their Siderophores, Reductants, and Exopolymers

Joseph Lack,[1] Hakim Boukhalfa,[1] Jennifer Forsythe,[1] Christy Ruggiero,[1] Delis Maldonado,[1] Samuel Traina,[2] Mary Neu,[1] and Larry Hersman[1] (PI)

[1]Bioscience and Chemistry Divisions, Los Alamos National Laborator[1]; [2]The University of California, Merced

Radionuclide-contaminated environments are often oxic, including the Rocky Flats Environmental Technology Site (RFETS), and the contaminated groundwater at the NABIR Field Research Center (FRC). Radionuclide distribution within such environments is affected by indigenous biogeochemical processes, including the metabolic activities of aerobic microorganisms, key members being the ubiquitous *Pseudomonas* and *Bacillus* genera. Because of the chemical similarities between the actinides uranium (U) and plutonium (Pu), and iron (Fe), the metabolic processes of these microorganisms that affect the biogeochemistry of Fe could also significantly affect Pu and U distribution. We are determining the extent to which metabolic processes involved in Fe acquisition and in exopolymer production affect the distribution of Pu and U between the aqueous and solid phases. Our focus is on determining the equilibrium distribution of Pu and U between these phases, in the presence and absence of microorganisms, and in relation to Fe bioavailability. Secondly, using transposon mutagenesis, we are determining to what extent microbial processes (including siderophore production and metabolism, and exopolymer and reductant production) directly or indirectly influence aqueous/solid phase distribution of Pu and U.

In addition to considering chelators and potential metal-reducing exudates, we are investigating the limits of direct metal reduction. The metal reductase activity of a microorganism is believed to be governed by its c-type cytochrome content. In order to catalytically reduce solid metal oxides such as iron(III) oxides and metal–ligand complexes, the redox potential of these compounds needs to be in the range accessible to cytochrome C. We have examined the redox properties of commonly used iron(III) complexes (such as NTA, EDTA, citrate, siderophores, etc.), which cover a wide range of redox potentials (+200 to 400 mV), and have started examining the ability of *Shewanella* and *Geobacter* sp. to use these iron complexes as terminal electron acceptors.

The results of the research program will contribute to NABIR's stated needs to understand both "the principal biogeochemical reactions that govern the concentration, chemical speciation, and distribution of metals and radionuclides between the aqueous and solid phases" and "what alterations to the environment would increase the long-term stability of radionuclides in the subsurface."