Poster #210

Natural-Organic Matter and Microbial Controls on Mobilization/Immobilization of I and Pu in Soils and Water Affected by Radionuclide Releases in the USA and Japan

Peter H. Santschi[1], Daniel I. Kaplan[2], and Chris M. Yeager[1]

[1] Laboratory for Environmental and Oceanographic Research, Department of Marine Sciences, Texas A&M University, Building 3029, Galveston, Texas 77551
[2] Savannah River National Laboratory, Aiken, SC 29808
[3] Los Alamos National Laboratory, Los Alamos, NM 87545

Contact: Peter Santschi [santschi@tamug.edu]

Background: a) ^{129}I is among the top three risk drivers at DOE Low Level Waste or High Level Waste disposal sites, such as Savannah River Site (SRS). Radioiodine's risk stems largely from its perceived high environmental mobility, large inventory (high fission yield), high toxicity (it is a thyroid seeker), and long half-life (16M years). ^{129}I exists as multiple species (usually iodide, iodate, and organo-iodine). b) The human and environmental risks associated with Pu disposal, remediation, and nuclear accidents scenarios stems mainly from the very long half-lives of several of its isotopes. The SRS, holding one16 third of the nation's Pu inventory, has a long-term stewardship commitment to investigation of Pu behavior in the environment.

Methods: a) Six different genera of Mn(II)-oxidizing bacteria were isolated from SRS soils, and examined for the ability to oxidize iodide directly through enzymatic catalysis, or indirectly through formation of reactive oxygen species (ROS) and/or biogenic manganese oxides. b) Humic substances (HAs) from different types of soils in various global regions, were re-suspended with groundwater at a far-field Pu concentration(10^{-14} M) to examine the influence of natural organic matter (NOM) on Pu partitioning during soil erosion events.

Results: Both extracellular enzymes and ROS play a role in microbial Mn(II)-oxidation. Iodide oxidation was not observed in cultures of the most active Mn-oxidizing bacteria. While substantial amounts of Mn(III/IV) oxides were only generated in cultures ≥ pH 6, iodide oxidation was only observed in the presence of Mn(III/IV) oxides when the pH was ≤ 5. Iodide oxidation was promoted to a greater extent by synthetic $Mn(IV)O_2$ than biogenic Mn(III/IV) oxides under these low pH conditions (≤ pH 5). Thus, the influence of biogenic manganese oxides on iodide oxidation and immobilization is primarily limited to low pH environments.
b) Under acidic condition (pH~5.5 as the global averaged soil pH), 29 ± 24% of organic matter was released from the HAs, carrying 76 ± 13% of total added Pu into mobile colloidal phase. Both Pu activity concentration and partitioning coefficients (LogKd) were strongly and positively correlated with nitrogen contents in both particulate and colloidal fractions. Results from solid state ^{13}C NMR suggest carboxylate functionalities contribute to the particulate-immobilization and colloidal-remobilization of Pu during HA37 groundwater resuspension. Carboxyl- and nitrogen-containing organic moieties in the bulk NOM pool served as the predominant Pu carrier, which is relevant to potential Pu mobility in natural soils during surface runoff events.