

Poster #21-36

Collaborative Research: Natural Organic Matter and Microbial Matter and Microbial Controls on Mobilization/Immobilization of I and Pu in Soil and Water Affected by Radionuclide Release in USA and Japan

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In collaboration with Japanese Universities, we carried our research targeting natural organic matter (NOM) compounds and microbial processes that are responsible for speciation and fate of different radionuclides in soils from Japan and the USA. NOM is among the key environmental factors that influences the fate and transport of radionuclides in the environment. While this has been known for decades, there still remains great uncertainty in predicting NOM-radionuclide interactions because of lack of understanding of radionuclide interactions with the specific organic moieties within NOM. Furthermore, radionuclide-NOM studies using modeled organic compounds or elevated radionuclide concentrations provide compromised information related to true environmental conditions. Thus, sensitive techniques are required not only for the detection of radionuclides, and their different species, at ambient and/or far-field concentrations, but also for potential trace organic compounds that are chemically binding these radionuclides. GC-MS, ESI-FTICR-MS and AMS techniques developed in our lab permitted us to define how two radionuclides, iodine (I) and plutonium (Pu), form strong bonds with NOM by entirely different mechanisms: I tends to bind to aromatic functionalities, whereas Pu binds to nitrogen (N)-containing moieties, likely, hydroxamate siderophores.

Microbially mediated chelation and incorporation reactions can control a number of radionuclides, leading to retardation or mobilization, depending on whether the carrier compound is in solution or particle-bound as a function of pH or redox conditions of the ambient environment as well as the molecular weight of the carrier itself (Santschi et al., 2017a,b). In a study with RFETS soils, Pu was found enriched in NOM fractions that contained elevated levels of polycarboxylated aromatic and condensed aromatic formulas. These fractions also contained greater abundance of CHON-type COO formulas, than fractions with lower Pu concentrations. N contents increased with the progression of purification and coincided with the trend of Pu concentration (DiDonato et al., 2017).

Based on humic acid (HA) samples from 10 soils collected from around the world, solid state ¹³C nuclear magnetic resonance (NMR), and C, N, and S elemental analysis, binding of low concentrations of Pu (10⁻¹⁴ M) was correlated to the concentration of carboxylate functionalities and N groups in the particulate and colloidal phases. The much greater tendency of Pu binding to colloidal HAs than to particulate HA has implications on whether NOM acts as a Pu source or sink during natural or man-induced episodic flooding (Lin et al., 2017). In a parallel study, uptake of six particle-reactive and/or redox-sensitive radionuclides (²¹⁰Pb, ²³⁴Th, ⁷Be, ⁵⁹Fe, ²³⁷Np and ²³³Pa) by 14 HA extracts was also investigated revealing the capacity of these HAs to bind strongly to these radionuclides, either in particulate or colloidal state of the HA in a mimicked groundwater slurry (Lin et al., 2018).

While the speciation of radionuclides (e.g., I) can be controlled by abiotic factors such as biogenic Mn oxides at low pH (Grandbois et al., 2018b), biotic factors extracellular enzymes, microbial metabolites and microbial processes that alter the chemistry (e.g. pH, redox conditions) of the immediate environment (Yeager et al., 2017). Grandbois et al. (2018a) showed that the activity of soil enzymes, such as oxidases and peroxidases, and microbial biomass (particularly that of the Actinobacteria) were both strongly correlated with uptake of I at environmentally relevant concentrations in forest soils from different regions and continents.

References:

- DiDonato, N., et al. 2017. A new perspective on structural components of Pu-mobilizing organic colloids from a contaminated soil. *ES&T*, 51, 4803–4811; DOI: 10.1021/acs.est.6b04955.
- Grandbois, R., et al. 2018a. Role of microbial enzymes in the uptake of iodine in soils. In review.
- Grandbois, R., et al. 2018b. Biogenic manganese oxides facilitate iodine oxidation at low pH. *Geomicrobiology*, in press.
- Lin, P., et al. 2017. Evidence for carboxyl-associated and nitrogen-containing organic compounds regulating the remobilization of plutonium in natural soils. *Environ. Sci. Technol.*, 51(20), 11742-11751; DOI:10.1021/acs.est.7b03409.
- Lin, P., et al. 2018. Radionuclide uptake by colloidal and particulate humic acids obtained from 14 soils collected worldwide. *Scientific Reports*, submitted.
- Santschi, P.H., et al. 2017a. Iodine and plutonium association with natural organic matter: A review of recent advances. *Applied Geochemistry*, 85, 121-127; DOI: 10.1016/j.apgeochem.2016.11.009.
- Santschi, P.H., et al. 2017b. Recent advances in the detection of specific natural organic compounds as carriers for radionuclides in soil and water environments, with examples of radioiodine and plutonium. *Journal of Environmental Radioactivity*, 171, 226-233.
- Yeager, C.M., et al. 2017. Microbial transformation of iodine: From radioisotopes to iodine deficiency. *Advances in Applied Microbiol.*, 101, 83-136.