

COLLABORATIVE RESEARCH: NATURAL ORGANIC MATTER AND MICROBIAL CONTROLS ON MOBILIZATION/IMMOBILIZATION OF I AND PU IN SOILS AND WATERS AFFECTED BY RADIONUCLIDE RELEASES IN USA AND JAPAN

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¹²⁹I is among the top three risk drivers at DOE Low Level Waste or High Level Waste disposal sites (such as SRS) due to its perceived high environmental mobility, large inventory (high fission yield), high toxicity, and long half-life (16M years). Additionally, it is one of the key radionuclides driving uncertainty with the secondary waste generated from the waste-water treatment facility at the Fukushima Daiichi Nuclear Power Plant (FDNPP). ¹²⁹I exists as iodide, iodate, and organo-iodine. 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 is holding one-third of the nation's Pu inventory. Release of Pu isotopes into the atmosphere and subsequent deposition on the Fukushima Prefecture after FDNPP accident has been reported.

Significant progress has been made over the last 10 years in understanding radioiodine chemistry in groundwater and waste streams as a result of our breakthrough in iodine species detection. This analytical method can detect stable or radioactive iodine at ambient concentrations using standard laboratory equipment. We have shown for the Savannah River Site, Hanford, and Fukushima, that each of the three iodine species has vastly different chemical properties, including their tendency to remain mobile, to adsorb to solids or co-precipitate. Ambient Pu concentrations and isotopic ratios were determined with ICP-MS and alpha spectrometry, in samples collected from both SRS and Fukushima Prefecture.

Sediment Pu concentrations in the SRS F-Area wetland were correlated to total organic carbon and total nitrogen contents and even more strongly to hydroxamate siderophore (HS) concentrations. HS was detected in the particulate or colloidal phases of the sediments but not in the low molecular fractions (< 1000 Da). Water-extractable Pu containing macromolecules were separated via isoelectric focusing (IEF) and assessed via ESI FTICR-MS. Results suggested the presence of HS functionalities in the Pu containing IEF extract. While HS is a very minor component in the sediment, its concentrations greatly exceeds that of ambient Pu. ^{239,240}Pu concentrations in soil samples with different land use types as well as iodine speciation in river water and rainwater northwest of the FDNPP were well correlated to soil organic matter (SOM) content. SOM concentrations and Eh were positively, and pH was negatively correlated to ¹²⁷I concentrations in surface water and rain samples.