

Poster #21-48

Plutonium Migration Behavior at Hanford – Trench 216-Z-9

Stefan Hellebrandt¹, Enrica Balboni¹, Carolyn Pearce², Vicky Freedman², Annie B. Kersting^{1*}, and Mavrik Zavarin¹

¹ Lawrence Livermore National Laboratory, Livermore, CA

² Pacific Northwest National Laboratory, Hanford, WA

Contact: kersting1@llnl.gov

BER Program: SBR

Project: SFA BioGeoChemistry of Actinides, LLNL

Project Website: https://doesbr.org/research/sfa/sfa_llnl.shtml

Located on the Columbia River, the Hanford Site (WA) was established in 1943 to produce plutonium (Pu) for the Manhattan Project and actively reprocessed Pu for nuclear weapons from 1944-1989. Over 184 kg of Pu along with other radionuclides were released into the environment during Hanford site operations. As part of the LLNL BioGeoChemistry of Actinides SFA, we are focusing on understanding the processes controlling Pu migration at the disposal site trench 216- Z-9, located at site 200 west. Over 4 million L of Pu-laden reprocessing waste was released directly to the sediments in this trench between 1955 and 1962. The acidic waste stream was rich in organics, high ionic strength and contained a number of different reprocessing related components (*e.g.* actinides, fission products, CCl₄, lard oil *etc.*).

In the Z-9 trench, most of the Pu precipitated immediately within the first decimeters of the trench as PuO₂ and Pu-polymers or -hydroxides. However, a small fraction migrated deep into the subsurface vadose zone to depths of 37 m. A correlation between Pu and organic components was found in some cases, as well as correlation with Fe and P. However, the mechanisms controlling subsurface migration have not been established. The goal of this project is to identify the Pu migration mechanisms and develop predictions for Pu mobility and potential groundwater contamination at environmentally relevant timescales.

Preliminary experiments have shown that the majority of Pu (approx. 99% from a 1×10^{-9} mol/L solution of Pu(IV)) coprecipitates with Al-hydroxides at $\text{pH} \geq 3$ and thus will be immobilized as a function of waste acid neutralization. Ongoing experiments are examining the structural incorporation of Pu into these Al/Fe co-precipitate phases and their stability in terms of Pu retardation, and possible formation of mobile Pu complexes with Al, Fe and/or P.

Furthermore, we are investigating the influence of organic waste components (*e.g.* TBP and its degradation products) on the migration behavior of Pu in the presence of high nitrate solutions in combination with organic solvents (*e.g.* kerosene, dodecane). Pu partitioning behavior is initially being examined in simple batch systems where pH, minerals, organic components, and ionic strength are varied. Knowledge gained from batch experiments will be applied in subsequent more complex column experiments that will mimic environmental conditions using Hanford sediments and an artificial waste solution containing relevant waste components. The goal of this effort is to identify the specific processes controlling the long-term behavior of Pu at the Z-9 trench and establish a conceptual models of actinide contaminant evolution at this site under relevant biogeochemical conditions.