

INVESTIGATIONS OF ACTINIDES IN THE VADOSE ZONE

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The United States Department of Energy (DOE) Savannah River Site (SRS), in collaboration with Clemson University scientists, is conducting field lysimeter experiments at the meso-scale dealing with the long-term fate and transport of radionuclides in the vadose zone. In this experiment, radionuclides are buried in 61 cm long x 10 cm diameter lysimeters packed with soil from the SRS and remain open to precipitation in order to mimic the conditions of the vadose zone. Effluent is collected from the lysimeters quarterly to provide a measure of radionuclide transport. Several Pu lysimeter sources were placed in triplicate to allow for destructive analysis of the source material and concentration profile within the soil after 2, 4, and 10 years. Several sources of Pu in varying oxidation states have been placed in lysimeters for these experiments. The results from destructive coring of the two-year Pu lysimeters are compared here with results from a previous set of Pu field lysimeter experiments conducted under similar conditions with 11 years in the field in the 1980's. In these experiments, soil is digested in concentrated HNO₃ and the liquid phase is analyzed by LSC for total alpha activity (^{239/240}Pu). Select samples were also analyzed by alpha spectroscopy to confirm that ^{239/240}Pu were the major alpha emissions in the samples. These two sets of experiments include various sources of Pu initially in the III, IV, V and VI oxidation states. The transport observed with variable oxidation states between the two sets of experiments qualitatively correlates with the theoretical expectations, IV < VI < III < V. After two years little transport of Pu(V) has been observed and greater than 99% of the Pu appears to have remained within the source. There was increased downward migration of the Pu^VO₂(NH₄)(CO₃)(s) source after just 2 years in the field compared with previously observed transport from Pu(IV) and Pu(III) sources after 11 years in the field. This behavior is consistent with both the enhanced mobility of pentavalent actinides relative to other actinides as well as reduction of Pu(V) to Pu(IV) leading to formation of Pu(IV) surface complexes or Pu(IV) (hydr)oxide precipitates. However, as transport of Pu(V) sources was slightly greater than other oxidation states, it is likely that the slightly enhanced transport observed within the lysimeters is due to transport of Pu as Pu(V) prior to reduction to Pu(IV). In addition, the presence of organic matter decreased the mobility of Pu(V). Although further investigation is needed, this could be due to: (1) enhanced reduction of Pu in the presence of organic matter leading to increased sorption to minerals or (2) complexation of Pu with organic matter bound to the bulk minerals. These field lysimeter observations highlight the importance of Pu reduction as a major factor limiting the environmental mobility of Pu in environmental systems.