Radioiodine/lodine Attenuation Mechanisms in Hanford Groundwater

Brady D. Lee (Brady.Lee@pnnl.gov), Mike Truex, James Szecsody, and Nik Qafoku (Pacific Northwest National Laboratory, Richland, Washington, USA)

Background/Objectives. During a time period lasting from 1944 to 1987, plutonium production and processing at the Hanford Site (Washington State, USA) generated liquid waste containing a variety of hazardous constituents including carbon tetrachloride, nitrate, and chromium, and radionuclides, such as uranium, technetium-99 (⁹⁹Tc) and iodine-129 (¹²⁹I). These waste streams were disposed of in single-shell tanks, as well as pits, cribs, and trenches. Leaking tanks and direct disposal into the subsurface has resulted in the majority of comingled contaminants found in the Hanford groundwater. Radioiodine is of particular environmental interest, due to a long half-life, mobility in groundwater and toxicity. Groundwater monitoring wells east of Waste Management Areas T and TX-TY within the 200-ZP-1 OU have shown ¹²⁹I concentrations exceeding the 1 pCi/L DWS; in some wells ¹²⁹I activity was greater than 20 pCi/L. Iodine speciation in the groundwater is dominated by iodate (~70%), followed by organo-iodine and iodide.

Approach/Activities. Radioiodine plumes in four different groundwater operable units in Hanford's 200 East and 200 West Areas were sampled for iodine speciation, as well as other contaminants. In addition, laboratory studies were performed to look at potential attenuation mechanisms that might explain the different iodine species found in the groundwater. Specific processes studied included, sorption to Hanford sediment, precipitation in carbonate phases and microbially-catalyzed redox cycling. Batch studies were performed and supernatant from the experiments was tested for total iodine, iodate, and iodide. Organo-iodine fractions were calculated from the difference between total and inorganic species.

Results/Lessons Learned. Iodine speciation analysis, indicated that iodate was still the dominant iodine species in groundwater from the radioiodine plume in 200-UP-1 OU. These results support results taken a number of years ago in the 200-ZP-1 OU.

Removal of iodate from solution appears to occur by sorption and precipitation. Late-spike experiments showed less iodate removal from solution than when iodate was added at same time as the calcite-forming solutions. These results suggest that iodate incorporation in precipitates occurred during the calcite precipitation process and late-spike removal of iodate was by sorption. The results of these recent experiments demonstrate iodate precipitation concurrently with calcite precipitation and that the water chemistry affects this process.

For iodide, batch sorption experiments over a range of sediment/water ratio showed very little sorption (Kd < 0.01 mL/g) over a time scale of 46 h (Figure 6). No oxidation of iodide to iodate was observed during these studies. Iodate sorption was difficult to measure due to relatively rapid biotic/abiotic iodate reduction to iodide that occurred in the sediment. Thus, limited batch experimental data could be used to quantify sorption.

When stimulated with simple carbon sources such as organic acids and sugars, bacteria enriched from a ¹²⁹I plume at the Hanford site were shown to be able carry out both oxidation of iodide and reduction of iodate. These results show that bacterial oxidation may be responsible for the prevalence of iodate in Hanford groundwater. In addition, bacterial reduction of iodate, can occur in the presence and absence of nitrate.

These results show that numerous attenuation mechanisms are likely working in parallel to affect fate and transport of radioiodine in the Hanford subsurface. These attenuation mechanisms also represent potential options for remediation of iodine.