Spectroscopic Confirmation of In Situ Biological Reduction of Uranium within Fractured Saprolite

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In situ immobilization of heavy metals, such as uranium, through biological reduction is a promising means for stabilizing contaminants within subsurface sediments. Species of U(VI) are highly mobile in groundwater systems, while those of U(IV) are only sparingly soluble. Stimulation of biological uranium reduction at the field scale presents several challenges, including heterogeneous sediment mineralogy, a complex and evolving community of bacteria, and the presence of multiple electron donors and acceptors. The NABIR Field Research Center at Oak Ridge National Laboratory is additionally complex owing to U concentrations of 1,000 ppm, pH values less than 3.4, and exceedingly high concentrations of nitrate (>0.1 M) and aluminum (>0.01 M).

Here, we present evidence of biological uranium reduction in a series of experiments of increasing complexity. First, uranium reduction was investigated in batch experiments using radionuclide-contaminated sediment from ORNL that were inoculated with a denitrifying bacterial community. Next, a packed column was used to mimic uranium reduction under field conditions. Finally, subsurface biological uranium reduction was stimulated at the field scale.

Successful stimulation of biological uranium reduction was accomplished in all experimental systems. However, even after long-term stimulation of biological activity, approximately 50% of uranium within the solid phase remained oxidized [U(VI)]. We are currently attempting to determine the speciation of the remaining U(VI) through micro-X-ray absorption spectroscopy (μ-XAS). Furthermore, U(IV) appears to be easily oxidized after the cessation of electron donor and exposure to air or other oxidants (NO_3^-). These factors complicate the long-term immobilization of uranium through in-situ stimulation of biological activity.