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)]. Furthermore, uranium(IV) was rapidly 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.

Discerning geochemical and biogeochemical metal reduction through gamma sterilization

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The adsorption of U(VI) and Cr(VI) onto sterilized and nonsterilized soil from the Oak Ridge Reservation was studied to distinguish biogeochemical versus geochemical effects on metal reduction. The Oak Ridge soil under investigation is a saprolite sequence of interbedded weathered shale and limestone with a pH near 7.6. Dried, crushed soil was sterilized using a Cobalt-60 source and a γ-ray dosage of 20kGy. Sterilization was greater than 99.99% successful.

U(VI) and Cr(VI) adsorption was studied through sterile and nonsterile batch shake reactions over a period of 3 days to 3 weeks. Absorption was studied over a U(VI) concentration range from 0.5-10 ppm and a Cr(VI) range from 0.5-50 ppm. Ethanol was used as an electron donor and an organic carbon source in all sterile and nonsterile experiments.

Results indicate that both U(VI) and Cr(VI) reduction decreases in sterilized reactions compared to the nonsterile soils. U(VI) adsorption coefficients (U k_D) decrease by as much as 25% in sterile soils while Cr(VI) k_D decrease up to 50%. The chemical environment and valence state of the adsorbed Cr and U species are being investigated by X-ray absorption spectroscopy (XAS). Investigations are underway to determine whether changes in metal adsorption are due to biogeochemical events or are caused by geochemical changes in the soil resulting from the sterilization technique.