Biotransformations Involved in Sustained Reductive Removal of Uranium in Contaminated Aquifers

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Microbial reduction of soluble U(VI) to insoluble U(IV) shows promise as strategy for *in situ* immobilization of uranium in uranium-contaminated subsurface environments. However, little is known about how this process works outside the laboratory and how to sustain this process for the long periods of time that may be needed to treat extensively contaminated sites.

In a previous study, acetate addition (1–3 mM) to a uranium-contaminated aquifer at a site in Rifle, Colorado, temporarily promoted removal of uranium from the groundwater via microbial reduction of soluble U(VI) to insoluble U(IV). Uranium removal continued while *Geobacteraceae* remained the predominant organisms, but as Fe(III) was depleted near the injection gallery, Fe(III) reducers could no longer out-compete sulfate reducers. Acetate was consumed via sulfate reduction and U(VI) concentrations increased in the groundwater.

In the 2004 field experiment, conditions were changed in an effort to prolong the activity of *Geobacteraceae* and removal of U(VI) from the groundwater. Acetate concentrations *in situ* were increased to levels comparable to or greater than the ambient sulfate concentration (ca. 9 mM) to prevent complete consumption of acetate by sulfate reducers and ensure acetate transport to downgradient areas containing available Fe(III). Bromide tracer data from monitoring well samples indicated delivery of acetate solution to the subsurface across a broad front but at varying concentrations. Loss of U(VI) after 109 days of acetate injection averaged 34%, 40 %, and 13% (n=5) in wells positioned at increasing distances from the injection gallery relative to control wells. However, the amount of U(VI) removed from the groundwater varied significantly from well to well with U(VI) losses as high as 91% observed in some wells. Loss of U(VI) was greatest (58–91%) in wells where both acetate and Fe(III) were still available, promoting the activity of *Geobacteraceae*. There was little or no U(VI) reduction at sites where Fe(III) was depleted and sulfate reduction predominated.

Analysis of the sediments from the field experiment, as well as studies with laboratory sediment incubations, demonstrated for the first time that the loss of U(VI) from the groundwater could be accounted for as an accumulation of U(IV) in the sediments. Surprisingly, a substantial amount (ca. 40%) of the U(VI) in sediments prior to treatment was adsorbed to the sediments. Unlike the soluble U(VI), the adsorbed U(VI) was not reduced over time, but did remain immobilized.

These results demonstrate that it is possible to promote microbial reductive precipitation of uranium in situ, but further research is required to devise strategies so that U(VI) will be reduced under sulfate-reducing conditions as well as under Fe(III)-reducing conditions.