Fate of Uranium During Transport Across the Groundwater-Surface Water Interface

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Discharge of contaminated groundwater to surface waters is of concern at many DOE facilities. For example, at the Savannah River Site (SRS), uranium contaminated groundwater is discharging into natural wetlands. The numerous sharp biogeochemical transitions occurring in wetlands have profound effects on the ultimate fate of redox-sensitive trace metals and radionuclides, including uranium.

The goal of this research is to provide new insights on how plant-induced alterations to the sediment biogeochemical processes affect key uranium reducing microorganisms, uranium reduction, and uranium spatial distribution. We have formulated the following hypotheses. (1) U(VI) discharged from ground- to surface-waters can be immobilized effectively as U(IV) in the sediments at the groundwater-surface water interface. The electron donor required to stimulate the microorganisms capable of reducing U(VI) is provided by wetland plants via their root exudates and root turnover. (2) Oxygen released into the sediments by plants reoxidizes Fe(II), forming iron oxy(hydroxi)des, which provide the bioavailable Fe(III) for long-term bacterial iron-reducing activity, which is key for a sustained biological uranium reduction.

To test the above hypotheses, we operated small-scale wetland mesocosms to simulate the discharge of uranium-contaminated groundwater to surface waters. The mesocosms were operated for 80 days, followed by analysis for solid-associated chemical species, microbiological characterization, micro-X-ray florescence (μ-XRF) mapping of Fe and U on the root surface, and U speciation via X-ray Absorption Near Edge Structure (XANES).

Bacterial numbers including Geobacter sp., Fe(III), as well as total uranium, were highest on roots, followed by sediments near roots, and lowest in zones without much root influence. Results from the μ-XRF mapping on root surfaces indicated a strong spatial correlation between Fe and U, while the analysis via XANES showed that a significant fraction of uranium was reduced to U(IV), indicating that iron cycling in the rhizosphere also results in uranium reduction and immobilization. Under high iron loading, results indicate that co-precipitation of U(VI) with iron might have been an important U removal process.

A series of depth-discrete dialysis samplers were set up in an uranium-contaminated wetland on the SRS to monitor seasonal U dynamics. Vertical profiles of dissolved U are consistent with our laboratory findings, indicating that during the growing season when there is more active iron reduction, the dissolved U in the sediment pore water is significantly lower.