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Uranium Immobilization in Wetland Soils

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In wetlands, which are a major feature at the groundwater-surface water interface, plants deliver oxygen to the subsurface to keep root tissue aerobic. Some of this oxygen leaches into the rhizosphere where it will oxidize iron that typically precipitates on or near roots. Furthermore, plans provide carbon via root exudates and turnover, which in the presence of the iron oxides drives the activity of heterotrophic iron reducers in wetland soils. Oxidized iron is an important electron acceptor for many microbially-driven transformations, which can affect the fate and transport of several pollutants. It has been shown that heterotrophic iron reducing organisms, such as Geobacter sp., can reduce water soluble U(VI) to insoluble U(IV). The goal of this study was to determine if and how iron cycling in the wetland rhizosphere affects uranium dynamics.

For this purpose, we operated a series of small-scale wetland mesocosms in a greenhouse to simulate the discharge of uranium-contaminated groundwater to surface waters. The mesocosms were operated with two different Fe(II) loading rates, two plant types, and unplanted controls. The mesocosms contained zones of root exclusion to differentiate between the direct presence and absence of roots in the planted mesocosms. The mesocosms were operated for several month to get fully established, after which a U(VI) solution was fed for 80 days. The mesocosms were then sacrificed and analyzed 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).

Results showed that 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. This correlation was stronger for the mesocosms with the higher Fe(II) load. Analysis via XANES showed that a fraction (up to $\sim$1/3) of uranium was reduced to U(IV), for mesocosms operated under low iron loading, indicating that iron cycling in the rhizosphere also results in uranium reduction and immobilization. For mesocosms operating under the higher iron loading, the fraction of uranium immobilized as U(IV) was much lower, indicating that uranium co-precipitation with iron might have been the dominant immobilization process.

In parallel to these mesocosm experiments, dialysis samplers have been deployed at the Savannah River National Laboratory near a creek with uranium contamination, to determine dissolved species, including Fe(II) and U(VI) in these wetland soils and their seasonal variability. The results show that there is a strong seasonal variability in dissolved iron and uranium, indicating a strong immobilization during the growing season, which is consistent with the mesocosm experimental results that the rhizosphere iron and uranium cycling are closely linked.