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Elemental Content, U Redox Dynamics, and Microbial Communities in Wetland Sediment Cores from Tims Branch, Savannah River Site

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The Argonne SBR SFA is transitioning to the study of wetland hydrobiogeochemistry with a focus on a riparian wetland field site (Tims Branch) at the Savannah River Site in South Carolina. As a basis for understanding current conditions at the site, sediment cores (30 cm) and stream water were collected for analysis of elemental distributions, U speciation, and microbial community composition. To minimize changes to the native redox state within the sediment, cores were sealed and shipped to Argonne immediately after collection and were sectioned under anoxic conditions within hours of arrival the following day. The sediment profile consisted of a dark brown, organic layer (OL) on top of an organic matter depleted, mineral layer (ML). The OL was enriched in U (44.5 ± 17.1 ppm) relative to the ML (6.38 ± 2.19 ppm). U L_{III}-edge XAFS spectroscopy indicated that in the unsaturated portion of the OL, ~80% of the U was present as U^{VI}. In the saturated portion of the OL, however, >95% of the U was present as mononuclear U^{IV} that rapidly oxidized when exposed to air: 70% U^{VI}/U_{total} was observed within three hours of exposure and > 95% after one month. U in the ML was predominantly U^{VI}. The cores contained diverse microbial communities, dominated by sequences from the phyla *Proteobacteria*, *Nitrospira*, *Chloroflexi*, and others. The stream water contained abundant reddish-brown flocs, consisting of Fe in the form of ferrihydrite (83%) and lepidocrocite (17%) as determined by Fe K-edge EXAFS spectroscopy. The flocs contained 320 ppm U, suggesting that they may provide a significant transport vector for U within the stream.

Stream water and core material were used to construct microcosms examining the effect of redox conditions (oxic versus anoxic) and C inputs (native C with and without a glucose amendment) on U speciation. Sediments incubated for 30 d under anoxic conditions showed >95% reduction to mononuclear U^{IV} regardless of the C input, whereas microcosms incubated under oxic conditions showed >95% U^{VI} regardless of C input. Anoxic microcosms generally showed an increase over time in fermentative (e.g., *Aeromonadaceae*, *Clostridiaceae*, and *Comamonadaceae*) and metal-cycling (e.g., *Geobacter* and *Geothrix*) organisms. Oxic microcosms were dominated by *Acidobacteriaceae*, *Sphingomonadaceae*, and particularly *Chitinophagaceae* sequences, which account for 20-40% of the total community in some microcosms.

Together these results demonstrate the potential for dramatic changes in U speciation/mobility with changes in the redox status of sediments at the Tims Branch site.