Effects of microbial communities on uranium oxidation and mobilization in the presence of nitrate, nitrite, and oxygen

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Throughout the upper Colorado River Basin, uranium (U) persists as a legacy contaminant of former ore processing activities. Elevated solid-phase uranium levels exist in fine-grained, organic-enriched sediments. Coupled with seasonal groundwater fluctuations that alter the subsurface redox conditions, recent evidence suggests this resupply of uranium may be controlled by biologically-produced nitrite and nitrate. Previous researchers have posited that nitrate and nitrite can oxidize U(IV). We hypothesize that when seasonal groundwater levels recede and the subsurface system becomes anoxic, the nitrate/nitrite diffuses into the reduced interiors of organic-rich sediments and becomes readily available for denitrification, the stepwise anaerobic reduction of nitrate/nitrite to dinitrogen gas. Denitrification may then be coupled to the oxidation of sediment-bound U(IV), forming mobile U(VI), allowing it to resupply uranium in the aquifer. Although both abiotic and biotic uranium oxidation have been documented, the rate of uranium oxidation coupled to bacterial nitrate reduction far exceeds observed abiotic oxidation rates. Thus, nitrogen-cycling microbial communities, specifically those involved in denitrification, may be key to understanding uranium mobility and long-standing environmental contamination. However, a paucity of knowledge exists regarding denitrifying communities in the subsurface.

Here we investigate how the indigenous microbial community from fine-grained, organic-enriched sediments responds to an oxidation event. Using flow-through column reactors packed with sterilized or unsterilized fine-grained sediment from a uranium plume at Riverton, WY, we simulated a laboratory-scale oxidation event by stimulating the columns with an oxidant (either 1mM nitrate, 100μM nitrite, or 268μM oxygen) for 24 days. The objectives of this study were to (1) identify how the type of oxidant introduced affects the denitrifying microbial community from Riverton, WY, and (2) assess over what timescales and to what extent do nitrate, nitrite, and oxygen oxidize uranium in reduced sediments. Preliminary uranium effluent data suggests that uranium mobilization occurred within the first five days and was greatest in the oxygen-stimulated columns. Uranium speciation results show that oxidation proceeded near the influent in the nitrite- [U(VI): 42%] and nitrate- [U(VI): 18%] amended columns; however, in the oxygen-amended columns uranium oxidation was greater [U(VI): 77-82%] and occurred throughout the entire column. Therefore, oxygen was the most efficient oxidant of uranium within this system and although, the nitrate- and nitrite-amended columns resulted in changes in microbial diversity and abundance, less uranium oxidation and mobilization was observed. These results provide important insights into the denitrifying community dynamics and suggest the dominant control over U(IV) oxidation is the oxidant introduced.