Diffusion-limited controls on U, Fe, and S cycling in organic-rich sediments

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Natural attenuation has been used as a remediation strategy at Uranium Mill Tailings Radiation Control Act (UMTRCA) Sites throughout the United States. At some of these sites, such as the Old Rifle Site in Colorado, uranium plume concentrations have not decreased at predicted rates. Accurate groundwater uranium predictions are necessary for long-term monitoring and evaluation groundwater plumes like Old Rifle.

Thin lenses of fine-grained, organic-rich sediments have been identified at the Ole Rifle Site and are characterized by elevated concentrations of organic carbon and trace metals. These Naturally Reduced Zones (NRZs) contain one third of the sediment organic carbon and up 95% of the total uranium in the aquifer. The formation of NRZ sediments and subsequent accumulation of uranium is not fully understood and is necessary for determining the long-term stability of sediment-associated uranium.

In this study, we simulate the development of NRZ sediments in diffusion-limited aggregates to investigate the physical and chemical conditions required for NRZ formation. Sediment aggregates were inoculated with the natural microbial biota from the Rifle aquifer and population dynamics monitored by 16S RNA analysis. Effects of sediment particle size and concomitant tortuosity in combination with particulate organic carbon (POC) loading on Fe, S, and U cycling on molecular and nano-scale are investigated with synchrotron-based extended X-ray absorption fine structure (EXAFS) spectroscopy.

The development of a homogenous reduced zone within highly porous, large pore-size sediment aggregates is contrasted by heterogeneous development of reduced zone within the finest grain sediments suggesting the importance of understanding the diffusion-limited system. 2-D X-ray elemental mapping of sediment cross sections illustrate the association between U, Fe, and S in relationship to POC.

The organic-rich NRZs within the aquifer have accumulated U and may in turn have become a source of uranium to the groundwater contributing to the plume persistence at the Old Rifle Site.