

## Fe and S redox cycling during a biostimulation episode at the Old Rifle, CO aquifer

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*In-situ* bioremediation studies have shown that adding electron donor to groundwater stimulates the activity of metal- and sulfate-reducing bacteria, profoundly impacting the cycling and speciation of redox-sensitive elements such as iron and sulfur, and changing the redox status of contaminants such as U(VI) to insoluble U(IV) products. While numerous studies have examined the impact of bioreduction in laboratory-based studies, very few have examined Fe and S speciation changes at the molecular to pore scales *in situ* in a biostimulated aquifer. In this study, we used in-well columns to obtain direct access to sediment speciation at different time points during a bioremediation experiment at the Old Rifle site, CO, transitioning dominantly from iron-reducing to sulfate-reducing regime.

We investigated Fe and S speciation at the molecular scale using X-ray absorption spectroscopy (XAS). Chemical extractions of reduced sediments revealed the rates of Fe(II) and sulfide formation. Bulk Fe EXAFS showed a significant change of Fe redox status with ongoing bioremediation. Results provide evidence for the reduction of the entire S pool to monosulfide and elemental sulfur. The speciation of FeS grain coatings that appear under sulfate-reducing conditions was probed using electron microscopy and micro-XAS. Results showed elemental sulfur accumulating in the coatings which appear to also concentrate uranium.