Sediments in Deep Fracture Networks Reveal Divergent Sulfur Cycles

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Chemical and mineralogical characterization of subsurface environments coupled with thermodynamic modeling will suggest metabolic niches that microorganisms can occupy, while genomic data collected from these organisms highlight their metabolic capabilities and their realized niches. Reduction and oxidation reactions involving iron and sulfur are especially important sources of energy to the anoxic and low-carbon environments of the deep continental biosphere. However, microbially mediated reactions compete with abiotic reactions catalyzed by minerals and aqueous chemical species. We sought to better understand the iron and sulfur cycles in isolated groundwaters within a 2.7 Ga banded iron formation (BIF) in northern Minnesota. Boreholes access fracture networks within the BIF and reach more than 700 m below the surface. We sampled groundwaters flowing from these boreholes and measured major cations and anions to characterize the water chemistry. Additionally, we sampled sediments along borehole walls tens or hundreds of meters into the boreholes. Sediments were characterized via X-ray diffraction (XRD) and scanning electron microscopy (SEM) and were taken to the scanning transmission X-ray microscope (STXM) at the Advanced Light Source for imaging and C and Fe X-ray absorption near-edge structure (XANES) spectroscopy. Microbial DNA was extracted from filtered borehole waters and borehole sediments; it was processed using a shotgun metagenomic sequencing approach. Metagenomes were probed for genes involved in iron and sulfur cycling. While the water chemistry of both boreholes was similar, the sediment in one borehole contained a mixture of the iron sulfide minerals mackinawite and greigite (reduced iron and sulfur) while the other borehole sediment contained only hematite (oxidized iron) and quartz. We theorize that hematite is oxidizing aqueous sulfide before FeS can precipitate, producing sulfur intermediates which fuel microbial chemosynthesis. Conversely, the sulfidic sediment may act as a sink, trapping both iron and sulfur in less-reactive mineral forms. Imaging of the sediment via SEM and STXM confirmed that microorganisms have colonized these sediments, and analysis of the borehole metagenomes confirmed that the microbial community compositions are statistically distinct. Further analysis of genes involved in sulfur cycling will provide a clearer picture of microbial and abiotic sulfur and iron cycles in the continental subsurface.