

Cryptic sulfur cycling coupled to iron and carbon – using metagenomics to inform environmental impact

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The biogeochemical cycling of iron (Fe) and carbon (C) exerts great control over the fate and transport of contaminants and nutrients in terrestrial and aquatic environments. There is increasing knowledge that sulfur (S) biogeochemical cycling, particularly in environments characterized by dynamic redox gradients that promote hotspots and hot moments of biogeochemical processes, can be coupled to Fe and C cycles that further influence contaminant fate. The environmental impact of the S cycle, however, is challenging to resolve using geochemical methods alone, as there can be fast cycling through highly reactive intermediate-valence S forms that may not be readily measured. Further complicating our understanding of the S cycle, the inorganic S cycle can be inextricably linked to the organic S cycle, of which the latter is largely overlooked and poorly resolved. To better understand coupled S-Fe-C biogeochemical cycling and resolve some of the “hidden or cryptic” S reaction pathways, we used field-based approaches including hydrogeologic flux measurements and sampling of water and sediments for shotgun metagenomic and geochemical analyses combined with reactive transport modeling using a PFLOTRAN code that incorporates “cryptic” S cycle reactions. In the sediments of a freshwater stream and wetland system, metagenomes unexpectedly revealed that thiosulfate oxidation to sulfate was a ubiquitous and predominant process, even under anaerobic conditions. When including these and other key reactions into our reactive transport model, we more accurately predicted the measured S geochemistry. Model simulations of a fluctuating flux conduction helped further explain the high level of aqueous Fe in the environment. In a study of the deep terrestrial subsurface, metagenome analysis revealed the significant contributions of organosulfur cycling. We anticipate that a metagenomics-informed modeling framework that includes key organosulfur reaction pathways could further enhance our ability to resolve these complex cycles and better understand and predict nutrient and contaminant fate and transport.