A geochemical and microbiological analysis of nitrate reduction at a hydrothermal vent and at a cold seep

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Nitrogen cycling in extreme environments is very poorly constrained even though N may be limiting in some systems. Cold seep and hydrothermal environments are characterized by advection of reduced substrates from the deep subsurface. Other than sulfate, acceptors for oxidation of these reduced substrates are not well characterized. Nitrate is abundant in bottom water (~30 μM) overlying cold seeps and hydrothermal systems, where it diffuses into sediments thereby providing an electron acceptor. To date the most significant proportion of dissimilatory nitrate reduction has been attributed to the activity of Vacuolate Sulfide Oxidizing Bacteria (VSOB). Through a combination of geochemical and microbiological tools, we show denitrification to N_2 to be the dominant nitrate reduction pathway in both VSOB hosting sediments and those without VSOB from the Mississippi Canyon, Gulf of Mexico and the Guaymas Basin, Gulf of California. In these sediments up to ~50% of nitrate was denitrified to N_2. Other pathways of nitrate reduction (i.e. reduction to ammonium and anaerobic ammonium oxidation) were less important. Rates of N_2 production were substantially lower in sediments that did not host VSOB (by 30-60%), and the activity of VSOB was suppressed by reducing the sulfide supply. At Mississippi Canyon, sediment nitrate turnover rates was on the order of days, which is comparable to sulfate turnover in relatively active cold seep sediments. Denitrification appears to be a vigorous process within these extreme environs, and may play an important role in nitrogen and carbon cycling.

Distinct uranium(IV) products result from uranyl reduction in different ferrous-ferric oxyhydroxide systems

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Iron-reducing conditions are often found in subsurface environments due to the presence of bacteria that respire on Fe(III) minerals in the absence of oxygen. The metabolic products of dissimilatory iron reducing bacteria (DIRB) can include soluble and adsorbed Fe(II), Fe(II) minerals, mixed-valence Fe(II)-Fe(III) oxyhydroxide salts (green rusts), and magnetite, and these phases can provide a reservoir of reducing capacity even after bacterial activity has ceased. There has been sustained interest in studying the reduction of U(VI) to U(IV) in subsurface environments, both from a fundamental and an engineering perspective. Uranium(VI) is relatively soluble and mobile, whereas uranium(IV) has low solubility in aqueous solutions and precipitates. The mechanisms that couple the oxidation of Fe(II) respiration products to the reduction of U(VI), as well as the resulting U(IV) phases are not very well understood.

Using X-ray Absorption Fine Structure spectroscopy (XAFS) we have examined a number of abiotic Fe(II)-containing systems in relation to their ability to reduce U(VI). Systems span the range of formal valence between Fe(II) and Fe(III), including aqueous Fe(II), Fe(II) adsorbed to carboxyl surfaces, synthetic green rusts with different interlayer anions, stoichiometric magnetite, and a range of magnetite-structure Fe minerals with decreasing Fe(II) content. Results indicate reduction of U(VI) to U(IV) in all systems where Fe(II)-O_6 octahedra are connected, with decreasing amounts of reduced U(IV) in the Fe(II)-poor magnetite series. Systems containing aqueous and adsorbed Fe(II)-O_6 monomers did not reduce U(VI). In the systems where U(VI) reduction did occur, the U(IV) products varied between systems and included: larger uraninite (UO_2) particles, nanoparticulate uraninite, and monomeric U(IV)-O_8 hexahedra embedded in another mineral. The environmental implication of these results is that the mobility and the reoxidation susceptibility of these distinct U(IV) phases may be quite different.