Formation of soluble organic-Fe(III) complexes during microbial iron reduction

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Iron is the fourth most abundant element in the Earth's crust and plays an essential role in the biogeochemical cycling of many elements on Earth. Microbial Fe(III) reduction is a relatively recent addition to the suite of anaerobic respiratory processes carried out by microorganisms. Microbial Fe(III) reduction is central to a wide variety of environmentally significant processes including the biogeochemical cycling of Fe, Mn, trace elements, and phosphate, degradation of organic matter, weathering of Fe(III)-containing clays and minerals, and biomineralization of Fe(II)-bearing minerals such as magnetite. Compared to the wealth of knowledge existing on the molecular mechanisms of aerobic respiration, denitrification, sulfate reduction, and methanogenesis, little is known about microbial Fe(III) reduction. Ironically, recent microbiological and geological evidence indicates that dissimilatory Fe(III) reduction may have been one of the first respiratory processes to have evolved on early Earth.

In this study, we used batch reactors and mercury-gold voltammetric microelectrodes to monitor the reduction of iron oxides and soluble Fe(III) in the presence of *Shewanella oneidensis* strain MR-1. Results show that electrochemically active soluble organic complexes of Fe(III) are formed during the reduction of iron oxides as well as during the reduction of a non-electroactive soluble Fe(III) model compound (i.e. ferric citrate). These data suggest that *S. oneidensis* releases organic ligands to solubilize Fe(III) oxides prior to reduction. Experiments performed with *S. oneidensis* mutants lacking the Type II protein secretion system that is postulated to transport the Fe(III) reductase to the outer membrane display completely different results. The implications of such findings on the mechanisms of microbial iron reduction and the existence of soluble organic-Fe(III) complexes in aquatic systems will be discussed.

Microbial Fe cycling in deep regolith

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Traditionally, most researchers have considered deep saprolitic regolith to be largely devoid of life due to a lack of bioturbation and organic carbon. Recently, a few researchers have documented a diversity of organisms in saprolite but the feedbacks between life and geochemistry in deep regolith remain unknown. We have completed microbial cell counts, DNA extractions, culture tests, and analyses of chemistry, moisture, and particle size on two saprolite cores augered in the Rio Icacos watershed in the Luquillo Experimental Forest (LEF), Puerto Rico. The 2–8 m of saprolite caps quartz diorite and is overlain by 0.5–1.0 m of soil. The bedrock-saprolite interface is a 20-60 cm-thick zone that weathers spheroidally. Direct cell counts for samples collected from 0-5 m were ~10^6–10^{10} cells g^{-1}. Cells with morphologies typical of the autotrophic iron oxidizer *Gallionella ferruginea* were identified. Culturable cell counts were <10^3–10^7 cfu g^{-1}. Cell numbers calculated from spectroscopic densities of extracted DNA were consistent with the direct cell counts. Total, culturable, and DNA-based counts indicate that cell densities decrease with depth over the upper several meters, but near the bedrock interface zone, we consistently observe an increase in cell densities. This increase does not correlate with moisture, organic carbon, or total Fe, but does correlate with HCl-extractable Fe, suggesting that cell densities are responding to availability of Fe at depth. Culture tests indicated aerobic iron-related bacteria (IRB) at <0.6 m, at 2-2.5 m, and at the bedrock-saprolite interface (~5 m). Based on the rate of advance of the bedrock-saprolite interface, the Fe(II) oxidized during weathering, the density of IRB at the interface, and the biomass yield per mole Fe(II) oxidized, we can calculate the rate of biotic Fe oxidation at the interface. Bacteria are an integral component of Fe cycling in the saprolite as they respond to the availability of Fe and also contribute to weathering by oxidizing Fe. Furthermore, the presence and activity of bacteria at the bedrock-saprolite interface may indicate a biological role in the disaggregation of bedrock and the formation of saprolite.