

Characterization of forms of sulfur that predominate during S reduction with enzymes, in microbial cultures and in sulfidogenic environments

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The ability of organisms to grow while respiring S_0 are widespread throughout the prokaryotes, especially in geomicrobiologically relevant organisms commonly found in sediment, hydrothermal and deep subsurface environments. Data from the mass-independent fraction of sulfur isotopes suggests that sulfur reduction was actually the predominant form of respiration on the early earth, making it one of the oldest energy-conserving strategies for life, while recent data suggests that on the modern earth rates of microbial sulfide production from elemental sulfur are a major factor in the overall chemical cycling of sulfur, which is largely unexplored.

At first glance elemental sulfur may seem to be a simple compound, as it is usually found in the form of a relatively insoluble S_8 ring, which can be reduced to soluble polysulfide (S_n^{2-}) or sulfide (S^{2-}). In reality, however, sulfur can form a range of different allotropes and can come in the form of particles with very different sizes and characteristics, the speciation of which will have a significant effect on the availability of this sulfur to microbes, enzymes, and abiotic reactants. In the studies described here we use cyclic voltammetry with solid-state mercury/gold amalgam electrodes to detect the sulfur compounds that form during the enzymatic and/or microbial reduction of sulfur. During these reactions, and in artificial sediment systems with large amounts of cycling sulfur, we observe high concentrations of "soluble" elemental sulfur that is electrochemically distinct from insoluble S_8 . The sulfur compounds observed are analogous to various nanoparticulate forms of sulfur that we have generated abiotically and characterized in the laboratory. These results suggest that chemically and structurally distinct forms of nanoparticulate sulfur are important in the cycling of zero-valent sulfur compounds, and control access of these compounds to enzymes and microbes.