

Does organic sulfur make a significant and overlooked contribution to sedimentary S cycling in low sulfate environments?

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Microbially mediated sulfate reduction mineralizes organic carbon, generates hydrogen sulfide, and mediates the geochemical cycles of iron, phosphorus, and other elements. Under the prevailing paradigm, the sulfate for this process is assumed to be supplied into sediments by diffusion from the overlying water column. Under low-sulfate conditions, however, a significant or even dominant contribution may come from organic sulfur that is buried into the sediment with the deposited organic matter. In deeply oxygenated sediments of Lake Superior, for example, the concentrations of porewater sulfate below the sediment-water interface are higher than in the overlying water column, and the sediment serves as a source rather than a sink of sulfate to the water column. Under such conditions, activities of sediment sulfur-reducing microbes must be supported entirely by the in-sediment production of oxidized S compounds from organic S. Modeling indicates that contributions from organic sulfur may be pervasive in environments such as oligotrophic freshwater lakes or the Earth's oceans during certain past geologic periods. The organic sulfur compounds buried into the sediment below the depth of sulfate penetration may also potentially support cryptic biogeochemical cycling there, affecting in particular the redox cycling of iron. Drawing on literature data compilation, we make modeling estimates for the contributions of organic S to sediment sulfate reduction in different types of sediments, simulate the expected geochemical and isotopic signatures of such contributions, and discuss current knowledge gaps.