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Sulfidic Conditions Transform Sinking Marine Particles

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The sulfur isotope compositions recorded in sediments and rocks describe overlapping global and local processes and their impacts on the biogeochemistry of sulfur, iron, and carbon. One major gap in our understanding of sedimentary sulfur-isotope records is that we do not fully understand when, where, and how organic sulfur forms in the environment. We recently demonstrated that organic matter in sinking marine particles can sulfurize within days under the anoxic conditions in modern marine O_2-deficient zones. Here, we follow up on that result by investigating how those same marine particles might be transformed if they were exposed to more strongly reducing, sulfidic conditions, such as those believed to generate organic S records in many ancient rocks. We exposed particles to environmentally relevant sulfidic conditions (20°C, 0.5 mM (poly)sulfide) for 48 hours and characterized the organic and inorganic products in detail using the S and C isotopes, elemental ratios, and micron-scale maps of the redox speciation of iron and sulfur (by x-ray absorption spectroscopy / x-ray fluorescence at the Stanford Synchrotron Radiation Lightsource).

Sulfurization roughly tripled the relative abundance of sulfur in both hydrolysable and hydrolysis-resistant organic materials, reaching values similar to those found in black shales and anoxic margin sediments (~3.3 mol% S:C). At the same time, approximately 25% of the newly formed solid-phase sulfur accumulated as pyrrhotite (FeS), and polysulfide oxidation to sulfate was negligible. Organic matter sulfurization products are primarily alkyl sulfides and disulfides, spatially and compositionally distinct from controls. Both organic and inorganic sulfides accumulated in a diffuse and ubiquitous matrix surrounding original biogenic materials, suggestive of EPS. Initial sulfurization products may be further transformed into a more stable form for long-term preservation, which could impact both speciation and S-isotope compositions. Although additional work is needed to connect organic S pools to long-term reservoirs, these experiments demonstrate that OM-rich particles that encounter sulfidic conditions in the environment should be expect to sulfurize rapidly, and that particle-derived organic S may preserve specific information about the distribution of S isotopes from the water column.