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Formation of Dissolved Organic Sulfur in Sediment Porewaters Under Different Redox Conditions

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The formation of dissolved organic sulfur (DOS) in marine sediments garners significant interest due to its role in enhancing the preservation of organic matter and the low-temperature formation of protokerogen. Recent studies reveal that the carbon to sulfur (C/S) ratio of solid-phase extracted DOS (SPE-DOS) from sulfidic sediment porewaters ranges from 18 to 53, suggesting a notably higher sulfur content in this dissolved organic matter (DOM) compared to phytoplankton biomass (C/S = 99) and SPE-DOS from the marine water column (C/S = 213 to 294). While both biotic and abiotic sulfurization mechanisms are responsible for the formation of DOS in porewater, the molecular structure of biotic organic sulfur and the mechanisms of abiotic sulfurization are not fully understood. In this study, we coupled liquid and ion chromatography techniques with a Fusion Orbitrap mass spectrometer to chemically characterize SPE-DOS in sediment porewaters collected from the top ~300 cm of the sediment column at four sites in the central and southern California continental margin. The sediments at these sites are fine-grained and relatively organic-rich yet vary in their degrees of oxygenation, bioturbation and bioirrigation, and organic carbon (OC) burial efficiency. Leveraging MS/MS fragmentation structural databases, we identified over 30 DOS microbial metabolites, offering new insights into the biotic contributions to the DOS pool. Furthermore, our innovative use of 3-D molecular network analysis sheds light on the abiotic addition of H_2S and H_2S_2 to DOM in sediment porewaters, pinpointing the functional groups of dissolved organic compounds involved in sulfurization. Notably, while many microbial DOS metabolites were common across all sites, the patterns of sulfurization differed significantly, suggesting site-specific drivers of sulfurization pathways and controls at the molecular level. This study advances our understanding of the molecular mechanisms and environmental factors influencing DOS formation in marine sediments, with implications for global sulfur cycling and organic matter preservation in marine environments.