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Multiple sulfur isotope constraints on microbial sulfate reduction in methane seep environments

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Microbial sulfate reduction is a key metabolic pathway on Earth that significantly impacts the global sulfur cycle by reducing sulfate to sulfide. Marine sediments rich in organic matter and methane along continental margins are global hotspots of microbial sulfate reduction. Depending on the electron donor available, sulfate reduction primarily occurs in two modes: organoclastic sulfate reduction (OSR) and sulfate-driven anaerobic oxidation of methane (SD-AOM). The hydrogen sulfide released during both processes can react with reactive iron, ultimately leading to the formation of pyrite in the sediments, which serves as a crucial archive for reconstructing microbial sulfur metabolism. However, distinguishing between OSR and SD-AOM based solely on the $\delta^{34}S$ values of pyrite is often challenging. This difficulty arises because both processes produce a wide range of isotope fractionations, and variable intensities of methane seepage can further complicate the diagenetic history.

Advances in multiple sulfur isotope analysis ($\delta^{34}S$, $\Delta^{33}S$, $\Delta^{36}S$) provide a useful tool for identifying specific microbial processes such as sulfate reduction, sulfur disproportionation, and sulfide oxidation. Such advances also allow us to effectively trace the mixing of different sulfur reservoirs in sediments. Here, we analyzed and compiled multiple sulfur isotope data ($n > 400$) for authigenic pyrite from modern and ancient methane seep environments, including the South China Sea, the Sea of Marmara, the North Sea, the Baltic Sea, and the Black Sea. Generally, in environments characterized by a high methane flux (i.e., methane seeps), where SD-AOM is the dominant pathway, pyrite exhibits $\delta^{34}S$ values ranging from -50‰ to 20‰ and $\Delta^{33}S$ values from -0.05‰ to 0.16‰. In contrast, in sediments with low methane flux, where both OSR and SD-AOM are active, pyrite shows much wider variations in both $\delta^{34}S$ (-50‰ to 60‰) and $\Delta^{33}S$ (-0.15‰ to 0.25‰) values. The differences between these isotopic patterns are not only controlled by methane flux, but also by the extent of OSR at shallower depth above the sulfate-methane transition zone. Our results highlight the value of multiple sulfur isotope composition of pyrite as a proxy for characterizing the different modes and intensities of microbial sulfate reduction during early diagenesis.