

Oxygen isotopic composition of sulfate in deep sea pore fluid: Evidence for rapid sulfur cycling

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Changes in the major element and related isotope profiles in porewaters of organic-rich sediments suggest that various microbial processes using a succession of electron acceptors are in play during the remineralization of organic matter. Of the electron acceptors, bacterial sulfate reduction (BSR) is responsible for most organic matter remineralization in sediments. In addition, nearly all the methane produced during methanogenesis below the sulfate minimum zone is oxidized anaerobically through sulfate reduction (anaerobic methane oxidation (AMO)). In places where AMO occurs, recent studies have demonstrated that the majority of the sulfate is reduced by methane. When sulfate is reduced through OMO or AMO the sulfur isotope profile of the residual sulfate pool ($\delta^{34}\text{S}_{\text{SO}_4}$) increases monotonically from seawater values of 20‰ to values as high as 60 or 70‰, reflecting the kinetic isotope fractionation for sulfur isotopes associated with BSR. We have measured oxygen isotopes in marine sulfate ($\delta^{18}\text{O}_{\text{SO}_4}$) in pore fluid profiles through organic-rich deep-sea sediments from eleven ODP sites around the world. In almost all sites studied sulfate is depleted with depth, through both OMO and AMO. The $\delta^{18}\text{O}_{\text{SO}_4}$ increases rapidly near the top of the sediments, from seawater values of 9‰ to maxima between 22 and 25‰, and remains isotopically heavy and constant at these values with depth. The $\delta^{18}\text{O}_{\text{SO}_4}$ in these pore fluid profiles is decoupled from the $\delta^{34}\text{S}_{\text{SO}_4}$ measured on the same pore fluid samples. This isotopic decoupling between the $\delta^{34}\text{S}_{\text{SO}_4}$ and the $\delta^{18}\text{O}_{\text{SO}_4}$ is hard to reconcile with the traditional understanding of bacterial sulfate reduction in sediments. Our data support the idea that sulfate or some sulfur intermediate and water isotopically exchange during sulfate reduction and that some of the isotopically altered sulfur pool returns to the environment. The rapid increase in the $\delta^{18}\text{O}_{\text{SO}_4}$ in the upper part of these sediments requires rates of this oxygen isotope exchange that are several orders of magnitude higher than the rates of net sulfate reduction calculated from the sulfate concentration profiles and the $\delta^{34}\text{S}_{\text{SO}_4}$. We will explore several mechanisms by which this may occur, including “net-zero” sulfur



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as well as further experiments through which
we can test and resolve these processes.