

From rift to off-rift: The sulfate cycle in the oceanic crust

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The formation of anhydrite (CaSO_4) has been shown to be critical for sulfate sequestration in the oceanic crust. Although the mechanism of anhydrite formation is well understood, the largest enigma – the fate and whereabouts of anhydrite and seawater-sulfate in the oceanic crust – remains a subject of debate. At mid-ocean ridges (MORs), seawater carrying dissolved sulfate (SO_4^{2-}) infiltrates the oceanic crust. However, hydrothermal fluid emissions from such systems predominantly contain H_2S with much lower $\delta^{34}\text{S}$ than seawater. The absence of SO_4^{2-} may be explained by the reduction of seawater-sulfate to H_2S and/ or by the immediate precipitation of anhydrite upon temperature increase. These contrasting hypotheses highlight the need to explore sulfate cycling in the oceanic crust.

Here, we combine a geochemical isotope modelling approach with natural datasets of chemical and stable isotope ($\delta^{34}\text{S}$, $\delta^{18}\text{O}$) compositions of anhydrite and pyrite from various locations along the submarine and on-land section of the Mid-Atlantic ridge to quantify the key variables that control the sulfate cycle in the oceanic crust. Comparison of the natural dataset with results from geochemical isotope modelling revealed that $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of anhydrite and pyrite were dependent on the isotope composition of the source fluid, extent of water-rock interaction, temperature, and redox conditions. Quantitative formation of anhydrite mainly occurred at temperatures $<150^\circ\text{C}$, whereas at elevated temperatures ($>200^\circ\text{C}$) reduction of seawater-sulfate to H_2S and subsequent pyrite precipitation were found to limit anhydrite formation.

Extending our calculations to the oceanic crust revealed that the majority of seawater-sulfate is sequestered into anhydrite in vicinity of MORs at $<200^\circ\text{C}$ at shallow depths (<1500 m), with only a small portion discharged by high-temperature hydrothermal vents. Sequestration of sulfur by anhydrite is not long-lasting due to retrograde dissolution of anhydrite as oceanic crust cools as it is rifted away from the spreading centers. The removal of anhydrite upon cooling and aging of the crust may result in a return back to the oceans of up to 60% of the sulfur originally sequestered in anhydrite upon hydrothermal alteration in vicinity of MORs.