

Multi isotopic ($\delta^{56}\text{Fe}$, $\delta^{34}\text{S}$, $\Delta^{33}\text{S}$) constraints on the origin and mineralization of sulfide in the oceanic crust

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Within the oceanic crust, sulfur originates from the mantle through volcanism and degassing as well as from seafloor alteration, sedimentation and microbial processes. Thereby, sulfide is the predominant oxidation state of sulfur in fluids and mineral deposits.

Here, sulfide-formation processes in the Icelandic crust – adopted as an analogue for the oceanic crust – were assessed using coupled $\delta^{34}\text{S}$ and $\Delta^{33}\text{S}$ in hydrothermal fluids and $\delta^{56}\text{Fe}$ and $\delta^{34}\text{S}$ systematics in sulfides. The hydrothermal fluids studied had a wide range of reservoir temperatures and salinity (~50–300 °C, Cl = ~20–21,000 ppm). The $\delta^{34}\text{S}$ and $\Delta^{33}\text{S}$ values for fluids were highly variable: $\delta^{34}\text{S-S}^{\text{II}} = -11.6$ to 10.5‰, $\delta^{34}\text{S-S}^{\text{+VI}} = -1.0$ to +24.9‰, $\Delta^{33}\text{S-S}^{\text{II}} = -0.12$ to 0.00‰, $\Delta^{33}\text{S-S}^{\text{+VI}} = -0.04$ to 0.02‰. Pyrite was the predominant sulfide mineral with $\delta^{34}\text{S} = -13.1$ to +19.7‰ and $\delta^{56}\text{Fe} = -1.48$ to +1.96‰.

In order to constrain the origin and reactions involving sulfur and iron, isotope geochemical modeling (PhreeqC and IsoGEM) was applied taking into account aqueous and gas speciation, phase separation, redox reactions and mineral-fluid interaction as well as the associated isotope fractionations. Comparison of the modeling results with the dataset revealed that the large isotope variations observed are consistent with multiple sources of sulfur and iron. Leaching of mantle-derived rocks with minor seawater contribution, is the main source of sulfur and iron. Additionally, close-to-equilibrium isotope fractionation between aqueous species, gases and secondary minerals also play an important role in controlling the S and Fe isotopic composition of the oceanic crust.