Paleoenvironmental constraints on the mechanism of Archean mass-independent sulfur isotope fractionation

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Despite more than fifteen years of research, the exact mechanism responsible for the production of mass-independent fractionation of sulfur isotopes (S-MIF) in Archean sedimentary rocks remains unclear. There are strong indications that S-MIF production is related to photolysis of SO_2 or other S-bearing molecules, but neither photolysis experiments nor calculations using the best available SO_2 absorption cross-sections have been able to convincingly reproduce observed $\delta^{34}S$-$\Delta^{33}S$ and $\Delta^{33}S$-$\Delta^{36}S$ relationships. In addition, the geologic record of S-MIF presents several unresolved phenomena, such as an apparent asymmetry between positive and negative $\Delta^{33}S$ magnitudes, a net positive value of preserved $\Delta^{33}S$, and disagreement bewteen estimates of seawater sulfate $\Delta^{33}S < 0$ from barite and volcanogenic massive sulfide deposits, and $\Delta^{33}S > 0$ from carbonate-associated sulfate.

If interpreted in light of insight into the biogeochemistry of S-bearing compounds, and an understanding of the preservation potential of marine sedimentary environments, these complexities of the geologic S-MIF record may be turned around to constrain the S-MIF formation mechanism(s) and to resolve some of the outstanding questions outlined above. Analyzing the paleoenvironmental dependence of the properties of existing S-MIF data, and using a model that accounts for the biogeochemistry of the relevant S-bearing compounds in the water column and during early diagenesis, I will attempt to peel back these effects and provide constraints on the identity and fate of the carriers of S-MIF from the atmosphere to the ocean. Knowledge of the carrier identity importantly constrains the ongoing experimental and theoretical search for the mechanisms responsible for Archean S-MIF generation.

With constraints on the carriers, I will use independent knowledge about the preservation potential of sedimentary environments, together with constraints on how these environments evolved during Earth's early history, to address some of the poorly understood observations arising from the geologic record of S-MIF.