Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.7206

Multiple local controls on pyrite sulfur isotopes and implications for reading the sedimentary pyrite record

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Sulfur isotope ratios in modern marine and sedimentary pyrite ($\delta^{34}S_{pyr}$) have been extensively used in investigations of sub-seafloor microbial activity, in reconstructions of the biogeochemical sulfur cycle and the tightly coupled cycles of carbon and oxygen, and in reconstructions of the redox evolution of Earth's surface environment. These applications are guided by the assumption that $\delta^{34}S_{pyr}$ variation is related to drivers in the global sulfur cycle and the aggregate fractionation imparted by microbial sulfur metabolisms. A growing number of observations from a wide range of natural environments, however, suggest that major $\delta^{34}S_{pyr}$ variation is often related to neither of these factors.

To elucidate the controls on $\delta^{34}S_{pyr}$ values, we combine (i) studies of well-dated, well-understood Pleistocene marine sediments, during the deposition of which the ocean's chemical composition has varied negligibly, and (ii) a coupled microbial-diagenetic model, which accounts for the transport and reaction of sulfur and iron species, and which is capable of predicting the fractionation imparted by microbial sulfate reduction. Rather than global sulfur-cycle drivers of major $\delta^{34}S_{pyr}$ variation, the studied sediment cores clearly demonstrate controls by local variations in sedimentation rate, organic matter and reactive iron content, and reworking by currents, on timescales between tens of thousands of years and tens of millions of years.

Using gridded (1°×1°) datasets of the relevant sedimentary and environmental factors (e.g., sedimentation rate, organic matter content, reactive iron content, porosity), our model reproduces $\delta^{34}S_{pyr}$ distributions compiled from modern marine sediments. The microbial fractionation remains uniformly large under the vast majority of modern marine conditions, and the main determinant of $\delta^{34}S_{pyr}$ is the sedimentation rate, which controls the relative importance of sulfur species' diffusion, advection and reaction ("openness"). The abundance and reactivity of organic matter and iron are important secondary controls on $\delta^{34}S_{pyr}$. Another important determinant of $\delta^{34}S_{pyr}$ values is the concentration of seawater sulfate, and we suggest that the trend of increasing Archean-Proterozoic-Phanerozoic sulfate-pyrite $\delta^{34}S$ offset reflects increasing sulfate concentrations, which affect the openness of the sedimentary system.

With this new framework, we understand the tectonic, biological and oceanographic factors that have governed $\delta^{34}S_{pyr}$ values in marine sedimentary rocks through time.