Goldschmidt 2024 Abstract
https://doi.org/10.46247/gold2024.23404

Quantifying key drivers of marine pyrite content and isotopic composition

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Microbial sulfate reduction (MSR) and subsequent pyrite burial in marine sediments plays a crucial role in Earth's carbon and oxygen budgets; by reducing sulfate and producing alkalinity, this process effectively increases atmospheric O_2 and lowers CO_2 levels. Given that MSR exhibits a large and reduction-rate-dependent sulfur-isotope fractionation, changes in pyrite sulfur-isotope compositions ($\delta^{34}S$ values) through geologic time have long been interpreted to reflect global sulfur cycling or microbial community behavior. However, recent research instead implicates local environmental and sedimentological factors as drivers of pyrite $\delta^{34}S$ values. Still, quantifying how these factors control pyrite formation rate and $\delta^{34}S$ value remains challenging due to complex interactions between controlling variables and the uncertainty regarding the importance of each variable involved. To provide mechanistic and quantitative constraints, we developed a non-dimensional diagenetic model that extracts the natural variables governing pyrite formation. Using only globally interpolated boundary values as inputs, our model accurately predicts all modern observations with an average root-mean square error of 0.3 wt% for pyrite content and 16.6‰ for $\delta^{34}S$. Extrapolating this, we estimate global pyrite burial to be 4.0 ´10^{14} g yr^{-1} with a weighted-average $\delta^{34}S$ value of -3‰ VCDT. This burial flux is substantially higher than recent estimates of terrestrial pyrite weathering, suggesting the modern sulphur cycle results in net pyrite burial and thus atmospheric O2 accumulation. Mechanistically, we conclude that pyrite formation rate is highly sensitive to local reactive iron input, whereas $\delta^{34}S$ value is primarily controlled by organic carbon reactivity-to-sedimentation rate ratio (termed Da*, a modified Damköhler number) and organic carbon-to-sulfate ratio (termed Γ_0). Importantly, $\delta^{34}S$ appears insensitive to microbial fractionation factor and bioturbation. These results indicate that the modern sulfur cycle requires an increase in Da* and decrease in Γ_0 since the Paleozoic, possibly driven by changing organic matter reactivity and sulfate concentrations through time.