Goldschmidt 2025 Abstract
https://doi.org/10.7185/gold2025.28706

The barite record of past seawater oxygen isotope composition

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The oxygen isotope composition of seawater ($\delta^{18}O_{seawater}$) is mostly governed by the relative fluxes of high- and low-temperature rock-water exchanges, thereby recording the long-term Earth system interaction. However, the history of $\delta^{18}O_{seawater}$ remains debated, particularly regarding whether it remained constant or varied over time, due largely to uncertainties in post-depositional alteration in all current mineral proxies. The oxygen atoms in the sulfate molecule resists alteration under most burial conditions, yet they rarely reach equilibrium with ambient water, rendering sulfate mineral useless as a proxy for geological $\delta^{18}O_{seawater}$. However, there is one exception. When residual sulfate's $\delta^{34}S$ rises due to microbial sulfate reduction in a closed or semi-closed system, the associated $\delta^{18}O$ value often reaches a plateau value, a value that is best interpreted to be in equilibrium with the $\delta^{18}O_{seawater}$, thus a proxy for geological $\delta^{18}O_{seawater}$.

We measured 289 paired $\delta^{18}O$-$\delta^{34}S$ data for sedimentary barite deposits spanning six periods of the Phanerozoic Eon. A plateau $\delta^{18}O_{sulfate}$ value is observed for each of the six periods, typically when its $\delta^{34}S$ exceeds 50‰. These trajectories resemble patterns seen in microbial culture experiments and modern field observations, further guaranteeing $\delta^{18}O_{sulfate}$ values in equilibrium with original seawater $\delta^{18}O$, as any alterations would have disrupted or distorted the trajectories. Our barite data suggests that the $\delta^{18}O_{seawater}$ has been measurably fluctuating over the Phanerozoic, recording changes in the fluxes of global high-temperature vs. surface-temperature water-rock interaction. The $\delta^{18}O_{seawater}$ is 4-6‰ lower than today before the Carboniferous Period, suggesting a dramatic relative increase in the global high-temperature rock-water interaction in the late Paleozoic. Our findings provide a platform for additional data from different paleolatitudes of each geological period to enhance both the temporal and spatial resolution of the plateau $\delta^{18}O$ value of barite, ultimately resolving the long-standing debate on seawater $\delta^{18}O$ history.