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Why is the $\delta^{18}O$ of the ^{17}O-depleted, post-Snowball-Earth sulfate so positive?

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In the earliest Ediacaran, *ca.* 635 million years ago, Marinoan glaciation swiftly ended. Subsequent seawater sulfate recorded a period of pronounced mass-anomalous ^{17}O depletion in atmospheric O_2. We compiled all published basal Ediacaran barite $\delta^{34}S$-$\delta^{18}O$-$\Delta'^{17}O$ data (Fig. 1), representing a series of snapshots of shallow-marine sulfate isotope signatures of the post-Marinoan-Snowball-Earth. Its $\delta^{18}O$-$\Delta'^{17}O$ space displays an outstanding feature: the $\Delta'^{17}O$ of barite samples of all sites converge at ~−1.3‰ while their respective $\delta^{34}S$ and $\delta^{18}O$ values at ~ 20‰ and ~ 17‰. This pattern is puzzling in many ways and does not fit the river model in which Ba^{2+} came from deep-water sources while sulfate was mainly supplied by rivers. The average $\delta^{34}S$ and $\delta^{18}O$ values of modern river sulfate—4.8‰ and 4.1‰, respectively— are markedly too low in comparison to those of the basal Ediacaran barite.

Here, we propose that H_2S (or HS^-) oxidation at the oxic-sulfidic redoxcline on the shelf, not the river input, was the dominant source of the ^{17}O-depleted SO_4^{2-}. One prediction of the H_2S model is that the $\delta^{18}O$ of SO_4^{2-} is ~ 17‰ to 22‰ higher than that of H_2O, assuming the early Ediacaran seawater $\delta^{18}O$ is at 0 to −5‰. Laboratory experiments and natural observations indicate that a $\delta^{18}O$ difference of this magnitude, as predicted by the H_2S model, is feasible but requires experimental validation. A second prediction is that the dissolved O_2 at redoxcline had air O_2 signal and was incorporated into the product SO_4^{2-} during H_2S oxidation. To date, no experiment has quantified the proportion of dissolved O_2 in the SO_4^{2-} produced this way, being biotic or abiotic.

The constraints provided by the high-dimensional isotope signature $\Delta'^{17}O$ has revealed our lack of understanding at two distinct levels. At the molecular level, there are unresolved biotic or abiotic pathways that incorporate dissolved atmospheric O_2 into sulfate during sulfide oxidation. At the paleo-ocean redox history level, we are prompted to consider a scenario in which H_2S-rich, SO_4^{2-}-free water masses could have been globally prevalent during the early Ediacaran or possibly throughout much of the late Proterozoic.

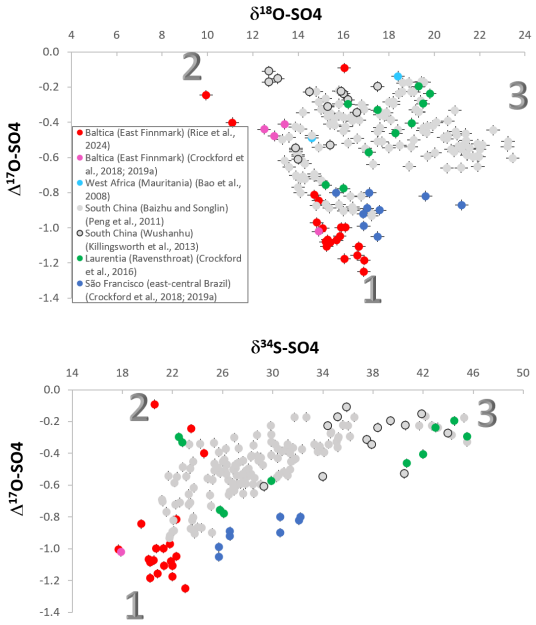


Fig. 1. Basal Ediacaran barite sulfate $\delta^{18}O$-$\Delta'^{17}O$ plot (upper panel) and $\delta^{34}S$-$\Delta'^{17}O$ plot (lower panel).