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# Proterozoic sulfur isotope excursions driven by systematic changes in sulfur isotopic composition of volcanic outgassing

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Sulfur isotope ratios recorded in sedimentary sulfur are a valuable tool to investigate sulfur cycling over geologic time. $\delta^{34}_{CAS}$ and $\delta^{34}_{pyrite}$ exhibit large excursions during the Proterozoic. Notably, there is a large, long-lived positive sulfur isotope excursion (SIE) recorded in both $\delta^{34}_{CAS}$ and $\delta^{34}_{pyrite}$ from around 800 to 550 Ma (Fike et al. 2015). During the Ediacaran, $\delta^{34}S$ values in marine sediments abruptly decrease to pre-excursion values, coincident with the Shuram negative carbon isotope excursion (Osburn et al. 2015). It has proven difficult to identify mechanisms to explain this coupled evolution of marine S and C records. Here, we expand on the C cycle model of Eguchi et al. (2021), which invokes longer mantle residence times for graphitized organic C compared to carbonates to explain the major carbon isotope excursions in marine carbonates of the Proterozoic. Similarly, sulfate minerals may have a longer mantle residence time compared to sulfide minerals because sulfates are more soluble in silicate melts and fluids compared to sulfides (Jégo & Dasgupta, 2014). We propose that increased deposition and subduction of sulfide and sulfate minerals in response to increasing ocean sulfate concentrations drove a long-lived positive SIE followed by an abrupt decrease in $\delta^{34}S$ in marine sediments. This is because sulfate minerals may be rapidly released after subduction at arc volcanoes (Muth & Wallace, 2021), shifting global $SO_2$ emissions to higher $\delta^{34}S$, initiating a positive SIE in marine sulfates and sulfides. The subducted slab will then be enriched in subducted sulfide relative to sulfate, as it subducts deep into the mantle. The slab may then be entrained in upwelling mantle plumes releasing the subducted sulfide as $SO_2$, ending the positive S isotope excursion due to the release of sulfide-derived $SO_2$ with low $\delta^{34}S$. We present results from our coupled surface-interior sulfur cycle and a synthesis of natural data to propose that the major SIEs of the Proterozoic result from changes in $\delta^{34}S$ of volcanic outgassing.

References

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