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The Precambrian S-cycle viewed through the lense of microscale isotopic observations

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Reconstructions of past environmental conditions and biological activity are often based on the use of stable isotope proxies, which in the vast majority of cases are measured on bulk rocks. This is particularly true of the multiple-sulfur isotopic compositions of sedimentary pyrite ($\delta^{34}S_{PYR}$-$\Delta^{33}S_{PYR}$), which are used to reconstruct ocean-atmosphere oxidation state and track the evolution of several microbial metabolic pathways.

In this presentation, we will change the scale of our isotopic investigations to harness previously published microscale $\delta^{34}S_{PYR}$-$\Delta^{33}S_{PYR}$ analyses between 3.8 and 2.2 Ga, to extract novel information on the structure and temporal evolution of Archean-Proterozoic surface environments. In doing so, we identified two arrays in the $\delta^{34}S$-$\Delta^{33}S$ isotopic space. In one array (Array 1), $\delta^{34}S_{PYR}$ values span a wide range from the lowest to the highest measured values while $\Delta^{33}S_{PYR}$values stay relatively uniform and close to 0‰. In the other array (Array 2) $\delta^{34}S_{PYR}$ and $\Delta^{33}S_{PYR}$ values covary positively along the known Archean reference array[1].

We found a striking resemblance between Array 1 and various microscale investigations that we have conducted in well-dated and well-understood modern marine sediments (e.g., Gulf of Lion and/or Black Sea). As such, we propose that Array 1 reflects mass-dependent processes related to microbial respiration of aqueous sulfate, its isotopic distillation, and the buildup of porewater sulfide over the timescale of pyrite precipitation. In addition, the magnitude of the isotopic fractionation between the lowest Archean $\delta^{34}S_{PYR}$ (and their associated $\Delta^{33}S_{PYR}$) and the expected Archean seawater sulfate composition appear to be consistent with the limited microbial fractionation (i.e., by few tens of ‰) we have found in low-sulfate natural environments.

Interestingly, this array is mostly, if not exclusively, observed in sedimentary rocks younger than ≈2.5 Ga, which might attest to the increasing microbial sulfur recycling leading up to the Great Oxygenation Event.

1. Farquhar J, Bao H, Thiemens M. Atmospheric influence of Earth's earliest sulfur cycle. *Science* **289**, 756-759 (2000).