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Micro-scale sulfur isotope analyses reveal microbial activity recorded in shallow-water 3.4 Ga colloform pyrite

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Sulfur isotopes are a foundational archive for reconstructing early microbial activity and redox cycling. However, sulfate availability in the Paleoarchean oceans is widely inferred to have been extremely low, implying that sulfate reduction and its isotopic expression may have been restricted to localized microenvironments. If so, key questions remain regarding where sulfur-metabolizing ecosystems operated and how their signatures were preserved in sedimentary rocks. In microfossil-bearing black cherts of the ca. 3.4 Ga Strelley Pool Formation (Pilbara Craton), pyrite occurs as rare, concentrically laminated colloform spherules (Fig. 1). We analyze these grains using NanoSIMS $\delta^{34}S$ profiles and ion mapping to elucidate links among sulfur-isotope trends, growth processes, and local sulfate limitation.

We investigated concentrically laminated colloform pyrite (<10 μm in diameter) hosted by shallow-water, non-marine cherts from the Strelley Pool Formation. Petrographic relationships, including the association between pyrite and early diagenetic silica, suggest that the pyrite formed prior to complete lithification. In situ NanoSIMS analyses reveal pronounced intra-grain $\delta^{34}S$ gradients exceeding 20‰. The colloform pyrite cores are strongly ^{34}S-depleted, whereas outer growth zones become progressively heavier, approaching near-zero to positive values. These low $\delta^{34}S$ values and the systematic layer-by-layer trend are consistent with Rayleigh-type isotopic evolution in a closed to semi-closed microenvironment where sulfate availability was limited and reduced sulfur was progressively removed into the mineral phase, as expected for microbially mediated sulfate reduction. Secondary-ion molecular maps of carbon and nitrogen further show repeated micron-scale alternations between organic-rich and pyrite-rich layers, indicating tight coupling between organic matter distribution and sulfide mineralization during early diagenesis. Together, these results indicate that Archean shallow-water settings could have served as habitats for sulfate-based ecosystems, and they demonstrate that high-spatial-resolution sulfur-isotope measurements provide a new approach for accessing sulfur-isotope archives preserved within micron-scale pyrite.



Fig. 1. Outcrop photograph and a BSE image of the concentrically laminated pyrite