³⁴S-depleted barite records oxidation of biogenic pyrite – evidence for microbial sulfur cycling in the terrestrial subsurface

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The terrestrial subsurface hosts a significant fraction of Earth's microbial biomass and has likely been a habitable environment for most of its history [1]. Reconstructing the evolution of subsurface habitats in deep time requires tools to track microbial activity in the geological past (i.e., biosignatures). Fracturehosted minerals can record such biosignatures in their stable isotope compositions. However, the stability of these minerals can be affected by in-situ and secondary interaction with metamorphic, hydrothermal, marine, and meteoric fluids in subsurface environments. To date, the preservation potential of stable isotope biosignatures associated with fracture-hosted minerals is poorly constrained. Here, we report in-situ (SIMS) δ^{34} S and δ^{18} O data in fracture-hosted barite and δ^{13} C and δ^{18} O of coeval calcite, crosscutting the Permian Paradox basin at Gypsum Valley (Colorado, USA) and Onion Creek (Moab, Utah, USA). Barite is 34 S-depleted (δ^{34} S_{CDT} = -16.55 ± 0.14‰) suggesting an origin from microbially cycled sulfur. Most likely, barite precipitated from ³⁴S-depleted sulfate, derived from the oxidation of pyrite formed via microbial sulfate reduction. Calcite shows δ^{13} C _{PDB} values of -12.23 \pm 0.37‰ and higher, providing no clear indication of microbial versus abiogenic carbon sources. Preliminary in situ (LA-ICP-MS) U-Pb dating of this calcite suggests a Cretaceous age of the crystal cores, with the barite-hosting outer regions being much younger (<2 Ma). This demonstrates that microbial activity, or at least barite precipitation, occurred recently when the strata were brought closer to the surface by incision of the Colorado River and related denudation, making temperatures more habitable (<121°C). Our results suggest that barite can record evidence for microbial sulfate reduction in terrestrial subsurface environments after oxidation of the primary biogenic pyrite. Thus, barite is a promising archive for reconstructing the evolution of subsurface microbial life in deep time.

[1] Drake, Ivarsson, & Heim (2020), Geosciences 10 (11), 461.

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