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Quadruple sulfur isotope signatures in microbialites from modern redox-stratified lakes

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Microbialites represent the oldest known traces of life on Earth and result notably from complex microbial interactions within biofilms harboring steep redox gradients. As sulfur is both a highly redox-sensitive element and ubiquitous in metabolic reactions, its $\delta^{34}S$ have been used from ancient to modern microbialites to characterize their formation mechanisms and environment of growth and the microbial diversity composing them. However, $\Delta^{33}S$ and $\Delta^{36}S$ signatures in microbialites have yet received very little attention. This is especially true in modern examples, although they offer the possibility to be more thoroughly constrained (*e.g.* absence of late and burial diagenesis, metagenomes characterization, isotopic compositions of the sources).

Here, we analyzed the quadruple S isotope compositions of bulk pyrite in living and subfossil microbialites, together with the lakes' dissolved sulfate (SO_4) and microbialite carbonate-associated sulfate (CAS) from several modern redox-stratified lakes from Mexico. The lakes show different SO_4 concentrations (from ~ 1 to 12 mM) and isotopic signatures ($\delta^{34}S_{SO4}$ from 0 to +19 ‰, $\Delta^{33}S_{SO4}$ ~ 0.01 ‰ and $\Delta^{36}S_{SO4}$ ~ -0.7 ‰, VCDT). The microbialite pyrites show a relatively large variation in $\delta^{34}S_{py}$ (-40 to 0 ‰), $\Delta^{33}S_{py}$ (+0.05 to +0.19 ‰), and $\Delta^{36}S_{py}$ (-0.2 to +1.1 ‰; all vs. VCDT, respectively). These isotopic signals vary according to the different studied microbialites mineralogies and facies, but not to the sulfate concentrations.

While $\delta^{34}S$ mostly reflects bacterial sulfate reduction, quadruple S isotopes also allow us to discuss the possible involvement of other metabolisms (*e.g.* sulfur disproportionation), different local conditions of formation (*e.g.* open *vs*. disconnected porewaters), and precipitation at different times in the lakes' history. A subset of samples also allows us to assess the effect of early sulfide oxidative alteration. Finally, the signatures of the CAS samples are either very close to the SO_4 composition or fall between SO_4 and pyrites suggesting different degrees of pyrite oxidation in our samples of potential abiotic *and* biotic origin.