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Microbialites from the seasonally anoxic lake Alchichica archive distinct sulfur and iron geochemical fingerprints

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The coupled biogeochemical cycles of iron and sulfur are central to understanding the microbial redox processes that drive microbialite formation. Iron mineral speciation can be used to track oxygen and free sulfur availabilities and distinguish between iron precipitated in the water column versus iron cycled by microbes within microbialites. Pyrite sulfur isotopes can further be used to assess euxinic conditions and microbial cycling. Here, we performed iron speciation and sulfur isotope measurements in microbialites and sediments from the Mexican volcanic crater lake Alchichica.

Lake Alchichica is fed by an aquifer composed of carbonate-volcanic sediments, from which it receives dissolved iron via subsurface inflow[1]. Dissolved S concentrations in the lake range from 10 to 12mM[2].

We analyzed vertical profiles of 2 cores drilled across fossil microbialitic reefs (MBF1 and MBF2), fossil microbialite fragments from around the lake (MBFi), living microbialites at different depths in the lake (MBL), including microbialites forming under seasonally anoxic conditions[2], and a gravity core from the lake-bottom sediments (SEDC2).

We show significant variation in $\delta^{34}S_{py}$ in drill core samples, from -23.9 to -46.6‰ (MBF1) and from -26.4 to -32.3‰, (MBF2). MBFi samples show variations from -14.6 to -36.2‰, while SEDC2 show an average $\delta^{34}S_{py}$ of -22±3‰. In general, the lowermost values indicate sustained bacterial reduction of sulfate, while ^{34}S-enriched values reflect the shrinking residual sulfate pool undergoing Rayleigh-type distillation as reduction proceeds.

According to the sequential extraction of iron in SEDC2 sediments, we observe that most reactive iron is associated with pyrite (23.5%). Pyrite content further increases with sediments depth, at the expense of oxide phases. For MBF1 and MBF2, iron is associated with oxides (such as Fe-bearing layered double hydroxides[1]) and carbonate phases and decreases with depth, while the pyrite and magnetite-iron increase conversely.

The Fe_{HR}/Fe_T ratio shows values >0.38 for all samples. While such "anoxic" values are expected in the bottom anoxic sediments and the deepest microbialites of the lake, a strong diagenetic overprint on the iron speciation of subsurface microbialites is evidenced.

[1] Zeyen *et al.* (2019), *GCA*-253, 201-230
[2] Caumartin et al. (in press) Advances in Geochemistry and Cosmochemistry