

The unbalanced Fe and S biogeochemical budgets in the ferruginous and sulfate-rich waters of a post-mining lake

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The redox structure of natural meromictic lakes features wide-ranging concentrations of dissolved Fe(II) but consistent SO_4^{2-} deficiencies. A few of these anoxic systems have been used as analogues to model the generalized redox structure of the Precambrian ocean. Nevertheless, mounting geochemical evidence suggest that some Proterozoic, out-of-equilibrium coastal environments associated with transiently elevated atmospheric O_2 are not accounted for by such deep time palaeoceanographic redox models. For instance, repeated Precambrian O_2 fluctuations were accompanied by increased dissolved sulfate delivery to ferruginous coastal mixing zones [e.g., refs. 1, 2]. Subsequent variations in C fluxes ultimately regulated the redox states of ocean interiors, alternating them between stable Fe(II)-rich (ferruginous) and $\text{H}_2\text{S}/\text{HS}^-$ -rich (euxinic) redox conditions [3]. A post-mining lake in NW Czechia represents a novel anoxic lacustrine system where an intermediate state between such purposely non-reversible, bistable redox conditions can be currently examined. Due to groundwater discharge, the ferruginous bottom waters ($[\text{Fe}^{2+}] = 1300 \pm 130 \mu\text{M}$) of the oligotrophic, artificial lake under examination (Lake Medard), are also sulfate-rich ($[\text{SO}_4^{2-}] = 14 \pm 1 \text{ mM}$). Dissolved inorganic carbon- $\delta^{13}\text{C}$, sulfate- $\delta^{34}\text{S}$ and $-\delta^{18}\text{O}$, gypsum- and pyrite- $\delta^{34}\text{S}$ isotope data, ion concentration gradients, volatile fatty acid contents, amplicon sequencing, and mineralogical analyses of the anoxic sediments suggest that microbial SO_4^{2-} -reduction occurs but S^{2-} species do not accumulate. The export and deposition of particulate Fe(III)-(oxyhydr)oxides formed at the redoxcline, the low amount and reactivity of metabolizable C_{org} , and the presence of redeposited Miocene siderite in the modern anoxic sediments stimulate the disproportionation of intermediate sulfur which prevents the stabilization of iron monosulfides while allocating the precipitation of gypsum. Several microbial community interactions consistent with modelled mineral equilibria can also be deduced.

[1] Blättler et al., *Science* **360**, 320–323 (2018) 20 (2018).

[2] Och, L.M. & Shields-Zhou G.A. *Earth-Sci. R.* **110**, 26–57 (2012).

[3] van de Velde, S.J. et al. *PNAS* **117**, 33043–50 (2020).