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Framboidal pyrite formation during the early diagenesis of inorganic sulfur in a small High Arctic lake

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Sulfur (S) speciation is highly sensitive to redox conditions, due to its eight oxidation states allowing the S cycle to structure microbial communities and to influence the cycling of other major (C, O, Fe) and trace elements (As, Pb, Hg). In High Arctic lakes, the biogeochemical cycle of S is still poorly understood. Indeed, High Arctic lakes differ from temperate ones in that they remain ice-covered for most of the year, receive lower fluxes of organic carbon and nutrients from their watersheds, and have short, truncated food webs that are dominated by microbial organisms.

Here, we studied Small Lake (Nunavut, Canada, 75 °N), which exhibits high under-ice algal productivity in the spring, despite a thick ice cover (2 m). We collected sediment cores and sampled the sediment porewaters. Measurements focused on inorganic S species. In the porewater, we analyzed sulfate (SO_4^{2-}), thiosulfate ($S_2O_3^{2-}$), elemental sulfur (S^0) and sulfide (ΣHS^-). In the sediments, we measured three fractions of reduced S (RIS): acid volatile sulfides (AVS; mostly iron monosulfide, such as $FeS_{(s)}$), chromium reducible sulfides (CRS; mostly pyrite, $FeS_{2(s)}$) and elemental sulfur (ES, mostly $S(\alpha)_{8(s)}$), along with the simultaneously extracted metals. Scanning electron microscopy (SEM) was used to ascertain the presence of $FeS_{2(s)}$.

We found elevated porewater concentrations of S species of intermediate and reduced redox states, with $S_2O_3^{2-}$ up to ~75 µM, S^0 up to ~16 µM and ΣHS^- up to ~440 µM. In contrast, SO_4^{2-} concentration were higher in the water column than in the porewater, with a maximum of ~130 µM. In the sediments, RIS were mainly composed of $FeS_{2(s)}$ (~80 %) to a concentration reaching up to ~370 µmol g^{-1} dry weight, pointing to diagenetic formation of $FeS_{2(s)}$, a result that is supported by the observation of framboidal pyrite by SEM. These results demonstrate unexpectedly dynamic S cycling in the sediment of a High Arctic lake, with ΣHS^- accumulation and rapid $FeS_{2(s)}$ formation, despite sustained ice cover and limited organic carbon inputs. The dominance of framboidal pyrite, coupled with persistent porewater ΣHS^- excess, indicates that iron availability, rather than SO_4^{2-} reduction potential, limits early diagenesis in these systems.

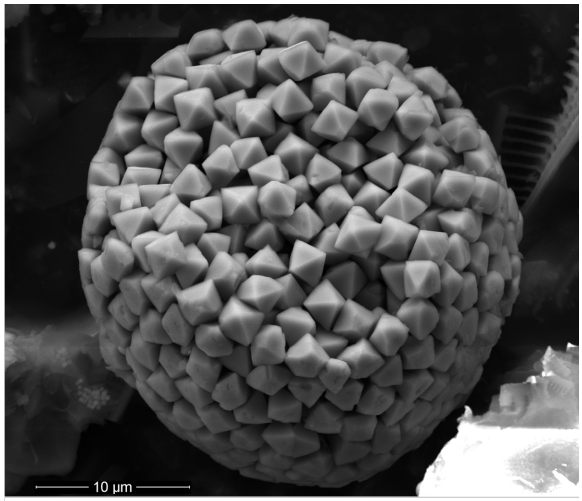


Fig. 1. Scanning electron microscopy image of a framboidal pyrite in the sediment of Small Lake (75 °N, Canada).