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Ice Amplifies Ligand-Controlled Mineral Dissolution in Microscale Hotspots

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Cold-region ecosystems are highly sensitive to climate change, yet the geochemical processes controlling iron mobilization in frozen environments remain poorly understood. This work challenges the assumption that ice is geochemically inert, demonstrating instead that polycrystalline ice acts as a dynamic reactor that systematically enhances ligand-promoted dissolution of goethite (α-FeOOH) nanoparticles through freeze-concentration into microscale reactive hotspots.

Using inorganic anions of varying iron-binding strength (chloride, fluoride, perchlorate, sulfate) under mildly acidic conditions and temperatures down to −30 °C, we show that dissolution rates scale with ligand binding affinity in both ice and liquid water, with ice consistently enhancing rates by ~0.4 log unit across all reactive ligands. Fluoride, the strongest complexing agent considered here, increased dissolution more than four-fold in ice, while weakly binding perchlorate produced no measurable dissolution in either phase. Quantification of residual liquid water reveals ten- to hundred-fold concentration enhancement as freezing progresses.Thermodynamic modeling shows that freeze-concentration activates dissolution pathways unavailable in liquid water, while imaging shows that freezing transforms dispersed nanoparticle suspensions into discrete micron-sized aggregates where dissolution reactions occur. Remarkably, dissolution persists below the eutectic temperature, sustained by minute volumes of liquid-like water stabilized within mineral aggregate networks.

A relationship between Gibbs free energy of Fe(III)-ligand complex formation and dissolution rates provides a predictive framework for mineral reactivity in frozen systems. As climate change intensifies freeze-thaw dynamics across permafrost soils, acid sulfate terrains, mine drainage systems, and polar aerosol environments, these findings suggest that current biogeochemical models may underestimate iron mobilization and associated nutrient fluxes in cold regions, with implications for carbon cycling and ecosystem feedbacks in a warming world.

