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A Nitrogen-Rich Molecular Vault: How Bio-silicifying Organisms Protect Polyamines in Opal

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Biogenic silica (bioSiO_2) produced by silicifying organisms is central to global silicon cycling and is intimately linked to carbon export via diatom-driven particle fluxes. Long-chain polyamines (LCPAs) and related cationic biomolecules are well recognized as key components of biosilicification in diatoms and have also been implicated in silica formation pathways in other silicifiers, such as sponges. However, the extent to which bioSiO_2 matrices can protect and preserve nominally labile, nitrogen-rich organic compounds and promote their persistence in sediments remains poorly constrained at the molecular level. Building on classical paradigms of organic matter preservation by mineral association and physical occlusion, we test an "opal entombment" mechanism in which electrostatic interactions between nitrogen-rich LCPAs and silica surfaces stabilize occluded organic matter against diagenesis. We combine Fourier transform ultra-high resolution mass spectrometry, tandem mass spectrometry, compound-specific radiocarbon (^{14}C) dating, and scanning electron microscopy imaging to investigate LCPAs associated with modern and ancient biosilica archives, including cultured diatom frustules, plankton tow material, sinking particles, sponge spicules, and opal-rich sediment cores from polar and lacustrine environments.

Across all systems, we observe a ubiquitous and structurally diverse suite of LCPA-like compounds associated with biosilica. LCPAs remain detectable from living diatoms through fossil diatomites, indicating persistence of diagnostic molecular features over millennial to million-year timescales. Compound-specific ^{14}C measurements of LCPAs extracted and purified from three diatom-rich Antarctic sediment cores indicate ages up to ~45 kyr, consistent with core stratigraphy and depositional chronologies, indicating persistence of these compounds within biosilica.

Consistent with atomistic predictions of strong polyamine-silica electrostatic interactions, our results suggest that biosilica can act as a widespread, previously underappreciated sink and stabilizing phase for organic nitrogen and carbon in marine and lacustrine sediments, with implications for nutrient sequestration and the interpretation of silica-rich paleoarchives.