Cryovolcanic plumes as a record of habitability: Fluid evolution and the fate of bioessential elements during freezing of simulated Enceladus ocean brines

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Saturn's ice-covered moon Enceladus may contain the requisite conditions for life. Its potentially habitable subsurface ocean is vented into space as large ice-rich cryovolcanic plumes that can be sampled by spacecraft, acting as a window to the ocean below. However, little is known about how Enceladus ocean fluids evolve as they freeze, and thus how key indicators of habitability, such as redox-sensitive metals and organic compounds, might be expressed in the plumes. We investigated fluid evolution and solid phase formation from simulated Enceladus ocean fluids during freezing at endmember cooling rates, using parallel thermodynamic modelling and experimental approaches. Cryo-imaging techniques showed that even under flash-freezing conditions (>10 K s^{-1}), Enceladus-like fluids undergo segregation, whereby the crystallisation of ice templates the formation of brine vein networks. The high solute concentrations and confined nature of these brine veins means that glass formation (vitrification) can occur at lower cooling rates than those typically required for vitrification of a bulk solution. Crystalline salts also form if flash-frozen fluids are re-warmed. The 10 µm-scale distribution and mineralogy of salt phases produced differ markedly from those of gradually cooled (~1 K min^{-1}) fluids, showing that they inherit a signature of their non-equilibrium formation conditions. Moreover, the mineralogy of cryogenic carbonates can be used as a probe for cooling rate and parent fluid pH. Implications for the incorporation of trace bioessential elements such as iron, which are expected in the ocean but have thus far evaded detection in plume particles, are explored using thermodynamic models. Our findings reveal possible endmember routes for solid phase production from Enceladus ocean fluids and mechanisms for generating compositional heterogeneity within ice particles on a sub-10 µm scale. Furthermore, our results have implications for how organic material, including biological or macromolecular organic structures, may be incorporated and delivered to space. Our findings permit more precise reconstructions of ocean chemistry and habitability from Cassini data, and provide a strong rationale for future non-destructive analyses of plume particles.