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Probing chemical properties of interstitial micro-fluids in ice

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Liquid is present as microscopic channels in polycrystalline ice at sub-freezing and even sub-eutectic temperatures. Not only do chemicals tend to concentrate substantially in this microscopic liquid phase, but local physicochemical properties may also differ widely from the bulk counterparts, therefore critically affecting the thermodynamics and kinetics of chemical processes occurring in frozen media such as snow, frost, and frost- flowers. This phenomenon has important implications in atmospheric chemistry such as affecting the composition of the atmospheric boundary layer in snow-covered regions. A method using con-focal laser scanning microscope equipped with a cryostat has been developed to measure physicochemical properties of the microscopic liquid phase in ice that are not readily extrapolated from the bulk data. The experimental setup allows for monitoring the freezing process of an aqueous solution with a sub- second time resolution and a submicron 3D spatial resolution. The physicochemical properties (e.g., viscosity, polarity, and acidity) can, in theory, be deduced from features of the fluorescence spectra of particular fluorescent indicators. For example, the acidity change during the freezing and melting process of electrolyte solutions has been monitored in real time by a pH-dependent dual emission fluorescent probe C-SNARF-1. The effects of temperature, freezing rate, and added electrolytes such as ammonium sulfate, sodium chloride and zwitterions are also examined. The findings complement the theory and previous experimental evidence of freezing hydrolysis.