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Characterization of Individual Ice Nuclei Collected During CARES and a New View on Immersion Freezing Kinetics

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1. Potential for Authentic Organic Dominated Particles to Form Cold Clouds

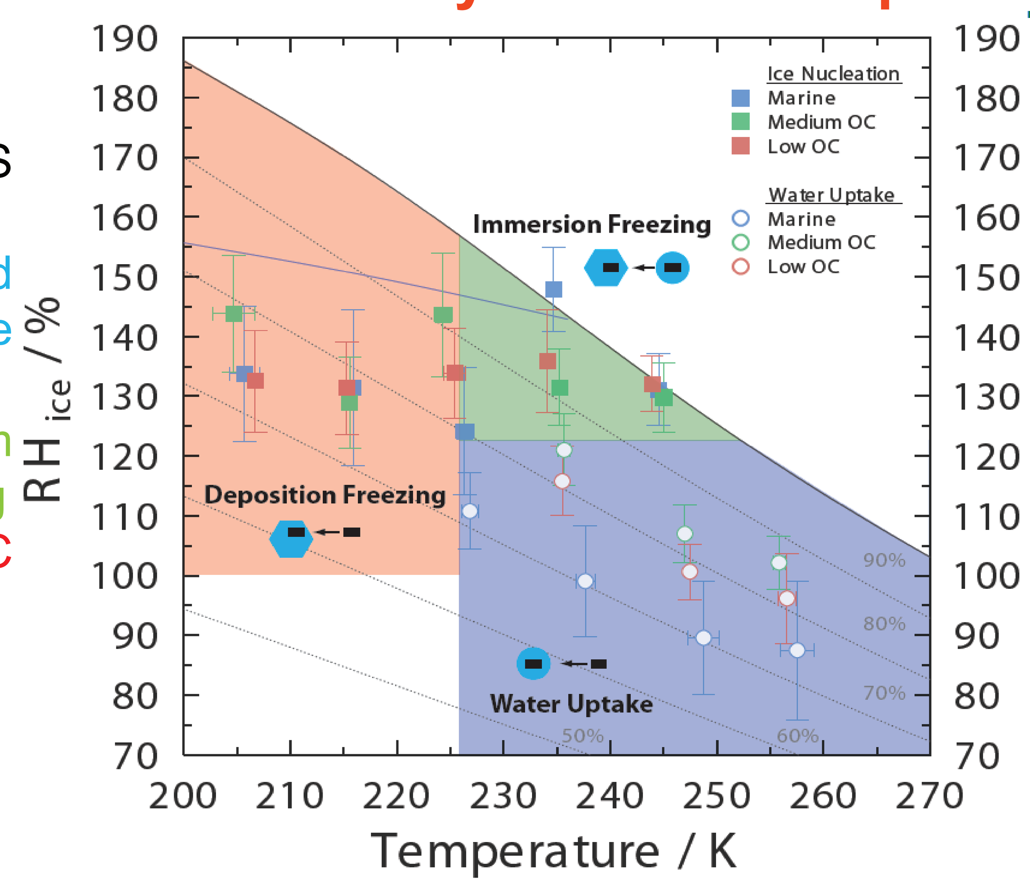
i) The ability of field-collected CARES particles to take up water and nucleate ice below 273 K is investigated.
ii) Individual ice nuclei (IN) are identified, then their chemical composition is probed with STXM/NEXAFS and CCSEM/EDX to relate chemical and physical particle properties to ice nucleation efficiency.
iii) A composition comparison is made between IN particles and the ambient particle population to discriminate similarities and differences.

A Novel Physical Model of Immersion Freezing Based on Water Activity (=RH)

i) Immersion freezing temperatures and heterogeneous ice nucleation rate coefficients are desrcibed solely by water activity independent of type of solute.
ii) This new model predicts frozen droplet fractions and ice crystal numbers for any atmospheric condition accounting for changes in IN surface areas and cooling rates.

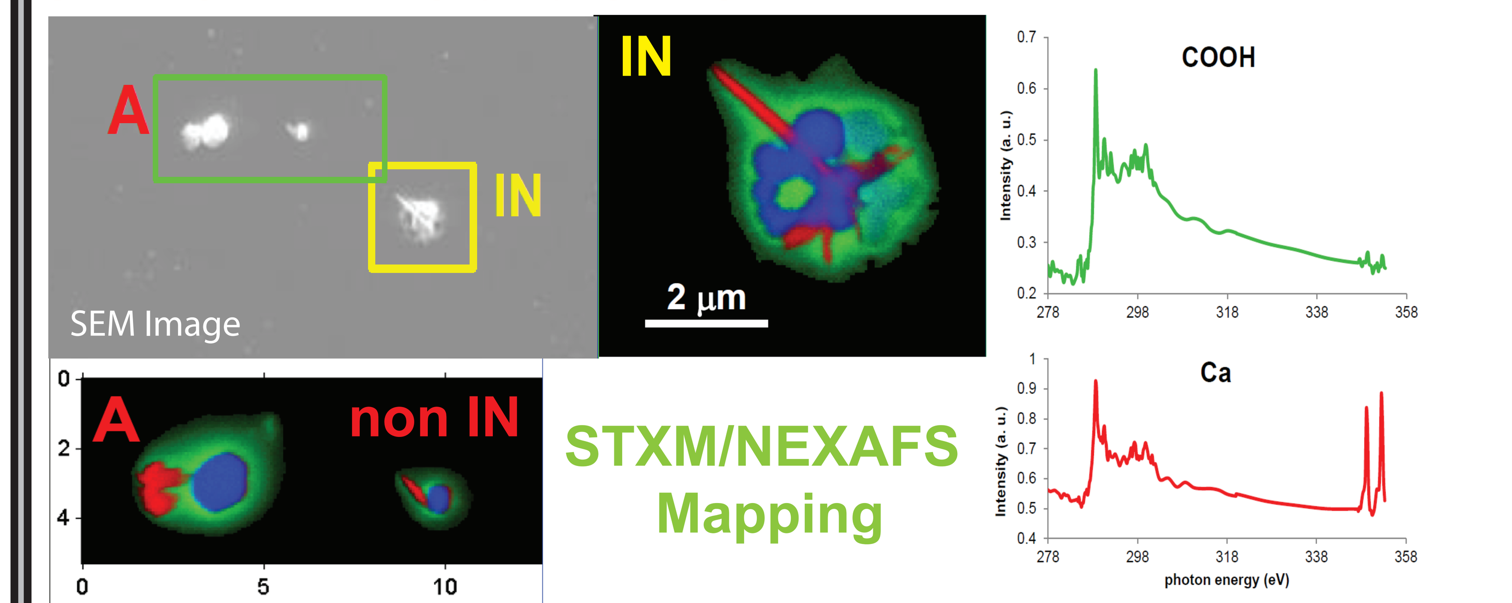
2. Ice Nucleation Pathways and Water Uptake

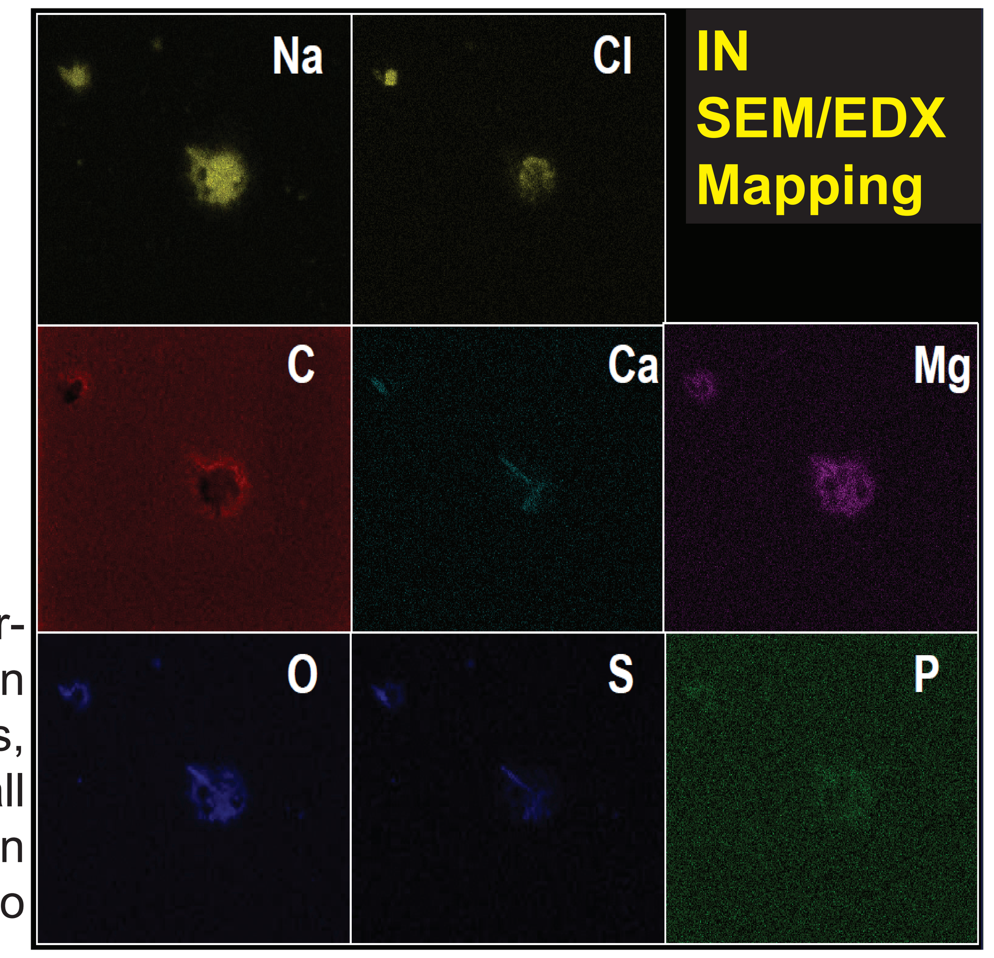
3 CARES samples:
i) affected by marine air
ii) medium OC loading
iii) low OC loading



Organic dominated particles can take up water until 230 K and efficiently induce immersion freezing (at subsaturated conditions) and depositon ice nucleation (below homogeneous freezing limit).

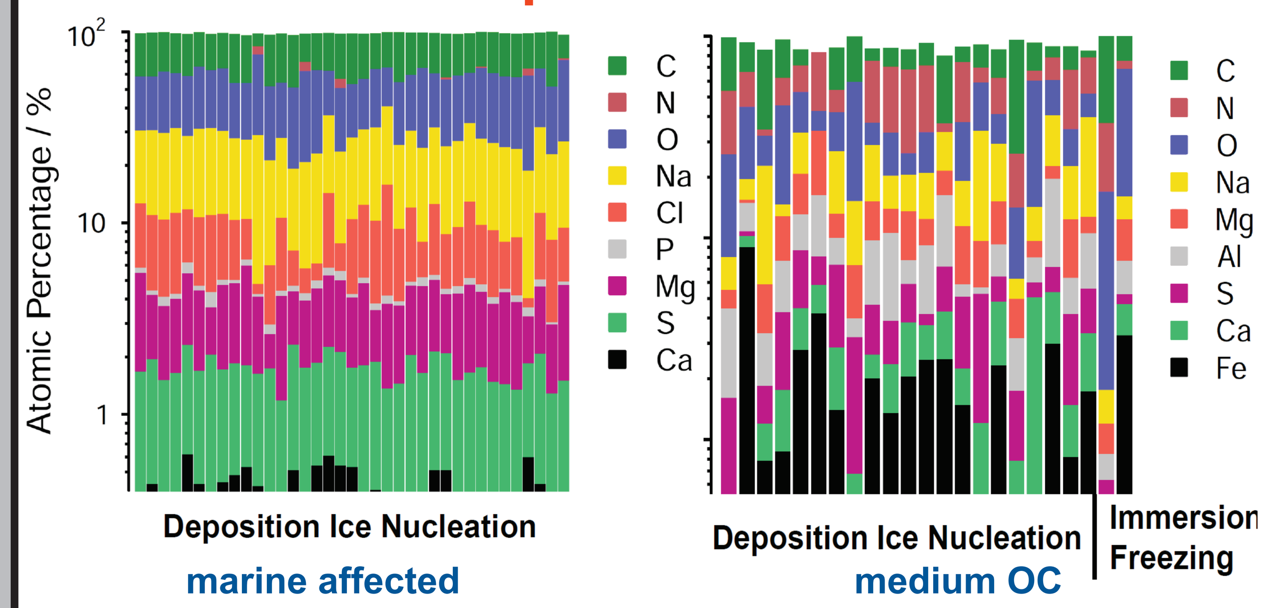
3. Composition Comparison of Ice-Forming and Non Ice-Forming Particles Using STXM/NEXAFS and SEM/EDX





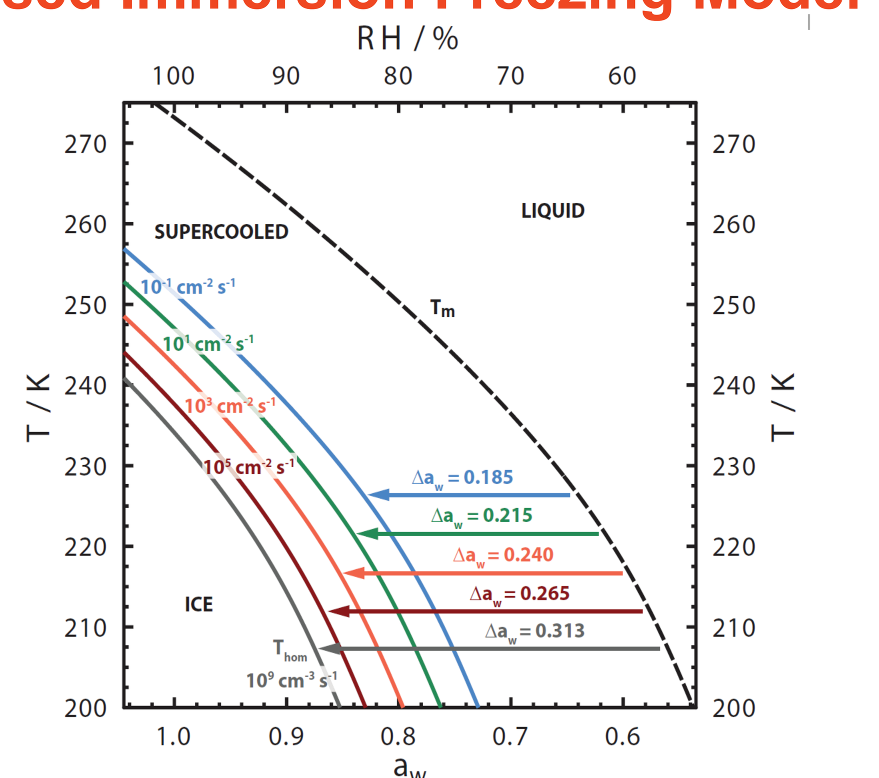
STXM/NEXAFS analysis: IN consists of an inorganic core coated by organic matter rich in COOH functional groups. Non-ice forming particles on the same sample (A) are morphologically and chemically similar. Thus, composition alone may not be sufficient to discriminate IN from the overall particle population. **SEM/EDX analysis:** Inorganic core has marine origin and also indicates presence of oxygenated carbon. Presence of P hints to a biological source (e.g. phytoplankton).

4. Elemental Composition of Ice Nuclei



More than 90% of IN consist of an inorganic core and presence of organic coating. Furthermore, most IN were greater than 1 µm in diameter. Preliminary particle type cluster analysis indicates that IN belong to a major particle type class for this size suggesting that IN may not be exceptional compared to overall particle population.

5. Concept of Novel Water Activity Based Immersion Freezing Model

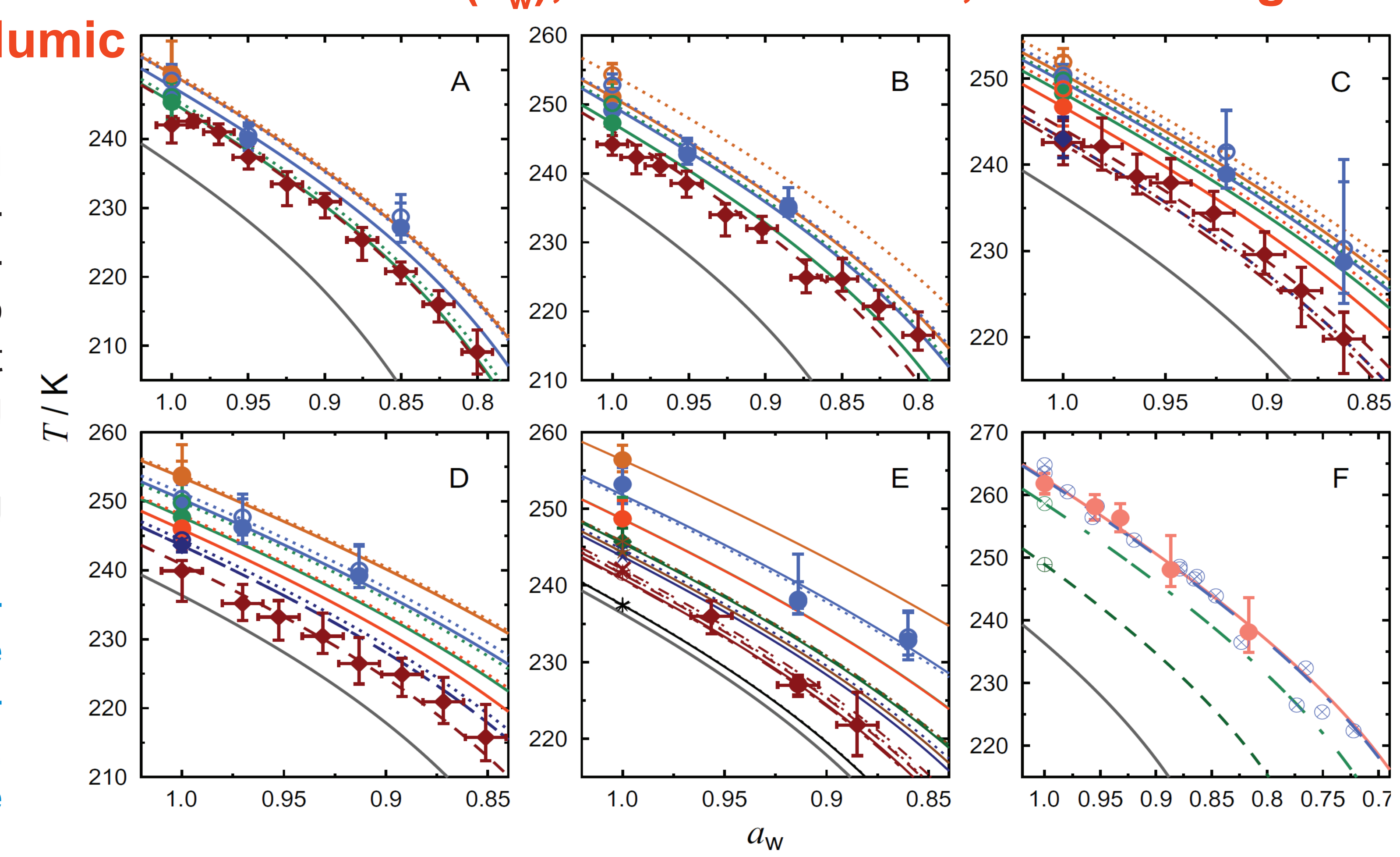


Δa_w implicitly defines a_w (=RH in equilibrium) and T and thus, provides holistic description of freezing temperatures and kinetics.

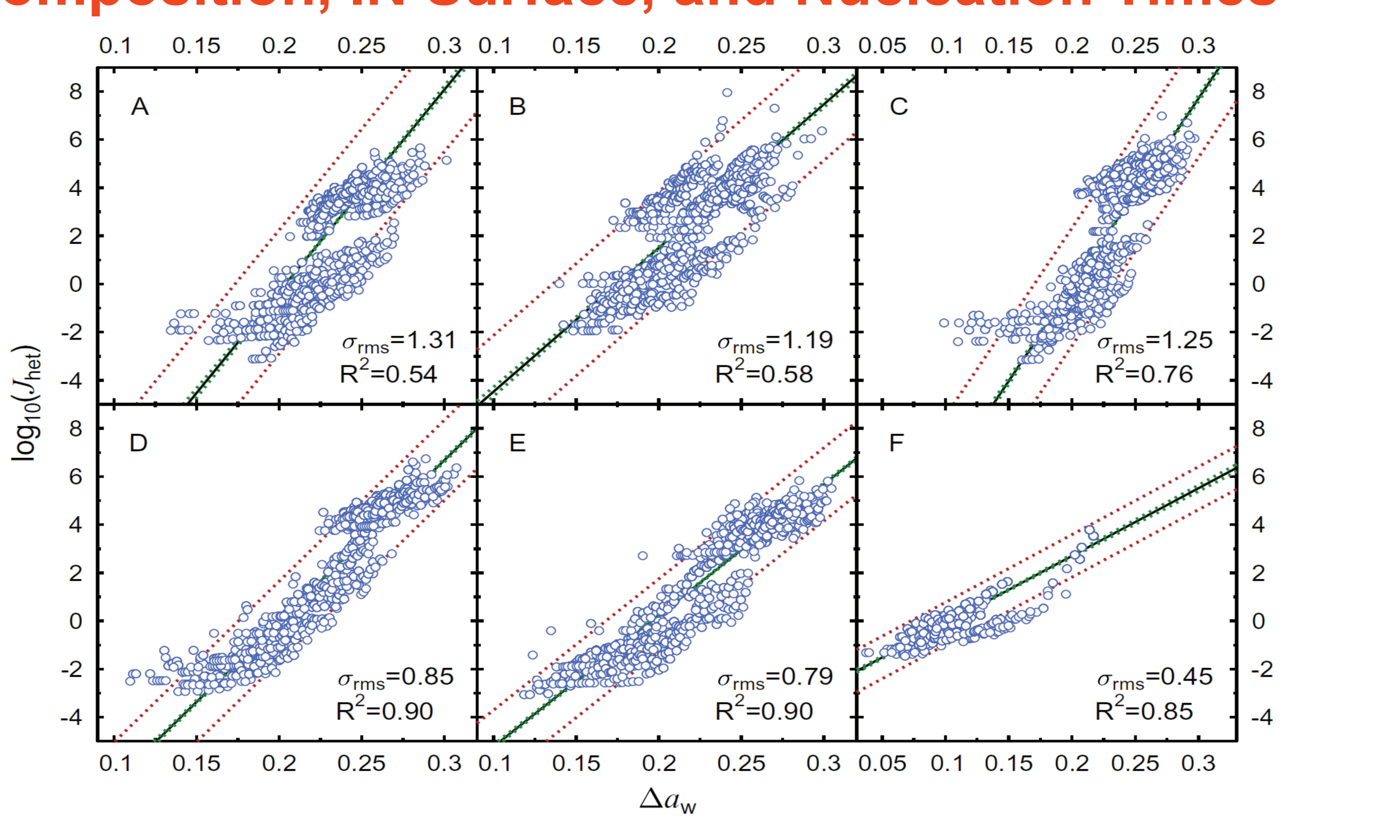
6. 18000 Individual Freezing Events as a Function of Solute (a_w), IN Surface Area, and Cooling Rate for 6 Different IN Types (Biogenic, Humic Acids, Mineral Dusts, Surfactants)

Freezing temperatures as a function of a_w for water and aqueous solution droplets containing (A,B) phytoplankton, (C) Pahokee Peat, (D) Leonardite, and (E) Illite particles, and (F) coated by 1-nonadecanol. Lighter to darker colors represent droplets with greater (light brown) and smaller (dark red) IN surface area ranging from about 1 cm^2 to 1×10^{-7} cm^2, respectively. Symbol/line combinations represent varying cooling rates ranging from1 K min^{-1} to 14.2 K min^{-1}.

This novel model successfully predicts immersion freezing temperatures accounting for changes in IN surface areas by 9 orders of magnitude and ice nucleation activation times by over 1 order of magnitude. Furthermore, freezing temperature predictions hold for a wide range of a_w, i.e. droplet composition and RH.

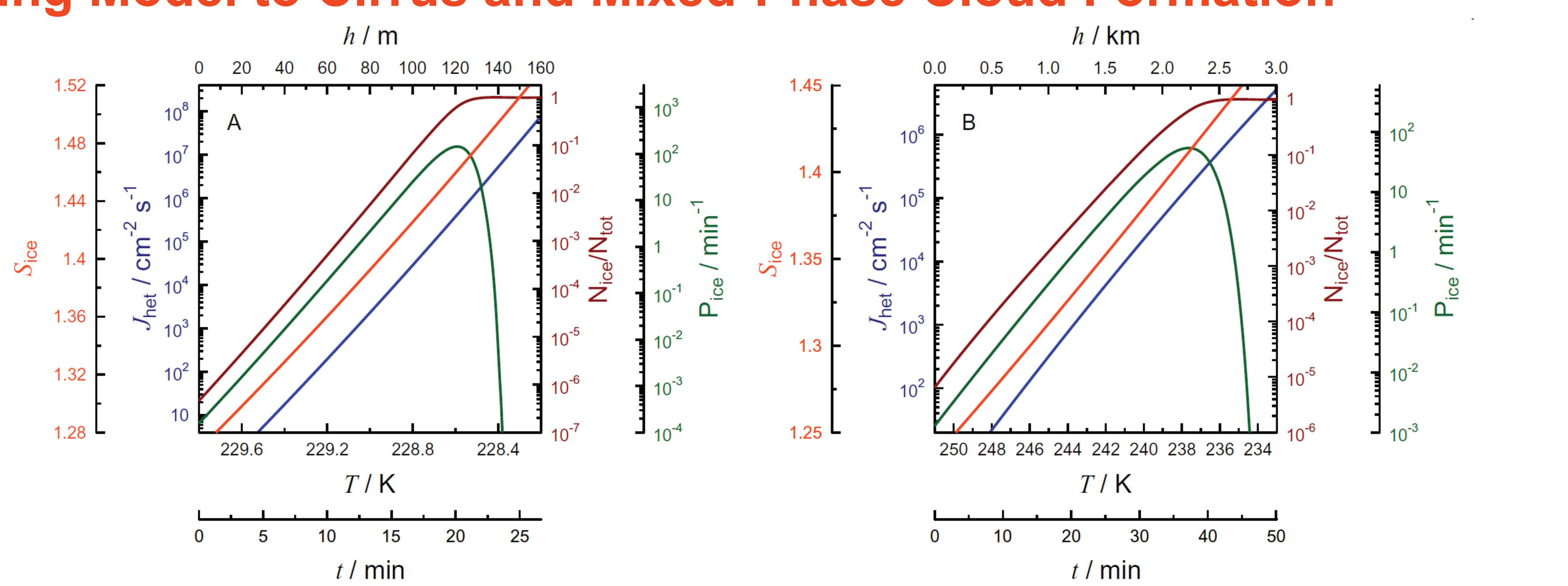


7. J_{het} as a Function of Δa_w for Various IN Types, Composition, IN Surface, and Nucleation Times



log(J_{het}) as a function of Δa_w. Uncertainy of linear parameterization is < ±2 orders of magnitude, although J_{het} spans > 10 orders of magnitude.

8. Application of Novel Water Activity Based Immersion Freezing Model to Cirrus and Mixed-Phase Cloud Formation



Ice nucleation calculations for typical cirrus (A) and mixed-phase (B) cloud conditions from Illite mineral dust. Calculations of ice particle production rate, P_{ice}, and frozen fraction, N_{ice}/N_{tot}, where N_{tot} = 0.5 cm^{-3} (air) is the total number of ice nuclei and N_{ice} is the number of particles that nucleated ice using the water activity based immersion freezing model to derive heterogeneous ice nucleation rate coefficients, J_{het}.

9. Summary and Conclusion I

i) Unique technique allows **identification of individual IN by SEM/EDX in combination with STXM/NEXAFS** with resolution of about 30 nm.
ii) **Most efficient IN do not appear unique** with regard to composition compared to overall particle population and **all IN are observed to be larger than 0.5 µm**.
iv) Common similarity between identified IN is organic coating.
v) Maybe **most abundant particles** govern ice nucleation and not most efficient.
vi) How to interpret ice nucleation studies using "simple" single component IN?

Summary Conclusion II

i) We have shown, using a variety of IN types suspended in various aqueous solutions, that parameterization of **immersion freezing temperatures and kinetics** can be described solely by temperature and solution **water activity (and thus RH)**.
ii) This new model is **independent of the nature of the solute** and accounts for varying cooling rate and IN surface area while allowing prediction of freezing temperatures, J_{het}, **frozen fractions, ice particle production rates and numbers**.
iii) The new model allows application of **laboratory generated freezing data to atmospheric conditions** (time scales and IN surface area).
iv) Can be **easily implemented in cloud-resolving models** that monitor T and RH.