INSTITUTE FOR TERRESTRIAL AND PLANETARY ATMOSPHERES

Ice Nucleation by Particles Collected in the Eastern North Atlantic and by Internally and Externally Mixed Mineral Dust Particles in Experiment and Model

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Motivation

Atmospheric ice nucleation still remains a challenge for our predictive understanding. Our research efforts make use of field-collected and laboratory generated aerosol particles for ice nucleation experiments and modeling studies. We focus on the physicochemical characterization of ice nucleating particles (INPs) and how different particle types impact ice formation. Currently we are working on the following tasks:

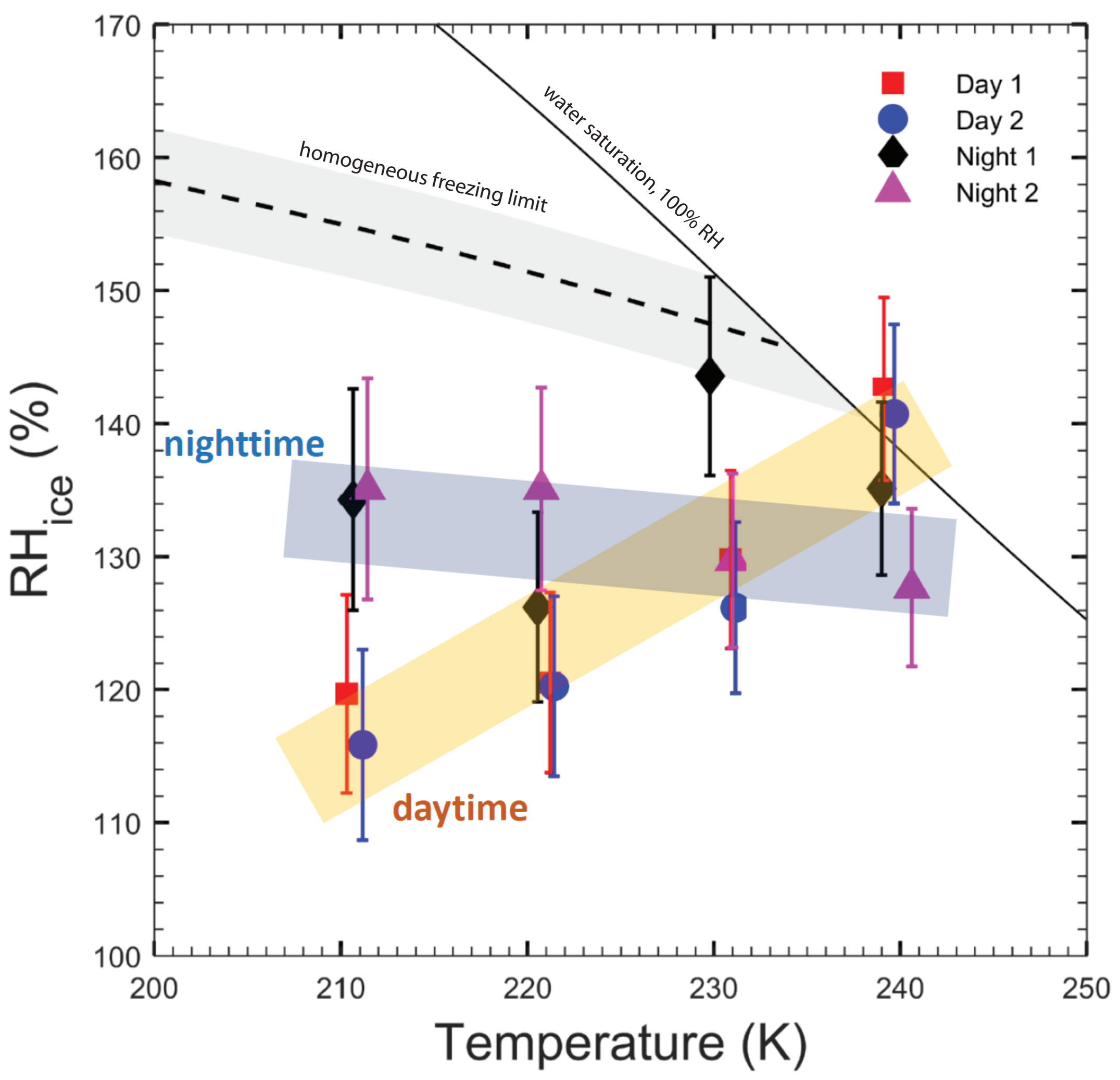
1. Use ambient aerosol particles collected during Aerosol and Cloud Experiments in Eastern North Atlantic (ACE-ENA) campaign for identification of INPs and evaluation of the particles' propensity for nucleating ice.

2. Physicochemically characterize ambient aerosol particles collected during ACE-ENA. We focus on micron- to supermicron-sized particles which have shown in the literature to be important as source of INPs.

3. Conduct constant cooling rate and isothermal immersion freezing experiments employing externally and internally mixed mineral dust particles to better mimic atmospheric aerosol populations. Investigate the applicability of various parameterization approaches. How important is the role of INP surface area for interpretation? (Alpert and Knopf, ACP, 2016; Knopf et al., ACS Earth & Space Chemistry, 2018).

4. Apply NASA GISS ModelE2.1 with dust module that distinguishes eight dust minerals and accretions of iron oxides with the other minerals (Perlwitz et al. 2015), while taking into account soil aggregation, partial fragmentation at emission (Kok, 2011) in the saltation range (< 20 μm particle diameter) as well as emission of large dust particles, based on empirical data (20-50 μm) (Kandler et al. 2009). We examine the importance of dust source distribution, dust size distribution, particle mixing state, and air temperature for simulating immersion freezing by mineral dust particles.

Ice Nucleating Particles From the ACE-ENA Ground Site



MOUDI Impactor:
Stage 6, cut-off: 560 nm

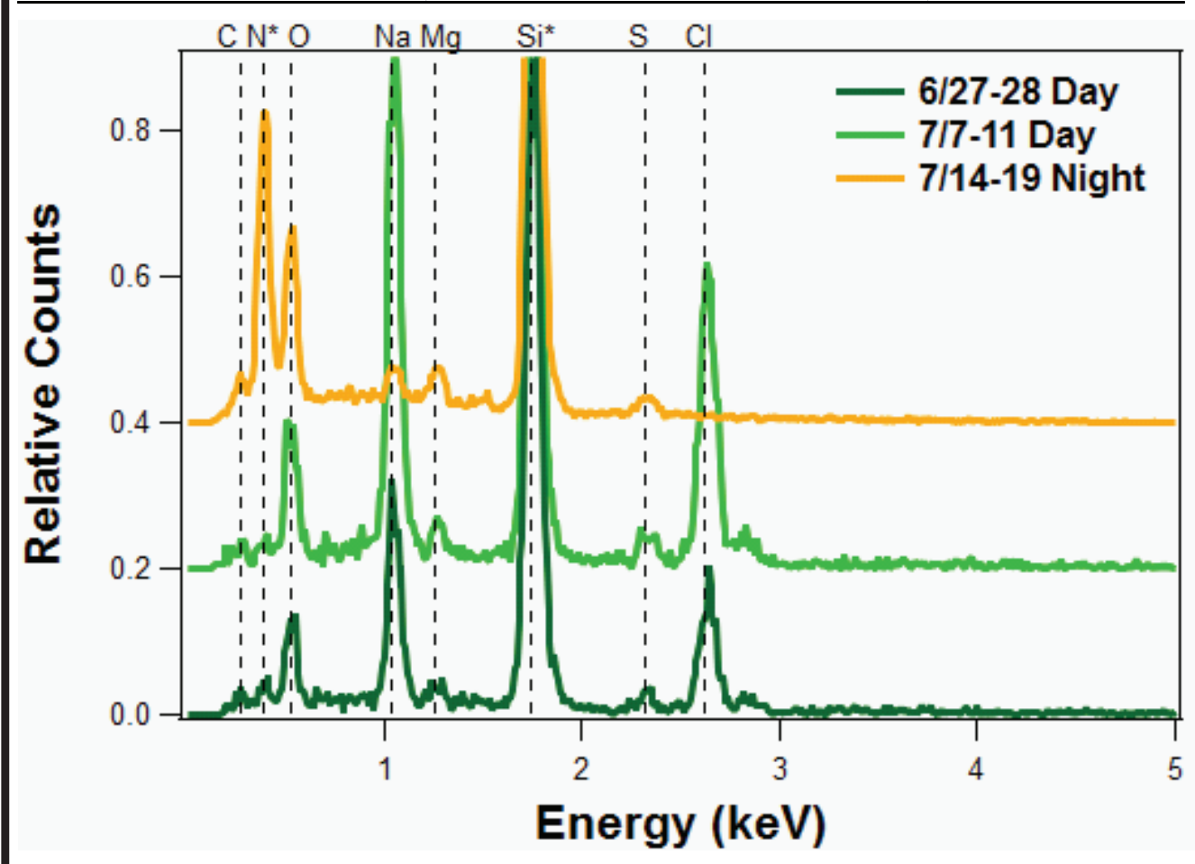
Day 1: 7/17, 7/18, 7/10, 7/11
Day 2: 6/27, 6/28
Night 1: 7/14-7/15, 7/15-7/16, 7/16-7/17, 7/18-7/19
Night 2: 7/2-7/3, 7/3-7/4, 7/7-7/8, 7/8-7/9

Sampled particles can induce ice nucleation at conditions (RH, T) relevant for mixed-phase and cirrus clouds.

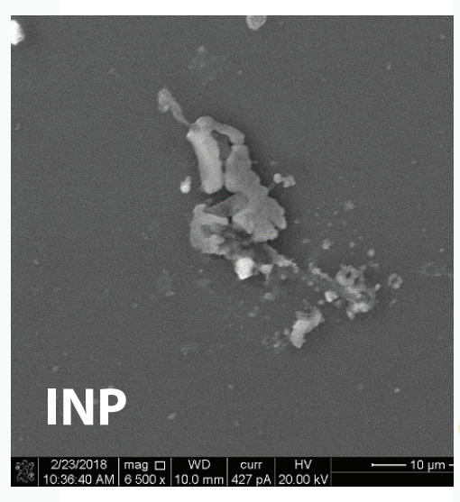
Data suggest different INP types are present at day and night.

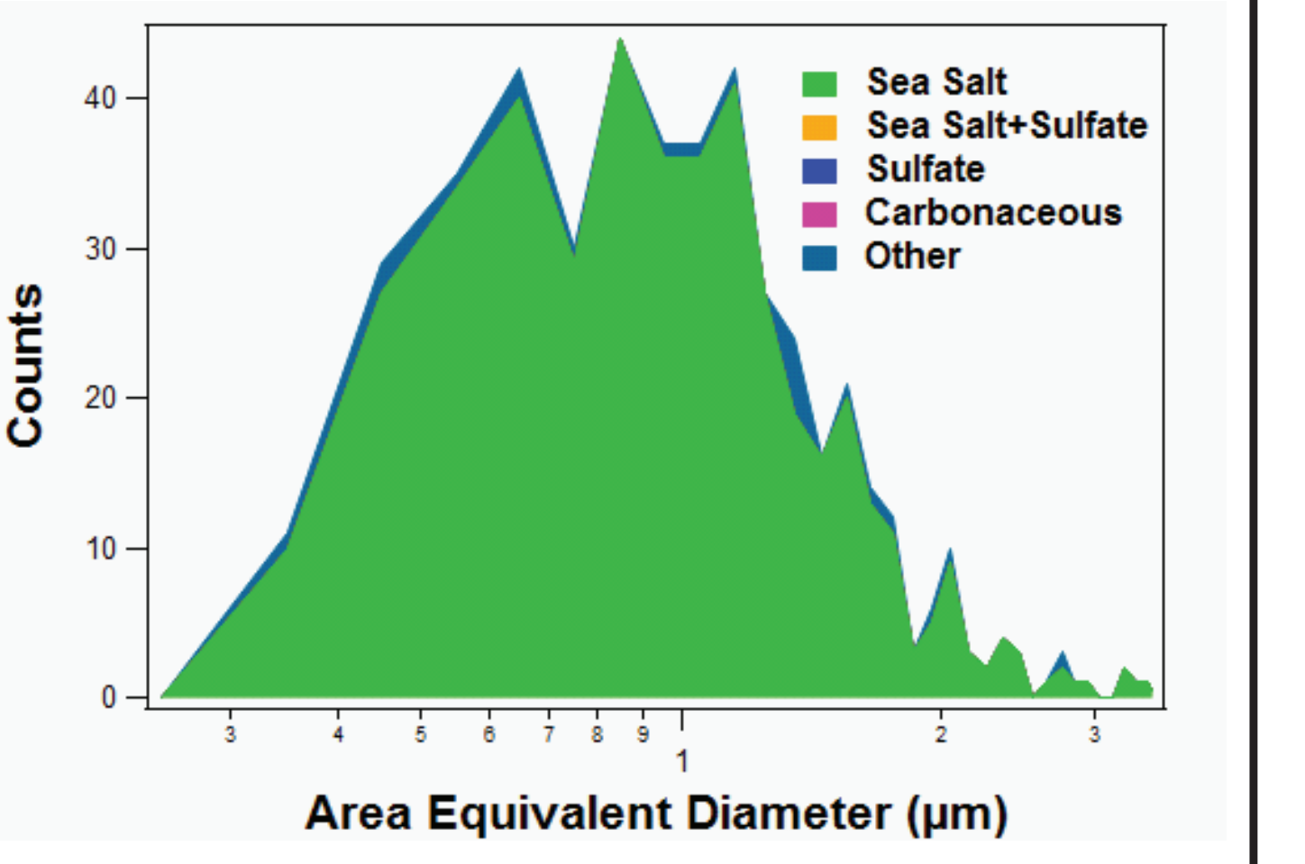
Ice Nucleating Particle Characterization (ACE-ENA Ground Site)

INPs are chemcially different between day and night:



Daytime INP and particle composition:

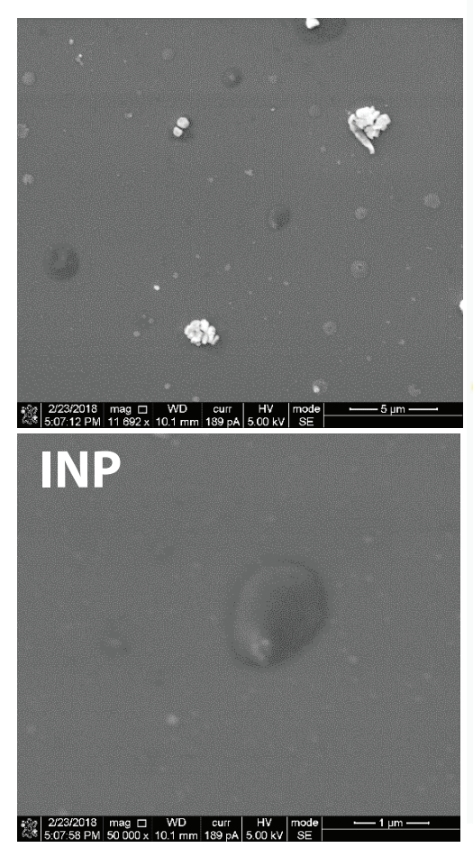


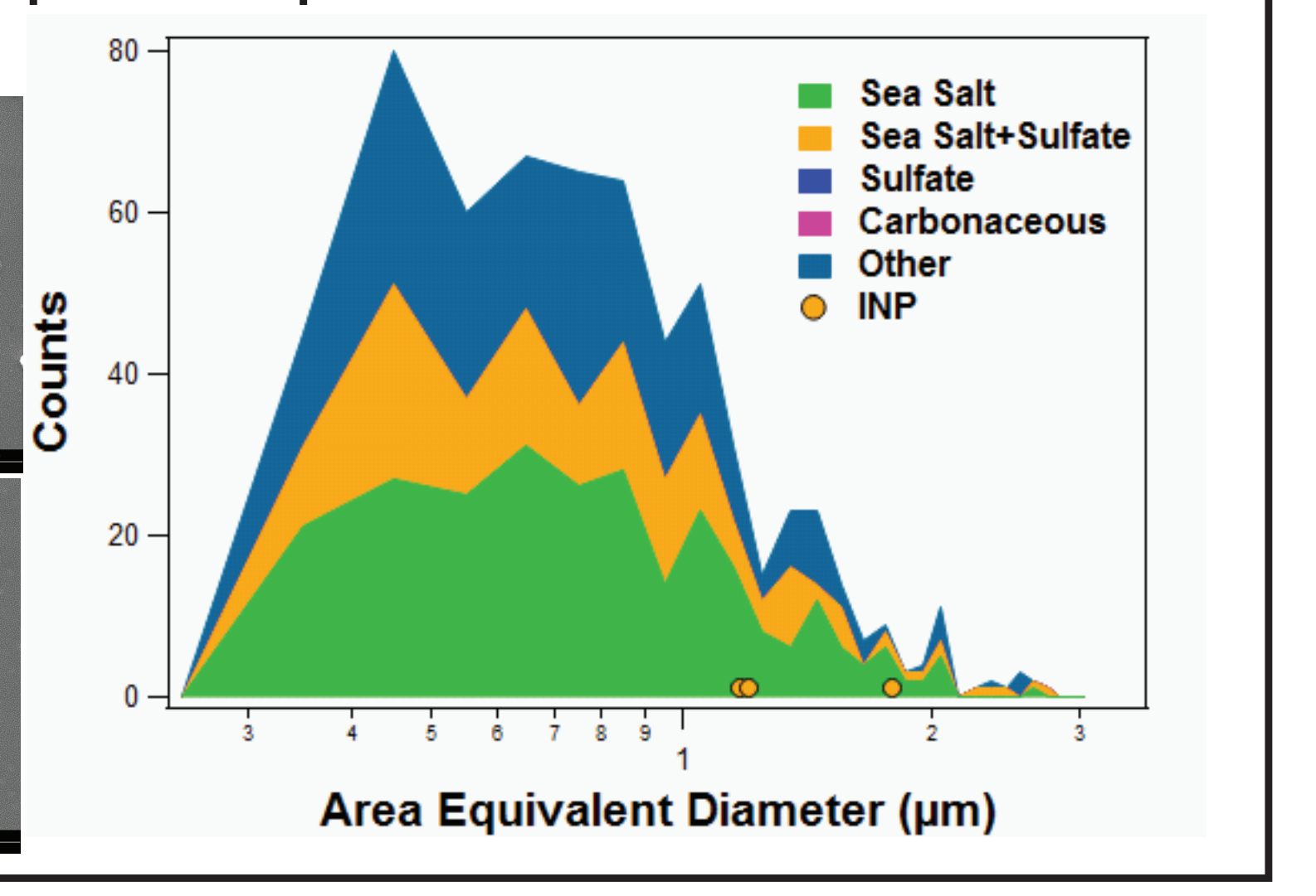


Nighttime INP and particle composition:

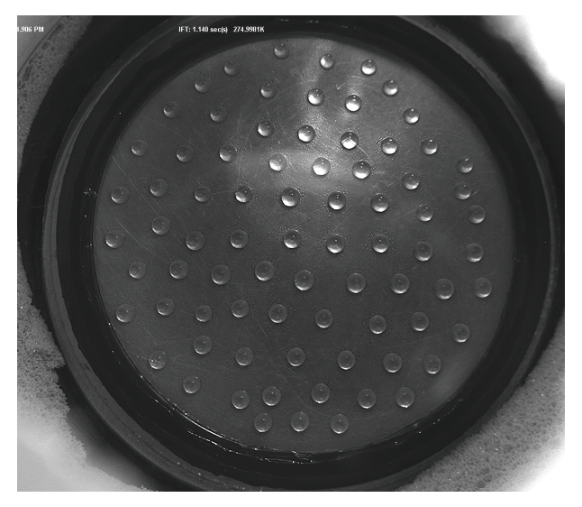
During daytime, particle and INP composition are dominated by sea salt, reflecting the marine environment.
During nighttime, particle and INP composition contain more organic material. Ice nucleation humidity threshold by nighttime aerosol particles does not show strong dependence on temperature (see panel to left).

INPs belong preferentially in the supermicron size population of particles similar to previous observations in literature (Knopf et al., ACS Earth & Space Chem., 2018).



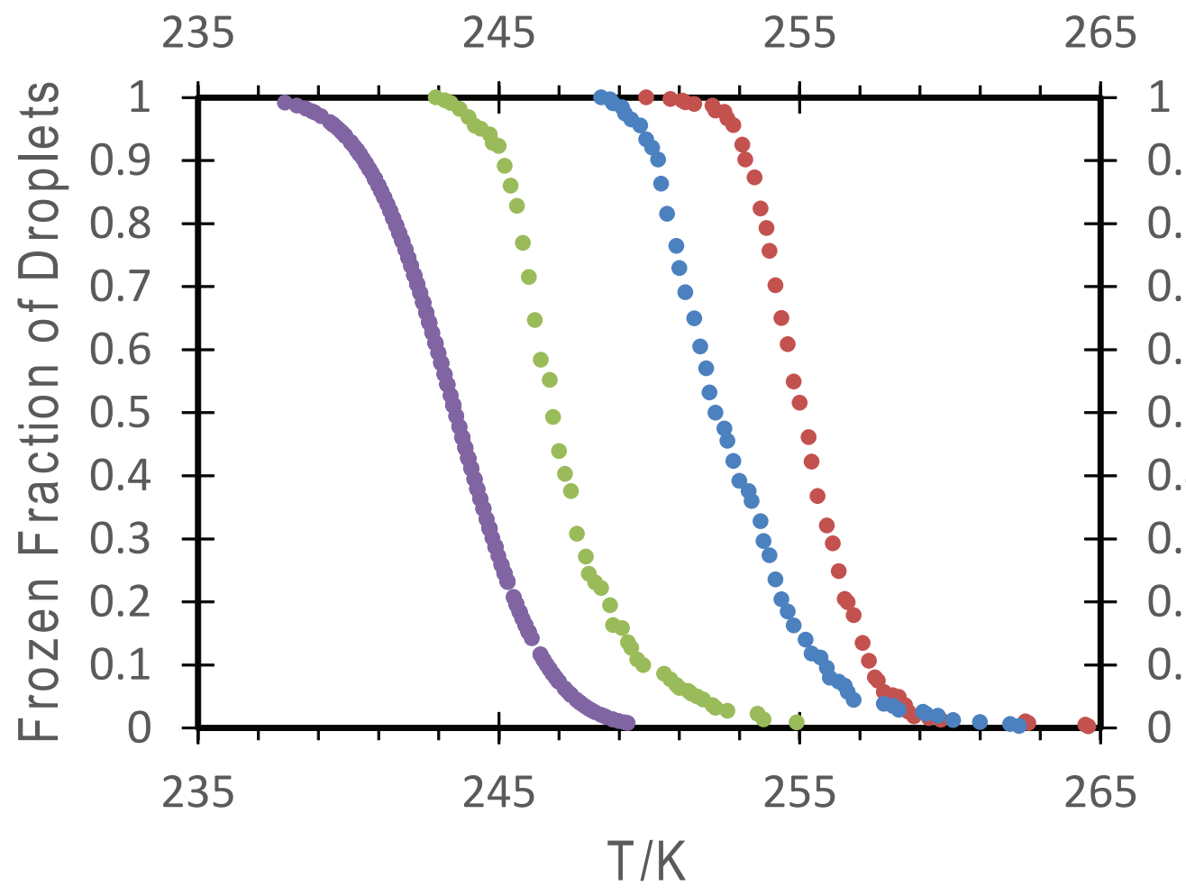


Externally - Internally Immersion Freezing - Feldspar, Kaolinite, Illite

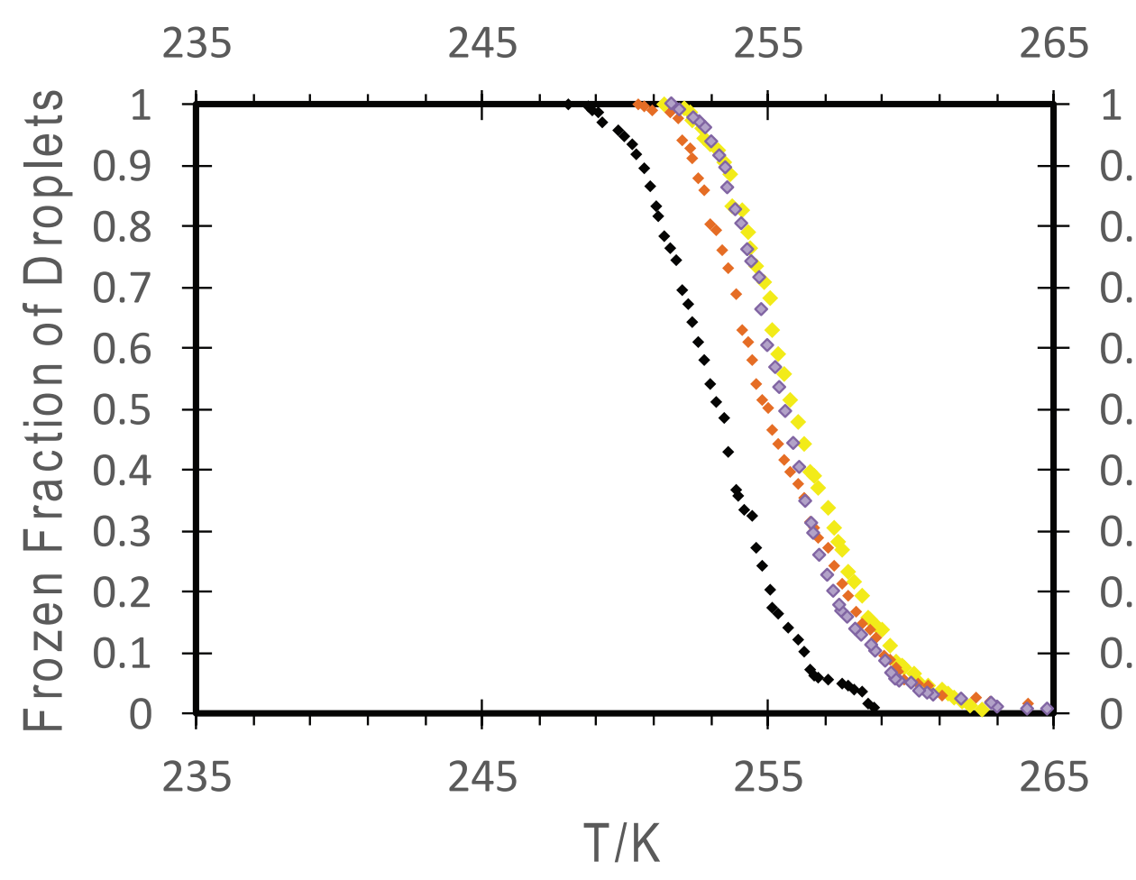


0.2 μL droplets containing either Feldspar, Kaolinite or Illite. Competitive experiments: Either externally or internally mixed droplets are present at same time on substrate.

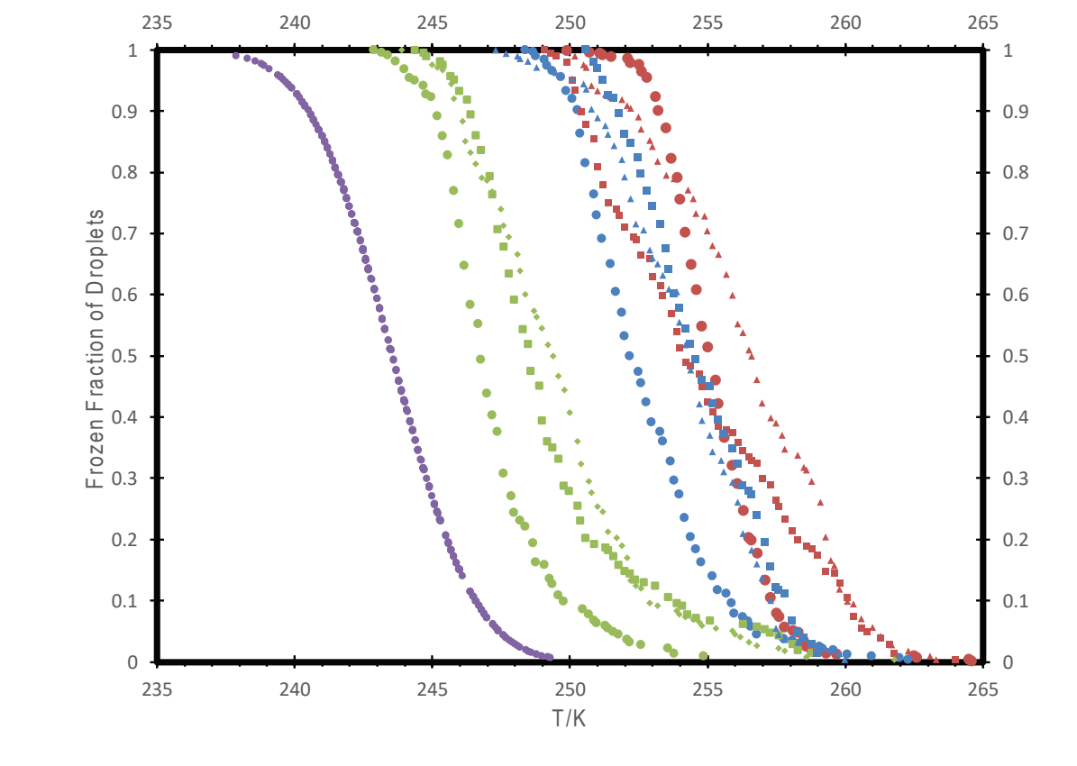
Water and Single Components Droplets



Internally mixed:



Externally mixed:



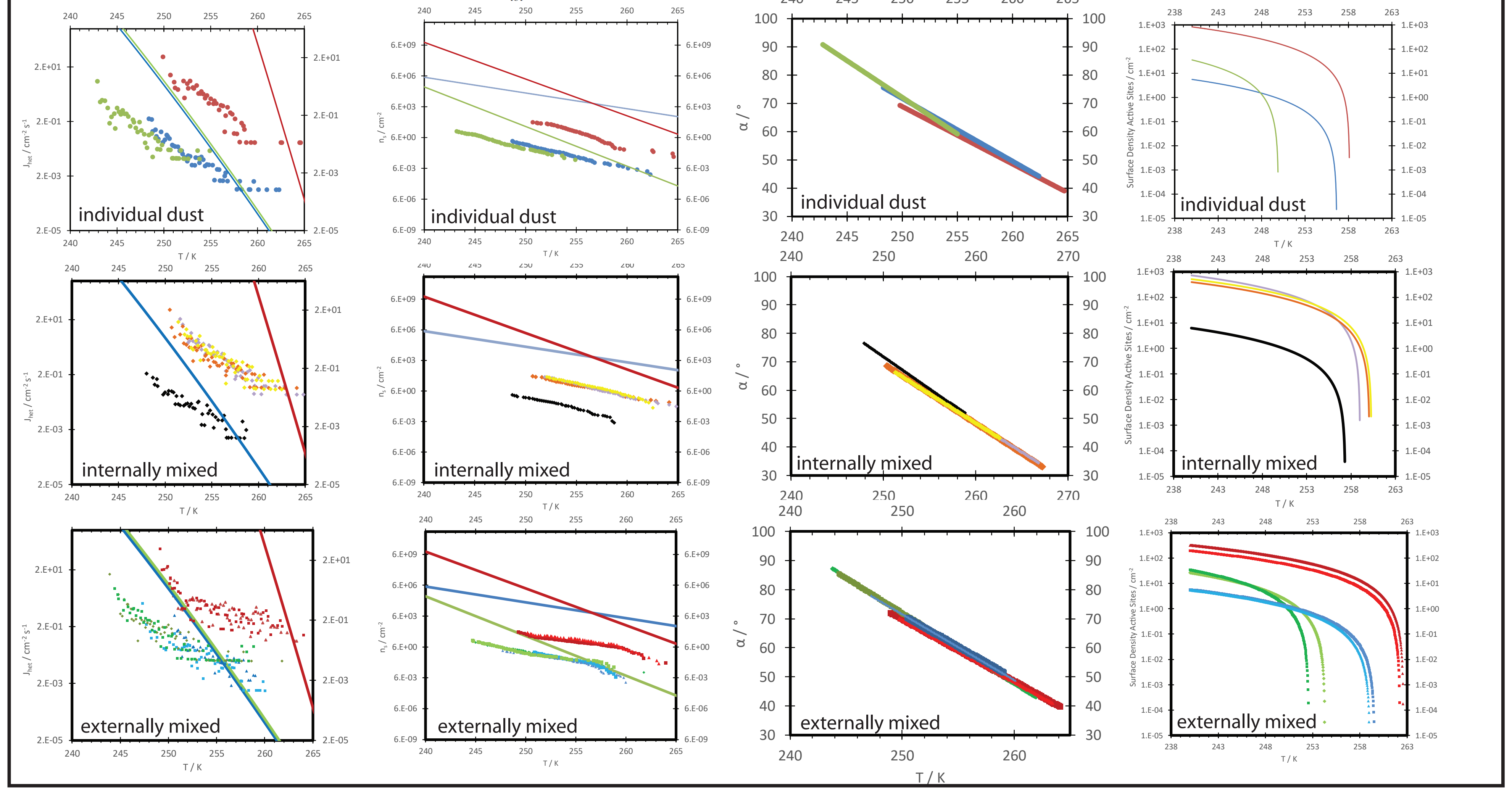
Immersion Freezing Descriptive Approaches - Constant Cooling Rate

$$\frac{N_{frz}}{N_{tot}} = 1 - e^{-J_{het} A_g t} \qquad J_{het}^{exp}\left(T^i\right) = \frac{n_{nuc}^i}{t_{tot}^i \cdot A^i}$$

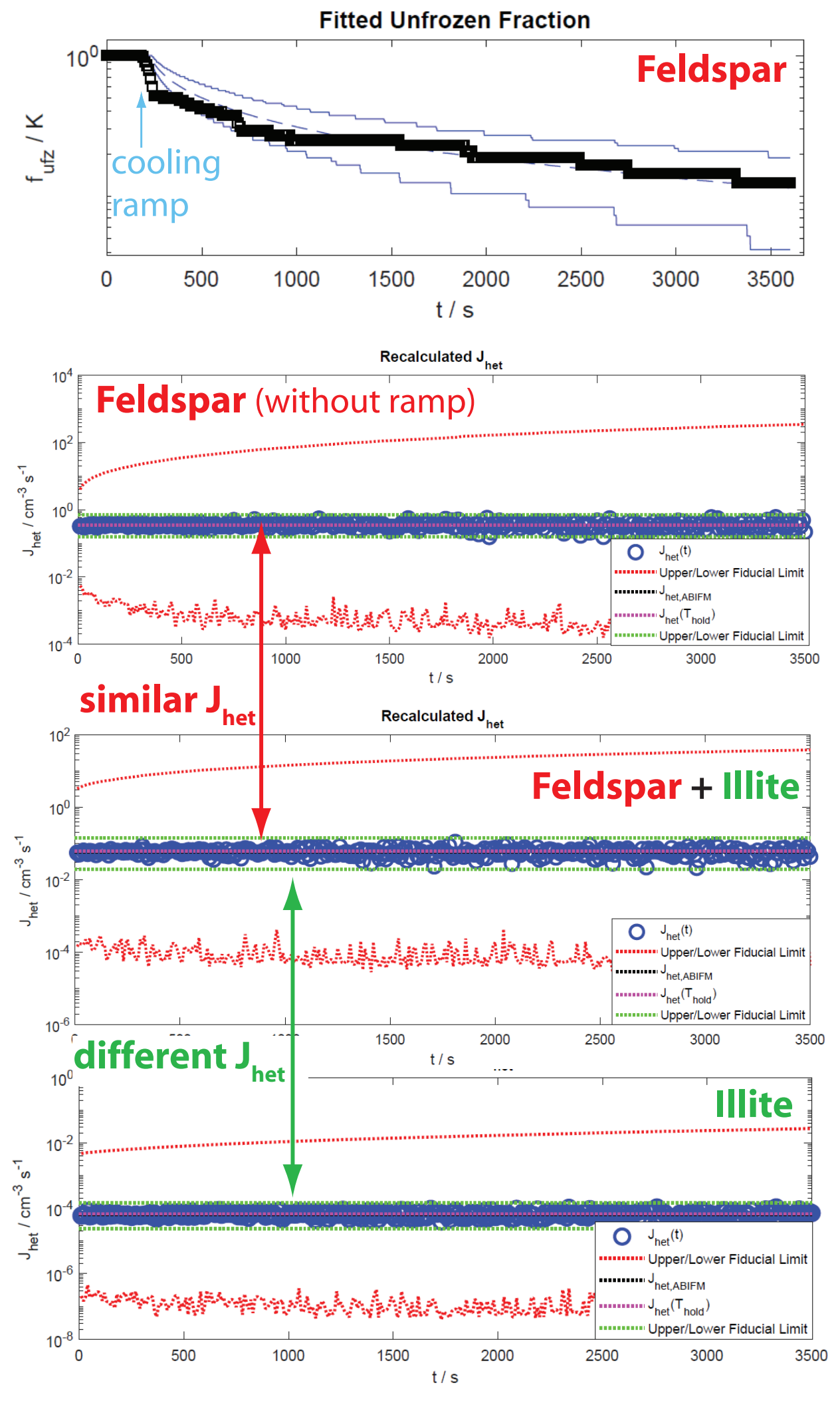
$$f_{ice}(T) = \frac{n_{ice}(T)}{N_{tot}} = 1 - \exp(-n_s(T)s)$$

$$J_{het}(T,\alpha) = \frac{kT}{h}\exp\left[-\frac{\Delta F_{diff}(T)}{kT}\right] n \exp\left[-\frac{\Delta G_{act}(T) f_{het}(\alpha)}{kT}\right]$$

$$\frac{dN_f}{dT} = (N_{tot} - N_f)\overline{A} K^{a_w}(T^i)$$



Isothermal Immersion Freezing



New Monte Carlo based code that accounts for INP surface area variability and stochastic nucleation process. It also includes initial cooling ramp to reach temperature of isothermal freezing experiment.

Feldspar and illite experiments can be described using single J_{het} (in agreement with J_{het} from cooling rate experiment) and variance in INP surface area.

Mineral dust in one 0.2 μL droplet. INP surface area varies by orders of magnitude from droplet to droplet.

Isothermal experiments of internally mixed Feldspar + illite can be described using particle surface area and J_{het} of Feldspar. In other words, in internal mixture Feldspar dominates the isothermal freezing behavior.

Earth System Model Study: INP Dependence on Feldspar Source and Size Distribution for Active Site Approach

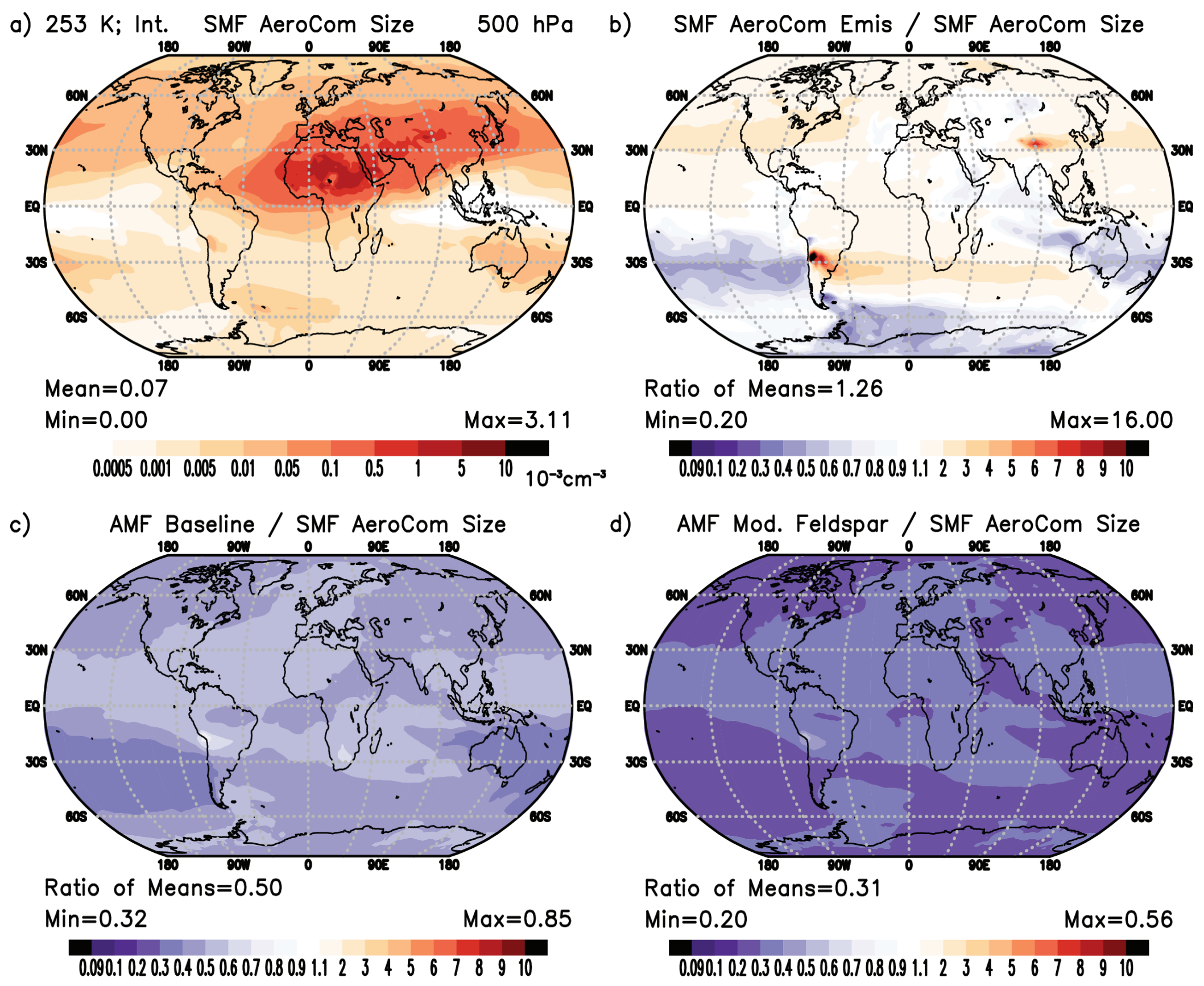


Figure 1. INP concentration at 500 hPa for activation temperature of 253 K and internal mixing.

For a fixed activation temperature (Fig. 1a, b), different source distributions between AeroCom and ModelE can lead to larger regional differences, e.g., exceeding 50% over some Southern Ocean regions. This indicates a significant sensitivity of the regional INP number to the assumptions on the dust sources between different models. Accounting for the various aspects that make the size distribution of emitted dust physically more realistic, compared to SMF Aerocom size, reduces the INP concentration by a factor of 2-3 for the global mean and up to a factor of 5, regionally (Fig. 1c, d).

Mineral Dust Mixing State Effect in Global Model

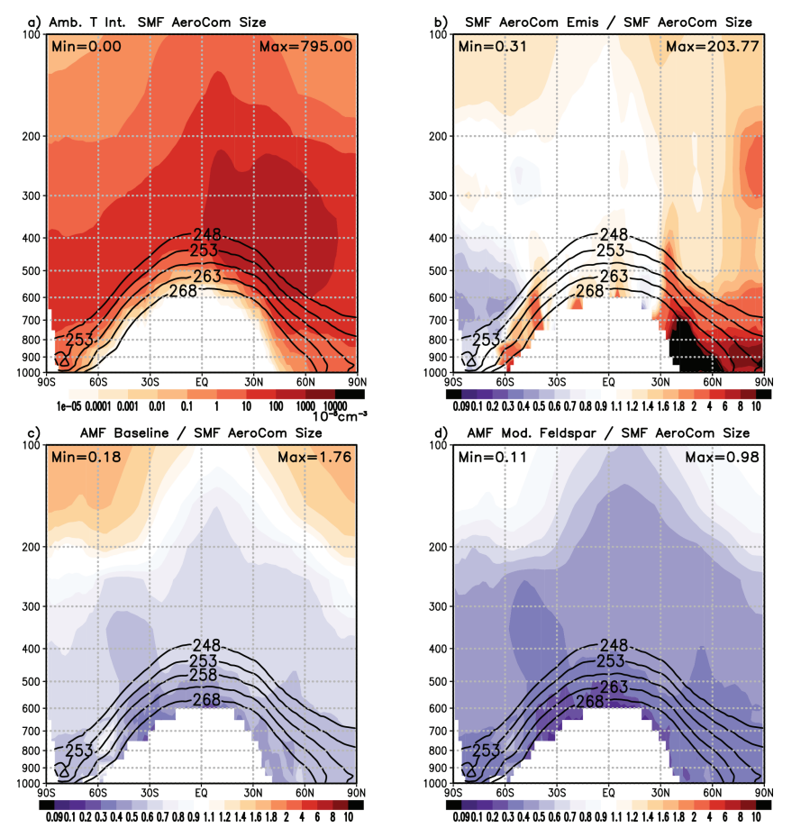


Fig. 1. Altitude – zonal mean cross section of INP concentration

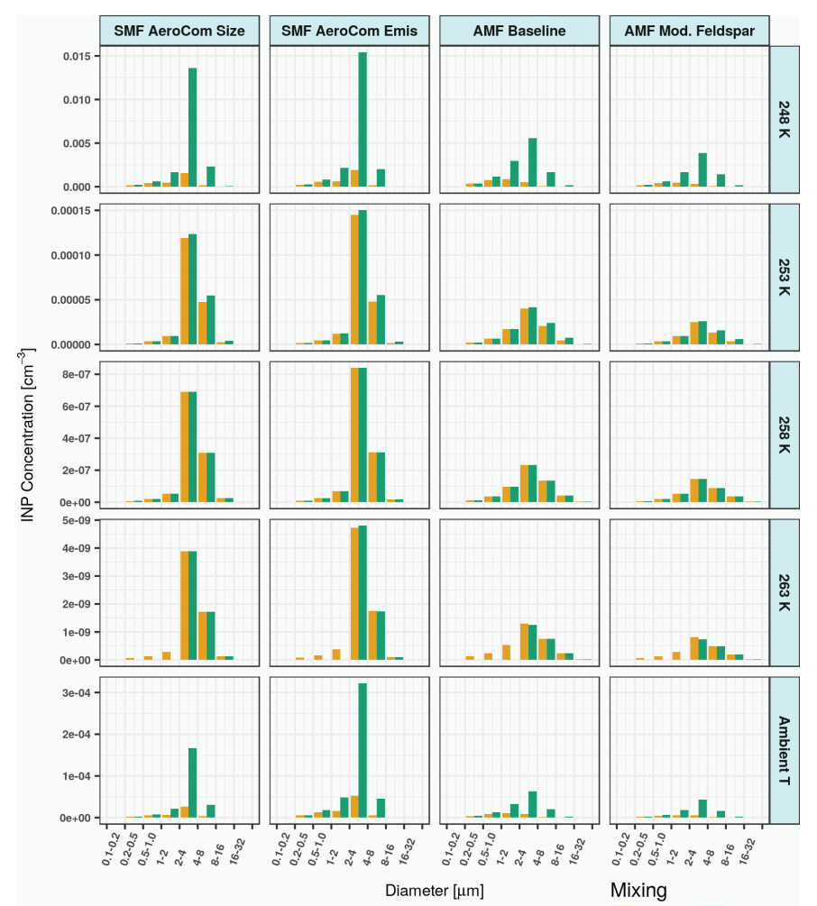


Fig. 2. Size resolved INP concentration at 500 hPa.

- The vertical distribution of the INP concentration shows strong sensitivity to the dust source distribution (AeroCom vs. ModelE), especially in the Northern Hemisphere (Fig. 1a,b) for ambient temperatures.
- The bias from assuming a simple mineral size distribution *vs.* more physically realistic mineral size distributions (Fig. 1c, d) leads to an overestimate of the calculated INP up to a factor of 10 in the tropics at altitudes where temperatures are at the upper end of the range for immersion freezing of K-feldspar (ca. 268 K).
- Fig. 2 displays the size resolved INP concentration at 500 hPa for the different fixed activation temperatures and for ambient temperature distribution both for external and internal mixture.
- The largest INP contribution comes from the diameter range of 2-4 μm, except at 248 K for external mixing in the AMF cases. Even though the total INP number is reduced in the AMF cases, a relatively larger INP number fraction relative to the total INP number is found for sizes greater than 4 μm.
- Results vary only a little with the activation temperature and mixing state of feldspar, except at the coldest temperature (248 K).
- The sensitivity of the INP concentration for ambient temperatures resembles the one for 248 K.

Summary and Conclusions

• Particles collected during ACE-ENA at the ground site can act as INPs under typical mixed-phase and cirrus cloud conditions. INP and particle population characterization indicate that during daytime particles are dominated by sea salt. However, during nighttime the chemical make-up of the particles changes and in turn alters their ice nucleation propensity.
• Immersion freezing experiments have been conducted characterizing the freezing of droplets with single component mineral dust surrogates and with externally and internally mixed mineral dust droplets with same abundance of mineral dust as in single component droplets.
• Single and external/internal mixture immersion freezing experiments were analyzed using $J_{het}(T)$, α(T), α-PDF, n_s, and deterministic model. Under same experimental conditions the freezing of the externally and internally mixed dust droplets can be predicted by single component characterization considering typical experimental uncertainties. Isothermal immersion freezing experiments can be described using CNT with constant J_{het} and realistic variation in particle surface area.
• An Earth System Modeling study reveals the importance of mineral dust source size distribution for prediction of atmospheric INPs. The relatively largest contribution to the total INP number comes from the size range 2-4 μm. Our model study reveals significant uncertainty (up to 2 orders of magnitude) in the calculated regional INP concentrations when the dust source distribution is changed from the ModelE to the AeroCom source distribution, everything else equal. The overestimate in the INP due to assuming a simple size distribution of dust at emission *vs.* a more physically realistic one may be up to a factor of 10, regionally in the tropics at the highest activation temperatures of K-feldspar for immersion freezing. That is, this bias may be up to five times larger than previously stated in the literature.

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