Ice Nucleating Particles from the Amazon Region

Amazon field-collected aerosol particles acting as ice nucleating particles (INPs) versus non-INPs for above and below forest canopy. Sampling Panels (a) and (b) show NEXAFS X-ray spectra of individual INPs and non-INPs. Panels (c,e) display binary component mapping of individual Amazon particles at different heights. Blue, green and red components correspond to inorganic material, organic material (COOH dominated), and carbon double bonding (C=C), respectively. Scale bar = 5 µm.

NEXAFS spectra of INPs are similar (a). Non-INPs can be similar to INPs (b) but also significantly different (d). INPs likely represent a major particle type class of which some nucleate ice. Amazon INPs have same NEXAFS spectrum as marine INPs dominated by polysaccharide material (see a,b).

Motivation

Despite much research effort, accurate prediction of atmospheric ice nucleation still remains a grand challenge. Reasons for this are manifold and include cloud dynamics as well as aerosol microphysical processes. Our research focuses on the physicochemical characterization of ice nucleating particles (INPs) and how different particle types impact ice nucleation. We approach this challenge employing modeling studies and laboratory experiments using collected and laboratory generated aerosol particles. Currently we are working on the following tasks:

1. Use Southern Great Plains ARM site collected ambient aerosol particles for identification of INPs, evaluation of the particles’ propensity for nucleating ice, and contrast physicochemical properties of INPs with the ambient particle population. Particular focus is placed on the role of airborne soil organic particles (ASOPs) that may act as INPs.
2. Physicochemically characterize ambient aerosol particles collected in the Amazon and examine the particles’ ice nucleation properties. We focus on supermicron-sized particles, potentially reflecting biological particles. The analyses include particles collected above and below the forest canopy.
3. Conduct immersion freezing experiments employing mineral dust particles. We investigate how sample treatment impacts INP surface area and thus interpretation of ice nucleation kinetics. Perform ice nucleation experiments with mixed mineral dust samples to better mimic the forest canopy.
4. Apply an Earth System Model with an improved aerosol mineral fraction to evaluate the atmospheric mineral dust aerosol population. The new model approach is based on brittle fragmentation theory which considers re-aggregation and partial fragmentation at emission.

Results – Testing Size Distribution and Source Distribution

- Figure 2a displays the geographical distribution of INP concentrations for 1- to 10-µm-sized soil mineral fractions to dust mineral fractions, Aerosolcom size distribution, and ModelL sources (reference experiment). Figure 3a shows the vertical and zonal-mean distribution for the same experiment. The INP concentrations are shown for an activation temperature of 273 K.
- Using the same size distribution, but the Aerosolcom-GOCART source distribution instead leads both to modifications in the geographical (Fig. 2b) and vertical (Fig. 3b) distribution of the INP concentration. Different source distributions can obscure the effect by different size distributions.
- Using the new version of the dust module for the prognostic mineral species with size distributions based on brittle fragmentation theory and empirical data (Figs. 2c, 3c), reduces the calculated INP concentration by 50% for the same source distribution. The decrease is not geographically uniform, but depends on the mineral composition of the soils.
- Assuming the same size distribution for Feldspar as for quartz, where the size distribution is shifted to larger particle sizes, decreases the INP concentration even more (Figs. 2d, 3d).

Competitive Immersion Freezing - Kaolinite vs. Illite

0.2 µL droplets containing either Kaolinite or Illite. Competitive experiments: Illite and Kaolinite droplets present at the same time on substrate; Duct filtered 0.8 µm - 11 µm.


Mixed population behaviors as expected from single droplet experiments.

 Differences to parameterizations: explainable by uncertainty in particle surface area.

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Different source size distributions result in significant changes of INP number concentrations

- In all experiments, fine silt Feldspar is most active of all sizes to serve as INP.
- Activation is strongly reduced with the new version of the dust module due to the shift of the size distribution to larger particle sizes.
- The INP concentration shows little dependence on the mixing state of Feldspar for warmer activation temperatures.
- In contrast, for the lowest activation temperature, the INP concentration is an order of magnitude larger for internal mixing than for external mixing of Feldspar.
- When saturation of active sites is reached for lower temperatures, the difference between external and internal mixing is dramatically reduced by up to 50% of dust particles available for activation as INP.

Summary and Conclusions

- Preliminary results suggest that SGP site collected aerosol contains inorganic and organic dominated particles. These active particles are dominated by inorganic material with potentially incomplete organic coatings. These particles do not represent very efficient INPs forming ice at the homogeneous ice nucleation limit.
- The population of supermicron Amazon particles show distinguishable chemical differences to other non-INPs. Interestingly, the Amazon INPs show a similar spectral feature as organic sea spray aerosol acting as INP. The INPs initiate ice formation before reaching the homogeneous freezing limit. For cirrus temperatures, these INPs show similar efficiency nucleating ice as Kaolinite dust particles.
- Immersion freezing experiments that apply droplets that contain either Kaolinite or Illite dust can be explained by nucleation kinetics derived from droplets containing only one dust species. Different sample preparation results in different particle surface areas present in droplets and likely explains differences to previous parameterizations.
- An Earth System Model study reveals the importance of mineral dust source size distribution for prediction of atmospheric INPs on the example of Feldspar. An improved dust module reduces INP concentrations by 50% compared to more simplified assumptions. Presence of larger Feldspar particles does not necessarily result in more INP numbers. At lower temperatures, INP concentrations are sensitive to the mixing state. INP numbers increase with internally mixed dust particles present.

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