The Ice Nucleation Pathway of Different Amorphous Secondary Organic Aerosol: The Role of Oxidation Level and Sulfate Content

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Motivation
Secondary organic aerosol (SOA) is ubiquitous in the atmosphere. Anthropogenic and biogenic SOA can exhibit various amorphous phase states ranging from liquid to solid (Virtanen et al., Nature, 2010; Saukko et al., ACP, 2012, Wang et al., JGR, 2012). We investigate the potential of laboratory generated SOA from biogenic precursor gases isoprene, α-pinene, and longifolene reacting with OH with and without SO2 present to take up water and nucleate ice for tropospherically relevant conditions. These data are set in context with estimated glass transition points.

Ice nucleation pathways investigated in this study.

Physical-Chemical Characterization of SOA Particles with STXM/NEXAFS and SEM
Scanning transmission X-ray microscopy coupled with near edge absorption fine structure spectroscopy (STXM/NEXAFS) and scanning electron microscopy (SEM) was used to characterize SOA particles at Lawrence Berkeley National Laboratory. STXM/NEXAFS allows:

i) discrimination of the inorganic and organic phase
ii) particle mixing state
iii) organic carbon speciation

Scanning electron microscopy (SEM): Rounded morphology or coalesced particles indicate a liquid like phase. While coagulated particles may be highly viscous or glassy.

Ice Nucleation on α-Pinene and Longifolene SOA Particles
Particles on various substrates, e.g. silicon wafer, hydrophobically coated glass slides, and 100 nm thick silicon nitride window membranes, were used in ice nucleation experiments.

Conclusions
- Multi-model approach including optical-, electron-, and X-ray microscopy on various substrates allows to characterize physicochemical particle properties in relation to the particles' cold cloud formation potential.
- These methods extend room temperature measurements of amorphous organic particles to temperatures as low as 200 K.
- SEM provides support for liquid or solid amorphous organic phase state
- STXM/NEXAFS reveals particle internal microstructure. Seed aerosol from SO2 vapors are coated with SOA, demonstrating a 2 phase separation.
- SOA from biogenic precursor gases take up water around estimated glass transition points.
- Biogenic SOA nucleate ice at conditions similar to homogeneous freezing.
- RHice rate dependent ice nucleation experiments reveal a viscosity dependent plasticizing effect of water. This results in different RHice onsets for different SOA types. This suggests that the time scales for glassy α-pinene SOA to (partially) dissolve is shorter than for glassy longifolene particles. In other words, the viscosity of longifolene may be larger than for α-pinene. As a result, the time scales to achieve equilibrium between an aqueous particle phase with sufficient volume to homogeneously nucleate ice and surrounding water partial pressure are different.

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