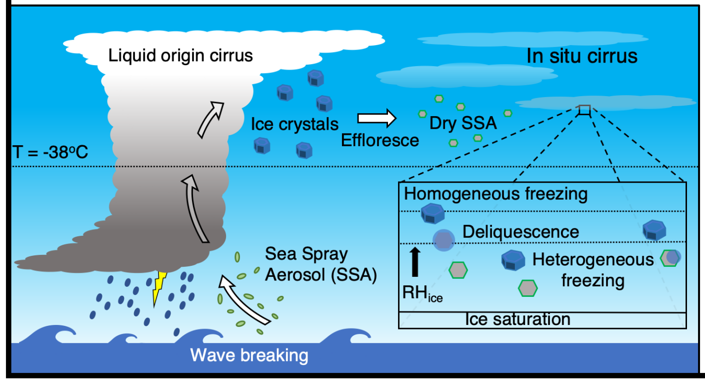
What controls the ice nucleating ability of sea spray aerosols at cirrus temperatures?

Ryan Patnaude, Russell Perkins, Sonia Kreidenweis, Paul DeMott

Colorado State University, Fort Collins, CO

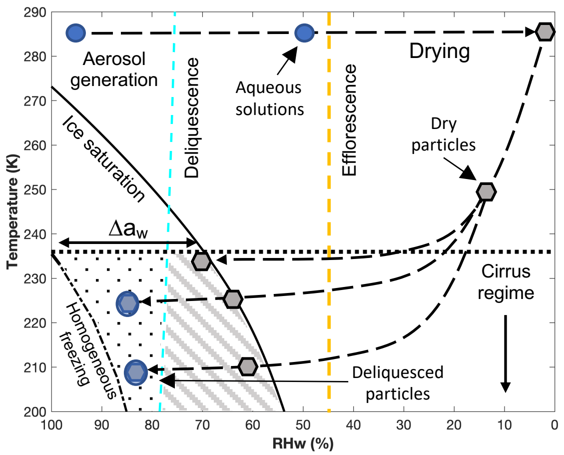
Motivation and background

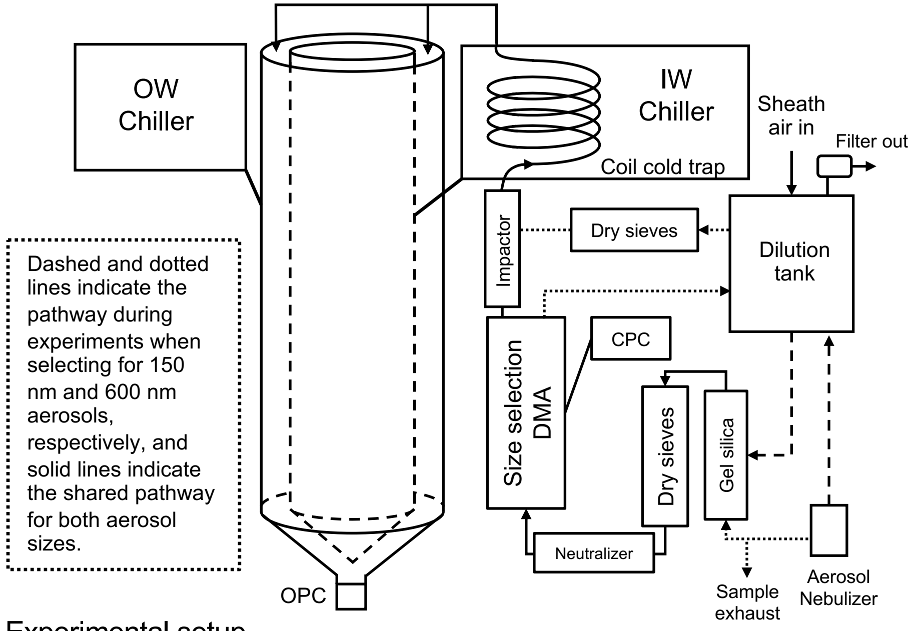


- Cirrus clouds are one of the most abundant cloud types composed entirely of ice below -38°C.
- Sea spray aerosols (SSA) generated by wave breaking may be lofted to cirrus levels through deep convection and detrainment in anvils.
- Wagner et al., (2018) showed that mixed-phase solid/liquid SSA freeze heterogeneously via immersion freezing below 220 K.
- Schill & Tolbert (2014) also observed heterogeneous freezing behavior, which they could not not determine as being either deposition nucleation or immersion freezing
- CSU CFDC modified to go down to lower temperatures < -70°C, making cirrus studies possible.

Methodology

- Continuous Flow diffusion chamber (CFDC) is a concentric cylindrical column with a "warm" outer wall (OW) and "cold" inner wall (IW)
- Linear temperature gradient between walls + nonlinear saturation vapor pressure produces supersaturation in focused lamina between ice-covered walls.
- Operates from ice saturation to water-supersaturated conditions depending on the wall temperature difference



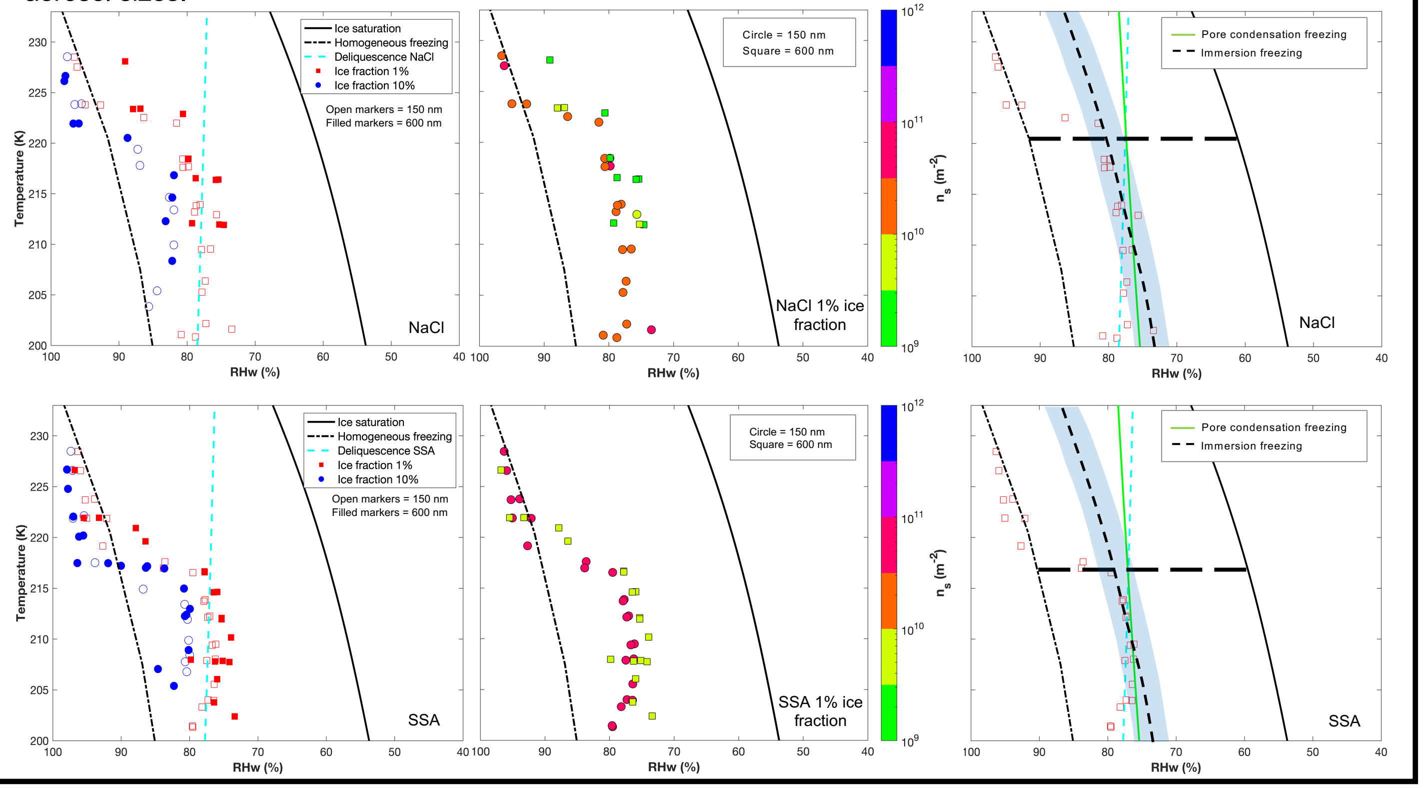


Experimental setup

- Used natural seawater collected at the Scripps Pier in La Jolla, CA for SSA and 3.5% wt NaCl solution.
- SSA and NaCl particles are generated by an aerosol nebulizer, and dried at room temperature.
- Used monodisperse aerosol distribution of ~150 nm and ~600 nm
- Particle phase trajectories (left) help describe the mode of freezing at cold temperatures

Results

- Homogeneous freezing at temperatures > 220 K.
- Heterogeneous freezing transition < 220 K; Lower onset RH_w for SSA particles.
- Similar freezing onsets for both aerosol sizes.
- Higher ice active surface site density (n_s) values for smaller particles.
- Regardless of size or surface area, nucleation onset remains the same → violates the n_s concept and suggests it is not applicable for low temperature SSA freezing.
- Two possible pathways to explain heterogeneous nucleation onset and lack of size dependence.
  1. Immersion freezing
  2. Pore condensation freezing
- Intersection of two lines coincides with onset of heterogeneous freezing.



Conclusions and atmospheric implications

1. NaCl and SSA experiments using CSU CFDC able to replicate heterogeneous nucleation results at cirrus temperatures, similar to previous cloud chamber studies.
2. SSA freeze via heterogeneous nucleation at RH_w ~75% below 220 K at high fractions (10%).
3. Multiple possible freezing mechanisms for NaCl and SSA particles depending on T and RH.
4. SSA particles may represent a significant source of INPs at cirrus levels (see Patnaude et al., 2021).

References: Schill & Tolbert (2014). *Journal of Physical Chemistry C*, *118*(50), 29234–29241. Wagner et al., (2018). *Journal of Geophysical Research: Atmospheres*, *123*(5), 2841–2860. Patnaude et al., (2021). *ACS Earth and Space Chemistry*, *5*(9), 2196–2211.

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