

Future Directions in Modeling Aerosol-Cloud Interactions

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Aerosol-Cloud Interactions and Climate Effects

- Aerosol modifications to microphysics:
 - Modification of cloud radiative properties
 - Modification of distribution of latent heating and precipitation
- Proposed for both warm and cold clouds
- This discussion will focus on cirrus and anvil cirrus, but is relevant to all clouds including the ice phase

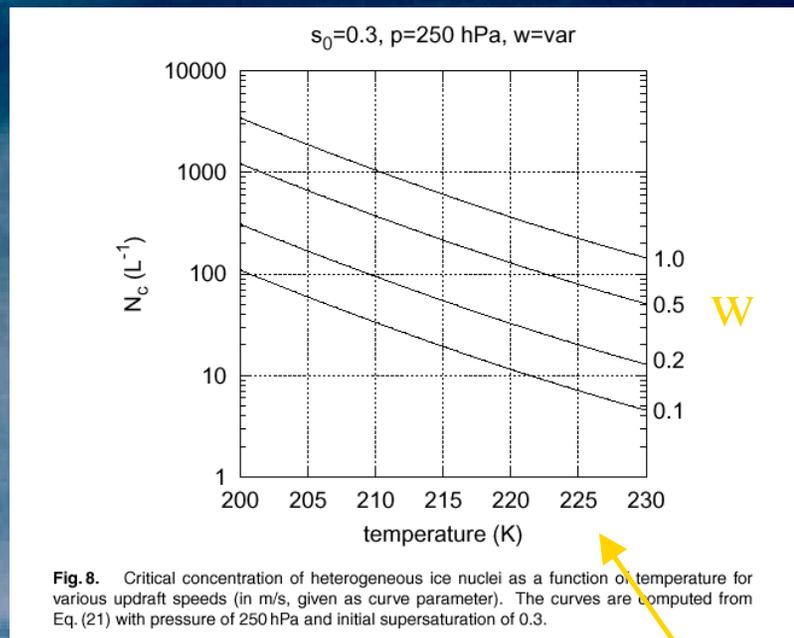
Why are aerosol-ice interactions of interest?

- *IPCC [2001] notes “The indirect radiative effect of aerosols also includes effects on ice and mixed phase clouds, but the magnitude of any indirect effect associated with the ice phase is not known.” Furthermore, “Anthropogenic ice nuclei may have a large ... impact on forcing”, “It is not possible to estimate the number of anthropogenic ice nuclei at the present time.”*
 - Is formation of the ice phase significantly modulated by aerosols?
 - Are a significant fraction of the ice nucleating aerosols anthropogenic?
 - How does the anthropogenic IN component vary spatially and temporally?

Potential effect of aerosols on cirrus are via mechanisms of cirrus ice formation

- Homogeneous freezing of aqueous solution drops, perhaps influenced by chemical phase transitions and other unexplored chemical and kinetic effects.
- Heterogeneous ice nucleation
 - Immersion freezing
 - Deposition nucleation
- Evidence for both processes occurring in the atmosphere, based on RH - T conditions of ice formation
- Can alter the microphysical properties, frequency and duration of high clouds and their forcing.
- Cirrus clouds are *most sensitive to* the abundance of heterogeneous nuclei, their activation characteristics (e.g., onset RH_{ice} and nucleation rate), and updraft (*DeMott et al., 1997; Jensen and Toon, 1997; Kärcher and Lohmann, 2003*)

What we know about aerosol impacts on cirrus forming in-situ

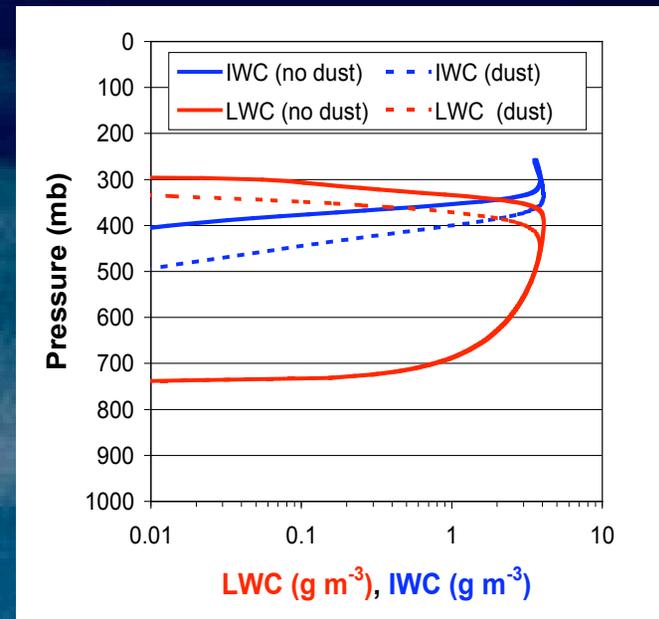
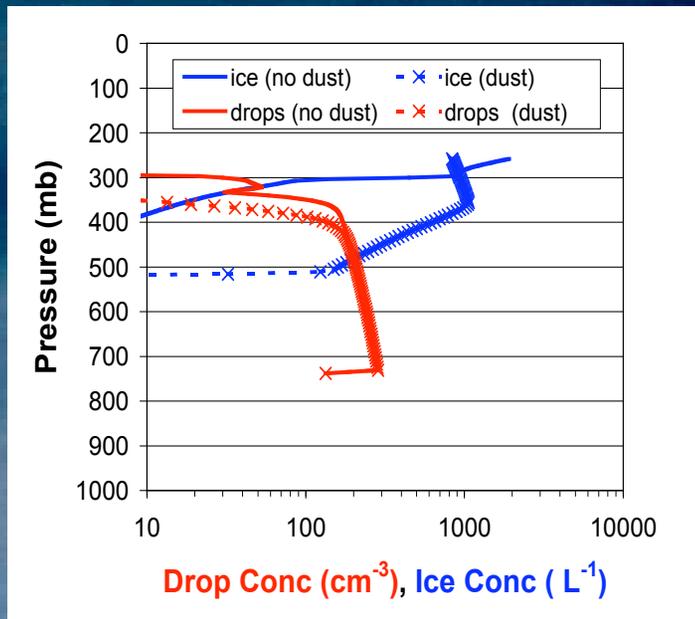


- Mechanism and ice concentration depend on updraft and [IN]
- Little sensitivity to increases in [CCN]
- Heterogeneous [IN] are probably a bigger factor in the NH

(Gierens, 2003)

Figure shows the “critical” concentration of heterogeneous IN triggering a switch in the dominant nucleation mechanism, as a function of T and updraft speed

Dust versus no dust adiabatic simulation of cumuli parcels (8 m s^{-1} updraft)



With dust,

- 1) increased ice concentration and ice mass loading at lower altitude
- 2) water depletion due to ice growth prior to homogeneous freezing
- 3) homogeneous freezing does not occur in dust case, so anvil ice conc. *lower*

In general,

Heterogeneous nucleation lowers UT RH and broadens ice crystal size distribution

Issues to be addressed

- Does homogeneous freezing dominate (everywhere?), or do heterogeneous IN play a role?
 - What are atmospheric sources and number concentrations of IN? How and when do IN compete with homogeneous freezing?
 - Run models with detailed microphysics + obs to elucidate
- Need improved understanding even to run “explicit” models
 - Note widely-varying model intercomparison results for homogeneous-freezing-dominated and homogeneous / heterogeneous competition
- CRMs and GCMs then need a way to predict not only aerosol mass and number concentrations, but IN characteristics as well, to drive any proposed realistic nucleation parameterizations

Advances in Aerosol Modeling in Regional and Global Scale Models

- First efforts aimed at predicting aerosol mass (e.g., global S models)
 - Early warm-cloud indirect-effects parameterizations responded to changes in aerosol mass (e.g., *Lohmann and Feichter, 1997*)
- Current models predict mass *and* number (e.g., PNNL Global Chemistry Model; Ga Tech/GSFC GOCART)
- Focus has been on direct effects and aerosol-warm cloud interactions (sulfate, carbon, dust)

Current Status and Outlook

- CRMs, forecast models, and climate models do not explicitly resolve the ice nucleation process
 - *Kärcher and Lohmann (2002)* propose a parameterization for homogeneous freezing
 - But even “explicit microphysics” models of homogeneous freezing have a wide range of predictions!
 - Model intercomparison by *Lin et al., 2002*
 - Discrepancies arise from various assumptions regarding nucleation rate expressions and crystal growth rates
 - [Aside: similar findings for warm clouds: model intercomparison for adiabatic parcel with SIMPLE input aerosol and sulfur oxidation chemistry, N_d varied by 30% (*Kreidenweis et al., 2003*)]

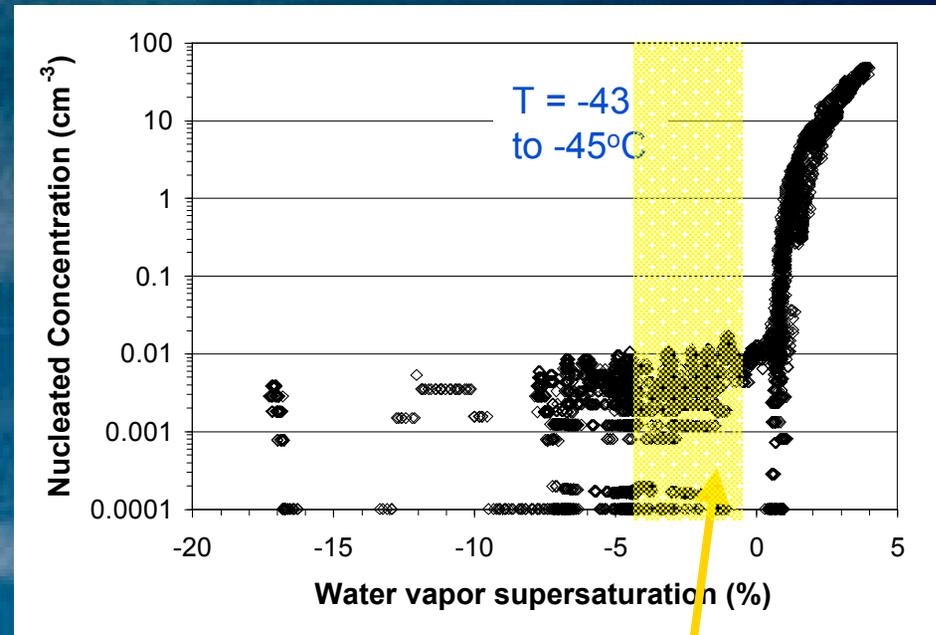
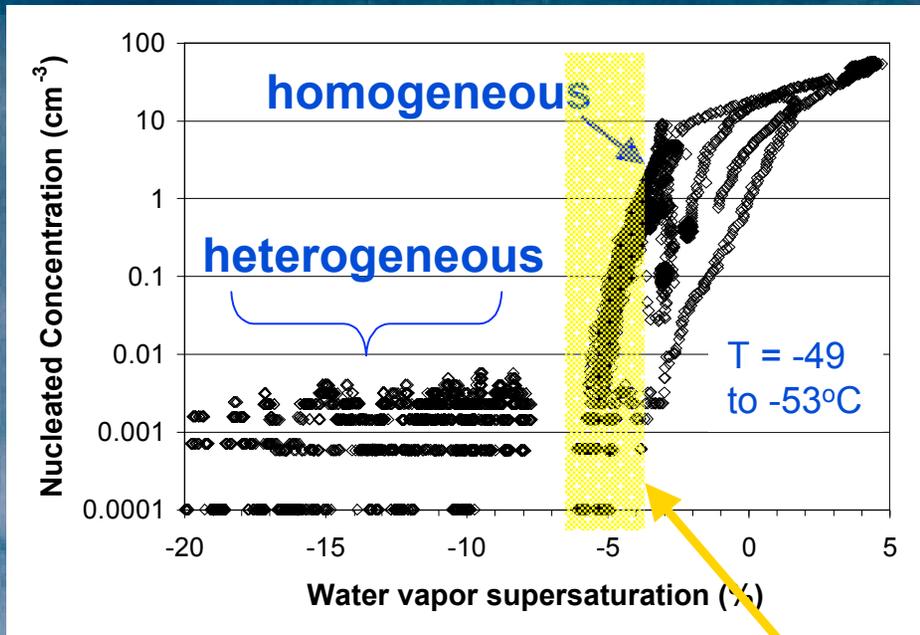
Current Status and Outlook, continued

- New insights on homogeneous freezing (cirrus and anvils)
 - Most studied in recent years
 - **For soluble species studied in the lab thus far, onset conditions for homogeneous freezing depend on size, but not so strongly on composition**
 - **The conditions of homogeneous freezing of background free tropospheric aerosols are relatively consistent with expectations for sulfates, but ...**
 - **Organic aerosol components appear to be associated with chemical and/or kinetic impedance of homogeneous ice formation (field and lab evidence)**
 - In principle straightforward to treat numerically
 - **Depends on water activity or melting point depression (organics...?)**
 - **Also vertical velocity dependent (as all activation processes)**
 - **May not be necessary to specify sensitivity to aerosol size distribution**

Variability noted in conditions required for homogeneous freezing

11/3/00, more typical for mission and expected

11/17/00, unusual



Predicted for sulfates in temperature range of sampling (shading)

Current Status and Outlook, continued

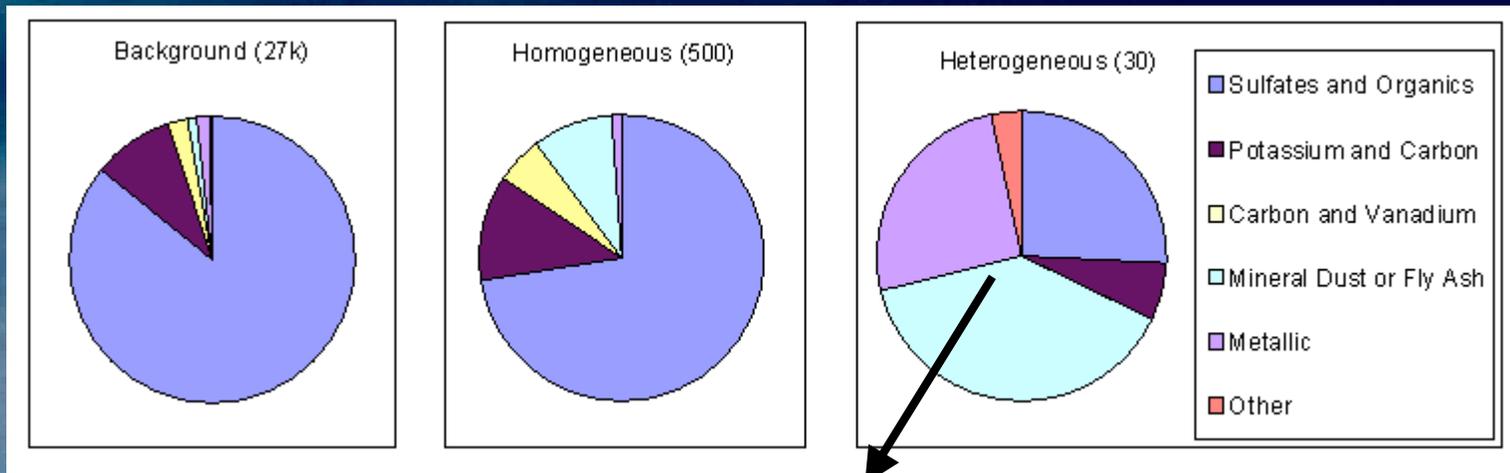
- Many models use *Meyers et al.* (1992) formulation for heterogeneous nucleation
 - Activated nuclei are a function only of supersaturation with respect to ice
 - Derived from limited data available in early 1990's
 - A 1-D cloud model (without prognostic supersaturation) showed strong sensitivity to T and RH traced back to this parameterization (*Benedetti et al.*, 2003)
 - Should be revisited with new information becoming available (more on this shortly)
- Poor constraint on heterogeneous ice nuclei in modeling studies thus far
 - *Kärcher and Lohmann* (2003) also propose parameterization of heterogeneous freezing for GCMs, but require definition of [IN] and whether more than one “type” is present (as suggested by CSU lab and field studies)

Current Status and Outlook, continued

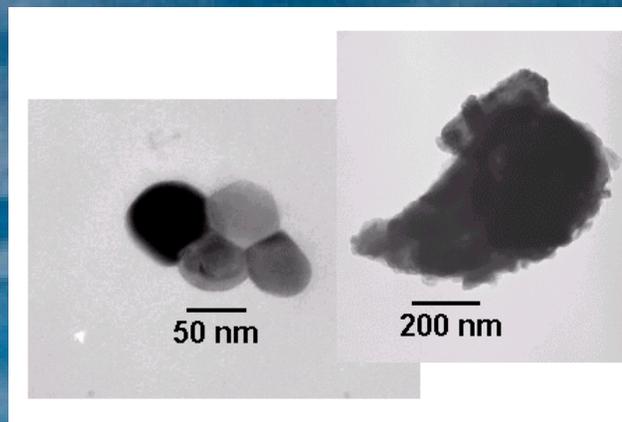
- New insights on heterogeneous freezing
 - Receiving renewed attention (*e.g.*, CRYSTAL-FACE)
 - The concentrations of heterogeneous IN effective at cirrus conditions are usually modest (not unlike assumptions made in some simulations).
 - Mineral dusts are major contributors to free tropospheric IN populations. Concentrations can be by factor of 1000 locally and ~10-50 seasonally.
 - Aerosols of anthropogenic origin or cause (industrial, combustion) are also potential major contributors to ice nuclei effective at cirrus conditions. Biological sources not yet well examined.
 - Suggests strong NH / SH differences in UT [IN] (*e.g.*, INCA results)
 - More complex to treat numerically
 - Activity of dust can be changed by exposure to pollutants
 - Multiple mechanisms, but cannot measure or treat all of these

What are important atmospheric heterogeneous nuclei for cirrus formation?

Statistics of PALMS cluster analyses of particle types



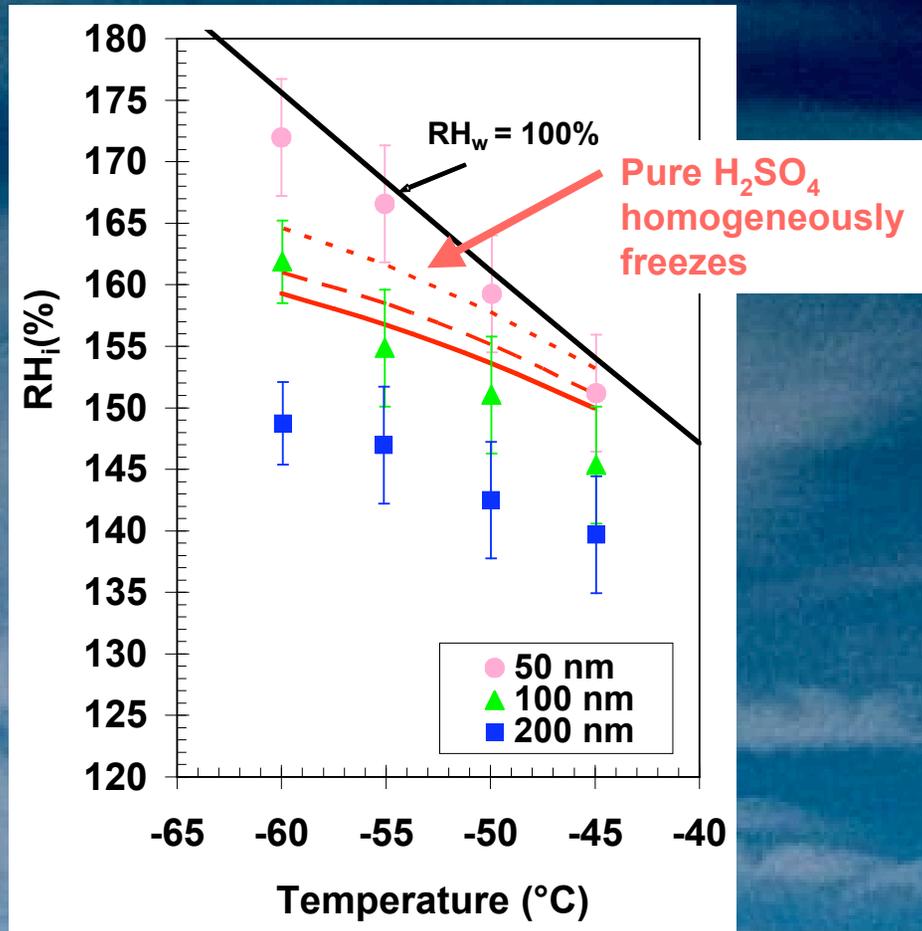
20%



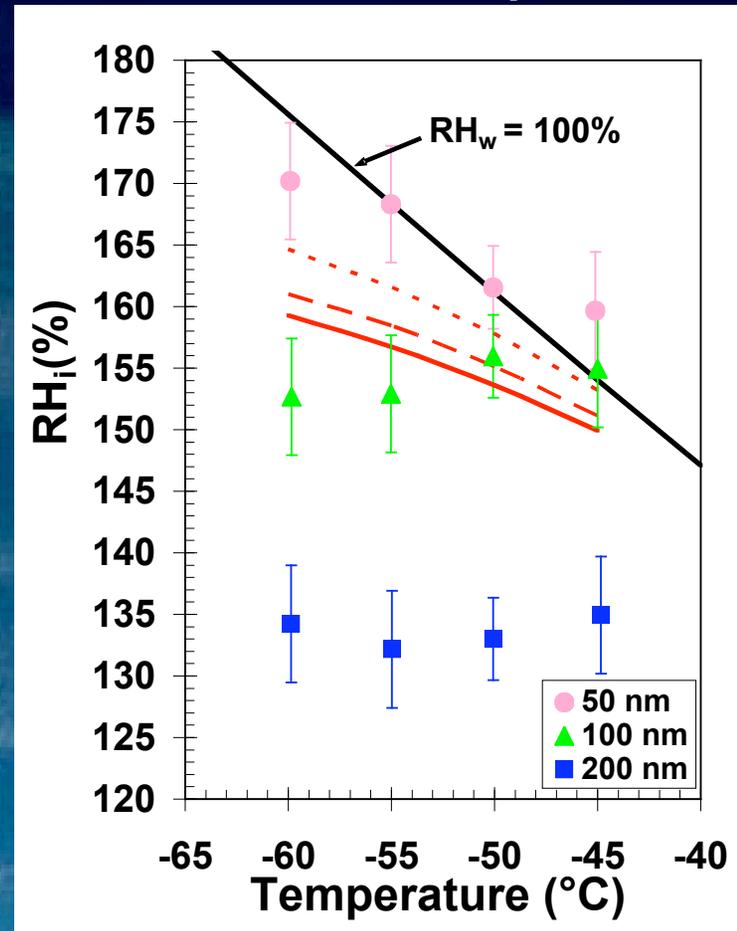
80%

Laboratory studies of ice formation on dusts

Fe_2O_3

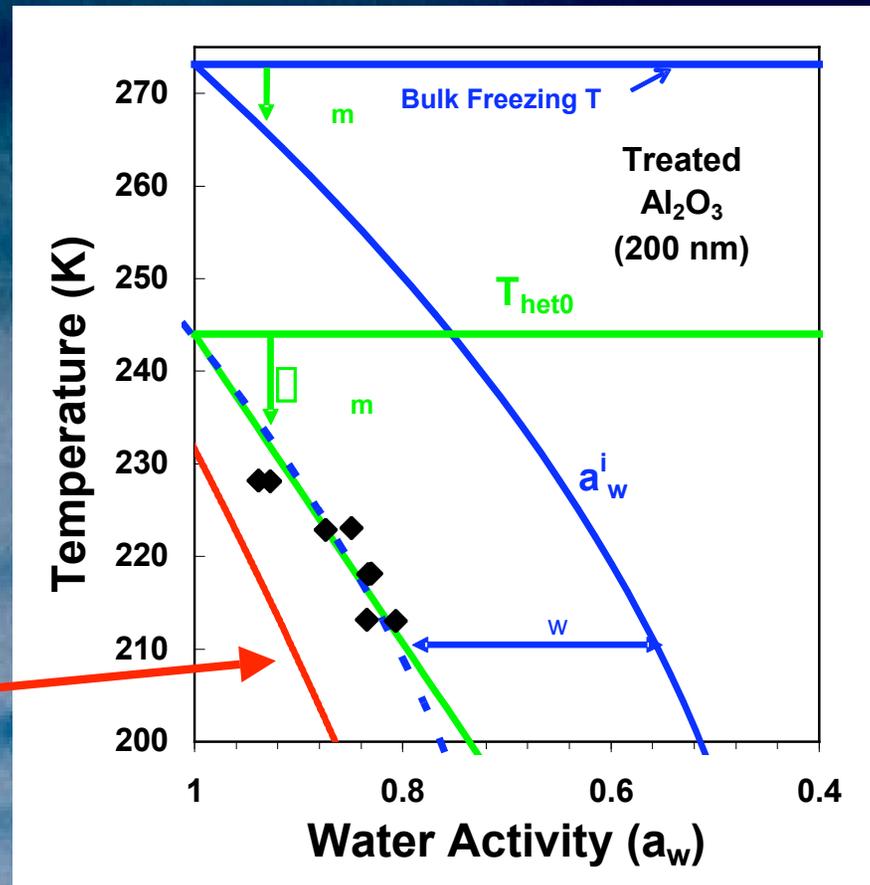


Asian dust sample



Note clear size effect

Parameterizing Heterogeneous Freezing



Coating freezing
homogeneously

Current Status and Outlook, continued

- For both nucleation pathways, particles should not be treated as a specified field (*i.e.*, particle-depleted regions exist in atmosphere after scavenging by cloud formation)
 - *e.g.*, this is ignored by Meyers et al. formulation if applied in bulk microphysics scheme
 - If [IN] has been lowered compared to initial condition, SS_{ice} must rise to activate remaining particles
 - But even prognostic aerosol \neq prognostic IN

Summary

- Lab and field studies are rapidly increasing our knowledge of how ice forms in the atmosphere
 - Homogeneous vs. heterogeneous nucleation is particularly important to understand and to model accurately
- New ice-microphysics parameterizations will be developed using these new insights, particularly for ice initiation
 - Development of prognostic aerosol number and mass treatments in larger-scale models is beneficial
 - But IN are even more difficult than CCN to determine source, sink, and transformation terms for
 - Vertical velocity will be important, so CRMs are potentially better suited than GCMs for application of new microphysical parameterizations
 - Note difficulty in incorporating in a global model, on a subgrid scale, the mesoscale motions largely responsible for cirrus formation