GMI simulations of:
Cirrus ice crystal number
Dust impacts on droplet number

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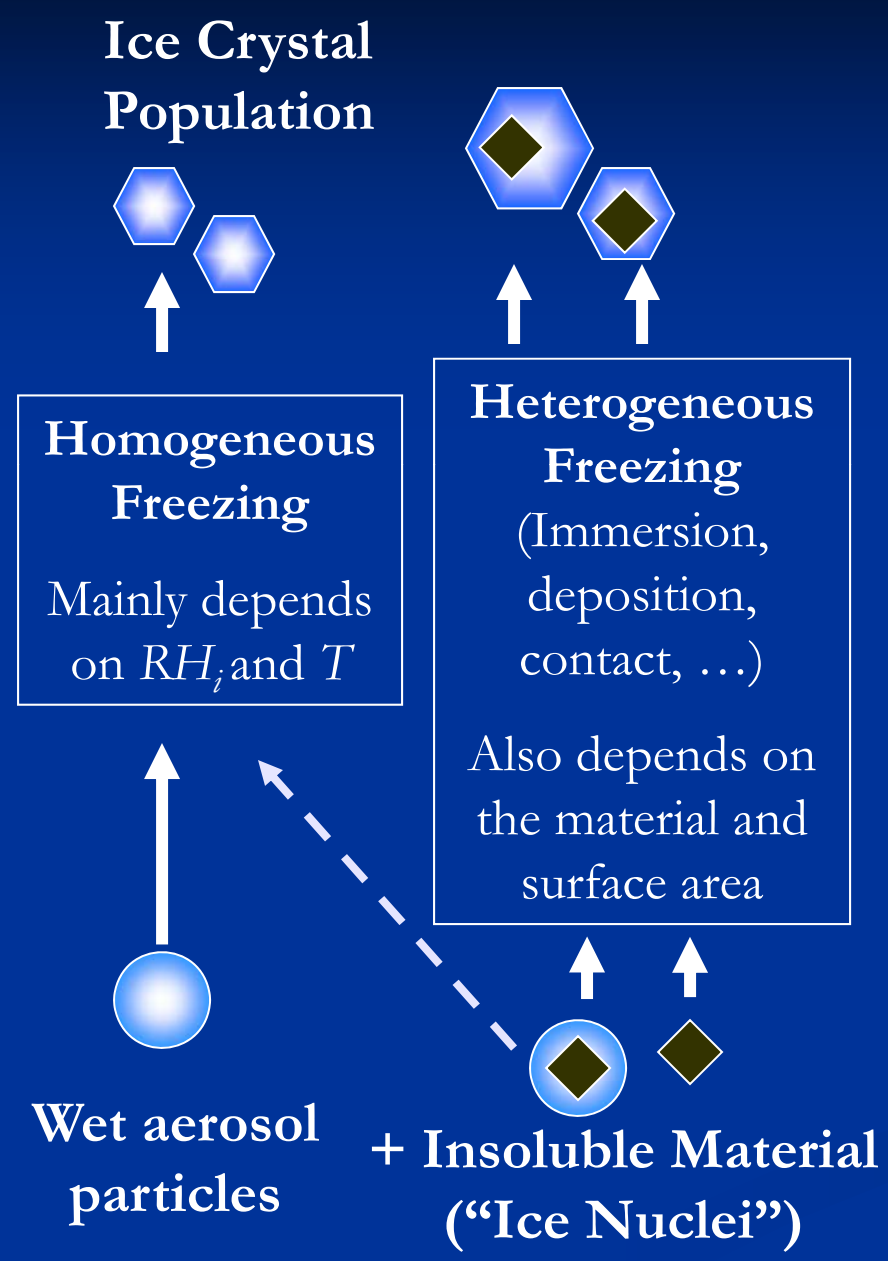
Part 1:

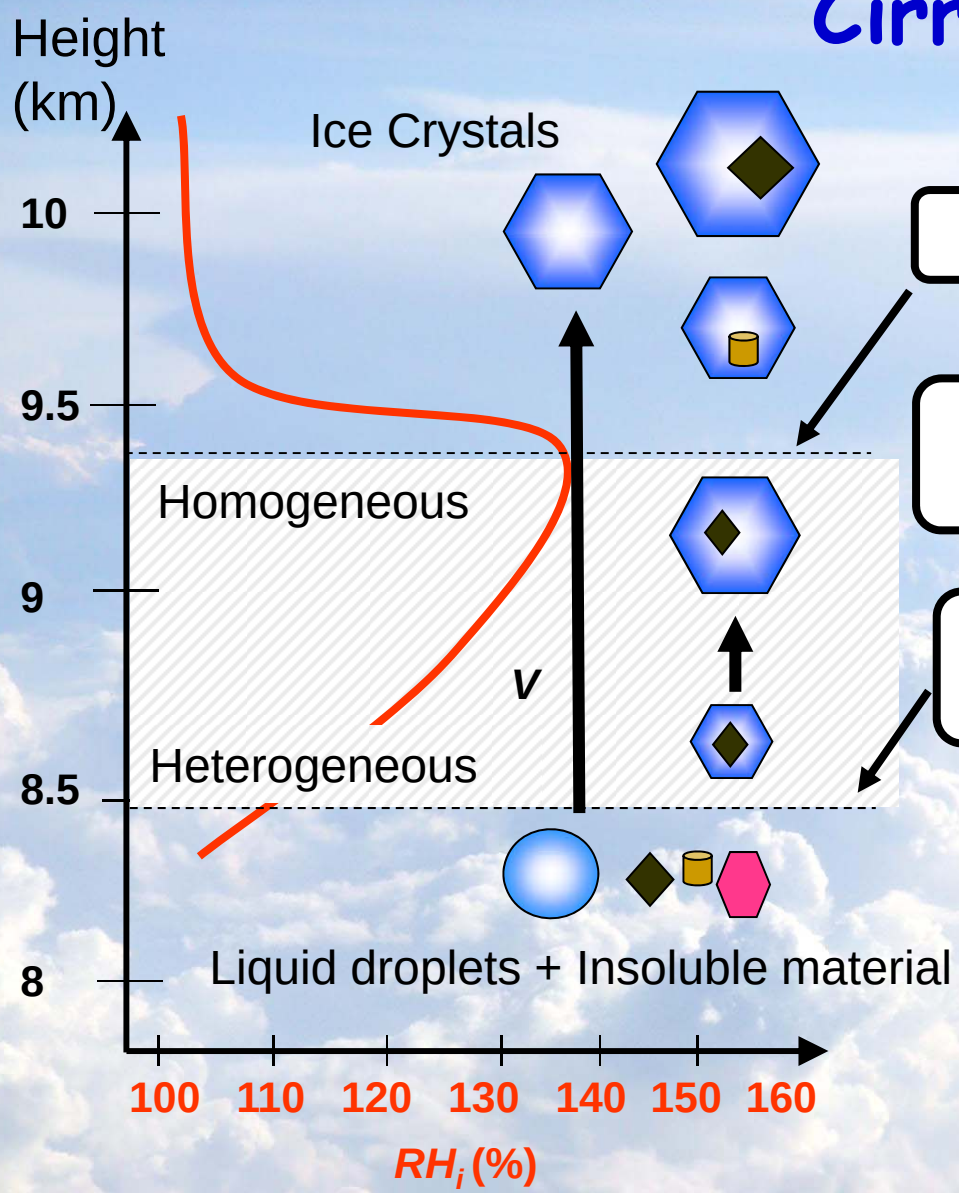
Sensitivity of the global distribution of cirrus ice crystal concentration to heterogeneous freezing

Cirrus (Ice) Clouds



Multiple mechanisms for ice formation can be active.

Conceptual Model of Ice Formation in Cirrus



Homogeneous freezing of droplets

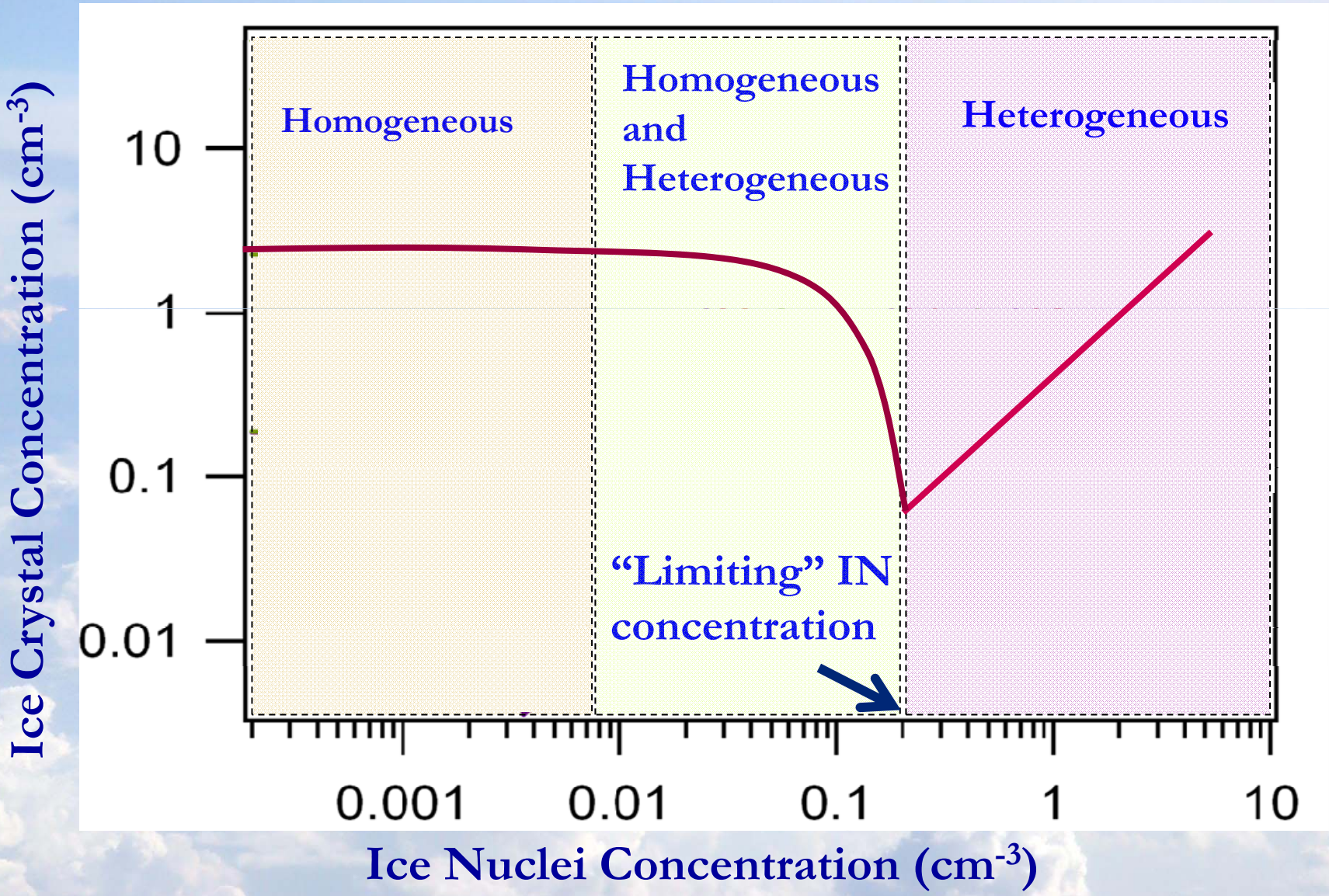
Crystal growth, fresh IN continue to freeze and deplete vapor

Heterogeneous IN freezing begin forming ice

Expansion cooling and ice supersaturation development

Soluble and insoluble aerosol initial distribution

The Effect of IN on Ice Crystal Number Concentration



Barahona and Nenes, *ACP*, 2009a.

Cirrus Formation in Global Climate Models: Current State of the Art

	Advantages	Disadvantages
Empirical	Very fast Give reasonable values	Limited Coverage Cannot be used to assess aerosol effects
Off-line solutions **(e.g., Liu and Penner, 2005)**	Fast Physically-based	Can't consider all the variability in formation conditions
Analytical-Numerical **(e.g., Kärcher, et al., 2006)**	Most of the physics included	Not fast enough

Analytical models based on cloud formation physics are needed!

Parameterizing ice formation: Solve the Mass and Energy Balances for cirrus

$$\frac{dS_i}{dt} = \frac{M_a p \; dw_i}{\ldots} + (1+S_i)\left[\frac{gM_a V}{\ldots} - \frac{\Delta H_s M_w \; dT}{\ldots}\right]$$

$$\frac{dT}{\ldots} = -\frac{gV}{\ldots} - \frac{\Delta H_s}{\ldots}\frac{dw_i}{\ldots}$$

$$\frac{dw_i}{dt} = \ldots$$

$$\frac{\partial n_c(D_c \ldots}{\partial t \ldots}$$

$$\frac{dD_c}{dt} = \frac{(S_i - S_{i,eq})}{\Gamma_1 D_c + \Gamma_2}$$

Condensation-type equations with dependencies on:

- **Temperature, Updraft Velocity**
- **Aerosol composition, size, and freezing properties**
- **Crystal growth rate**

- **Numerical solution is possible however challenging (we developed a new approach)**

- **We want to find an analytical solution!**

IC: Initial droplet size distribution, relative humidity, and temperature.

Ice Crystal Growth

Barahona and Nenes, *JGR*, 2008; *ACP*, 2009

Final expression: A Unified Description of Particle Formation and Growth

The solution of the PDE system reduces to:

Ice Crystal Concentration →

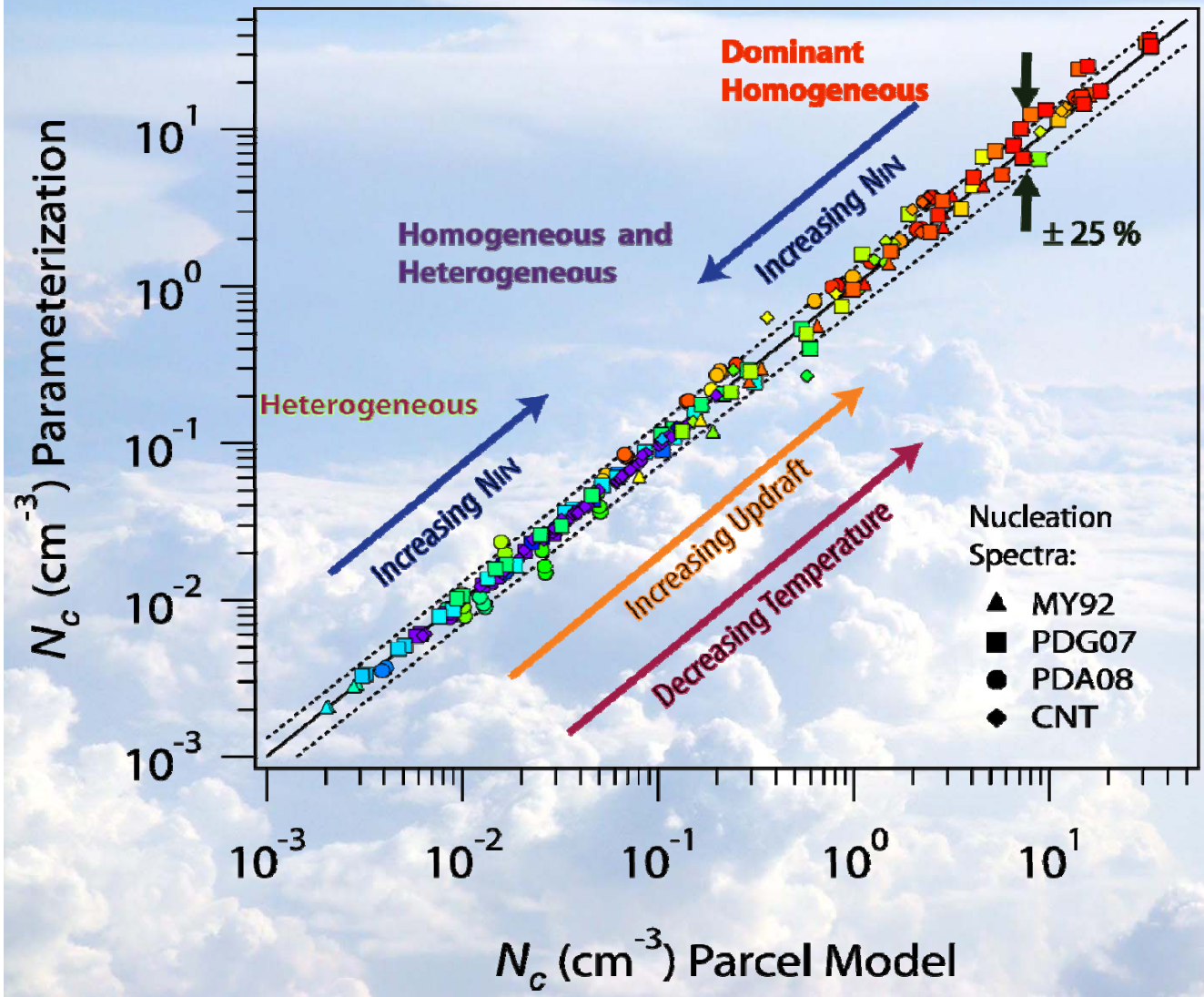
$$\frac{N_{het}(s_{\max})}{N^*} = \frac{1}{\sqrt{\Delta s^*_{char}}} \frac{(1+s_{\max})}{s_{\max}} e^{\frac{2}{\lambda s_{\max}}}$$

← **Condensation integral**

- Simple and physically based. Completely theoretical and analytical (i.e., robust). **Very fast!**
- Accounts for homogeneous and heterogeneous freezing
- Works with a general definition of heterogeneous freezing:
  - Can take into account the contribution from several freezing modes and aerosol species.
  - Allows direct incorporation of theoretical and empirical data into large scale models.

Barahona and Nenes, *ACP*, 2009b.

Parameterization evaluation: Compare Against Numerical Solution



Evaluation over a broad range of cloud formation conditions, aerosol concentrations, and freezing parameters.

Average error:
5 ± 12 %.

Orders of magnitude faster than the numerical solution

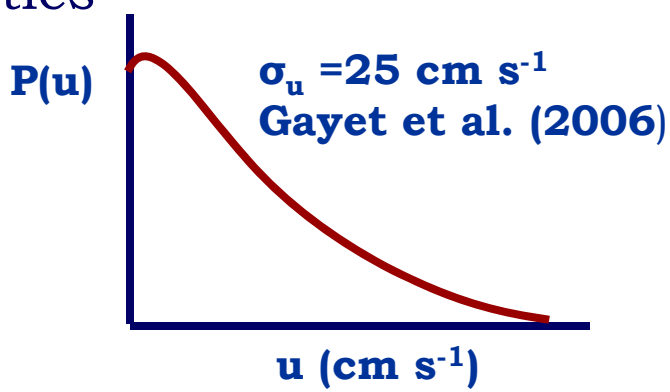
Barahona and Nenes, *ACP*, 2009b.

Including the ice formation parameterization in GMI

- NASA GMI Chemical and Transport Model. Aerosol model: Liu et al. (2005).

- Implementation:
  - Wind fields derived from GISS II' GCM
  - **Dust and black carbon** as IN precursors
  - Cirrus allowed for T<235 K. Time step 1h, resolution 4°×°5

- Dynamical forcing: Integrate over a Gaussian distribution of updraft velocities



Description of Aerosol Freezing

- Homogeneous (sulfate): Koop et al. (2000)
- Heterogeneous (dust and black carbon): Nucleation Spectrum:

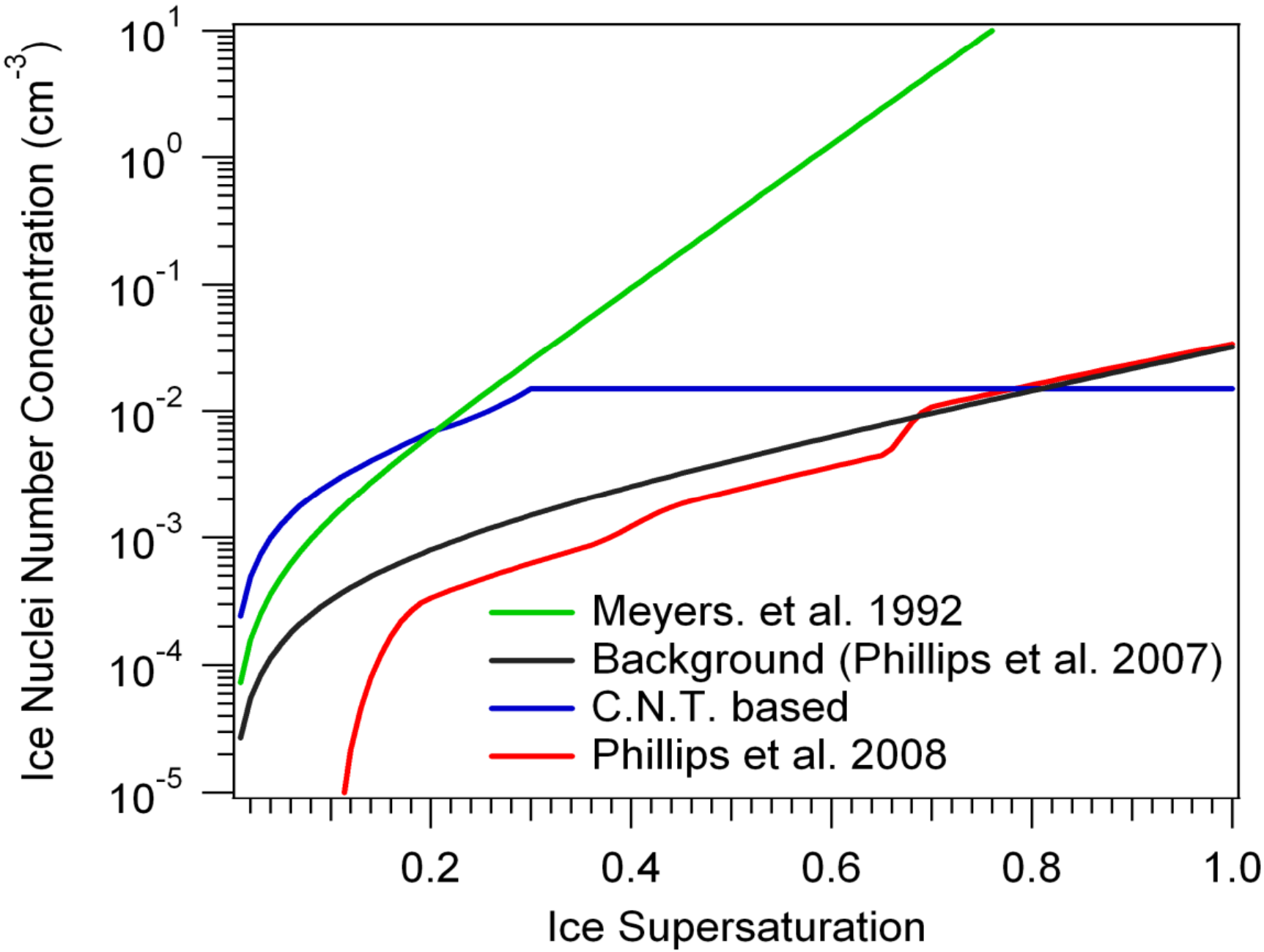
Aerosol size, chemical composition, surface properties

$$N_{IN}(Supersaturation)\big|_{T,N_{aerosol}}$$

Dust and Black carbon freezing fraction as a function of supersaturation

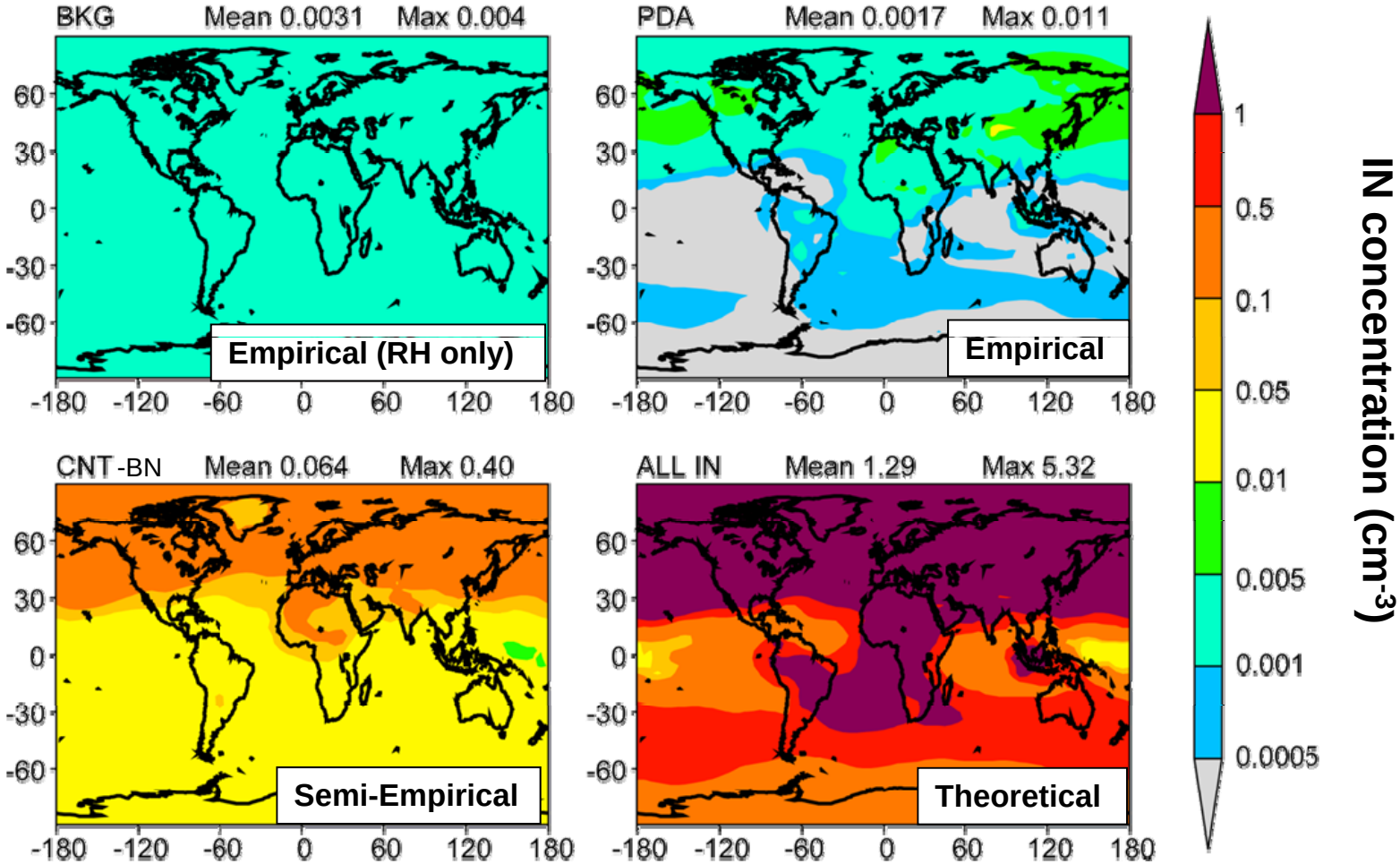
There is still uncertainty in the form of $N_{IN}(S_i)$; we tested several definitions currently used in cloud studies.

IN Spectra used in this Study



Heterogeneous IN Concentrations

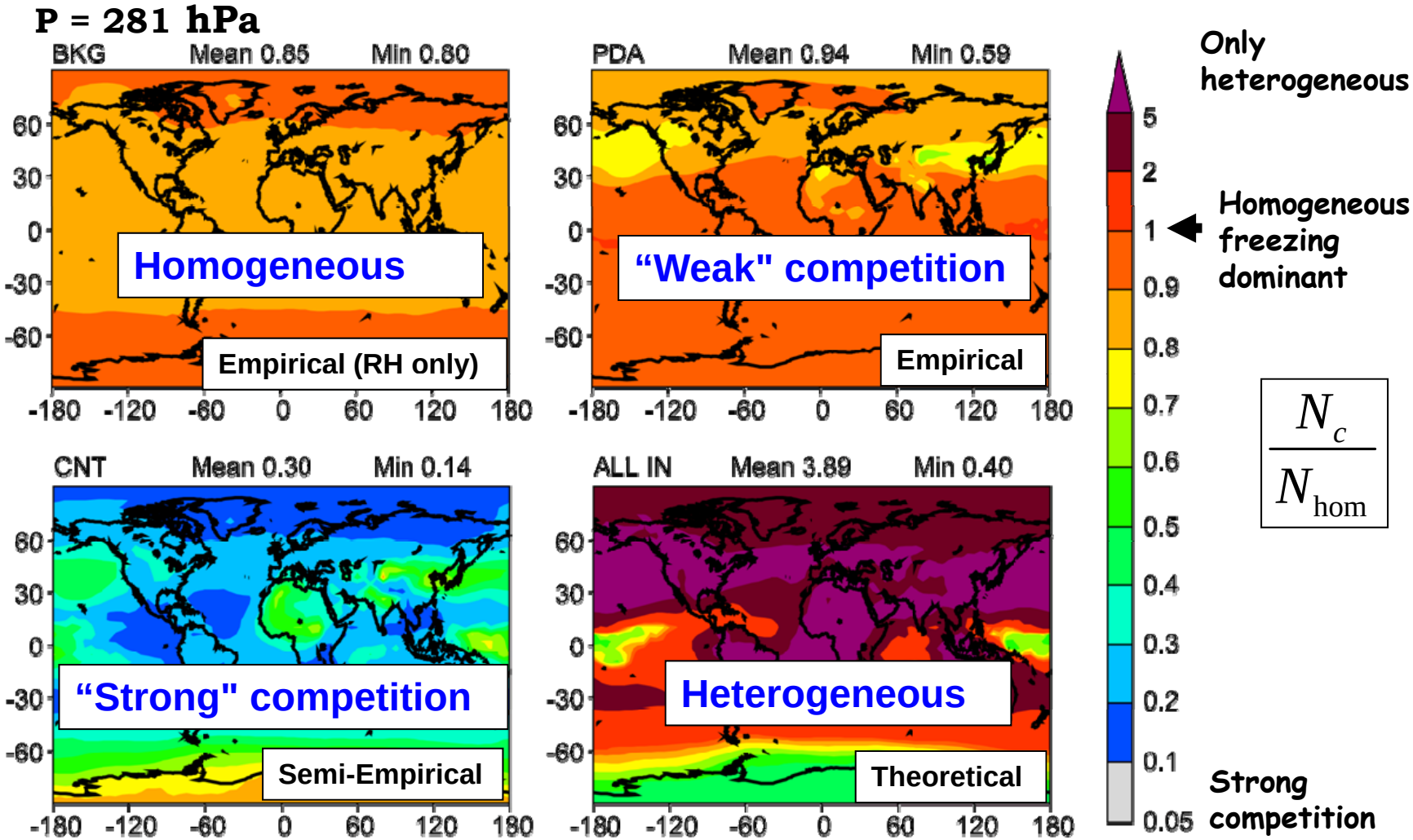
P = 281 hPa



About two orders of magnitude difference in IN concentration

Barahona, Rodriguez, and Nenes,JGR, *in press*.

Comparison against Homogeneous Freezing



At least a factor of 10 variation in global mean ice crystal concentration. Most significant in Northern Hemisphere

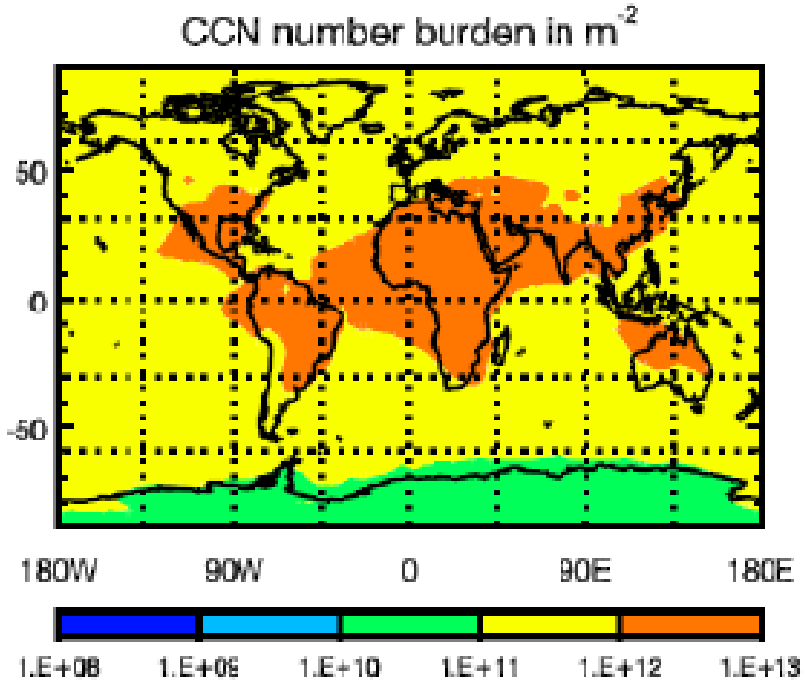
Barahona, Rodriguez, and Nenes,JGR, *in press*.

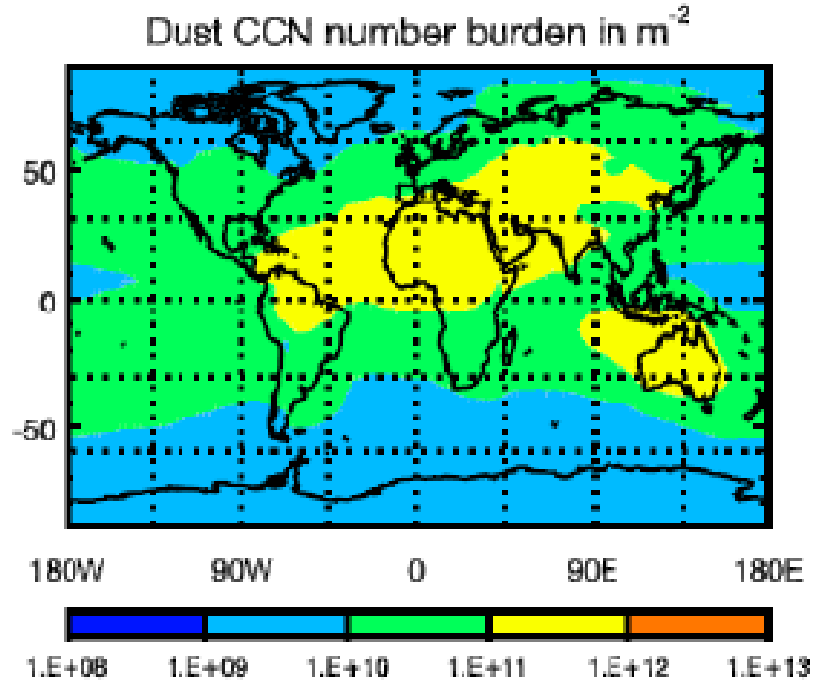
Cirrus cloud take home points

- A simple, analytical and fast parameterization of cirrus formation is presented which accurately reproduces numerical results.
- Considers the dependency of the ice crystal number on cloud dynamics, supercooled liquid aerosol size and number, deposition coefficient, and ice nuclei concentration.
- Accounts for the competition between homogeneous and heterogeneous nucleation and between different nucleation modes in pure heterogeneous freezing.
- Implemented in GMI and used to test the sensitivity of ice number to IN parameterization spectra available in literature.
- 2 orders of magnitude IN variability $\rightarrow$ 10fold change in crystal number. This sensitivity is preserved across met fields.
- BC does not really contribute to IN; predominant ice production mechanism however depends on the IN parameterization. Potentially important for predicted IWP.

Part 2:
Effect of insoluble (fresh) dust particles on global CCN and droplet number

Mineral Dust as CCN





Hoose and Lohmann (2008)

- ❑ Dust contributes to ~ 10% global CCN 'burden' (regionally larger).
- ❑ Dust acts as giant CCN, and its wet size controls its efficiency as a collector drop.
- ❑ Limited number of studies include dust-cloud interactions
- ❑ CCN activity of dust (espcially freshly-emitted) is not well understood.

Adsorption Activation:
A new theory of droplet formation

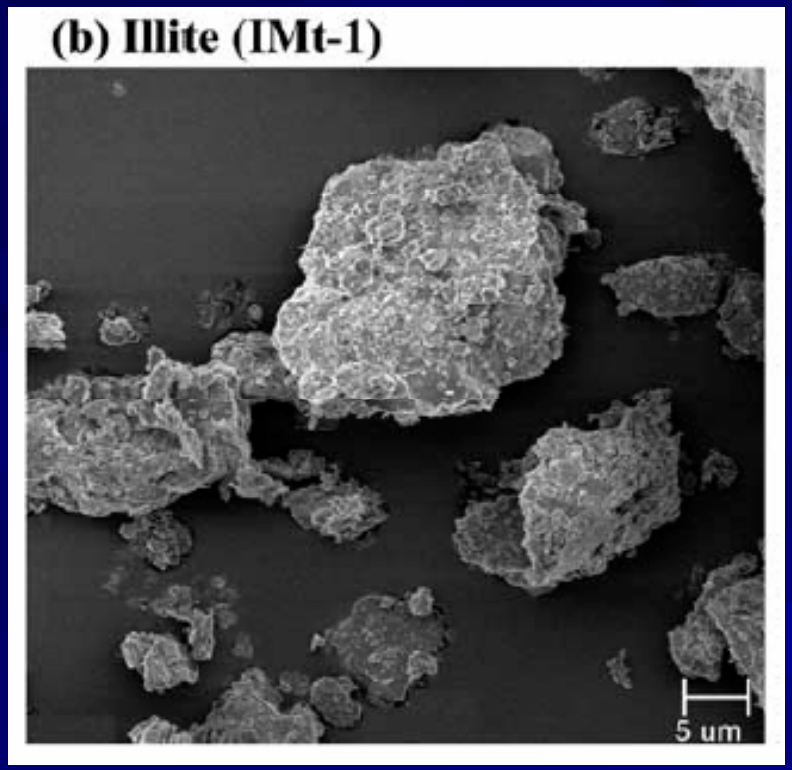
Current CCN theory makes very important assumptions about insoluble particles (dust).

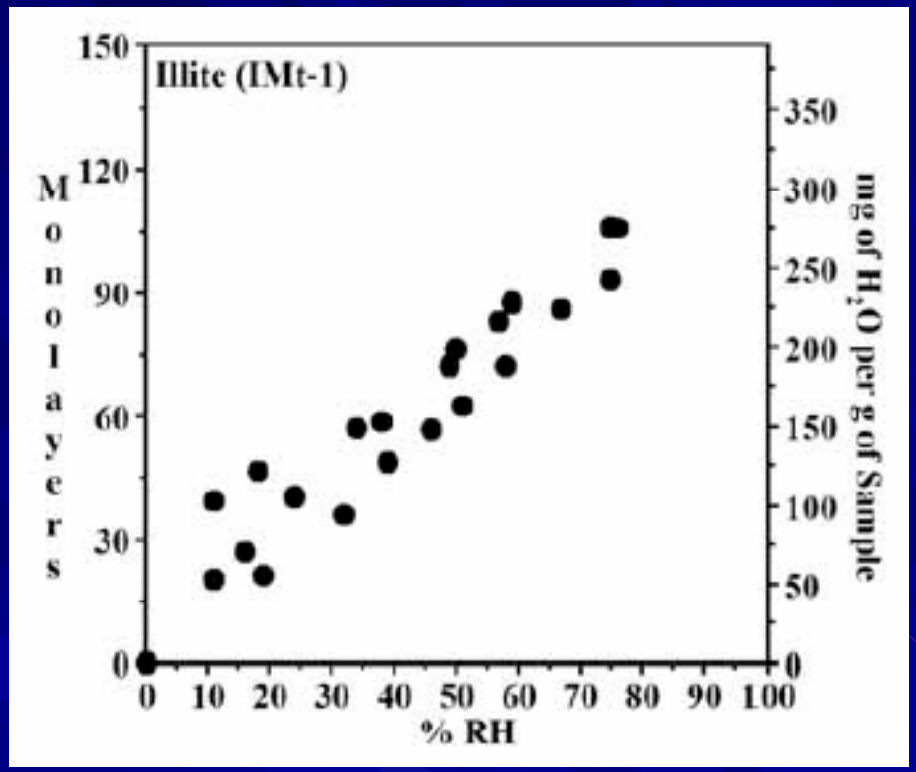
- Insoluble substances (like dust) are **not good** CCN.
- The CCN activity of solubles depends *solely* on soluble "coatings" from atmospheric processing.
- Theory works well when there is a fair amount of soluble solute in the particles (~ >10% by mass).

Does this mean fresh dust (no solute) are not CCN?

- Dust particles are wettable, and can swell. This means they do interact with water, but only on their surface.
- Look at the process of *adsorption* of water on dust, and its potential impact on droplet formation.

Example: adsorption/desorption curves of Illite





Adsorbed water increases with RH

Semi-reversible (hysterysis)

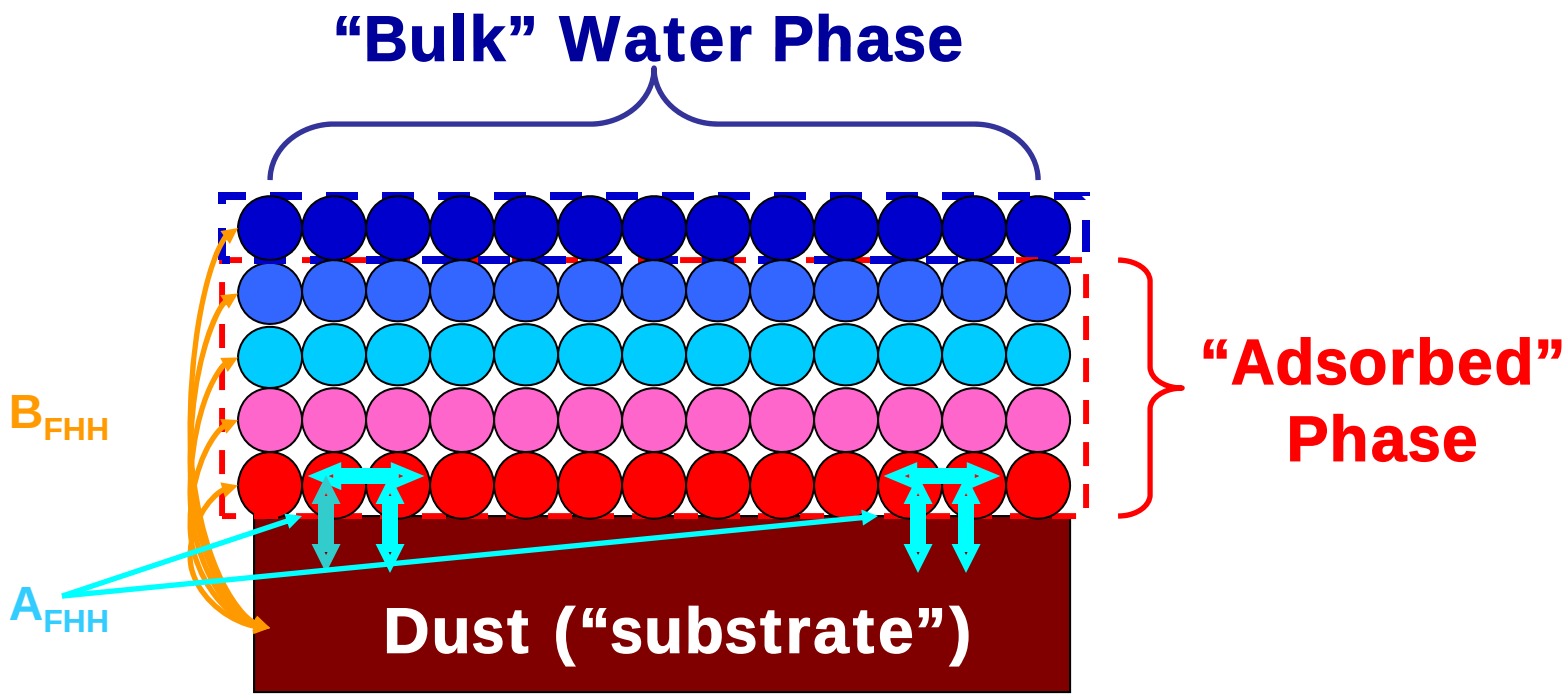
Substantial amounts of water can adsorb at equilibrium

Same for other clays.

Schuttlefield et al., JGR, (2007)

Describing water adsorption on Dust

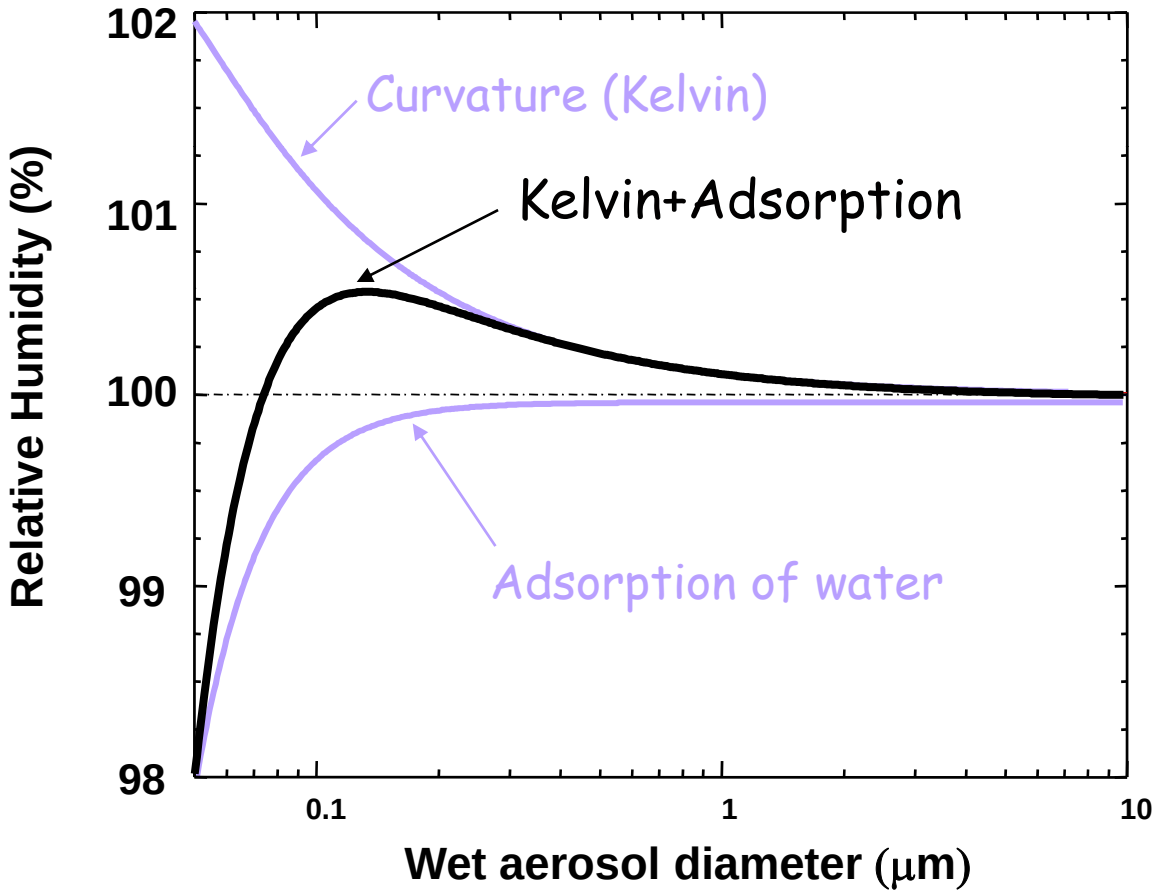
Use the **Frenkel-Halsey-Hill** (FHH) multilayer adsorption isotherm can be used to describe the interaction of water vapor with the dust surface.



- Long-range Van-der-Walls interactions between surface and upper layers are considered by **A_{FHH}** & **B_{FHH}**.
- **A_{FHH}** accounts for interactions between the first monolayer and substrate.
- **B_{FHH}** quantitatively accounts for the adsorption strength of each additional H_2O monolayer.

Can water adsorption turn particles into CCN ?

Plot the equilibrium vapor pressure of the dust particle as more and more water adsorbs on it…



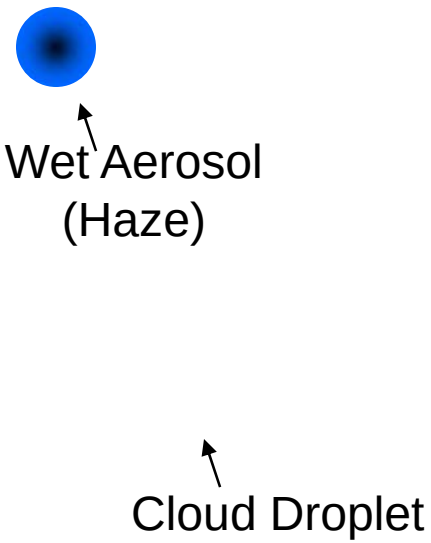
The combined curvature and effects determines the wet diameter at equilibrium.

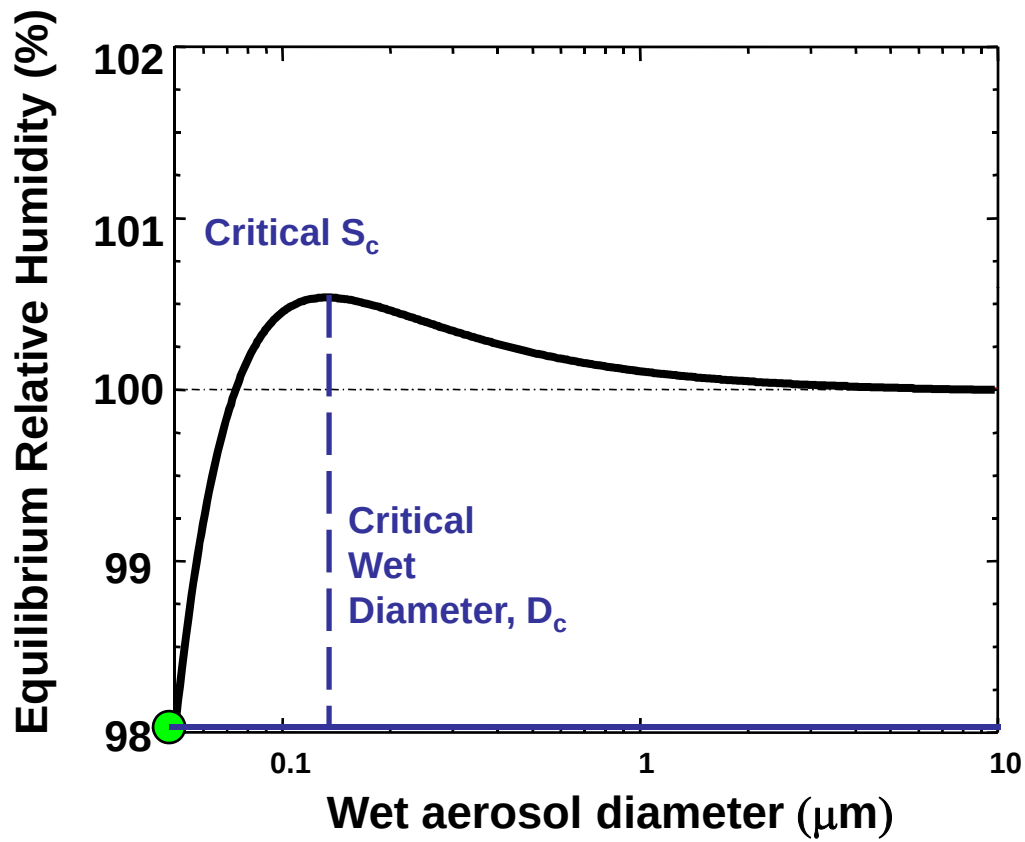
Looks like a "classical" CCN.

Here is why…

When does an aerosol particle act as a CCN ?

Dynamical behavior of a dust particle in a variable RH environment.



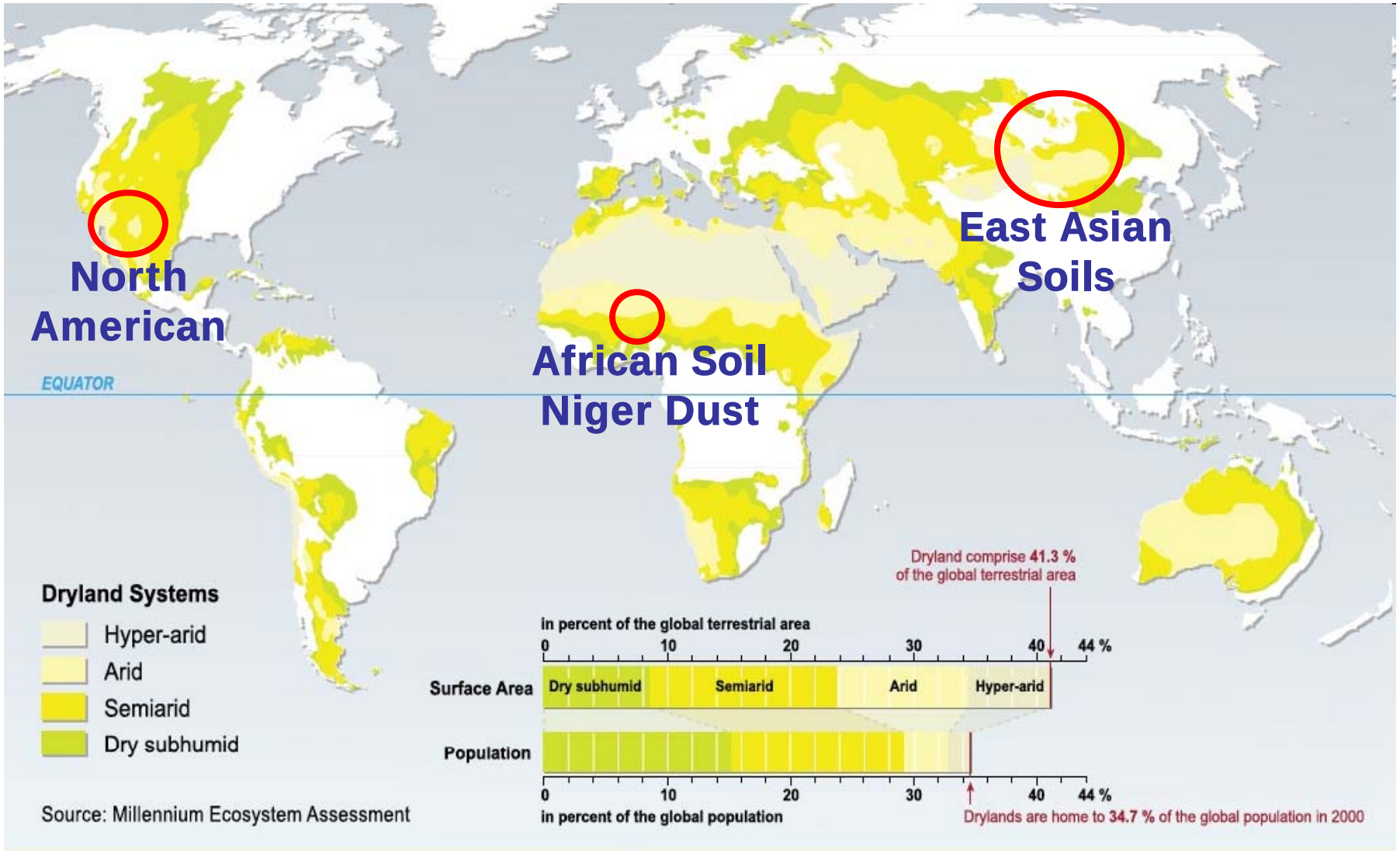


When ambient saturation ratio exceeds S_c, particles act as CCN.

CCN = f (Size, Adsorption parameters)

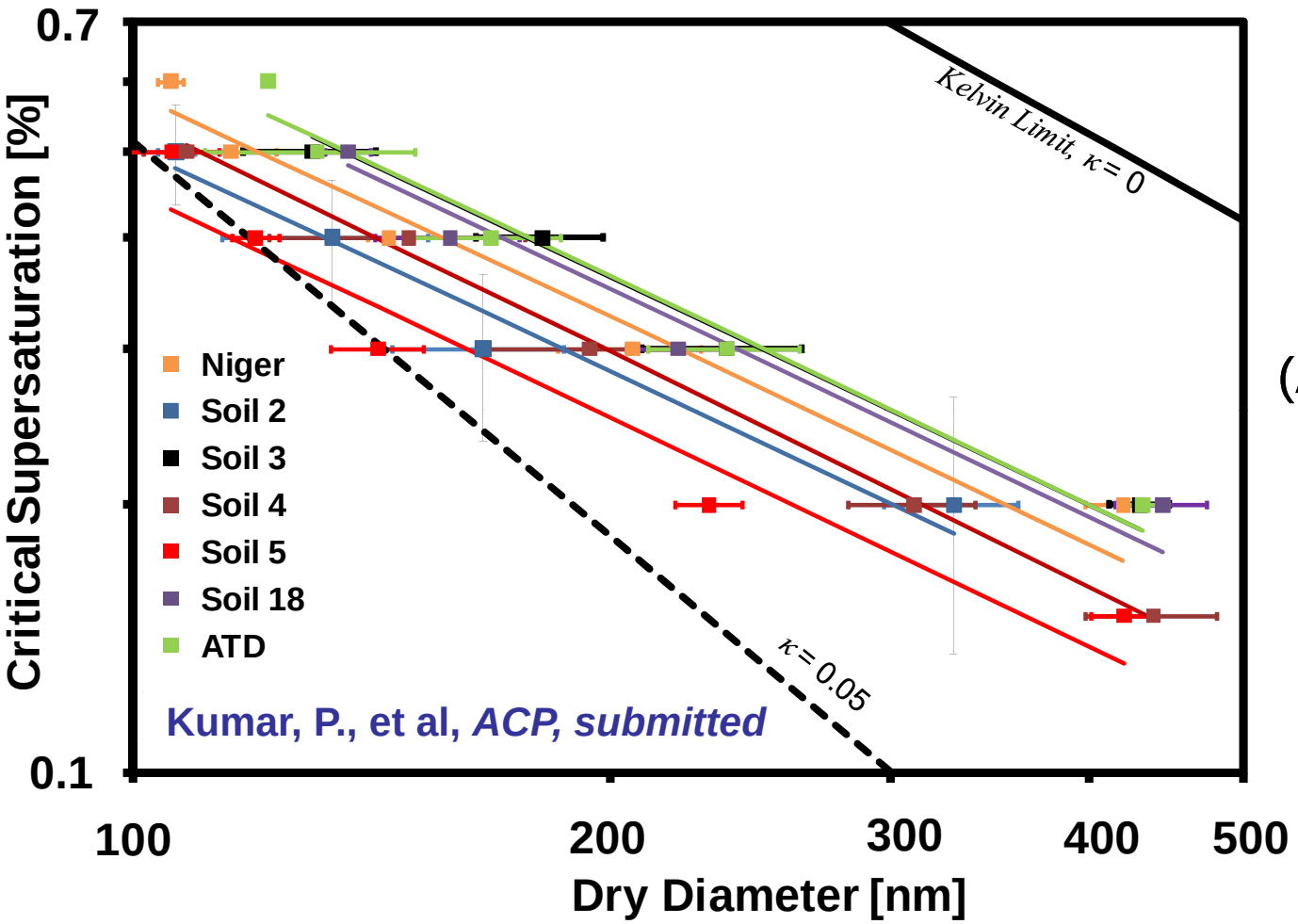
CCN activity of dry-generated dust (in the lab)

Collect soil samples from active dust source emission regions



Kumar, P., et al, *ACP, submitted*

CCN Activity of Regional Dusts



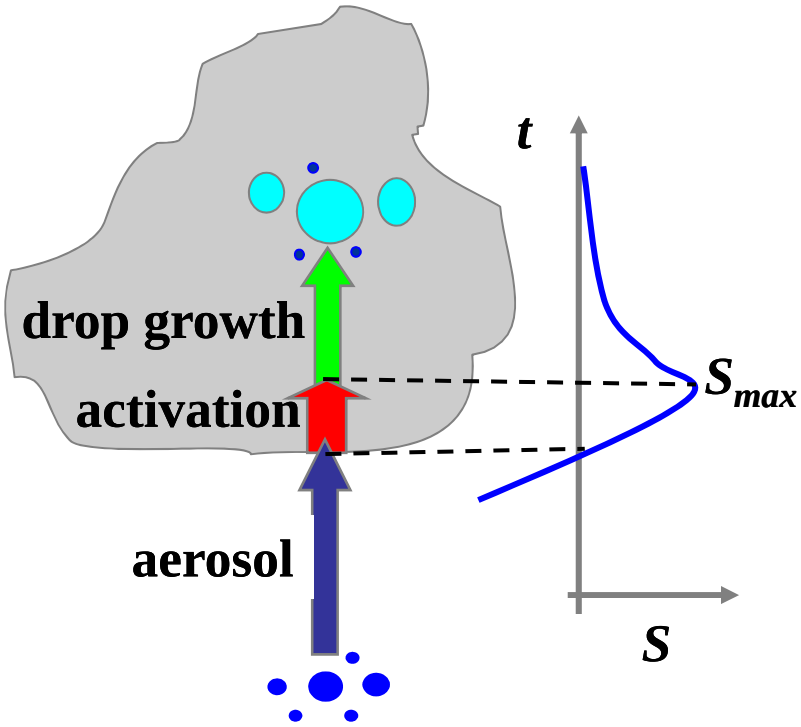
CCN activity is region dependent (Asian>Niger>ATD)

... but not terribly variable

- Fine mode dust is CCN active – equivalent to having up to 10% vol. fraction of ammonium sulfate (@ 100 nm). Smaller particles even higher.
- "Average" dust: $A_{FHH} \sim 2.50 \pm 0.50$ $B_{FHH} \sim 1.20 \pm 0.10$

Including Adsorption Activation in droplet formation parameterizations

Basic Idea: Solve conservation laws for energy and mass balances for water vapor condensing on aerosol particles in the ascending parcel



The final result is the number of activated droplets N_d that is calculated at the parcel maximum supersaturation, s_{max}.

This is a two step process

1. Obtain parcel s_{max}
2. Determine droplet number, N_d by counting particles with $s_c < s_{max}$

Kumar, P., Sokolik, I. N. and Nenes, A., *ACP*, 2009

Including Adsorption Activation in droplet formation parameterizations

Supersaturation Balance Equation: solved for s_{max}

Cooling from Expansion

Entrainment / mixing effect

Condensation growth from activated CCN

$$\frac{\alpha V + eV\left[\frac{\Delta H_v M_w}{RT^2}(T - T') - (1 - RH)\right]}{\gamma \frac{\pi}{2}\frac{\rho_w}{\rho_a}} - I_e(0, s_{max}) = 0$$

$I_e(0,s_{max})$ is the '**rate**' of condensation of water vapor on the activated CCN and contains contributions from soluble and insoluble particles

$$I_e(0, s_{max}) = I_{K,1}(0, s_{part}) + I_{K,2}(s_{part}, s_{max}) + I_{FHH}(0, s_{max})$$

Condensation integral

Soluble (Köhler) Contribution (Nenes and Seinfeld, 2003)

Insoluble (FHH) Contribution

Kumar, P., Sokolik, I. N. and Nenes, A., *ACP*, 2009

Including droplet formation from adsorption activation in GMI

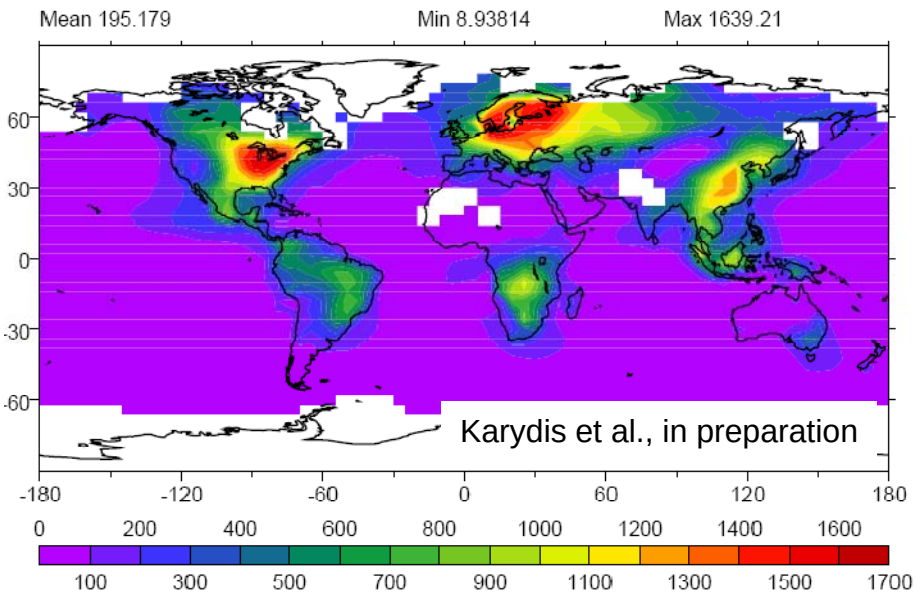
Emissions

- University of Michigan current day (Liu et al., 2007)

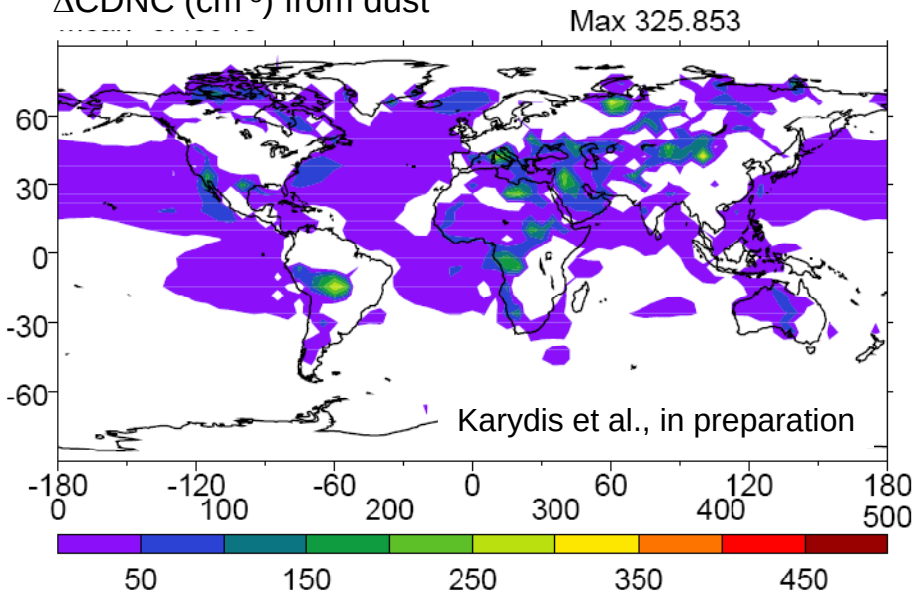
Aerosol-CDNC link

- Sulfate, OC, Seasalt (soluble), Dust (insoluble) using experimentally-defined activation properties

CDNC (cm^{-3}) sulfate, seasalt, primary organics



ΔCDNC (cm^{-3}) from dust



Globally: Dust not as important

Regionally: can dominate CDNC.

Dust CCN take home points

- Application of KT and FHH-AT leads to very different predictions in CCN and cloud droplet number for the same aerosol size distribution. FHH-AT seems to be a better description of CCN activity

- CCN measurements for dust generated from soils give on average:

$$A_{FHH} \sim 2.50 \pm 0.50 \quad B_{FHH} \sim 1.20 \pm 0.10$$

- Fresh dust ~ 100nm acts like a particle with κ~0.025-0.05. Strong size-dependence of the "k" though. This is not from solute, but from the surface-water interactions alone.

- The new theory can directly be introduced in droplet formation parameterizations, especially since the parameters are to first order constant.

- Initial simulations with the GMI suggest that dust near their sources can have an important contribution to droplet number.

THANK YOU !!
Photo: A.Sorooshian