Comprehensive, rapid cloud drop & ice crystal formation parameterizations: Developments and evaluations

Athanasios Nenes
School of Earth & Atmospheric Sciences
School of Chemical & Biomolecular Engineering
Georgia Institute of Technology

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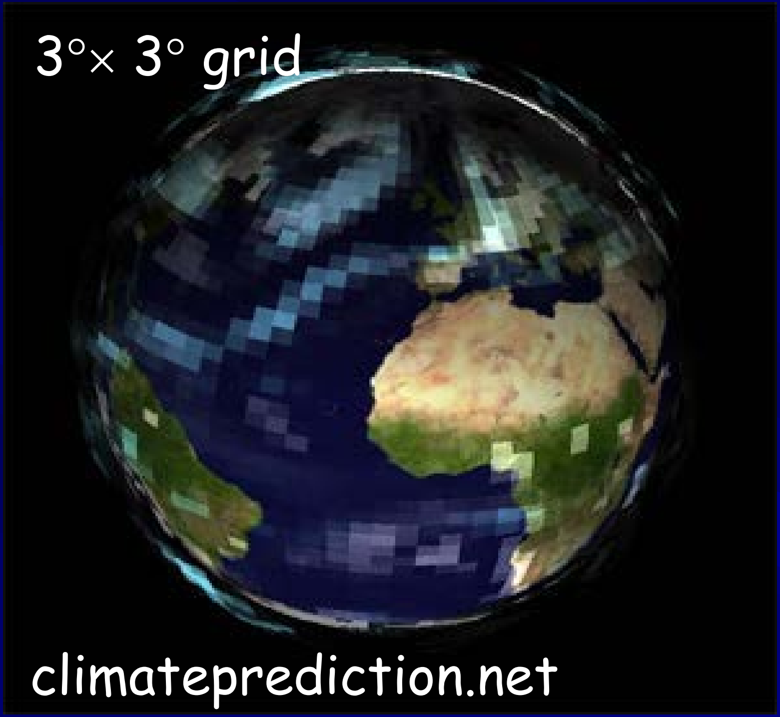
Problems with GCM assessments of aerosol indirect effect



climateprediction.net

- Cloud formation happens at smaller spatial scales than global climate models can resolve.
- Aerosol-cloud interactions are complex.
- Climate models provide limited information about clouds and aerosols.
- Describing cloud formation explicitly in global models is VERY expensive. These calculations need to be simplified (“parameterized”).

GCMs Need Fast Physics: Simple expressions capturing important cloud physics

Goal: Predict drop/ice number concentration in "characteristic" cloud types.

Dynamics

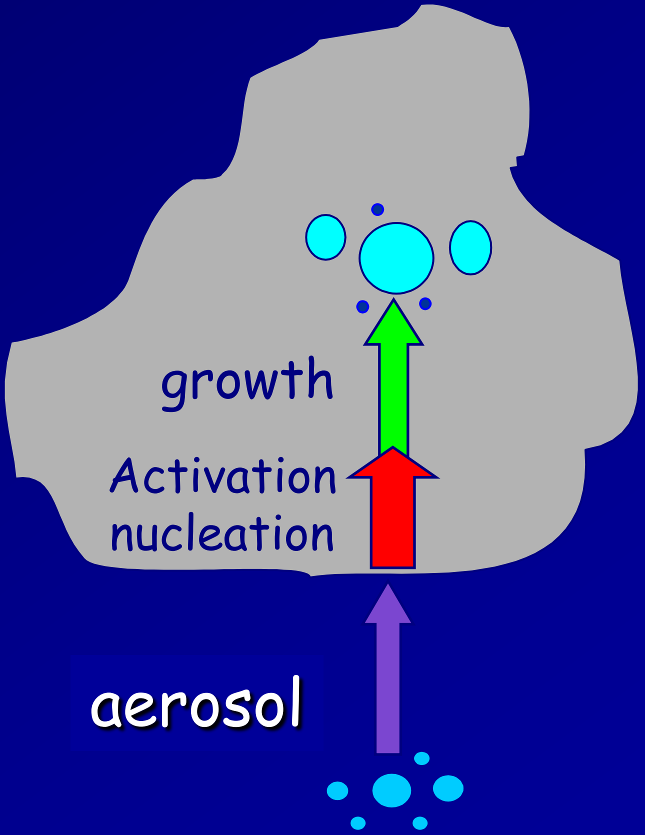
- Updraft Velocity
- Large Scale Thermodynamics

Particle characteristics

- Size & Concentration
- Chemical Composition

Cloud Processes

- Cloud droplet formation
- Ice crystal formation
- Effects of entrainment/mixing
- Collision/coalescence
- "Scaleup" of processes

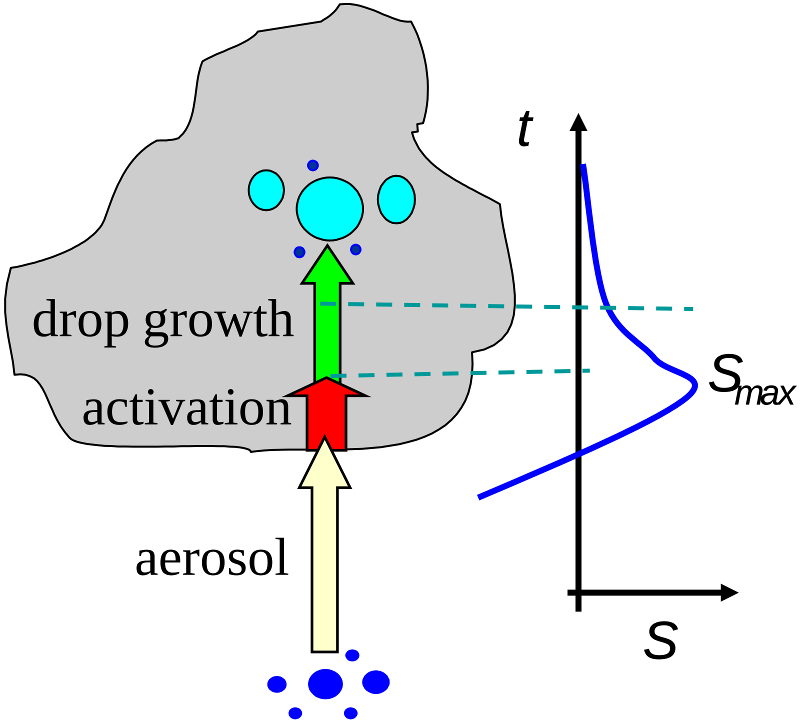


Links/feedbacks need to be incorporated (at appropriate scales). VERY challenging problem (Stevens and Feingold, 2009)

Liquid Phase Clouds

Approach: use the "simple story" (1D parcel theory)

Basic ideas: Solve conservation laws for energy and the water vapor condensing on aerosol particles in cloudy updrafts.



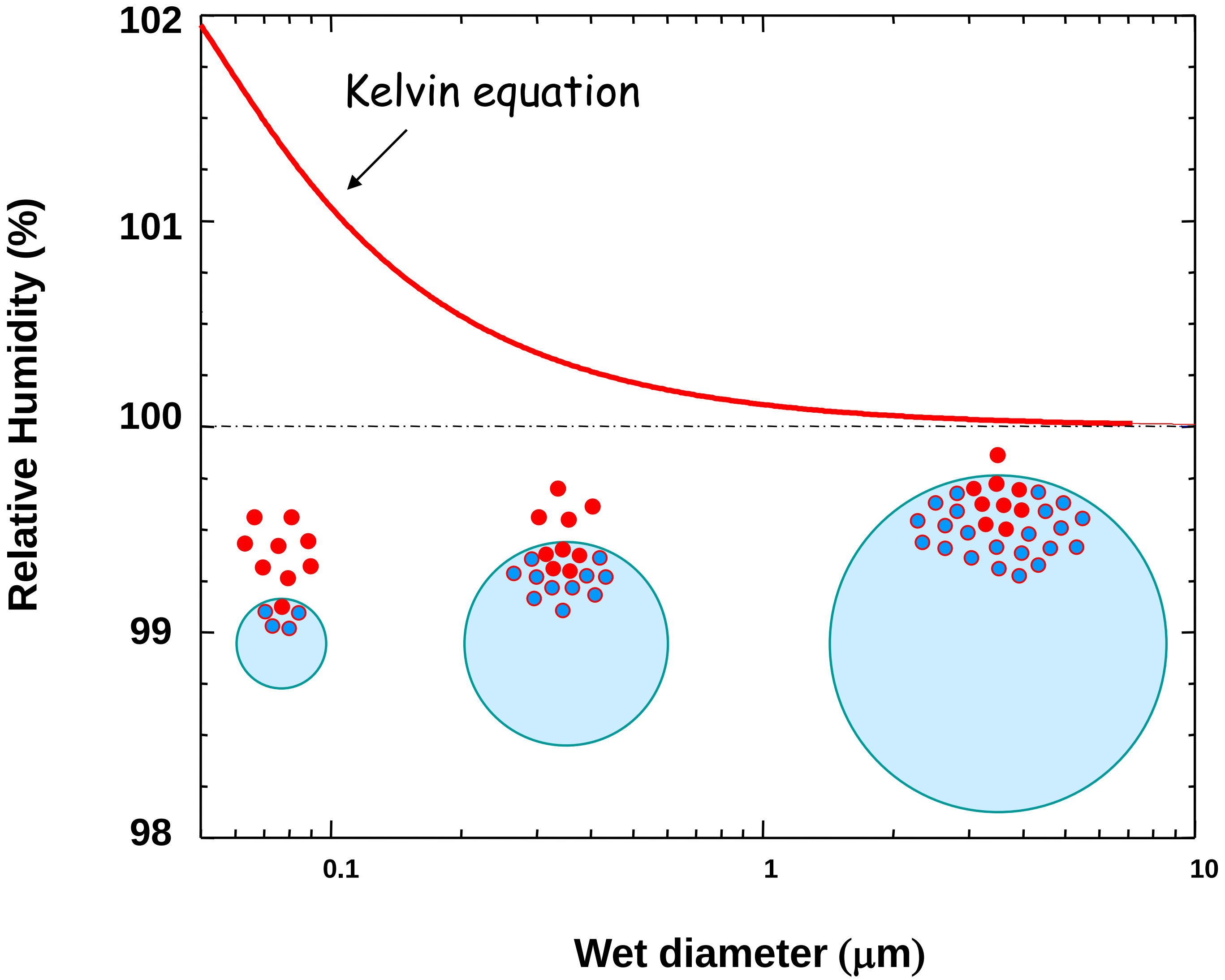
Conceptual steps are:

- Air parcel cools, exceeds dew point
- Water vapor is supersaturated
- Droplets start forming on existing CCN.
- Condensation of water on droplets becomes intense.
- *S* reaches a maximum
- No more additional drops form

A "classical" nucleation/growth problem

So… when does an aerosol particle act as a CCN ?

Start from a pure H_2O drop

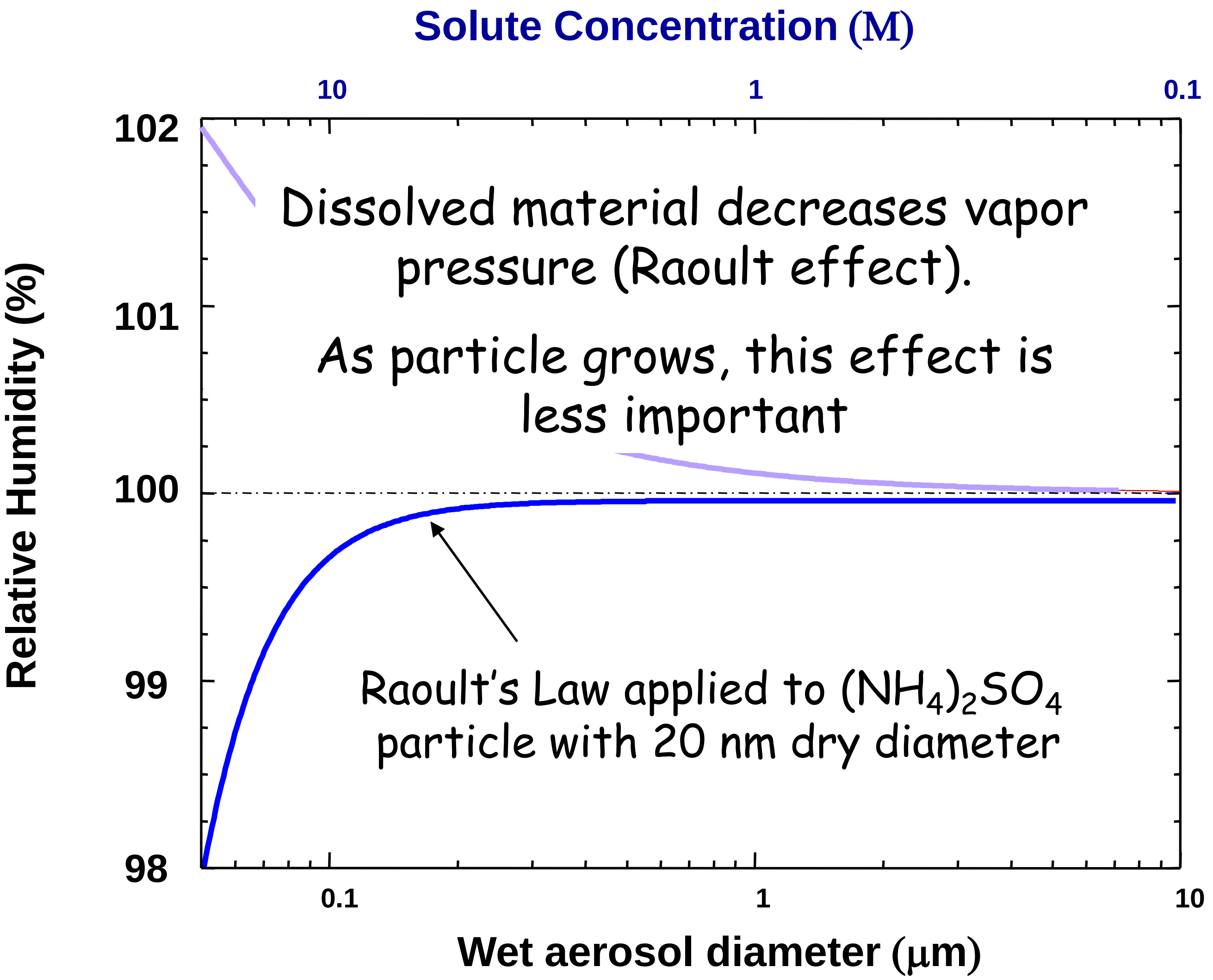


As the droplet size decreases, its equilibrium vapor pressure increases (Kelvin effect).

Less molecules around in small drops to "pull" H_2O in the droplet phase

When does an aerosol particle act as a CCN ?

Take same drop and add some solute; e.g., $(NH_4)_2SO_4$



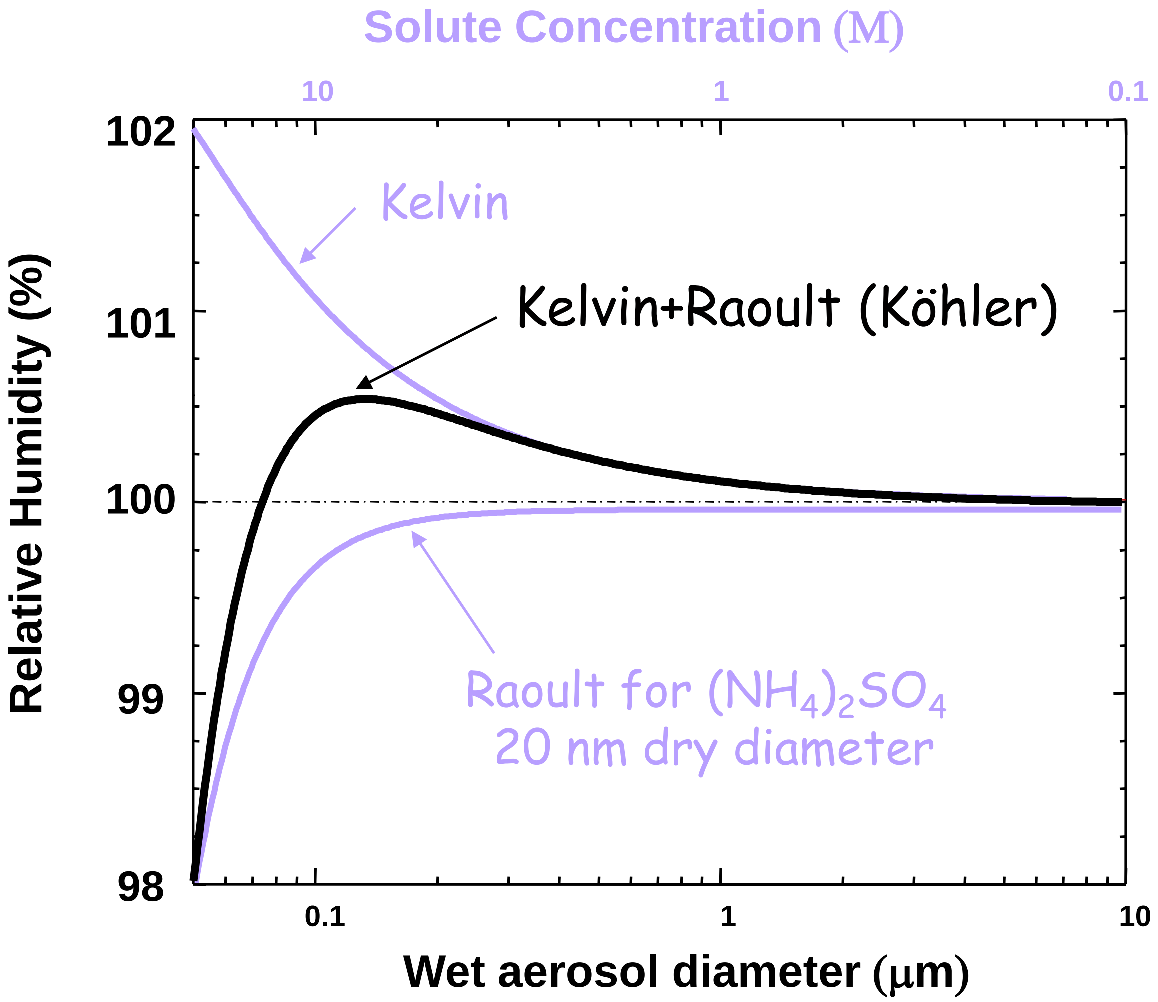
Dissolved material decreases vapor pressure (Raoult effect).

As particle grows, this effect is less important

Raoult's Law applied to $(NH_4)_2SO_4$ particle with 20 nm dry diameter

When does an aerosol particle act as a CCN ?

Put both effects together: You get the equilibrium vapor pressure of a wet aerosol particle.

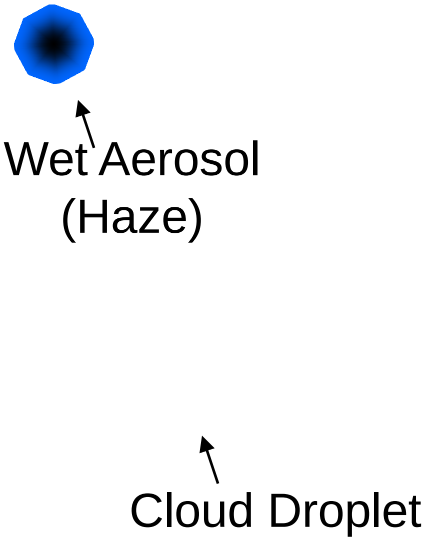


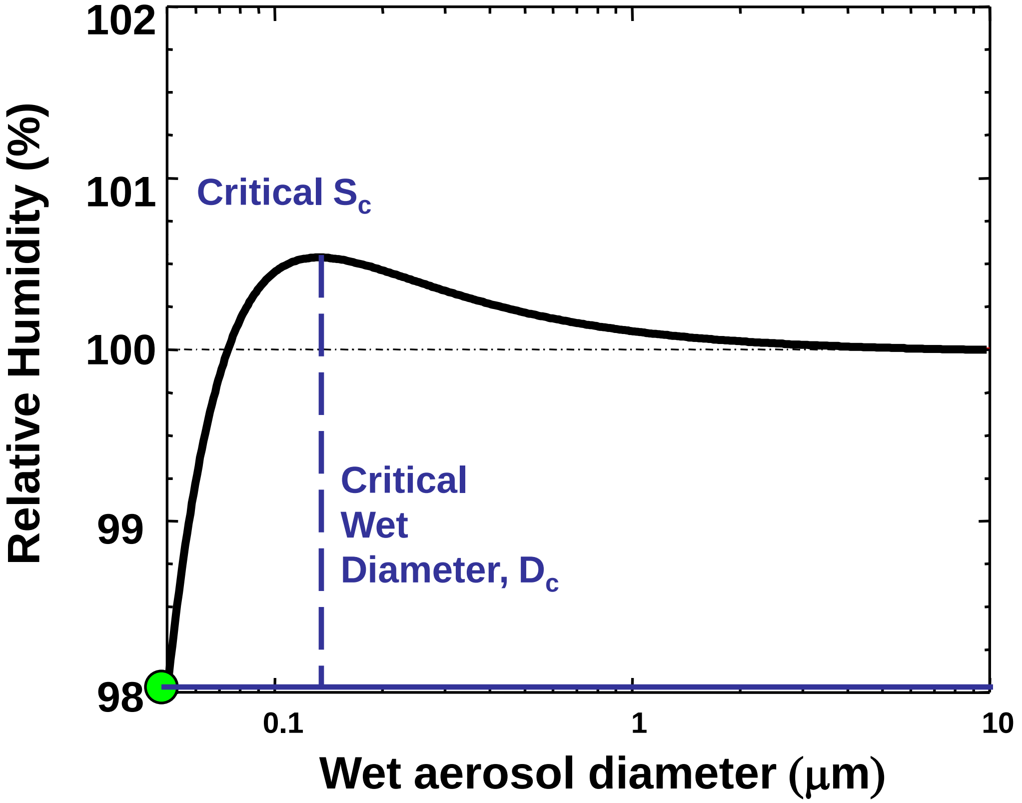
The combined Kelvin and Raoult effects is known as the **Köhler equation (1922)**.

You can be in equilibrium even if you are above saturation.

When does an aerosol particle act as a CCN ?

Dynamical behavior of an aerosol particle in a variable RH environment.

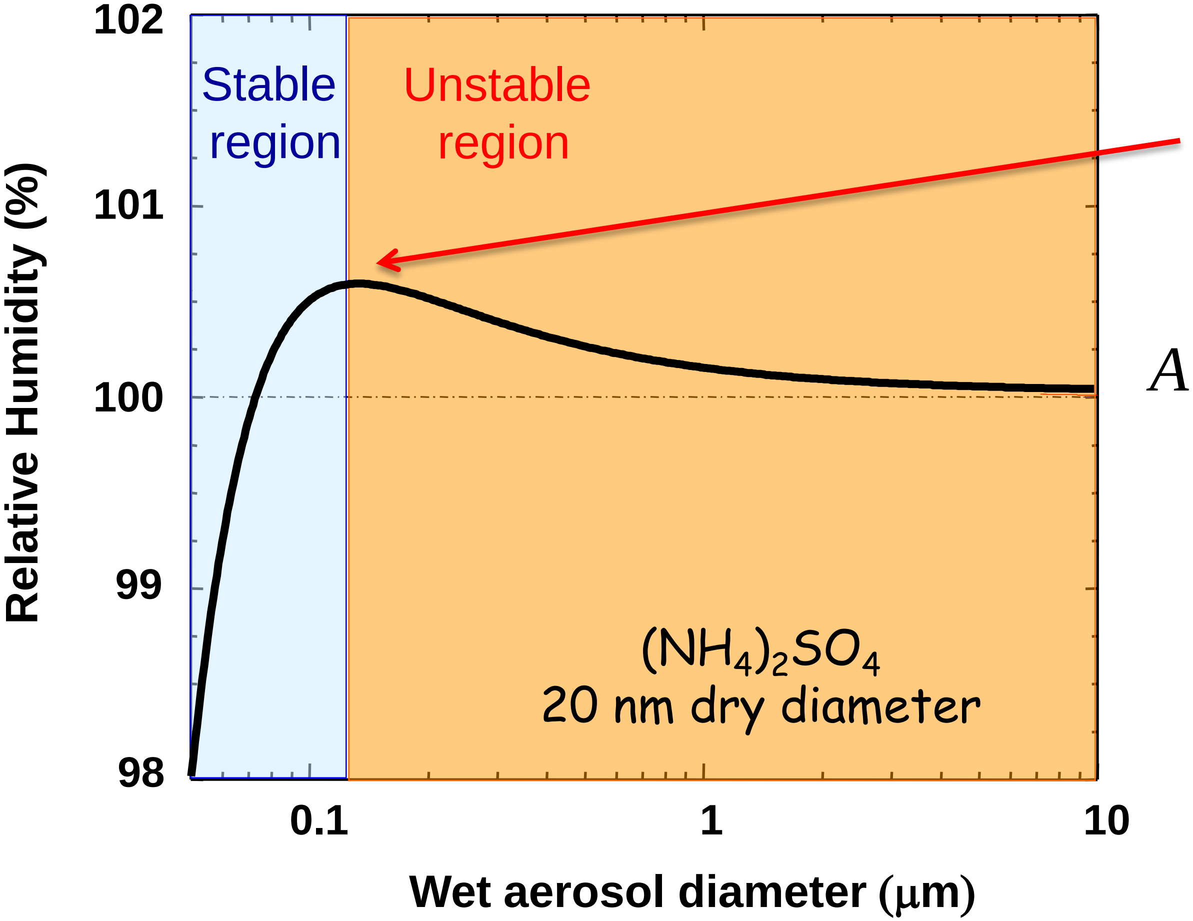




When ambient saturation ratio exceeds S_c, particles act as CCN.

When does an aerosol particle act as a CCN ?

When the ambient saturation ratio $S > S_c$ AND the wet size is larger than D_c. ($S > S_c$ sufficient; Nenes et al., 2001).



$$s_c = \left(\frac{4A^3}{27B}\right)^{1/2}$$

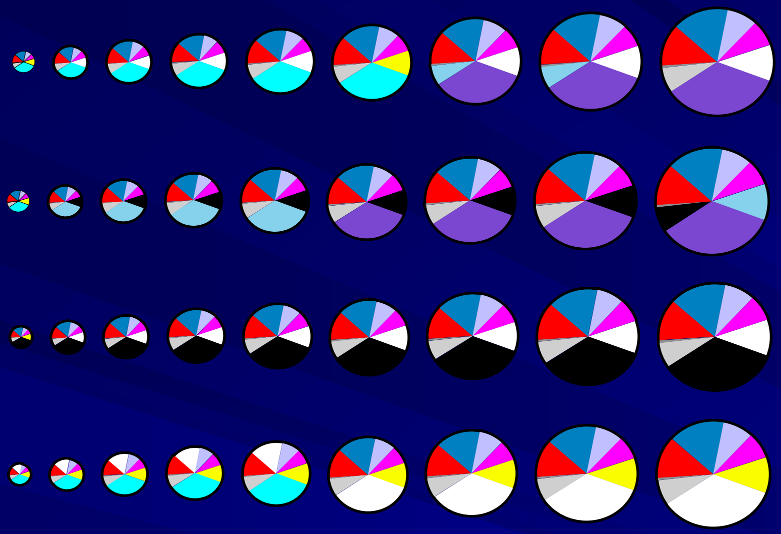
A:surf.ten. parameter

B:solute parameter

$$s_c \sim d_{dry}^{-3/2}$$

$$s_c \sim \varepsilon_{\mathbf{soluble}}^{-1/2}$$

Aerosol Problem: Complexity



An integrated "soup" of

- Inorganics, organics (1000's)
- Particles can have uniform composition with size…
- … or not
- Can vary vastly with space and time (esp. near sources)

Organic species are a headache

- They can facilitate cloud formation by acting as surfactants and adding solute (hygroscopicity)
- Oily films can form and delay cloud growth kinetics

In-situ data to study the aerosol-CCN link:

Usage of CCN activity measurements to "constrain" the above "chemical effects" on cloud droplet formation.

Understanding & parameterizing CCN activity…

Petters and Kreidenweis (2007) expressed the solute parameter in terms of a “hygroscopicity parameter”, κ

$$s_c = \left(\frac{4A^3}{27B} \right)^{1/2} \Longrightarrow s_c = \left(\frac{4A^3}{27\kappa} \right)^{1/2} d^{-3/2}$$

$\kappa \sim 1$ for NaCl, ~ 0.6 for $(NH_4)_2SO_4$, ~ 0-0.3 for organics

κ rarely exceeds 1 in atmospheric aerosol

Simple way to think of κ: the “equivalent” volume fraction of NaCl in the aerosol (the rest being insoluble).

κ ~ 0.6 ⇨ particle behaves like 60% NaCl, 40% insoluble

Quantifying hygroscopicity:
size-resolved CCN measurements

Polydisperse
Aerosol

Scanning Mobility Particle Sizer

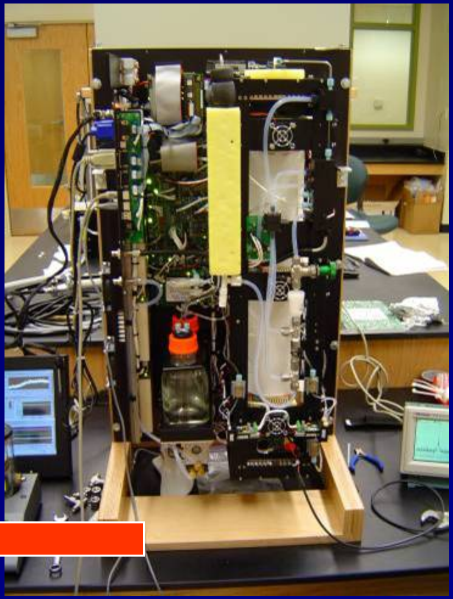
Size Selection

Monodisperse
Aerosol

Condensation Particle Counter, 3010

Count CN

Particle Detection

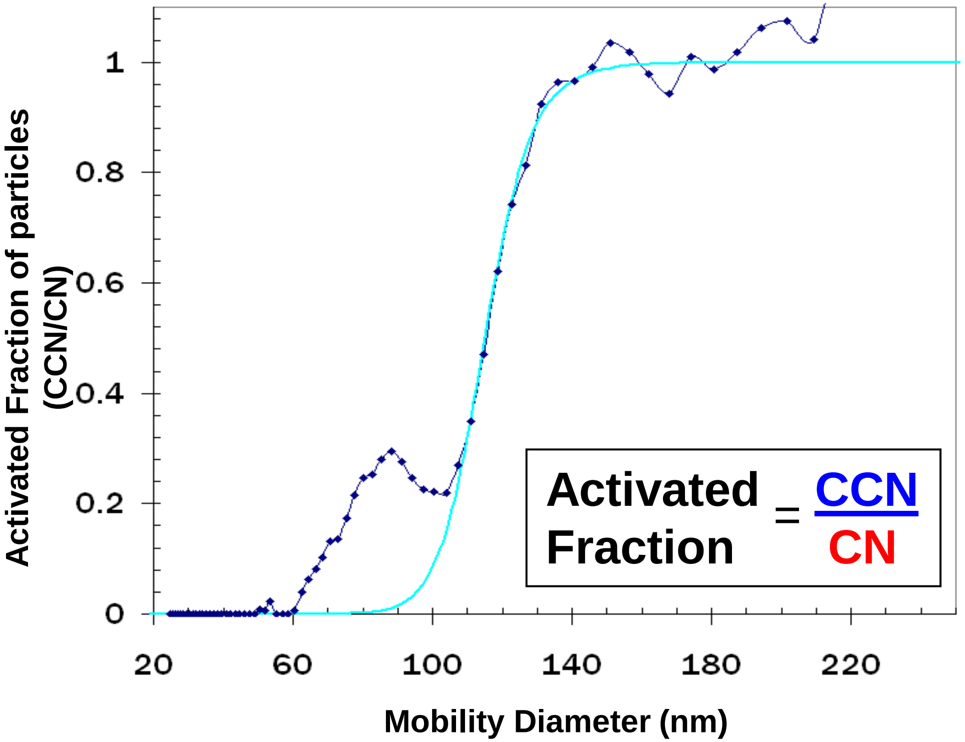


**Continuous Flow Streamwise
Thermal Gradient Counter**

Count
CCN

Quantifying hygroscopicity:
size-resolved CCN measurements

Results: "activation curves" CCN/CN as a function of *d*



rticle Counter, 3010

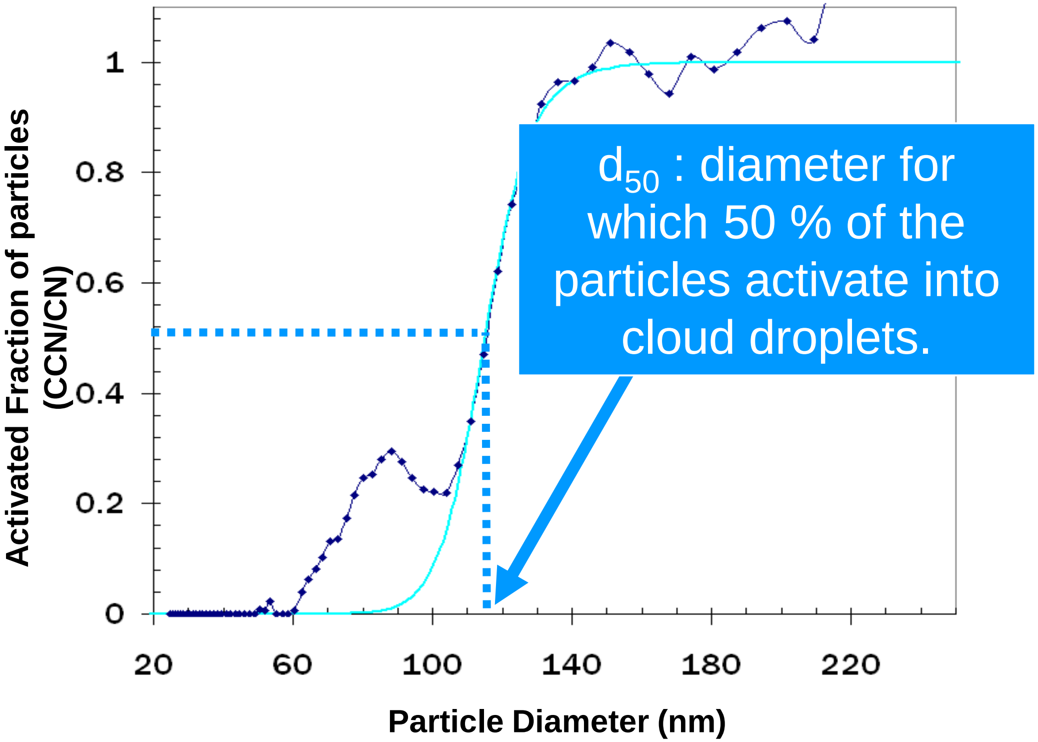
Count CCN

Continuous Flow Streamwise Thermal Gradient Counter

Count CN

Determining κ: size-resolved CCN measurements

$d_{50}\downarrow$: aerosol becomes more hygroscopic



d_{50} : diameter for which 50 % of the particles activate into cloud droplets.

Count CCN

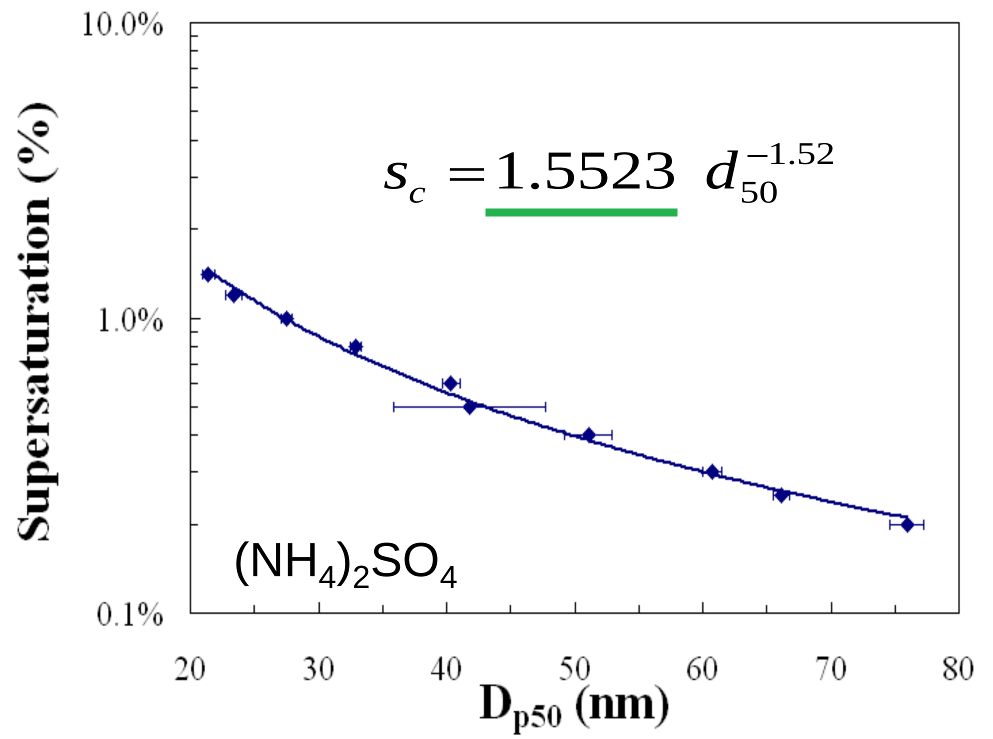
Continuous Flow Streamwise Thermal Gradient Counter

rticle Counter, 3010

Count CN

Parameterizing the CCN activity data using methods based on Köhler-theory

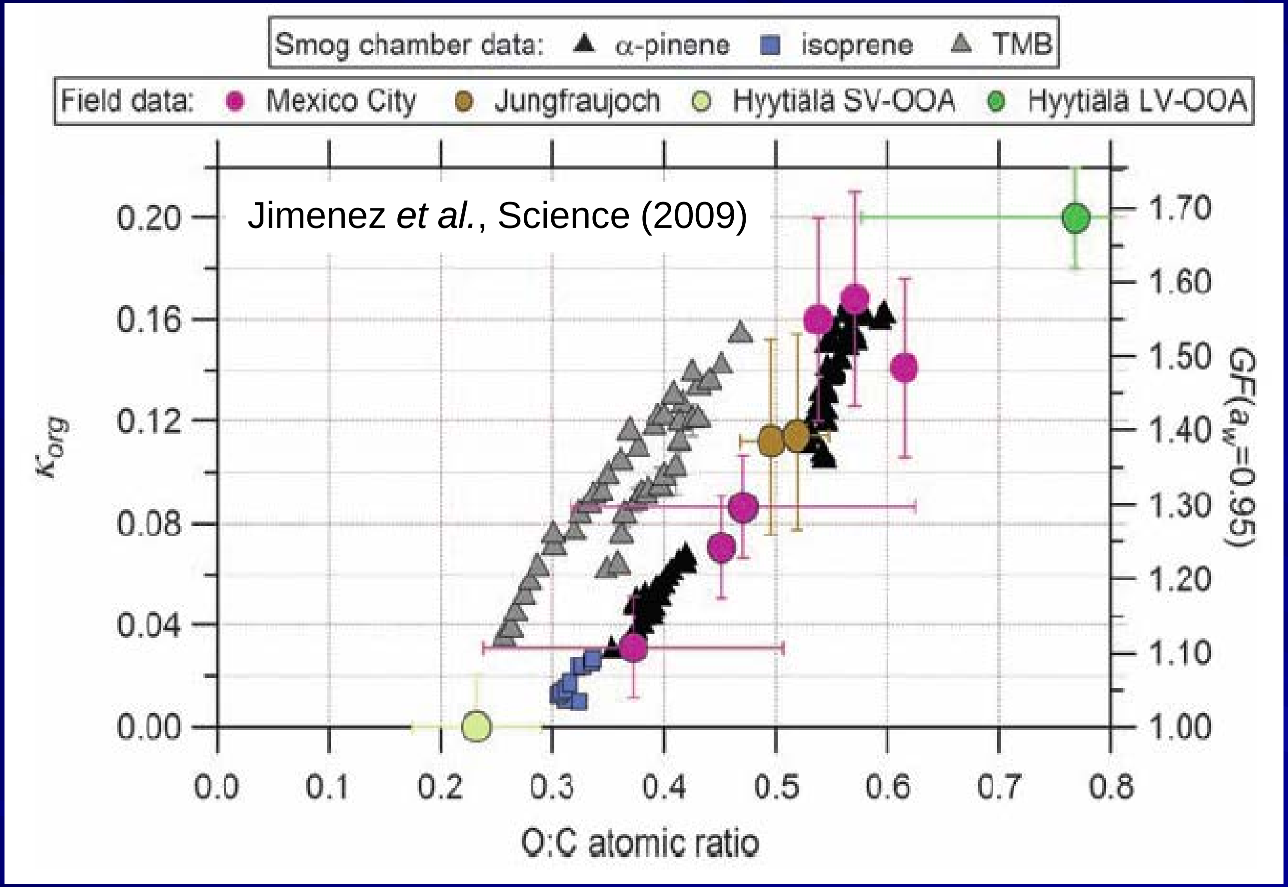
- Determine d_{50} dependence on supersaturation.
- Fit the measurements to a power law expression.
- Relate fitted coefficients to aerosol properties (e.g. hygroscopicity parameter κ) by applying theory:



$$S_c = \underline{\omega} d_{50}^{-3/2} = \left[\frac{4A^3}{27\underline{\kappa}}\right]^{1/2} d_{50}^{-3/2}$$

… κ can also be related to an average molecular weight of the solute in the aerosol (Padró et al., *ACP*, 2007).

Understanding & parameterizing CCN activity…
… of organic aerosol



κ_{org} depends on oxidation state and precursor.

O:C is related to the water-soluble fraction of organics,

$$\kappa_{org} \sim \varepsilon_{sol}\,\kappa_{sol}$$

Can most of the above be explained as variation in ε_{sol} ?

Look at the κ of water-soluble organics…

The link between κ_{org}, O:C and WSOC

- **Aged organics in Mexico City aerosol from MILAGRO (Padró et al, 2010).**

  κ_{sol} = 0.28 ± 0.06, regardless of location and time !

- **Organic SOA from biogenic VOCs**
  α-pinene, monoterpene, isoprene oxidation.
  κ_{sol} ~ 0.28 (Engelhart et al., *ACP*, 2009, 2011)

  β-caryophyllene (Asa-Awuku et al., *ACP*, 2009).

  κ_{sol} ~ 0.26

- **SOA from Anthropogenic VOCs** (Asa-Awuku et al.,*ACP*, 2010)
  terpinolene, cycloheptene, 1-methylcycloheptene ozonolysis

  κ_{sol} ~ 0.26-0.33

- **Biomass burning samples** (Asa-Awuku et al., 2008)

  κ_{sol} ~ 0.33

The link between κ_{org}, O:C and WSOC

Many “aged” soluble organics (SOA) from a wide variety of sources have a remarkably similar hygroscopicity.

Speciation across samples **varies considerably**, but their cumulative effects on CCN activity are about the same.

Changes in surface tension partially compensates for shifts in average molar volume to give the constant κ_{sol}

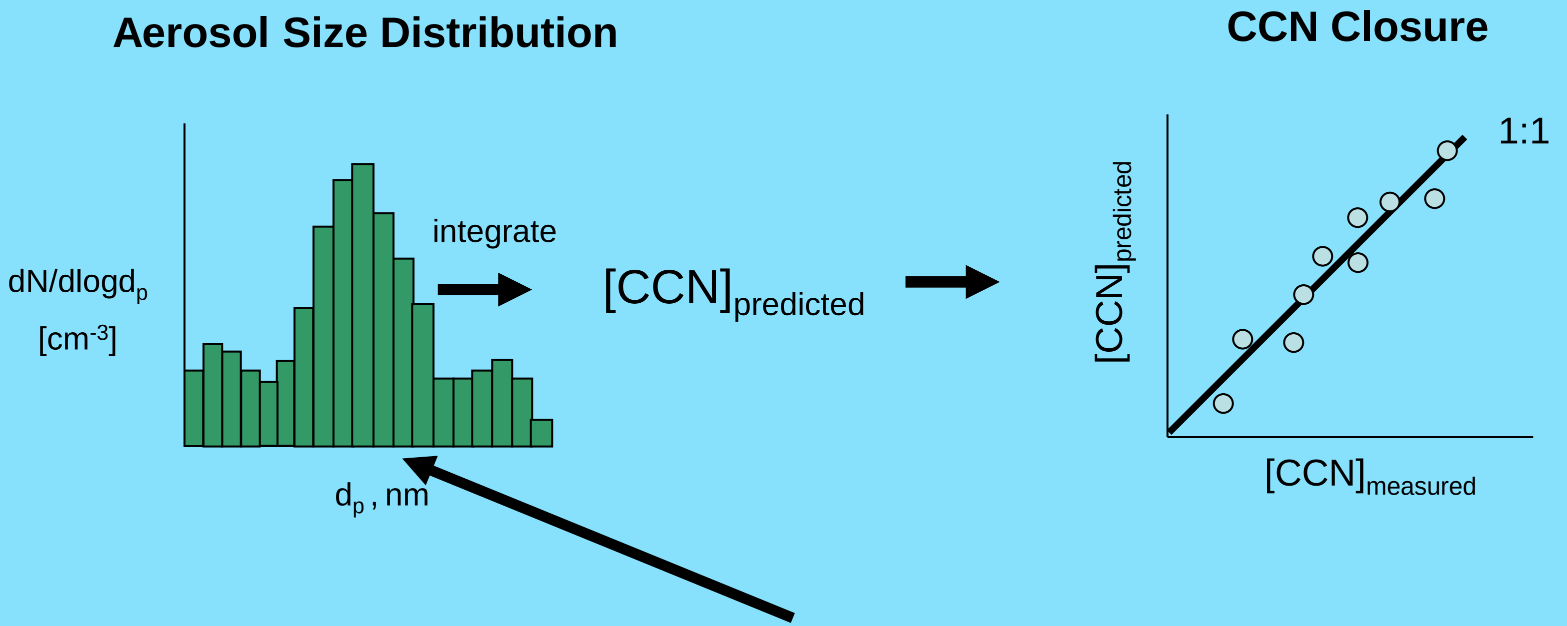
What matters is the fraction of soluble organic – which is consistent with κ_{org} correlating with O:C.

$$\kappa_{org} = (0.25 \pm 0.05)\ \varepsilon_{sol}$$

Complexity sometimes simplifies things for us.

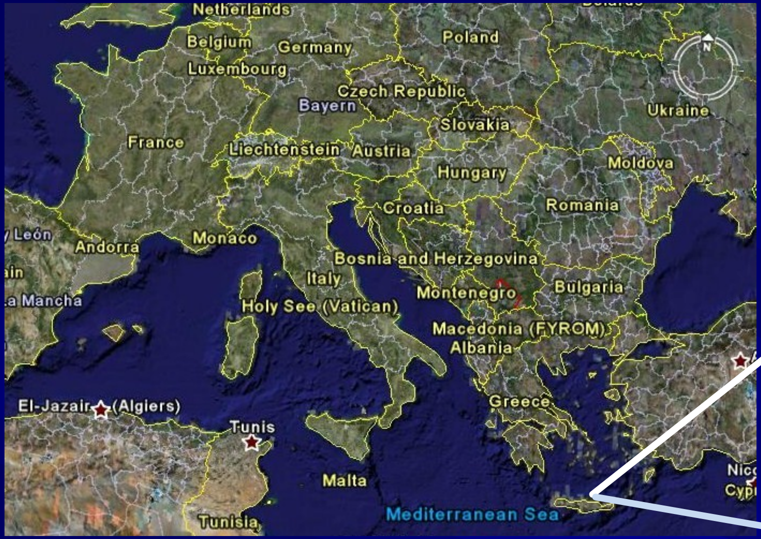
Testing CCN activation theory: CCN “Closure” studies

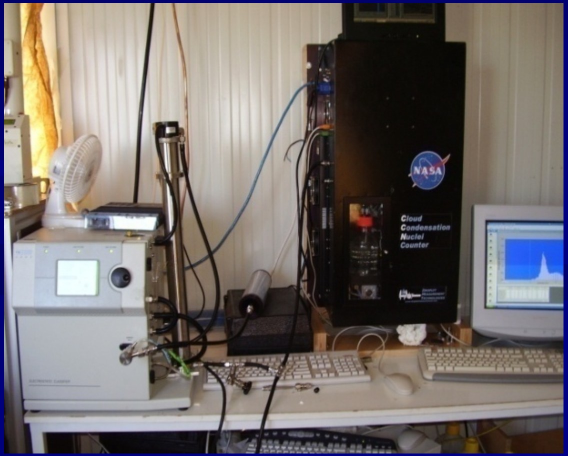
Compare measurements of CCN to predictions using Köhler activation theory and κ description



Use theory to predict the particles that can act as CCN based on measured chemical composition and CCN instrument supersaturation.

Finokalia Aerosol Measurement Campaign (FAME-07) – Summer 2007





DMT CCN counter
Supersaturation range: 0.2-1.0%

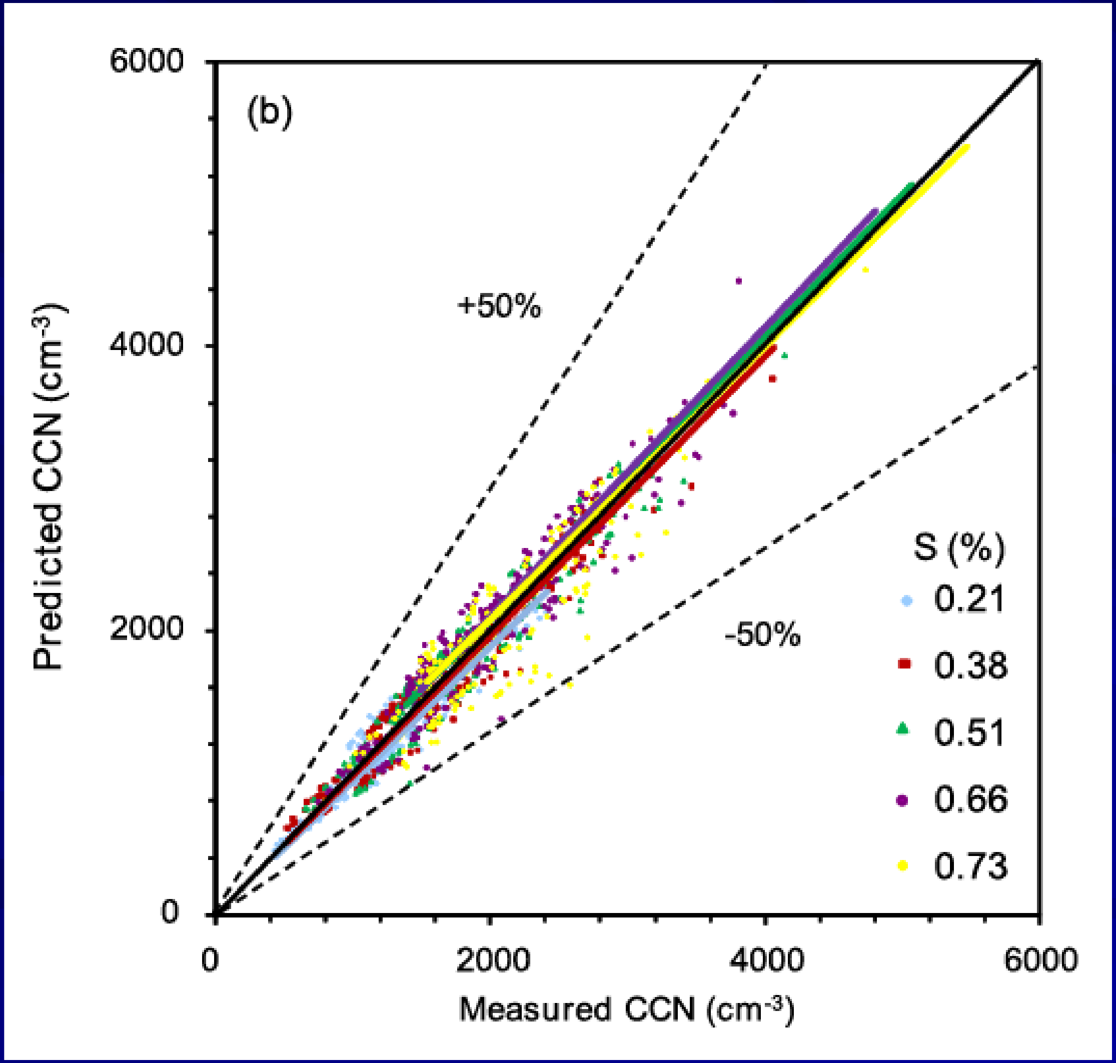
TSI 3080 SMPS
Size range: 20-460 nm

Low-vol impactor
Ionic composition measured via IC

WSOC/EC/OC also measured

(Bougiatioti et al., ACP, 2009)

Finokalia Aerosol Measurement Campaign (FAME-07) – CCN closure



2% overprediction (on average).

Introducing comprehensive composition into CCN calculation gives excellent CCN closure.

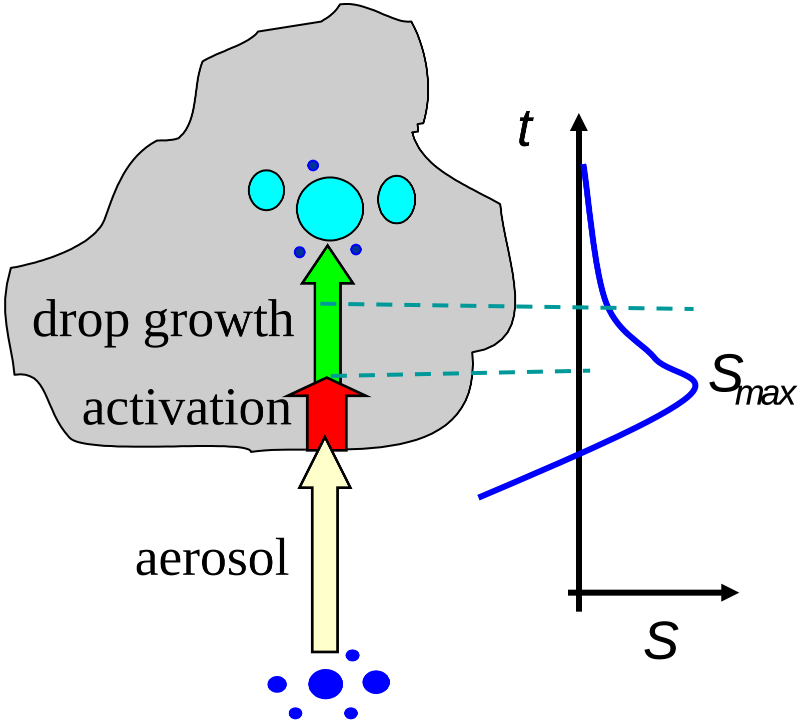
Köhler (CCN activation) theory *really* works.

(Bougiatioti et al., ACP, 2009)

CCN activation requires knowledge of cloud RH…

Approach: use the "simple story of droplet formation"

Basic ideas: Solve conservation laws for energy and the water vapor condensing on aerosol particles in cloudy updrafts.



Steps are:

- Air parcel cools
- Eventually exceeds dew point
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- Droplets start forming on existing CCN.
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- *S* reaches a maximum
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A "classical" nucleation/growth problem

Liquid Phase Clouds

Approach: use the "simple story" (1D parcel theory)

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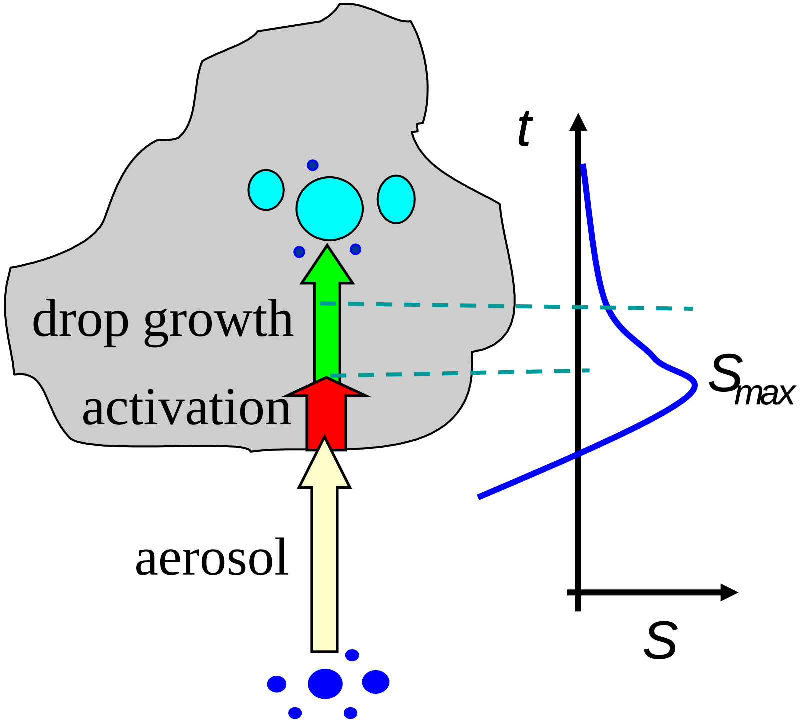


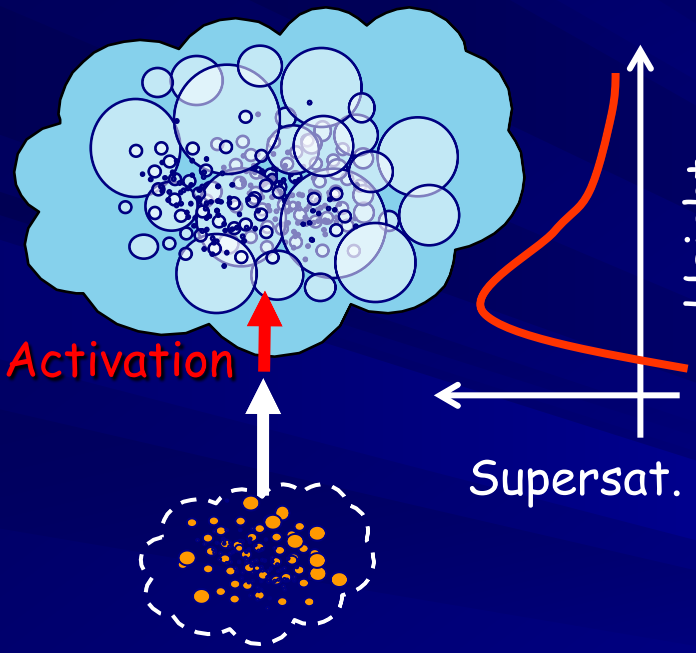
Parameterization goals:

Determine the number of droplets N_d that can activate at the parcel maximum supersaturation, s_{max}.

This is a two step process:

1. Obtain parcel s_{max}
2. Determine N_d by counting Cloud Condensational Nuclei (CCN) with $s_c < s_{max}$
3. CCN are determined from an appropriate theory (Köhler, Adsorption activation, etc).

Cloud Droplet Formation in GCMs
State of the art



Aerosol in an
adiabatic parcel

Mechanistic Parameterizations: Twomey (1959); Abdul-Razzak et al., (1998); Nenes and Seinfeld, (2003); Fountoukis and Nenes, (2005); Ming et al., (2006), and others.

Input: P,T, vertical wind, particle size distribution,composition.

Output: Cloud properties (droplet number, size distribution).

How: Solve/apply **one algebraic equation** (instead of ODE's).

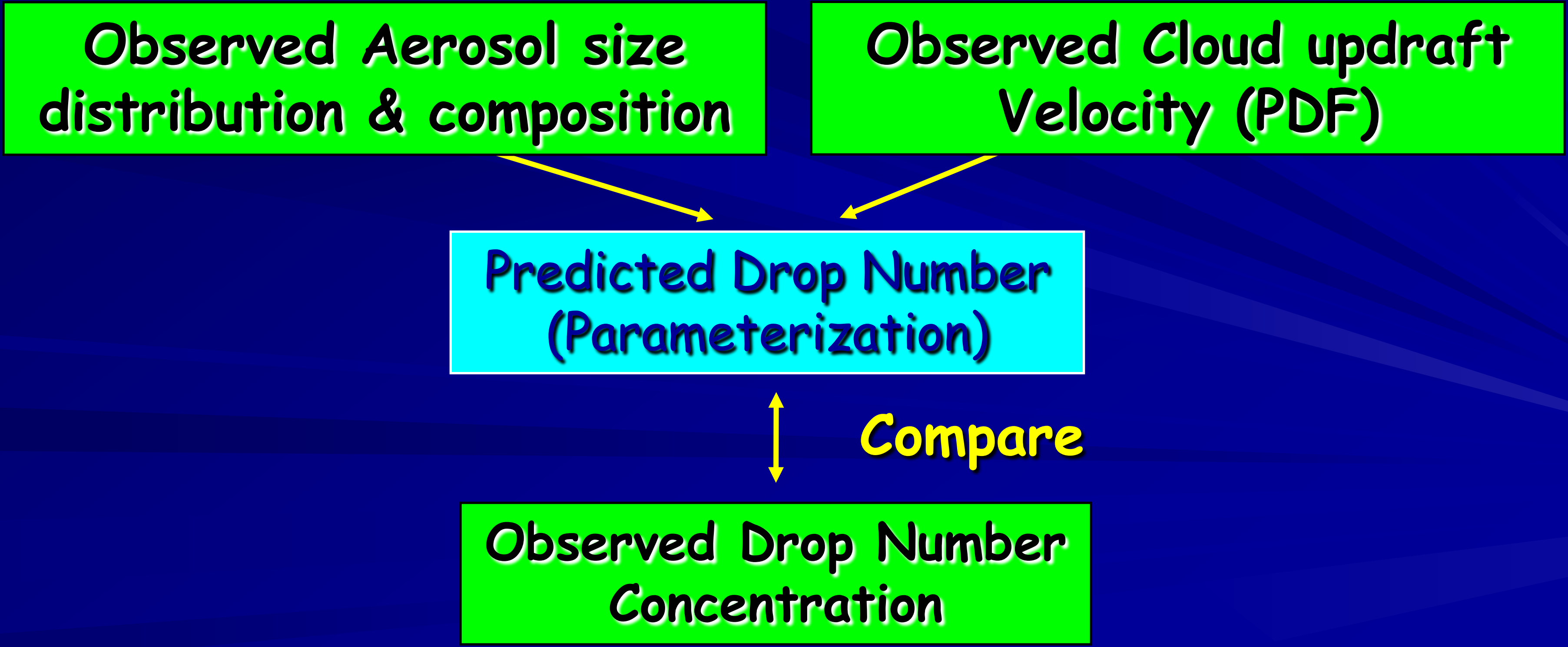
Basic Assumption: Adiabaticity

Comprehensive review & intercomparison:
Ghan, Abdul-Razzak, Nenes et al., *Rev.Geoph.*, in review

Are these parameterizations "good enough"?

Evaluate them with in-situ data from airborne platforms

CIRPAS Twin Otter



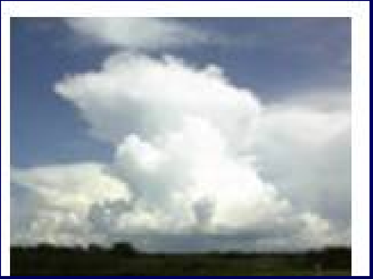
Observed Aerosol size distribution & composition

Observed Cloud updraft Velocity (PDF)

Predicted Drop Number (Parameterization)

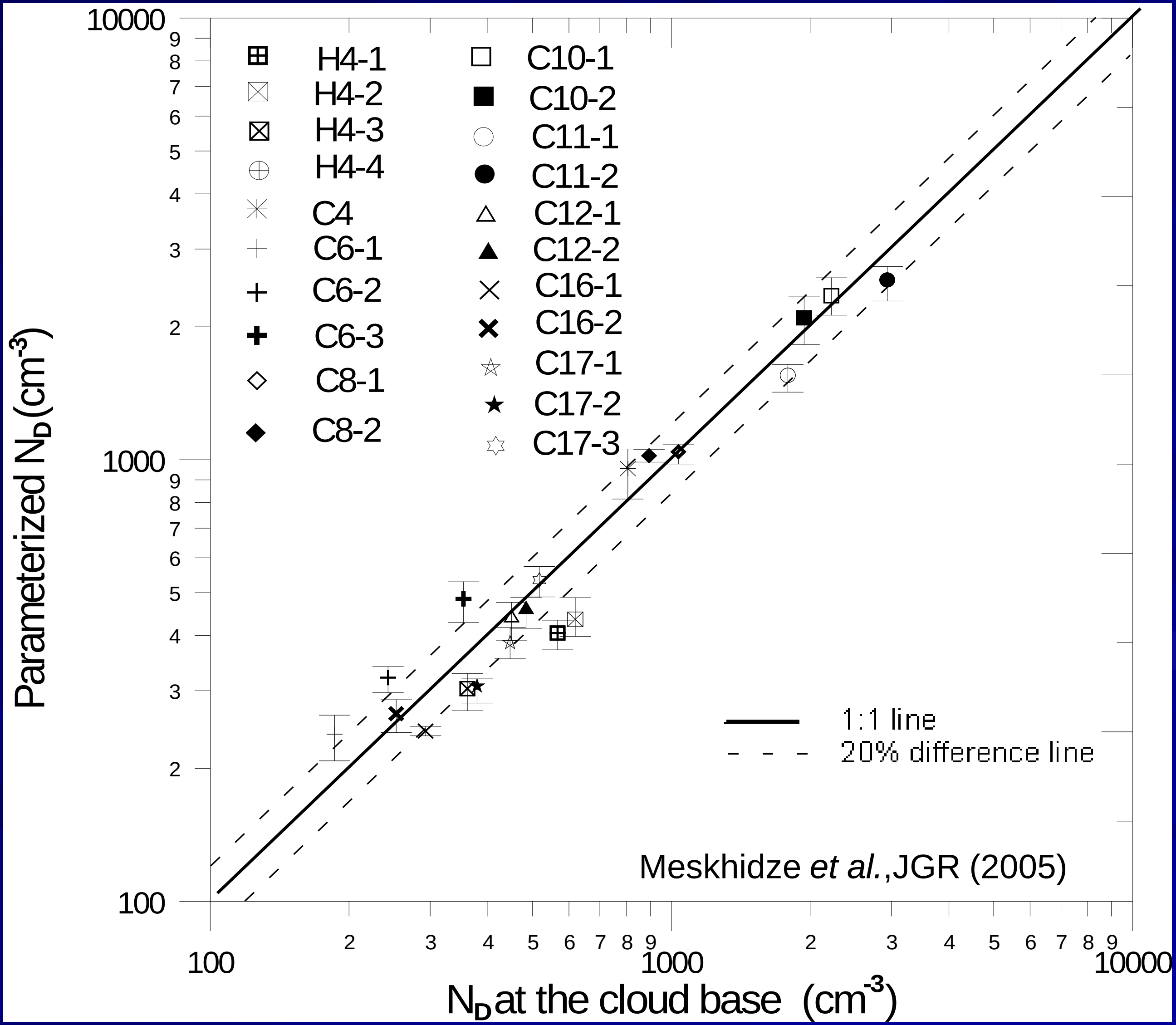
Compare

Observed Drop Number Concentration



CRYSTAL-FACE (2002)
“Adiabatic” Cumulus clouds

CIRPAS Twin Otter



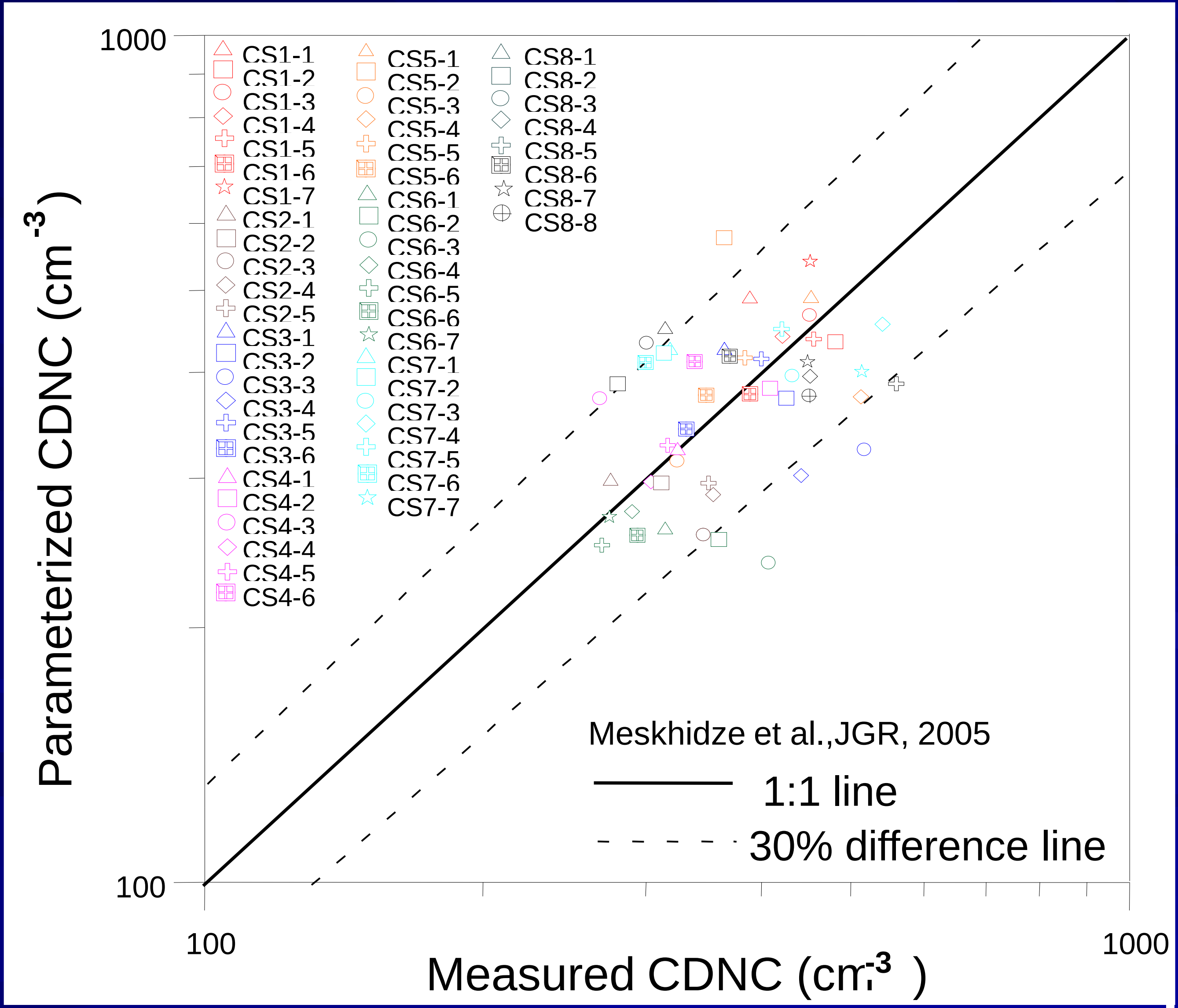
Meskhidze *et al.*,JGR (2005)

Paramet’n agrees with observed cloud droplet number *in “adiabatic-like” parcels.*

Agreement to within a few % (on average)!

CSTRIPE (2003)
Marine Stratocumulus

CIRPAS Twin Otter

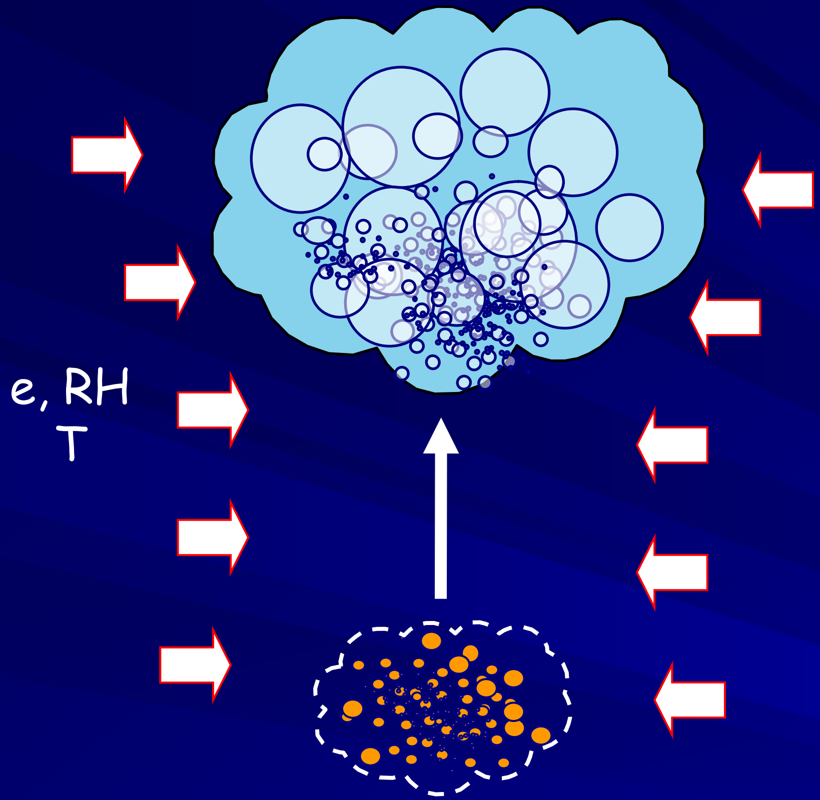


Meskhidze et al.,JGR, 2005

Paramet'n agrees with observed cloud droplet number *in "adiabatic-like" parcels.*

Agreement to within a few % (on average)!

Ambient clouds are not usually adiabatic…



- Barahona and Nenes (2007): Droplet parameterization for clouds continuously entraining in dry air.
- Equations are similar to adiabatic activation – only that mixing of outside air is allowed.
- "Outside" air with (RH, T) is assumed to entrain at a rate of *e* (kg air / kg parcel / m ascent)

Very important point finding:

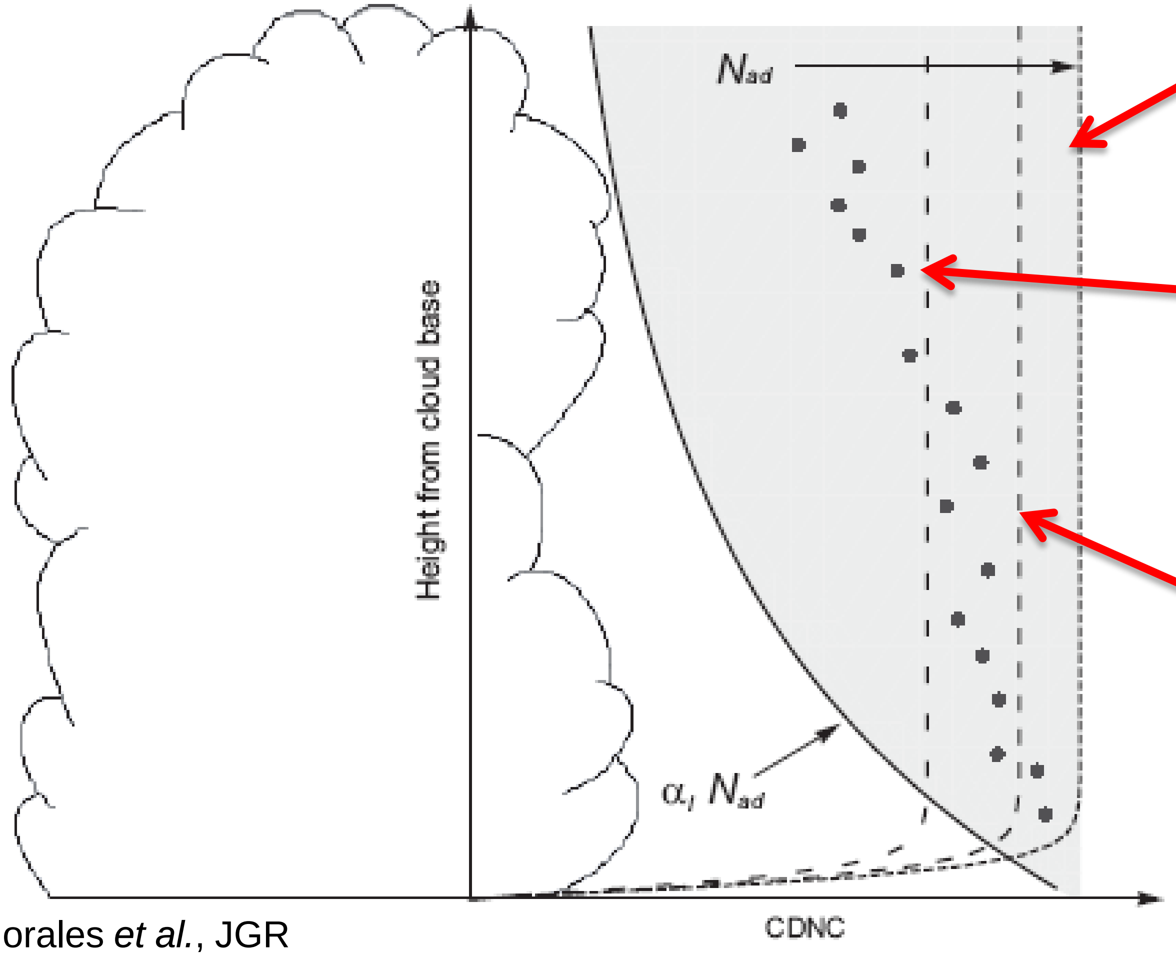
- Any adiabatic parameterization can be modified to consider entrainment - just replace **w** with $w(1-e/e_c)$!

vertical velocity

entrainment rate that completely dissipates cloud

Expressing entrainment effects on N_d

Approach: N_d predicted from the entraining parameterization represents cloud average. Link e,e_c to liquid water profile.

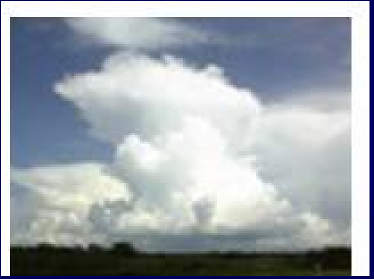


Morales *et al.*, JGR

Adiabatic N_d Overestimation

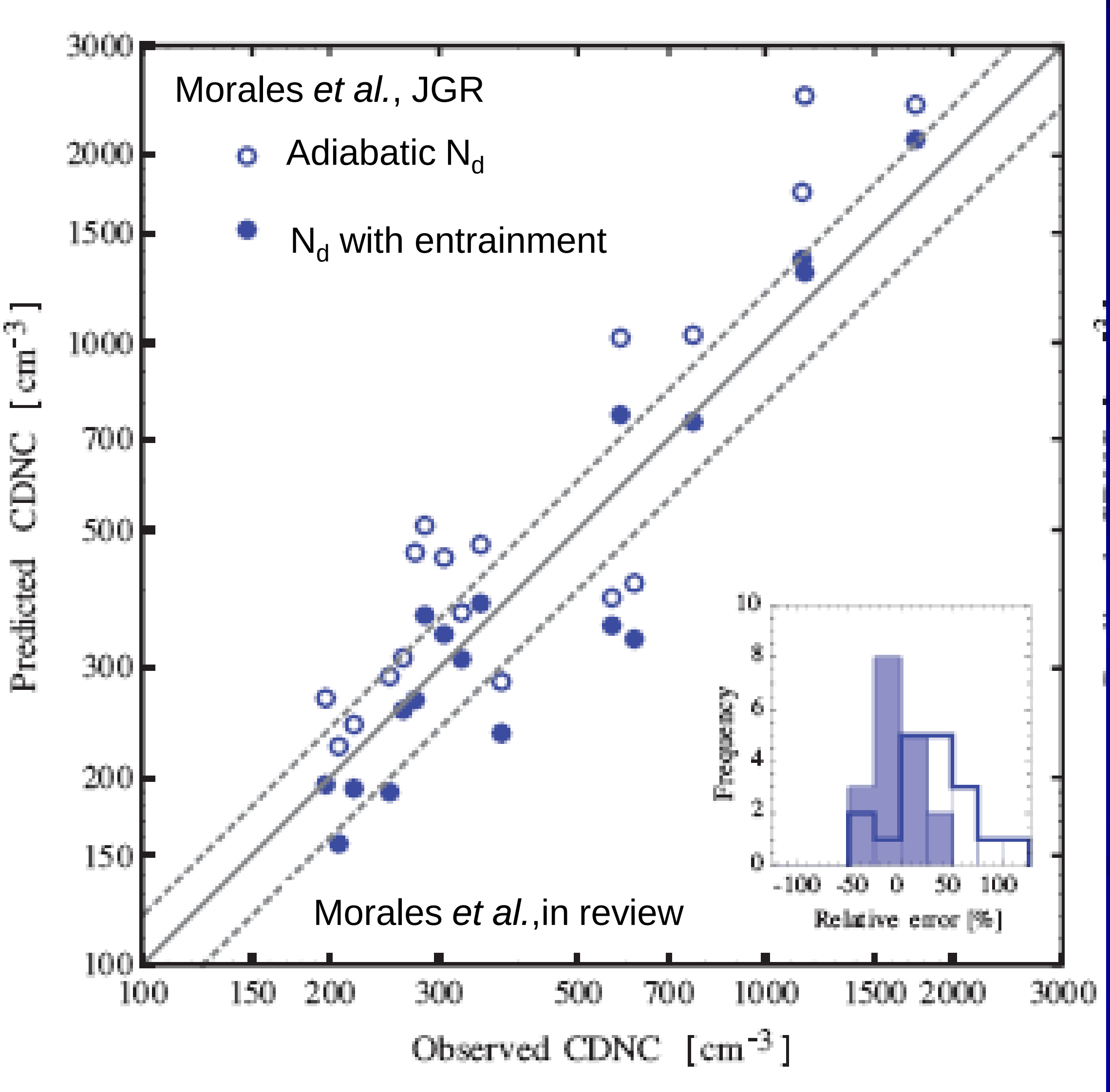
N_d with $1-e/e_c$ diagnosed from the average dilution ratio (LWC/LWC_{ad})

N_d with constant entrainment rate diagnosed from average dilution ratio



CRYSTAL-FACE (2002)
Entraining Cumulus clouds

CIRPAS Twin Otter



Adiabatic N_d:
45% overprediction

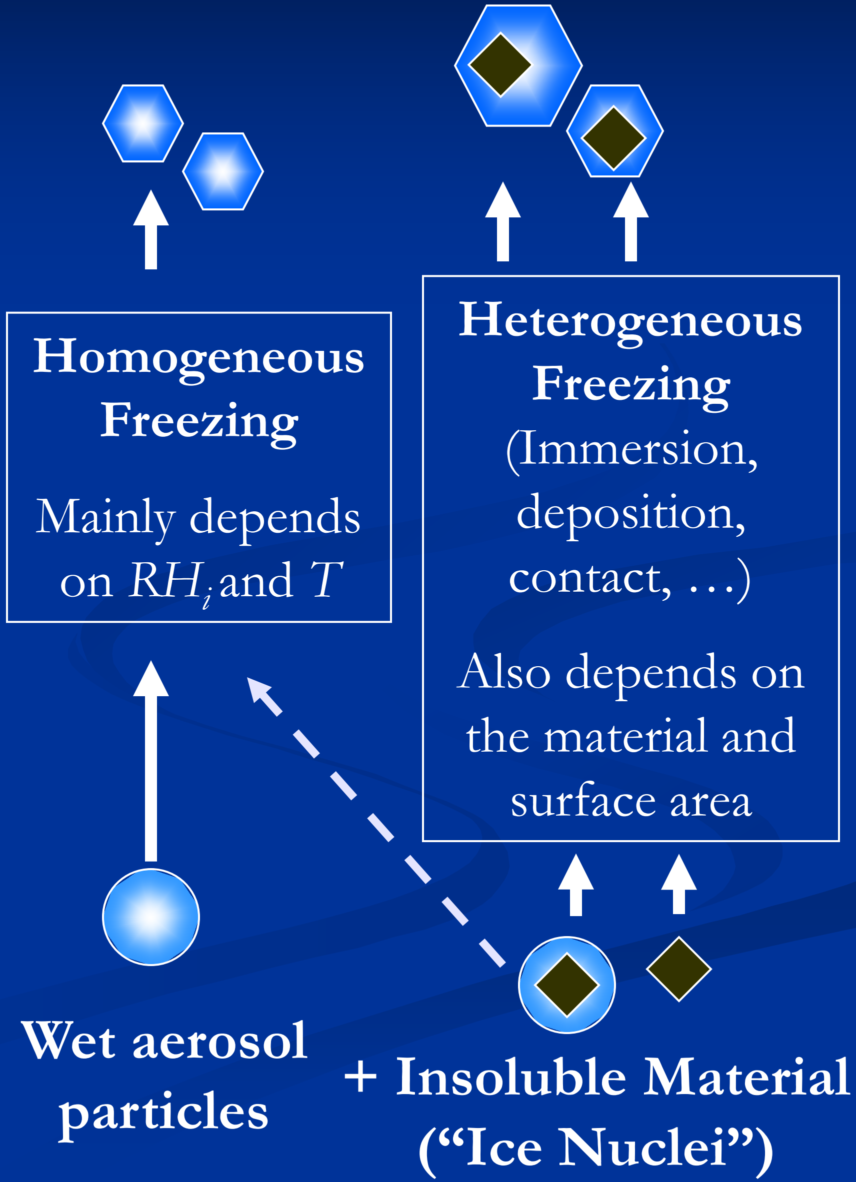
N_d with entrainment:
3.5% error when Column-average adiabaticity ratio (LWC/LWC_{ad}) used to diagnose $1-e/e_c$

N_d with pure heterogeneous mixing
45% underprediction

Cirrus (Ice) Clouds

Multiple mechanisms for ice formation can be active.

http://www.alanbauer.com



Homogeneous Freezing

Mainly depends on RH_i and T

Heterogeneous Freezing

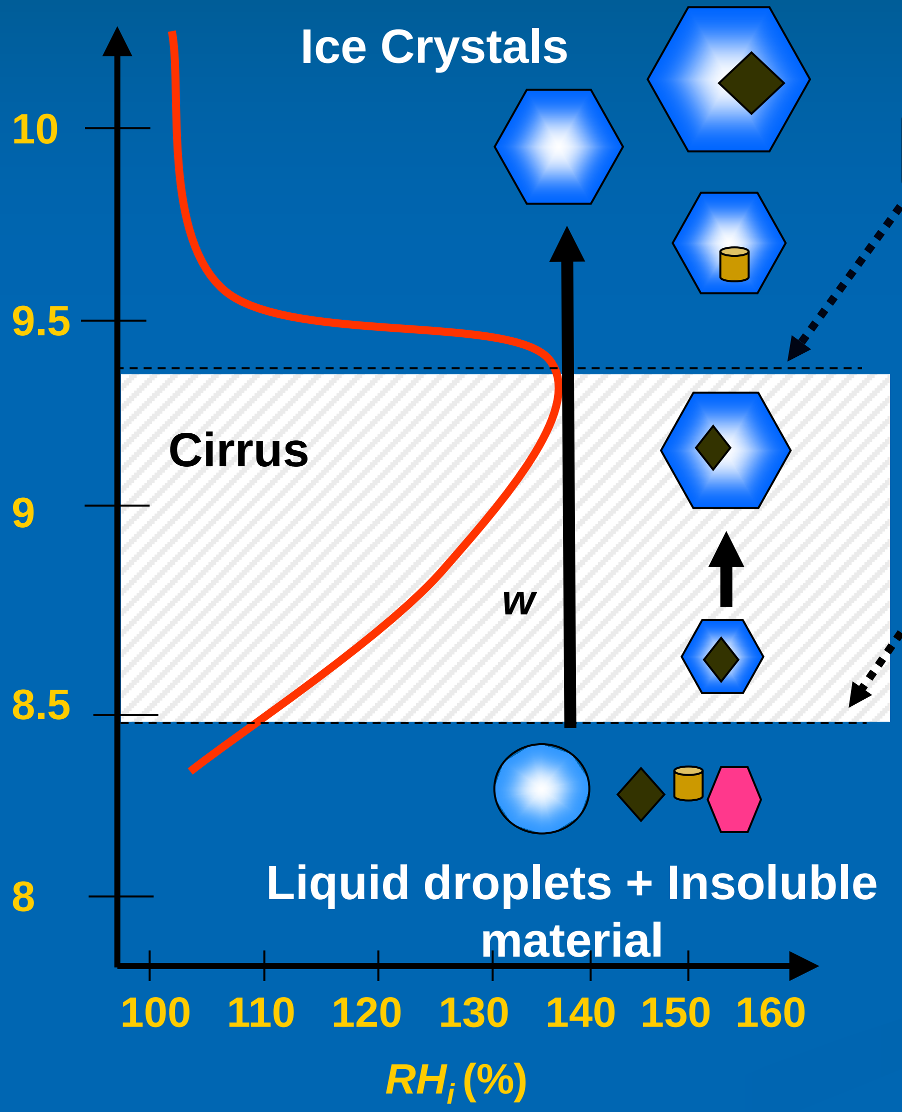
(Immersion, deposition, contact, …)

Also depends on the material and surface area

Wet aerosol particles

+ Insoluble Material (“Ice Nuclei”)

Cirrus (Ice) Clouds



Conceptual steps are:

Homogeneous freezing of droplets

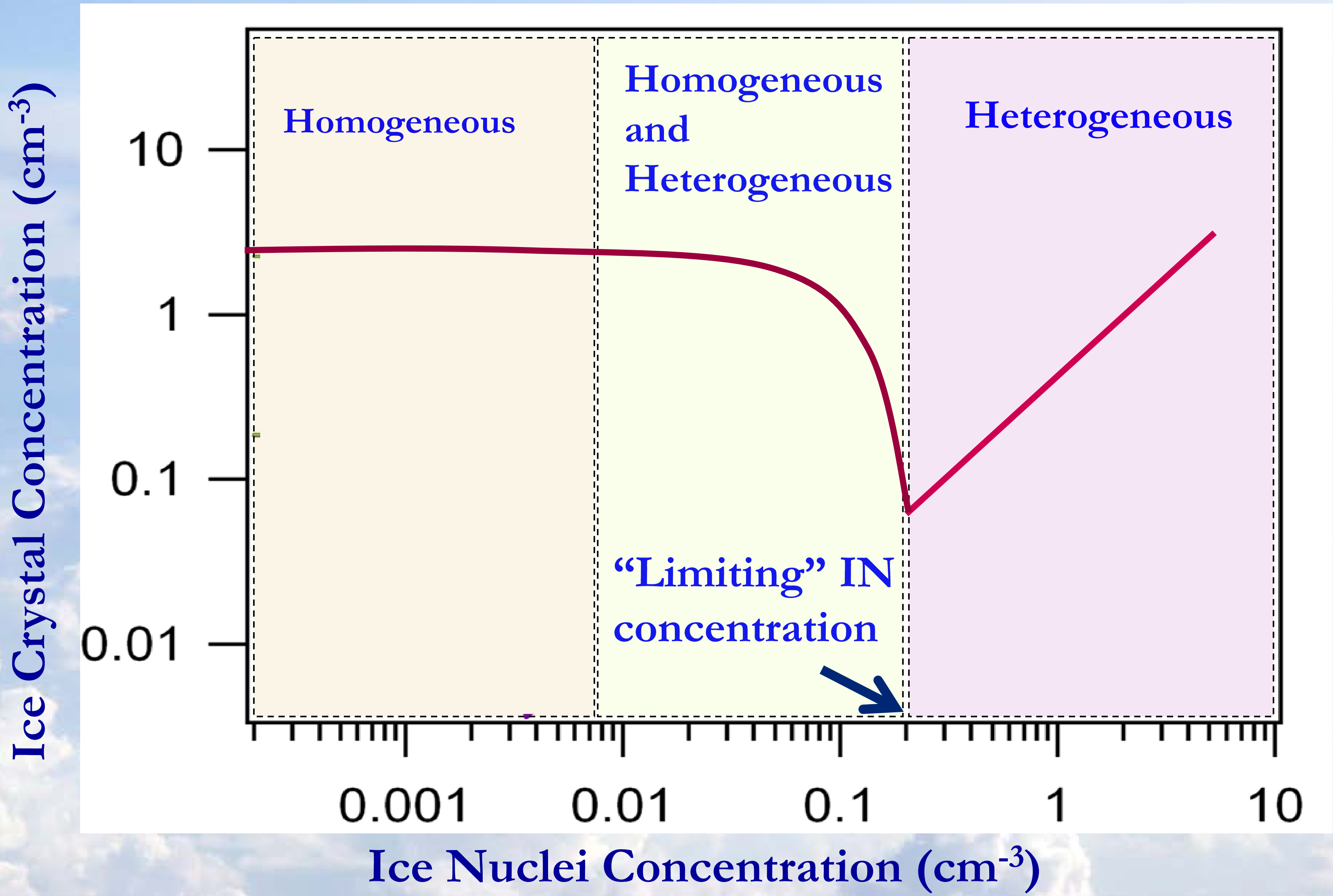
Crystal growth, fresh IN continue to freeze and deplete vapor

Heterogeneous IN freezing begin forming ice

Expansion cooling and ice supersaturation development

Soluble and insoluble aerosol initial distribution

Source of strong nonlinearity:
IN effects on Ice Crystal Concentration



Barahona and Nenes, *ACP*, 2009a.

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

Approach	Advantages	Disadvantages
Empirical	Very fast Representative values	Limited Coverage Cannot be used to assess aerosol effects
Off-line solutions (Liu and Penner, 2005)	Fast Physically-based	Consider only a limited range of conditions
Analytical-Numerical (e.g., Kärcher, et al., 2006)	Most of the physics included.	Simplistic description of IN (Single freezingthreshold”).

Analytical models based on cloud formation equations are needed!

Ice Parameterization Development
Solving the parcel equations…

$$\frac{dS_i}{dt} = -\frac{M_a p}{M_w p_i^o}\frac{dw_i}{dt} - (1+S_i)\left[\frac{\Delta H_s M_w}{RT^2}\frac{dT}{dt} - \frac{gM_a}{RT}V\right]$$

Global water vapor balance

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

Energy balance

$$\frac{dw_i}{dt} = \frac{\rho_i}{\rho_a}\frac{\pi}{2}\int \dots \int_X D_c^2 \frac{dD_c}{dt} n_c(D_c, D_{IN}, m_{1,\dots,nx}, t)\, dD_c\, dD_{IN}\, dm_{1,\dots,nx}$$

Ice water vapor condensation

$$\frac{\partial n_c(D_c, D_o)}{\partial t} = -\frac{\partial}{\partial D_c}\left(n_c(D_c, D_o)\frac{dD_c}{dt}\right) + n_o(D_o, S_i) v_o J(t) \exp\left(-\frac{\pi}{6}\int_0^t D_o^3 J(t)\, dt\right)$$

Ice crystal size distribution evolution = nucleation + growth

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

Ice crystal growth

… lots of math and scaling…

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

Analytical Parameterization for Cirrus Ice Formation and Growth

The analytical solution of the parcel equations :

Ice Crystal Concentration →

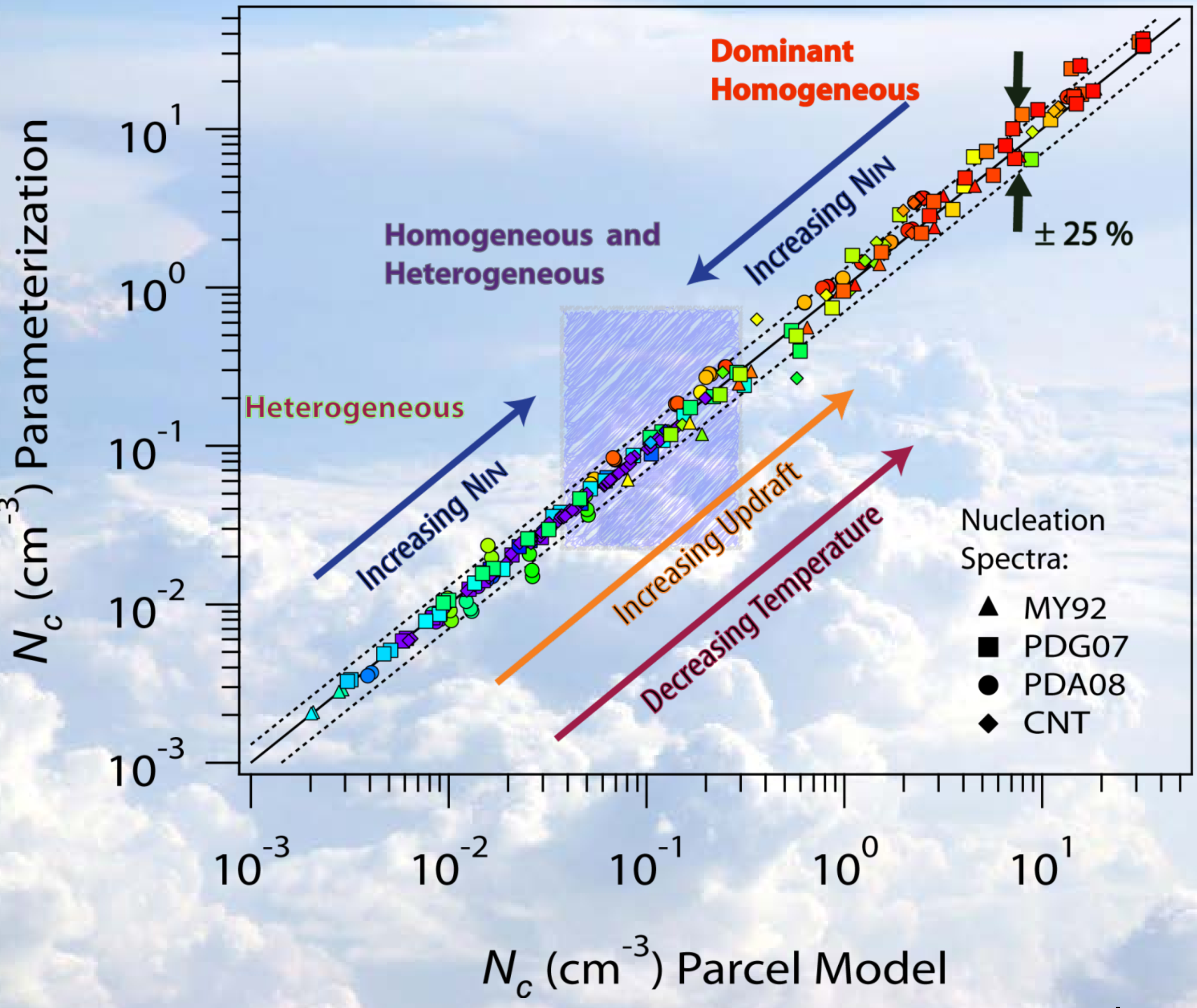
$$\frac{N_{het}(s_{\max})}{N^{*}} = \frac{1}{\sqrt{\Delta s^{*}_{char}}}\,\frac{(1+s_{\max})}{s_{\max}}\,e^{\frac{2}{\lambda s_{\max}}}$$

← **Condensation integral**

Barahona and Nenes, *ACP*, 2008,2009ab.

- 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 (i.e., ranges of freezing thresholds).
    - Allows direct incorporation of theoretical and empirical data into large scale models.

Cirrus parameterization evaluation: Compare Against Numerical Solution



Average error over a broad range of conditions: **5±12 %.**

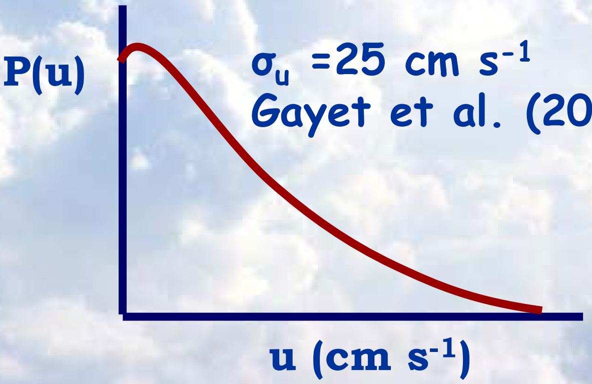
Orders of magnitude faster than the numerical solution

In-situ datasets are much needed to evaluate these relationships.

Barahona and Nenes, *ACP*, 2009b.

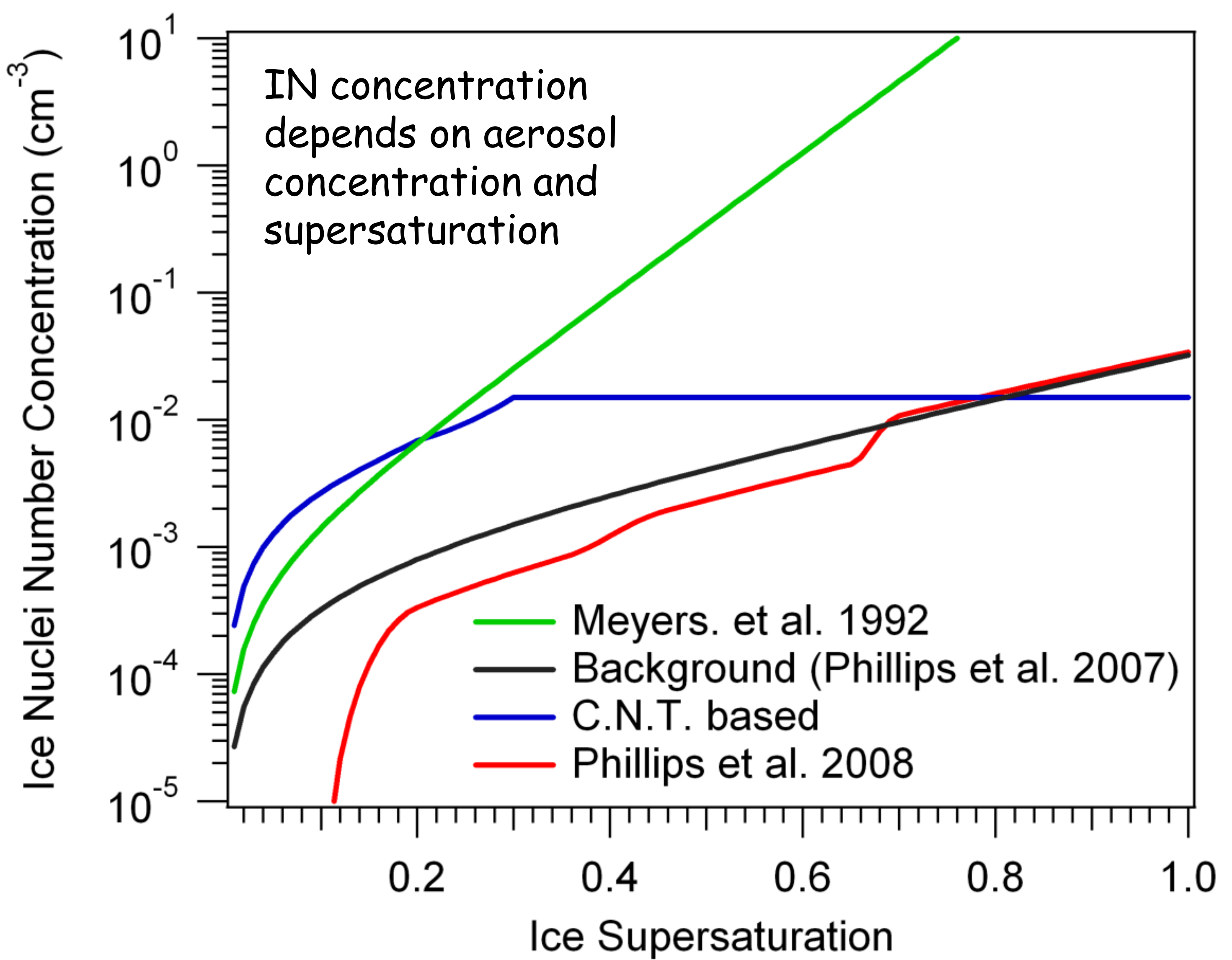
Application: Sensitivity of global ice crystal concentration to IN

- 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



σ_u =25 cm s^{-1}
Gayet et al. (2006)

Heterogeneous Freezing Spectra Considered

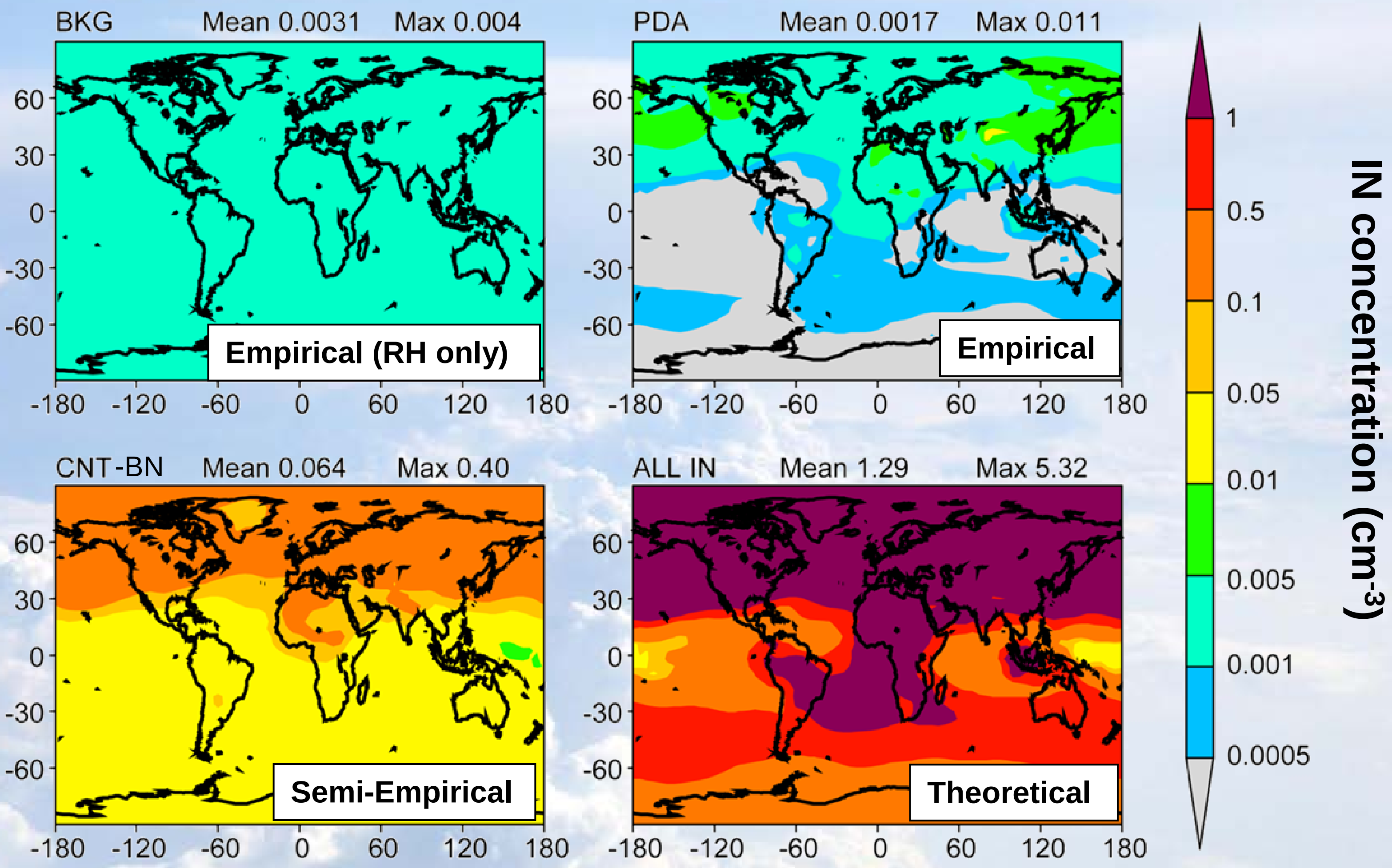


IN concentration depends on aerosol concentration and supersaturation

Barahona, Rodriguez, and Nenes,JGR, 2010.

Heterogeneous IN Concentrations

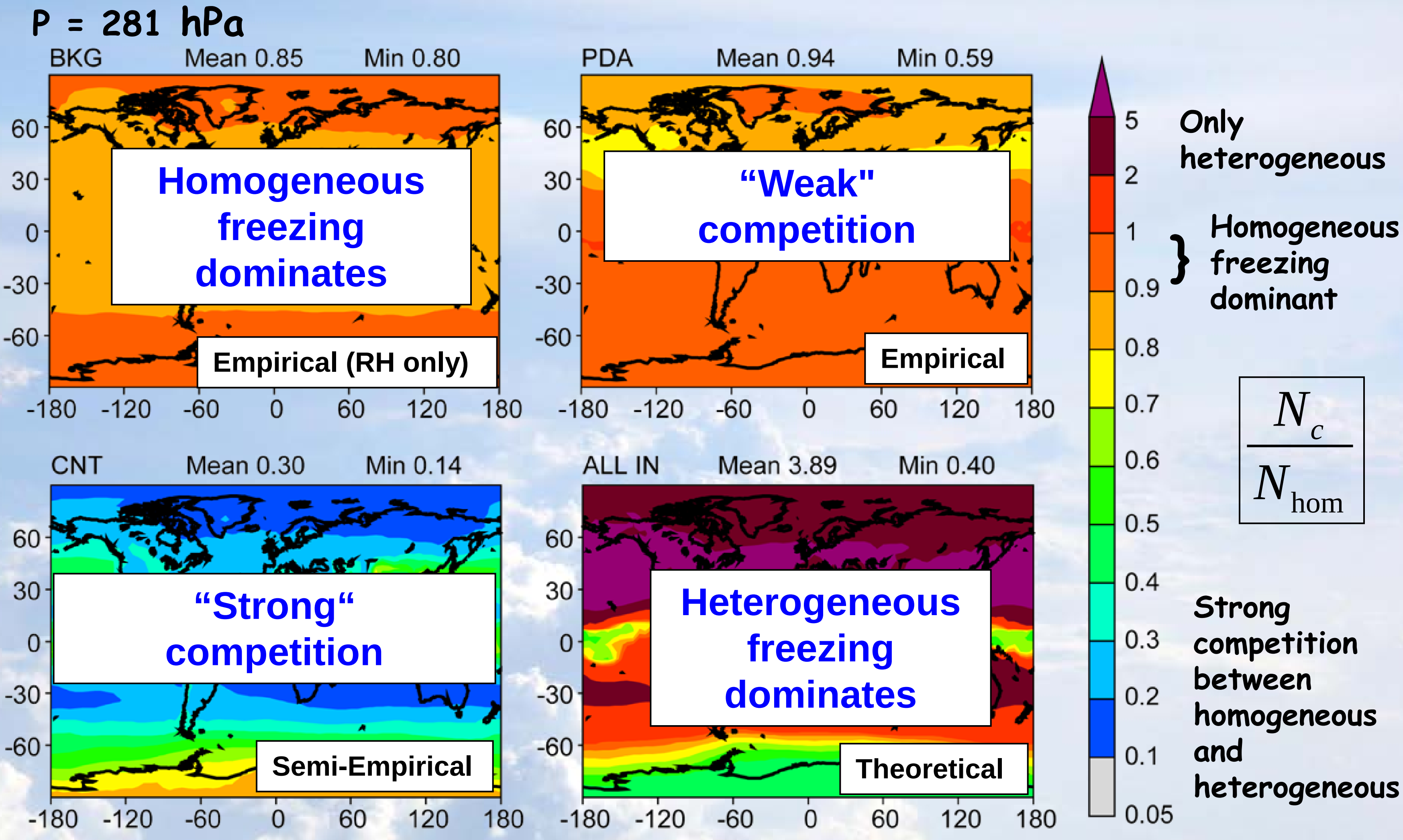
P = 281 hPa



About three orders of magnitude difference in IN concentration
Between IN parameterization expressions

Barahona, Rodriguez, and Nenes,JGR, 2010.

IN impacts on Ice crystal number



P = 281 hPa

Only heterogeneous

} Homogeneous freezing dominant

$$\frac{N_c}{N_{\mathrm{hom}}}$$

Strong competition between homogeneous and heterogeneous

- **A factor of 5-10 variation in global mean ice crystal concentration. Most significant in Northern Hemisphere**

Barahona, Rodriguez, and Nenes,JGR, 2010.

Fast & Comprehensive Physics: Take-home messages

- Physically-based representations of droplet and ice formation in GCMs is now becoming sophisticated… but still very fast.
- With simplified aerosol composition treatment, activation parameterizations can do a good job of predicting N_d in ambient clouds.
- A simple treatment of entrainment/mixing of air seems to capture N_d in diabatic clouds (on average).
- Ice formation in cirrus can now be comprehensively treated, using wither observational data or heterogeneous nucleation theory for IN predictions.
- These expressions need to be continuously evaluated with model/in-situ data (especially ice) but are very promising for linking aerosol with clouds.

THANK YOU !!

For more information and PDF
reprints, please go to
http://nenes.eas.gatech.edu