Representing and Constraining Cloud Droplet Formation

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The complexity of aerosol-cloud interactions

Everything depends on everything across multiple scales

Dynamics

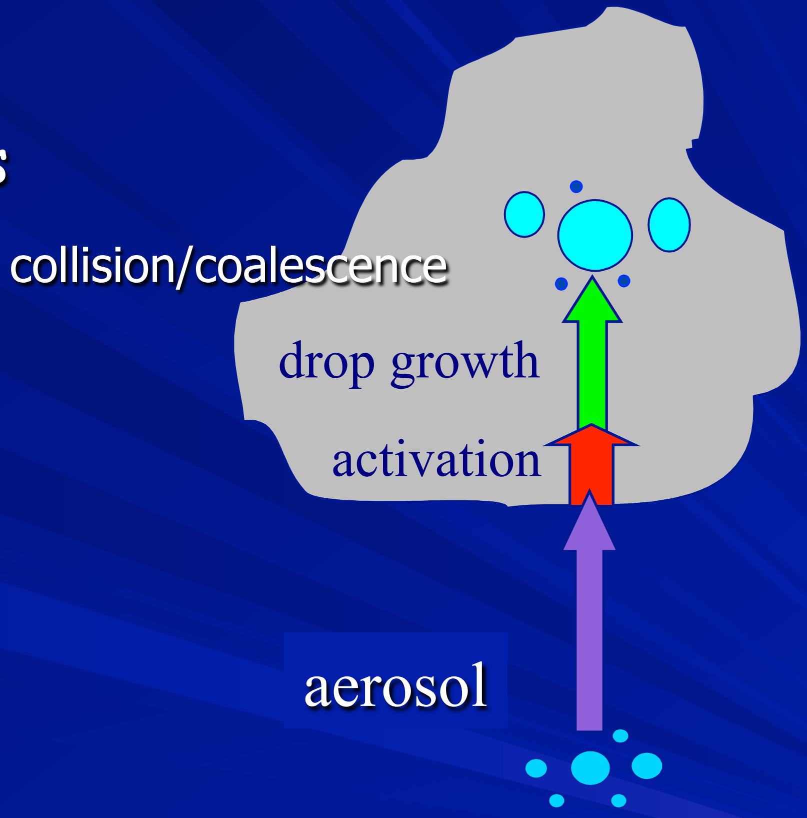
- Updraft Velocity
- Large Scale Thermodynamics

Particle characteristics

- Size
- Concentration
- Chemical Composition

Cloud Processes

- Cloud droplet formation
- Drizzle formation
- Rainwater formation
- Chemistry inside cloud droplets



All the links need to be incorporated in global models
The links need to be COMPUTATIONALLY feasible.

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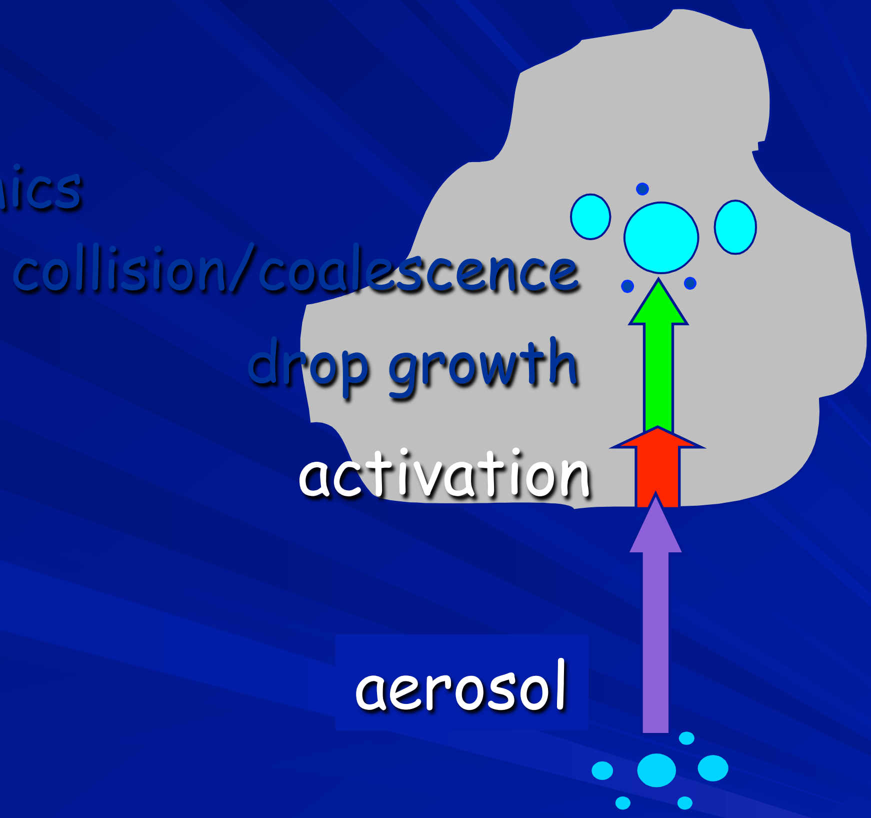
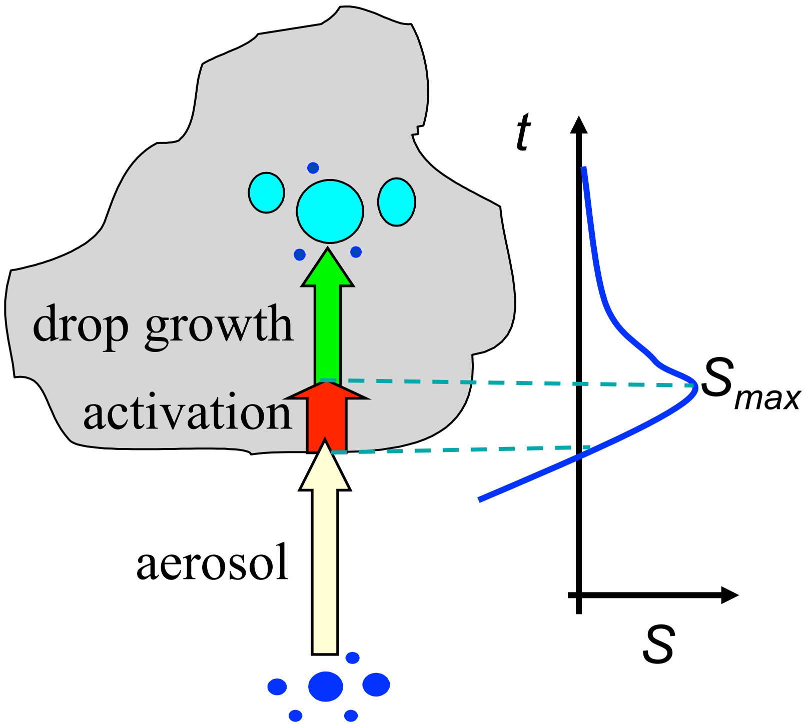


We focus on the aerosol-CCN-droplet link

The "simple" story of cloud droplet formation

Basic idea: Solve conservation laws for energy and water for an ascending Lagrangian parcel containing some aerosol.



Steps are:

- Parcel cools as it rises
- Exceed the dew point at LCL
- Generate supersaturation
- Droplets start activating as S exceeds their S_c
- Condensation of water becomes intense.
- S reaches a maximum
- No more droplets form

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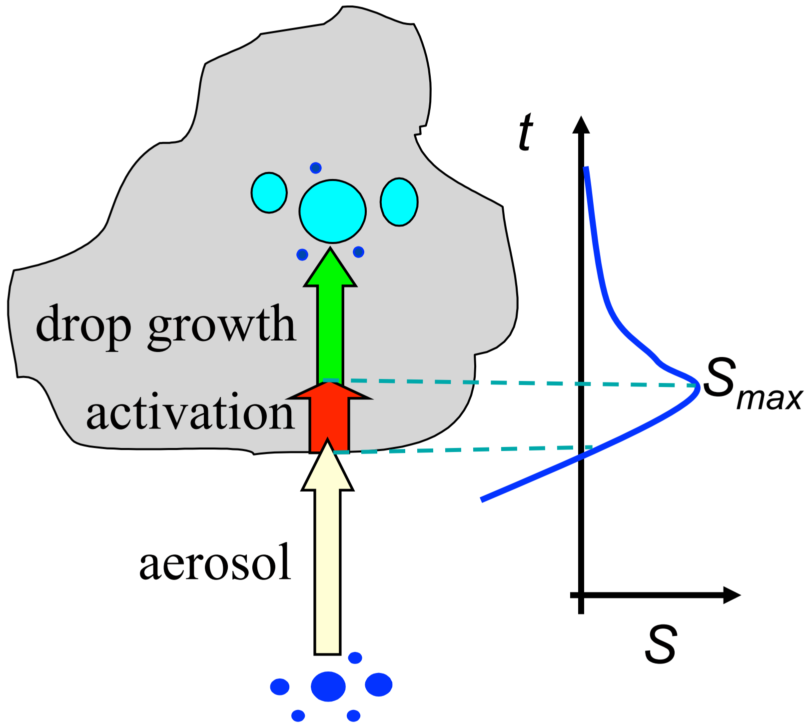

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Theory known for many years. Too slow to implement "completely" in large scale models

"Mechanistic" Cloud Parameterizations efficiently solve the drop formation problem

Input: P,T, vertical wind, particle characteristics.
Output: Cloud properties (droplet number, size distribution).
How: Solve an algebraic equation (instead of ODE's).

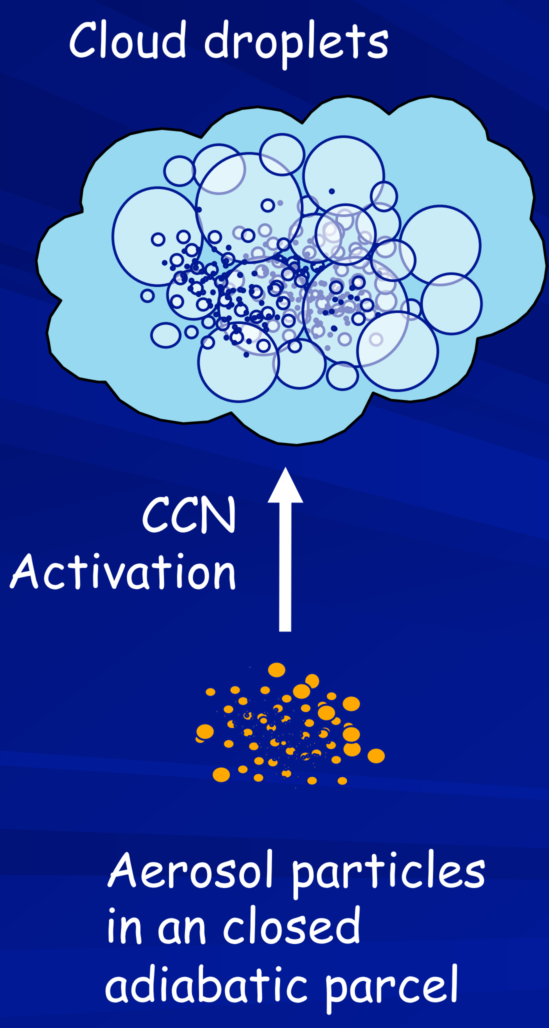
Examples:

Abdul-Razzak et al., (1998); Abdul-Razzak et al., (2000); Nenes and Seinfeld (2003), Fountoukis and Nenes (2005), Ming et al., (2007); Barahona and Nenes (2007)

Characteristics:

- 10^3-10^4 times *faster* than numerical parcel models.
- some can treat very complex chemical composition.
- have been evaluated using in-situ data with large success (e.g., Meskhidze et al., 2006; Fountoukis et al., 2007)

Mechanistic Parameterizations Current state of the art in GCMs



- Physically-based prognostic representations of the activation physics.
- Cloud droplet formation is parameterized by applying conservation principles in an ascending adiabatic air parcel.
- All parameterizations developed to date rely on the assumption that the droplet formation is an adiabatic process.

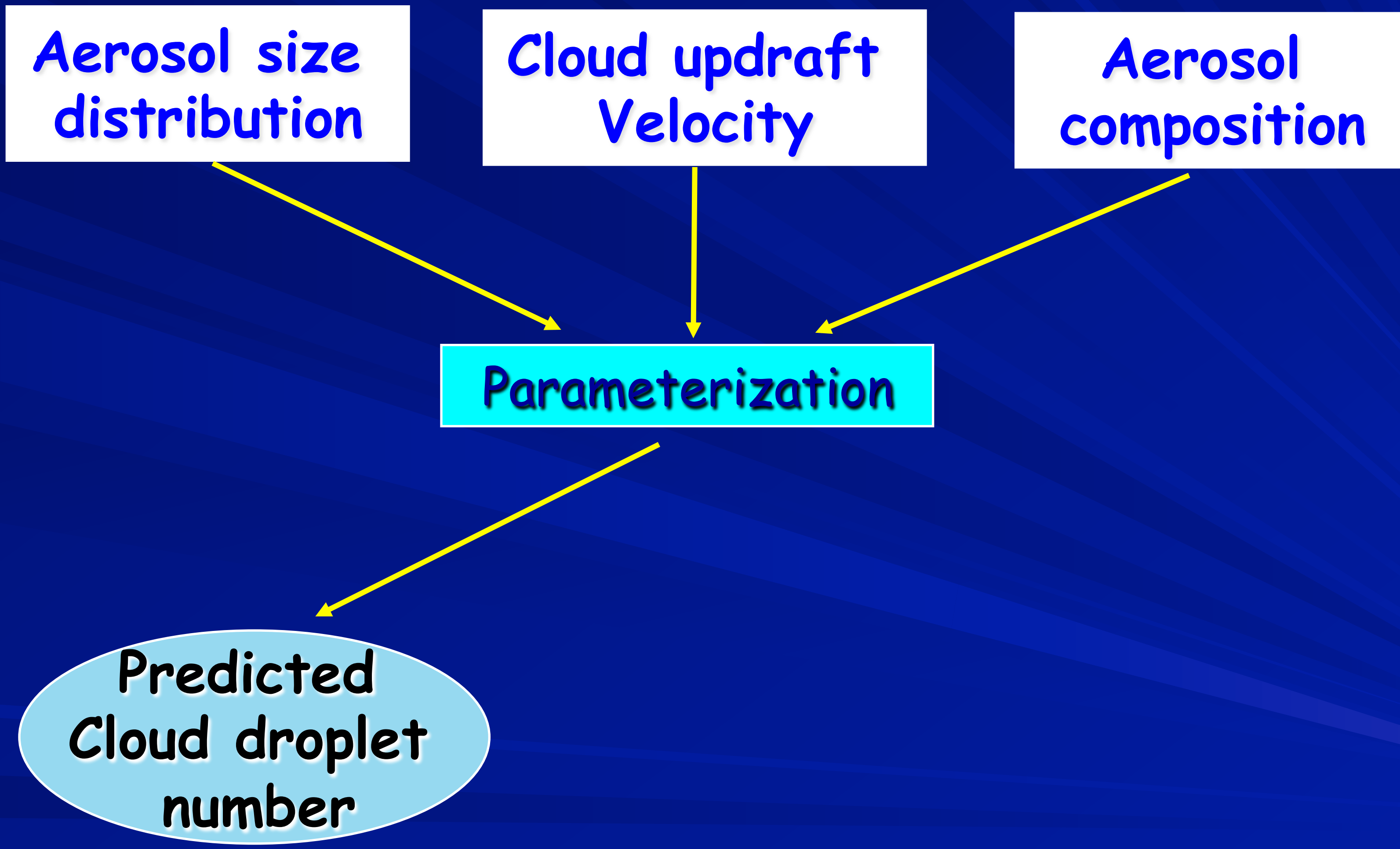
In-situ airborne platforms

Major “workhorse” for producing the aerosol-cloud datasets we need for parameterization evaluation and development.

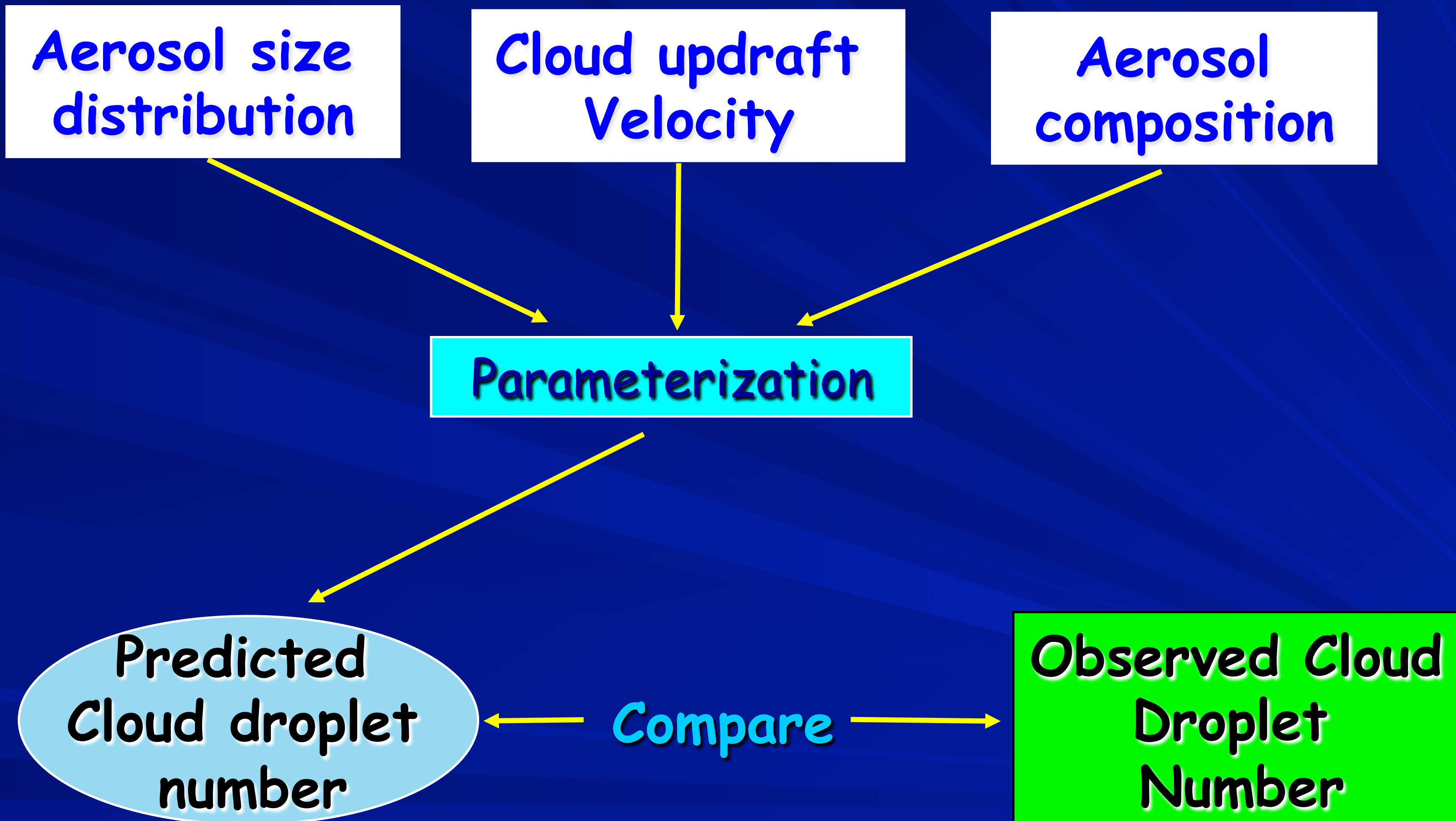
CIRPAS Twin Otter

NOAA P3

Cloud Drop Parameterization Evaluation CDNC “closure”



Parameterization Evaluation
CDNC "closure"



Adiabatic Cloud Formation Parameterization: Nenes and Seinfeld, 2003 (and later work).

Input: P,T, vertical wind, particle characteristics.
Output: Cloud properties.
How: Solve an algebraic equation (instead of ODE's).

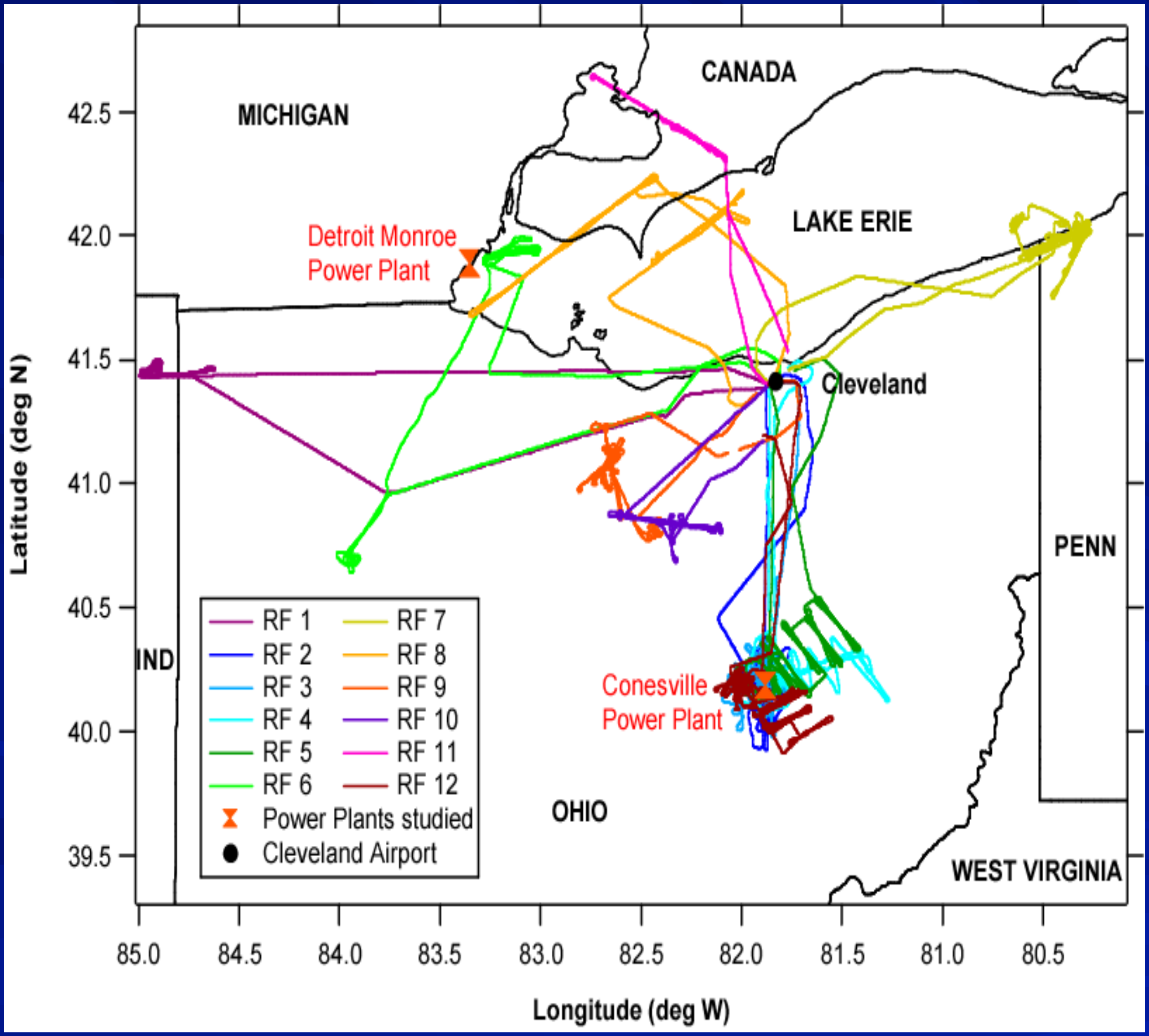
$$\frac{\pi}{2}\frac{\gamma\rho_w G S_{\max}}{aV}\left\{C_1\int_0^{s_{part}} f_1(s)ds + C_2\int_{s_{part}}^{s_{\max}} f_2(s)ds\right\} - 1 = 0$$

Features:

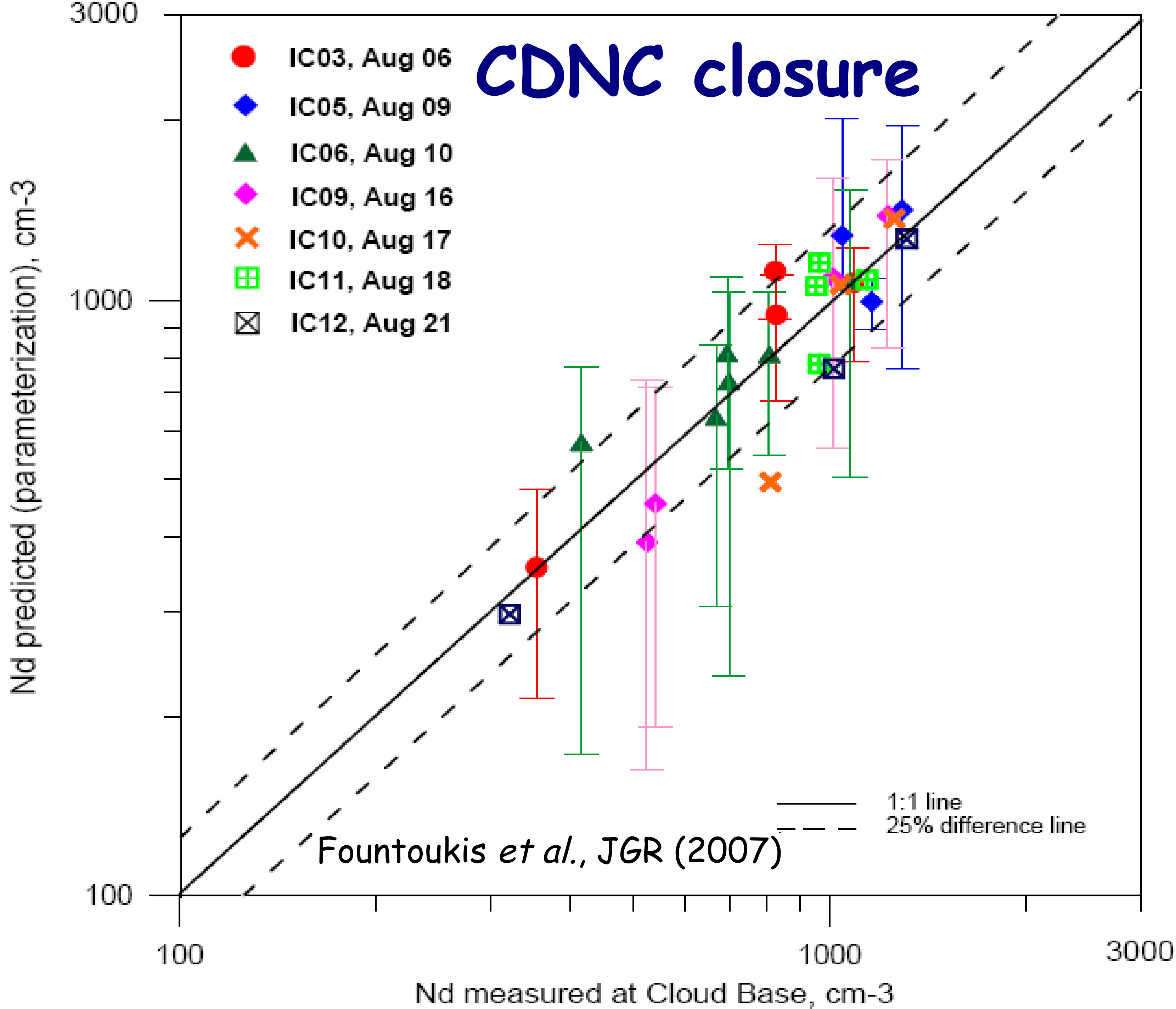
- 10^3-10^4 times *faster* than numerical cloud model.
- can treat very complex chemical composition.
- FAST formulations for lognormal and sectional aerosol is available

We evaluate this with the in-situ data.

CDNC closure during ICARTT (Aug.2004)

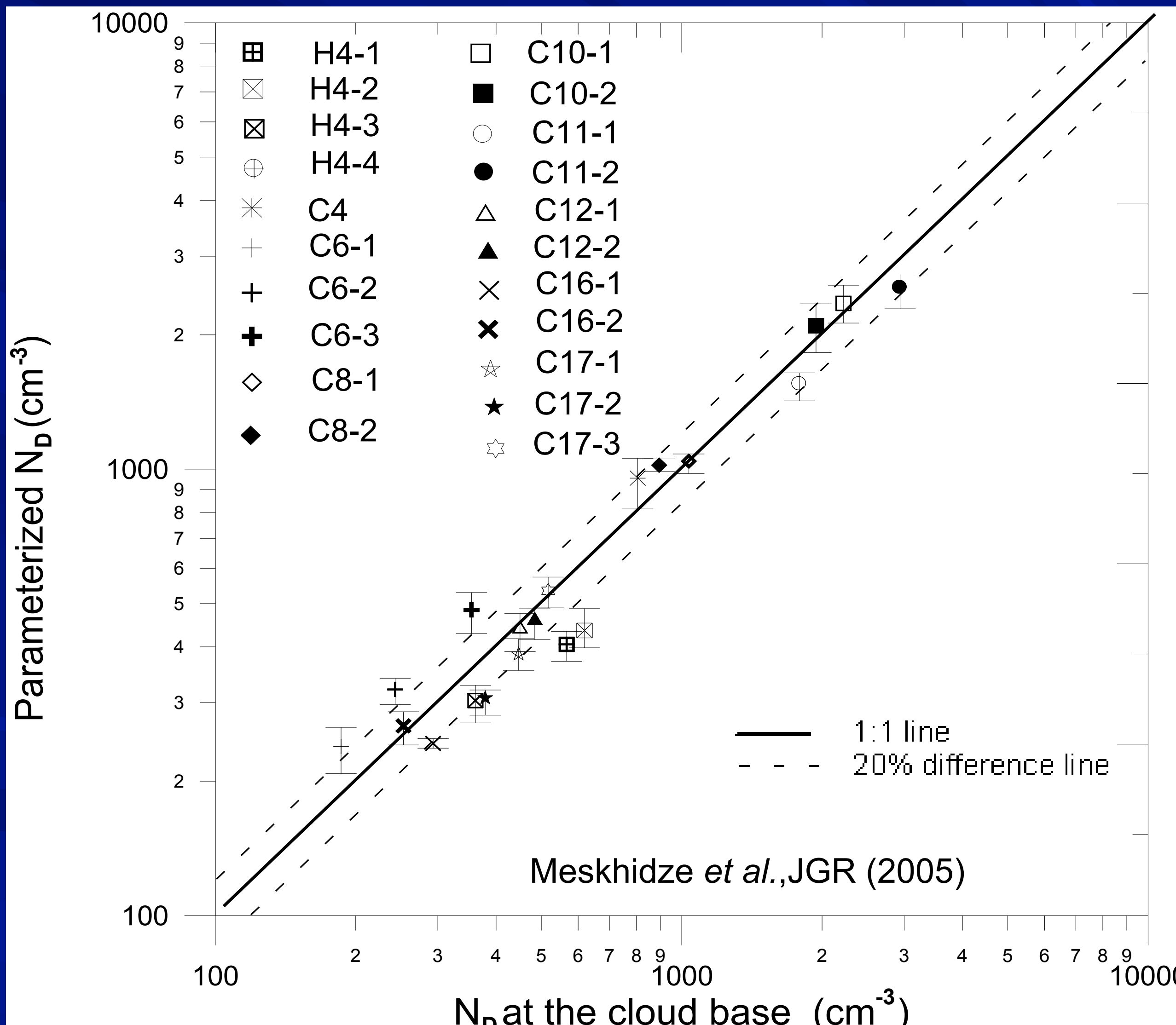


- Cumuliform and Stratiform clouds sampled
- Investigate the effect of power plant plumes on clouds

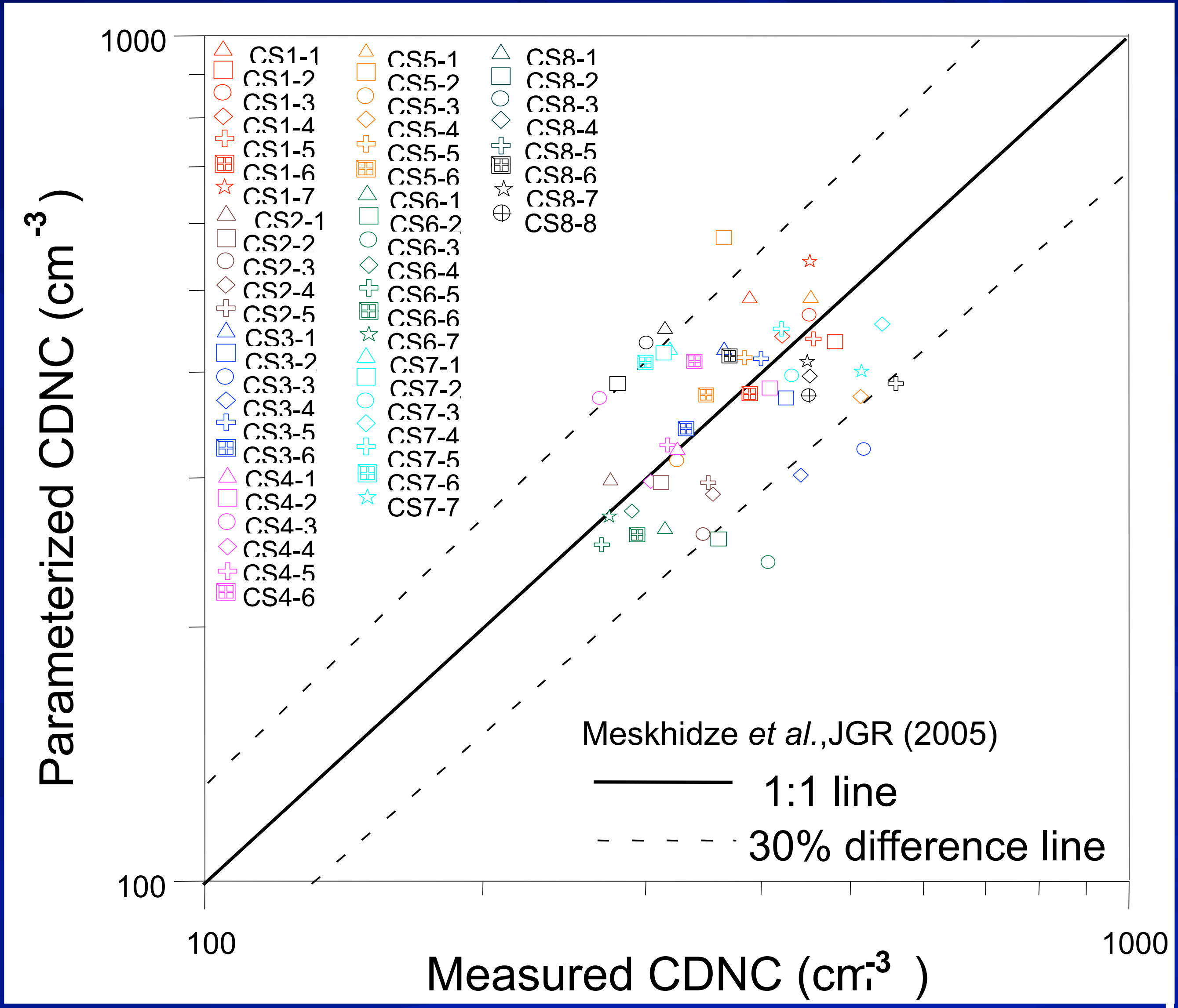


CDNC closure
IC03, Aug 06
IC05, Aug 09
IC06, Aug 10
IC09, Aug 16
IC10, Aug 17
IC11, Aug 18
IC12, Aug 21
1:1 line
25% difference line
Fountoukis et al., JGR (2007)
Nd predicted (parameterization), cm-3
Nd measured at Cloud Base, cm-3
3000
1000
100
100
1000
3000

CRYSTAL-FACE (2002)
Cumulus clouds



CSTRIPE (2003)
Coastal Stratocumulus

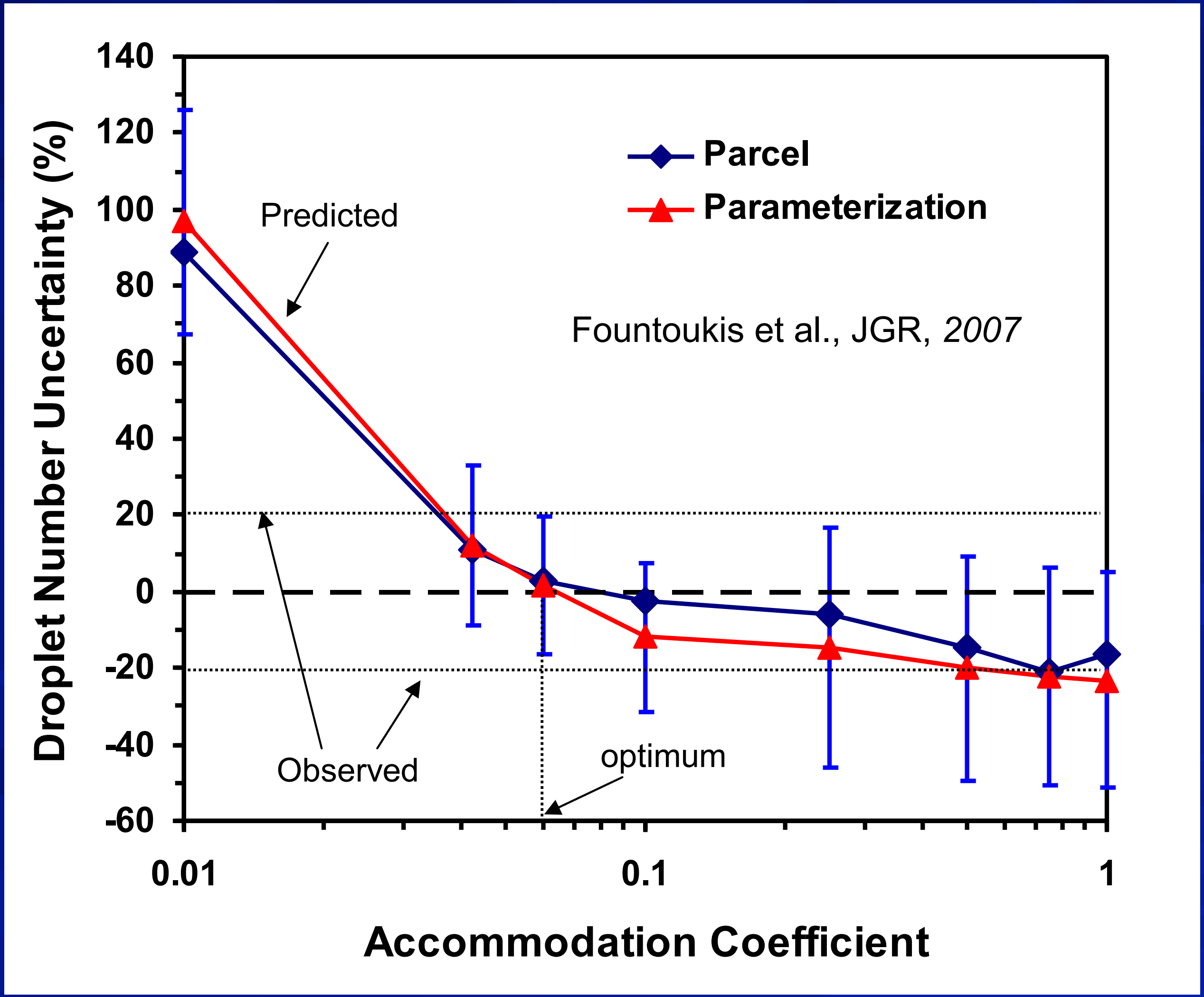


What we have learned from CDNC closure studies

- "Mechanistic" parameterizations do a good job of capturing droplet number for nearly adiabatic clouds and… when you know the input (they capture the physics).
- Gaussian PDF of updraft velocity is sufficient to capture average CDNC.
- In fact, the average updraft velocity does *equally well* (and is much faster) in predicting CDNC, compared to integrating over a PDF.
- CDNC closure studies also can be used to infer a range of droplet growth kinetic parameters ("water vapor mass uptake coefficient").

Range of *a* inferred from *in-situ* droplet closure studies

CIRPAS Twin Otter



ICARTT (2004)

Optimum closure obtained for *a* between
0.03 – 1.0

Same range found in CSTRIPE, CRYSTAL-FACE and MASE studies

We' ll get back to this point later on

Issues of Parameterizations

- Highly idealized description of clouds. Most often they are adiabatic (few feedbacks)…
- They require information not currently found in most GCMs (cloud-base updraft velocity, aerosol chemical composition, etc.).
- Few processes are represented and are largely decoupled from other processes or interact at the "wrong scale" (e.g., dynamics, entrainment and autoconversion/drizzle)
- Very difficult to address… but not impossible.

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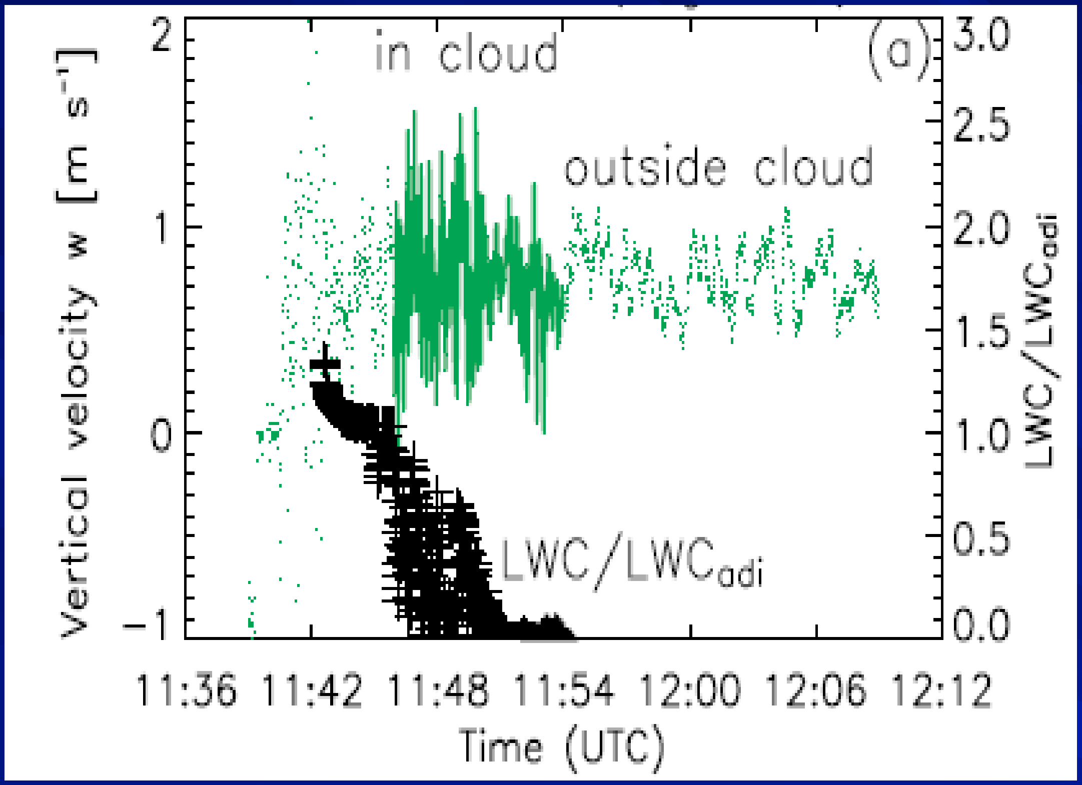
Real Clouds are not Adiabatic



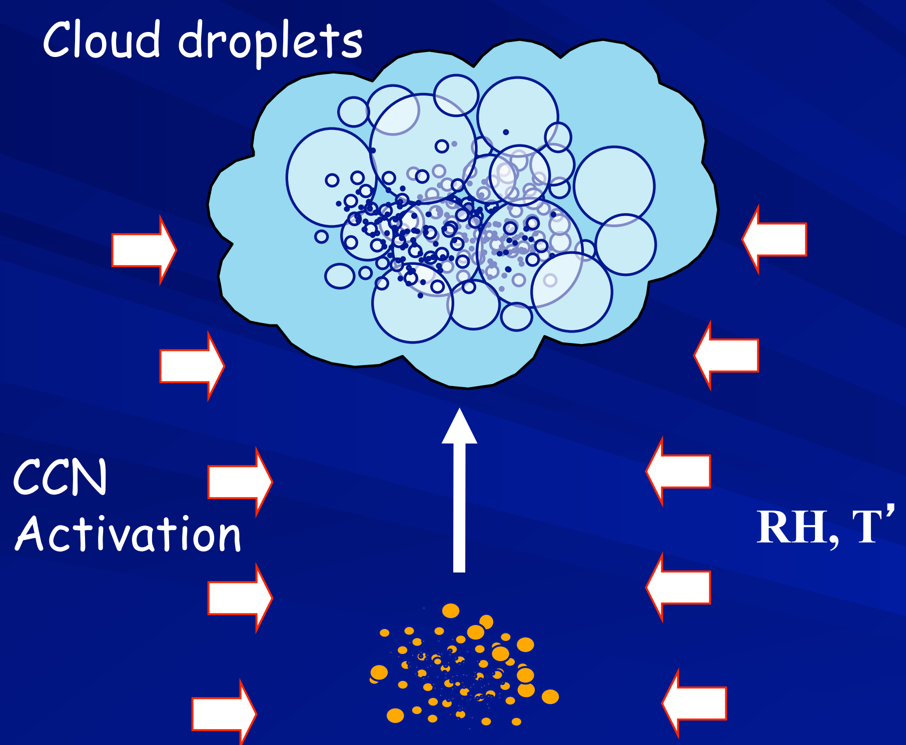
Peng, Y. et al. (2005). J. Geophys. Res., 110, D21213

- Entrainment of air into cloudy parcels decreases cloud droplet number relative to adiabatic conditions
- In-situ observations often show that the liquid water content measured is lower than expected by adiabaticity.

Neglecting entrainment may lead to an overestimation of in-cloud droplet number biasing indirect effect assessments

We need to include entrainment in the parameterizations

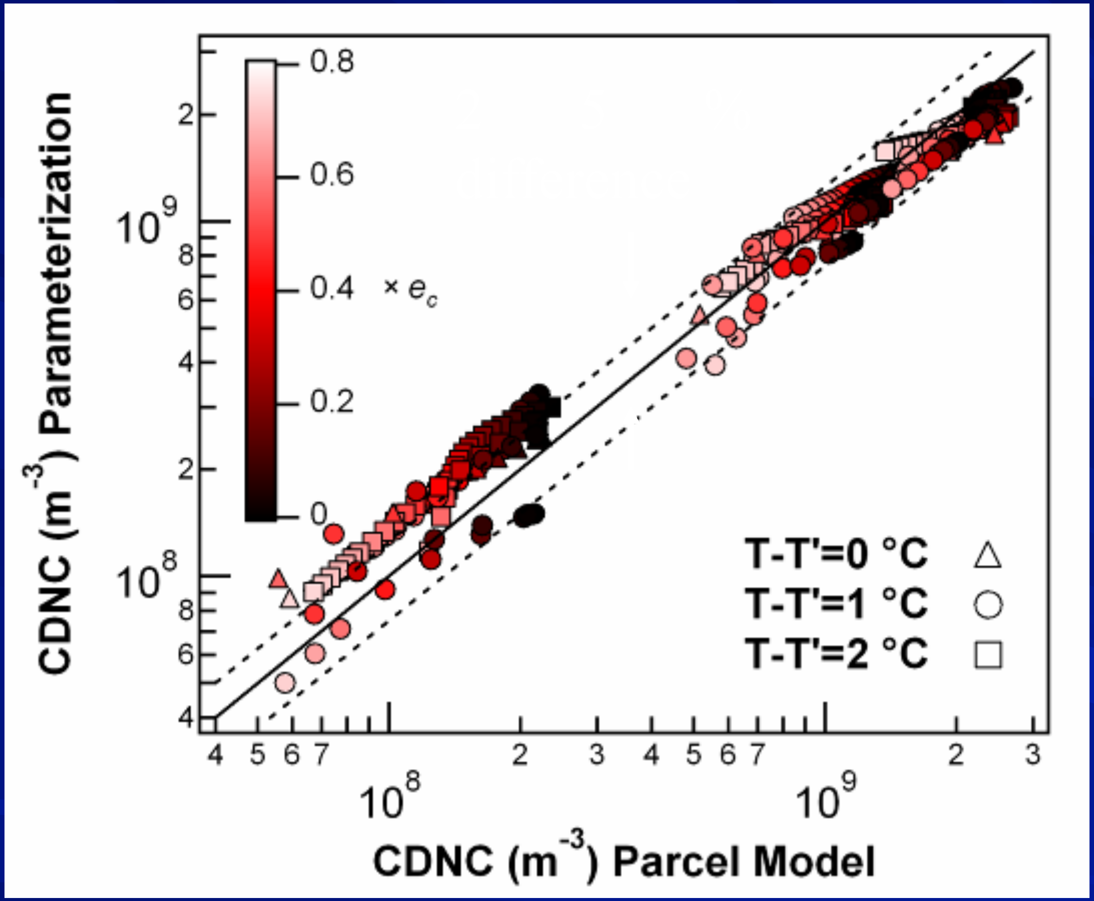
Barahona and Nenes (2007)
Droplet formation in entraining clouds



- Cloud droplet formation is parameterized by integrating conservation principles in an ascending entraining air parcel.
- 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)$^{-1}$(m ascent)$^{-1}$

- The formulation is the first of its kind and can treat all the chemical complexities of organics (which we will talk about in a bit).
- Formulations available for either lognormal or sectional aerosol.

Entraining Parameterization vs. parcel model



- Comparison with detailed numerical model.
- Parameterization closely follows the parcel model
- Mean relative error ~3%.
- 10^4 times faster than numerical parcel model.

V=0.1,1.0, and 5.0 ms^{-1}. *T-T'*=0,1,2 °C. *RH*=60, 70, 80, 90 %. Background aerosol. 2000 simulations.

Barahona and Nenes, (2007)

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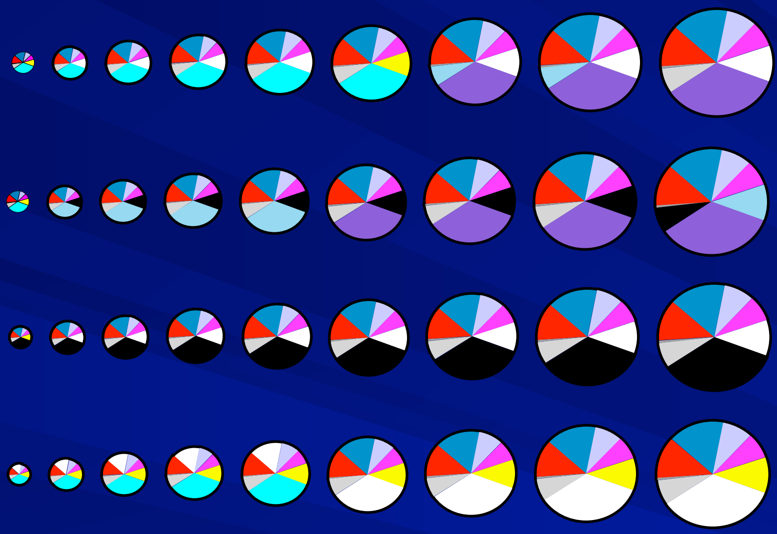
Aerosol Problem: Vast Complexity



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)

Predicting CCN concentrations is a convolution of size distribution and chemical composition information.

CCN activity of particles is a strong function ($\sim d^{-3/2}$) of aerosol dry size and (a weaker but important) function of chemical composition ($\sim$ salt fraction^{-1}).

Aerosol Description: Complexity range

The … headache of organic species

- They can act as surfactants and facilitate cloud formation.
- They can affect hygroscopicity (add solute) and facilitate cloud formation.
- Oily films can form and delay cloud growth kinetics
- Some effects are not additive.
- Very difficult to explicitly quantify in any kind of model.

The treatment of the aerosol-CCN link is not trivial at all.

CCN: Looking at what's important

How well do we understand the aerosol-CCN link?

What is the level of aerosol complexity required to "get things right"?

How much "inherent" indirect effect uncertainty is associated with different treatments of complexity?

Use in-situ data to study the aerosol-CCN link:

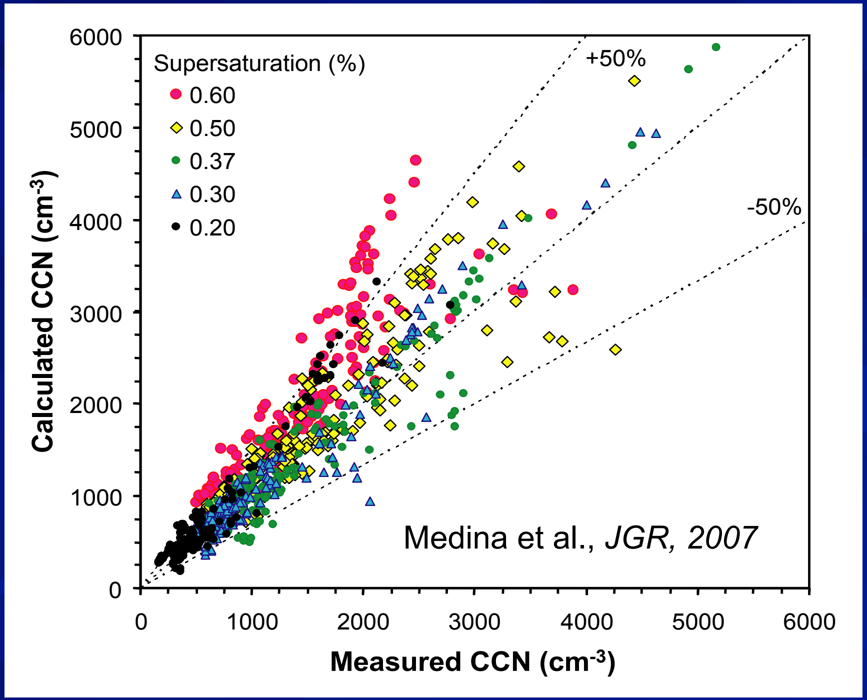
- Creative use of CCN measurements to "constrain" what the most complex aspects of aerosol (mixing state and organics) are.

- Quantify the uncertainty in CCN and droplet growth kinetics associated with assumptions & simplifications.

Do we understand the aerosol-CCN link?

Test of theory: “CCN Closure Study”

CCN MEASUREMENTS ⟷ CCN PREDICTIONS



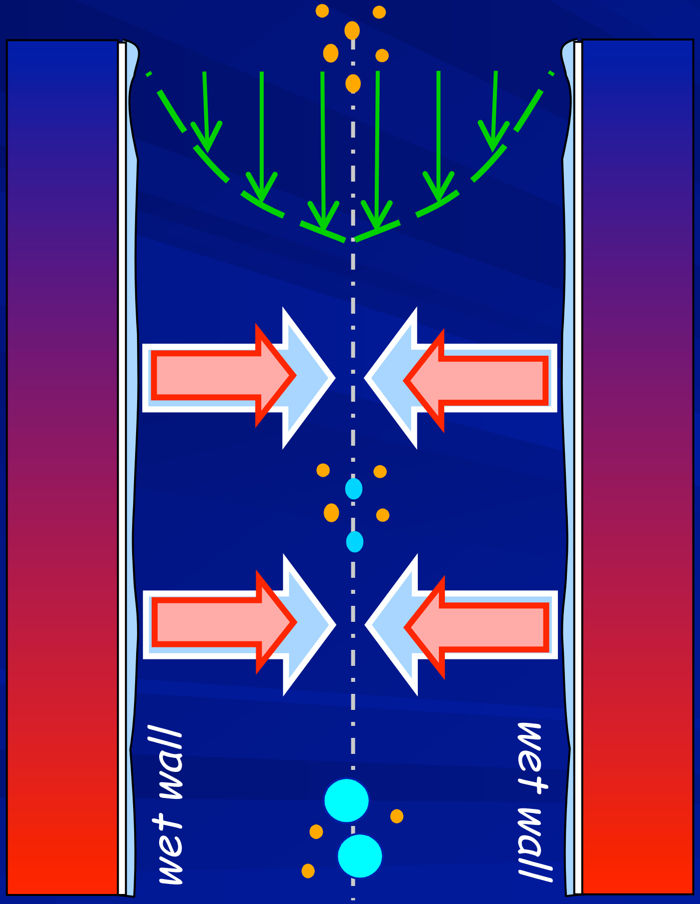
How is it done?

- Measure aerosol size distribution and composition.
- Introduce this information Köhler theory and predict CCN concentrations.
- Compare with measured CCN over a supersaturation range and assess closure.

CCN closure studies going on since the 70’s.
Advances in instrumentation have really overcome limitations

Measuring CCN: a key source of data

Goal: Generate supersaturation, expose CCN to it and count how many droplets form.



Continuous Flow Streamwise Thermal Gradient Chamber

Metallic cylinder with walls wet. Apply T gradient, and flow air.

- Wall saturated with H_2O.
- H_2O diffuses more quickly than heat and arrives at centerline first.
- The flow is supersaturated with water vapor at the centerline.
- Flowing aerosol at center would activate some into droplets.

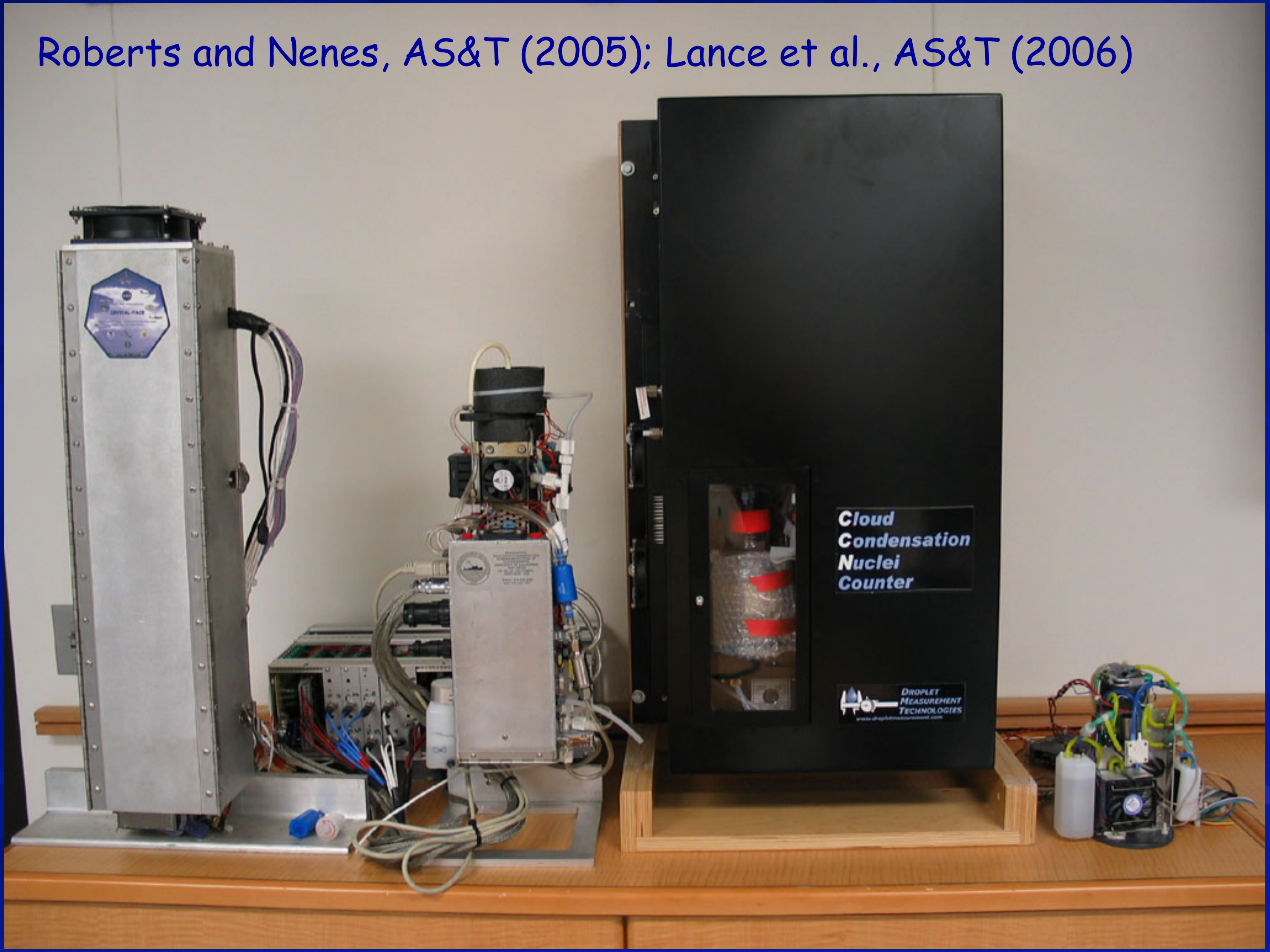
Count the **concentration** and **size** of droplets that form with a 1 s resolution.

Roberts and Nenes (2005), Patent pending

Development of Streamwise TG Chamber

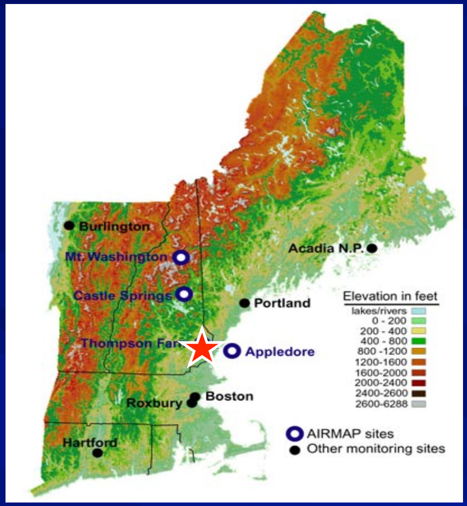
Roberts and Nenes, AS&T (2005); Lance et al., AS&T (2006)

scale = 1 m



1st version	2nd version	DMT	mini version
April 2002	January 2003	July 2004	August 2006

An example of a CCN closure



AIRMAP Thompson Farm site

- Located in Durham, New Hampshire
- Measurements done during ICARTT 2004
- Air quality measurements are performed on air sampled from the top of a 40 foot tower.

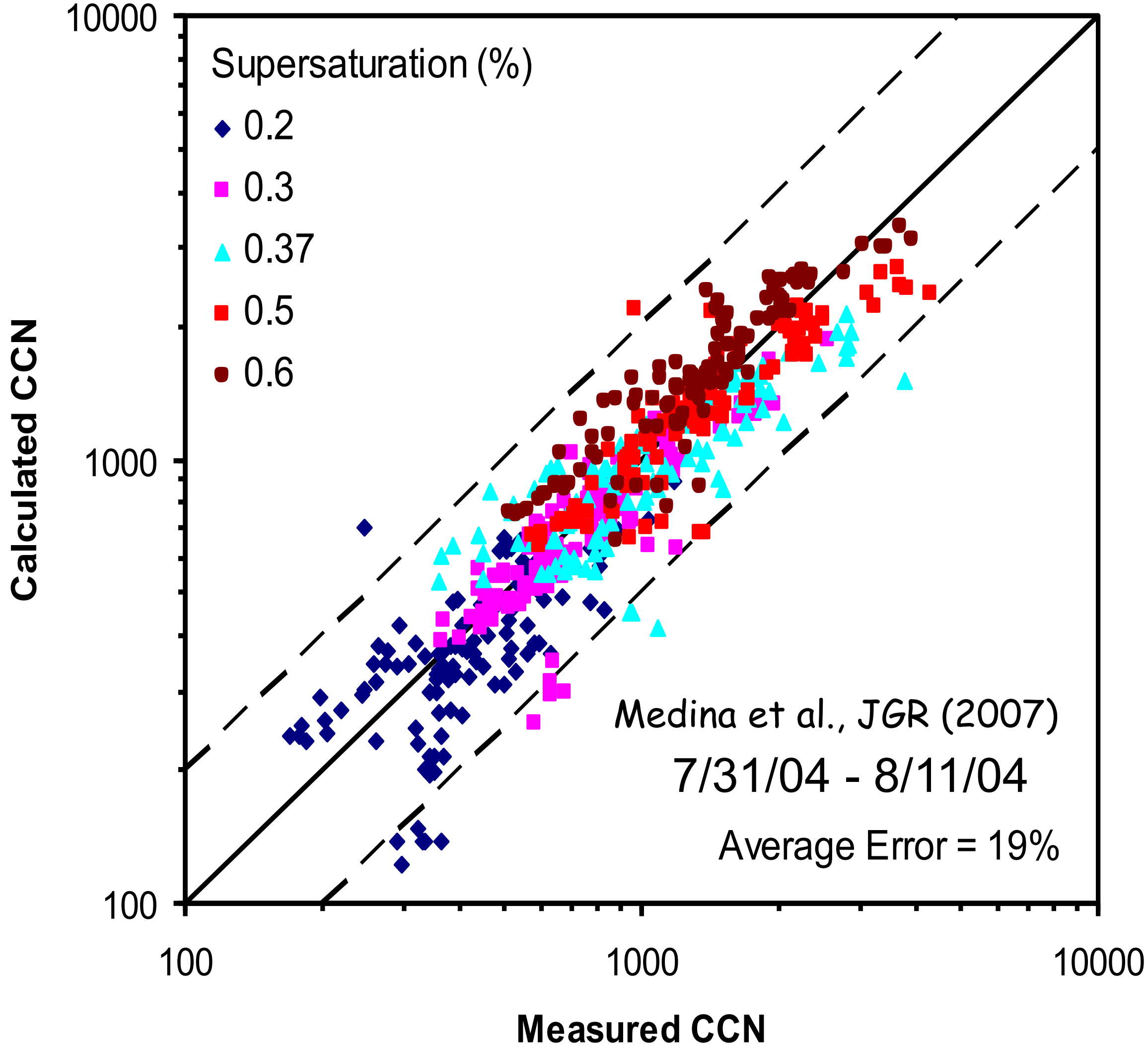
Two DMT CCN counters (Roberts and Nenes, AST, 2005; Lance et al., AST, 2006)

TSI SMPS, for size distribution

Aerodyne AMS, for chemical composition

2 weeks of aerosol and CCN data (0.2 - 0.6 % supersaturation)

CCN Measurements: "Traditional" Closure



20% overprediction (average).

Assuming **uniform composition with size** rougly **doubles** the CCN prediction error.

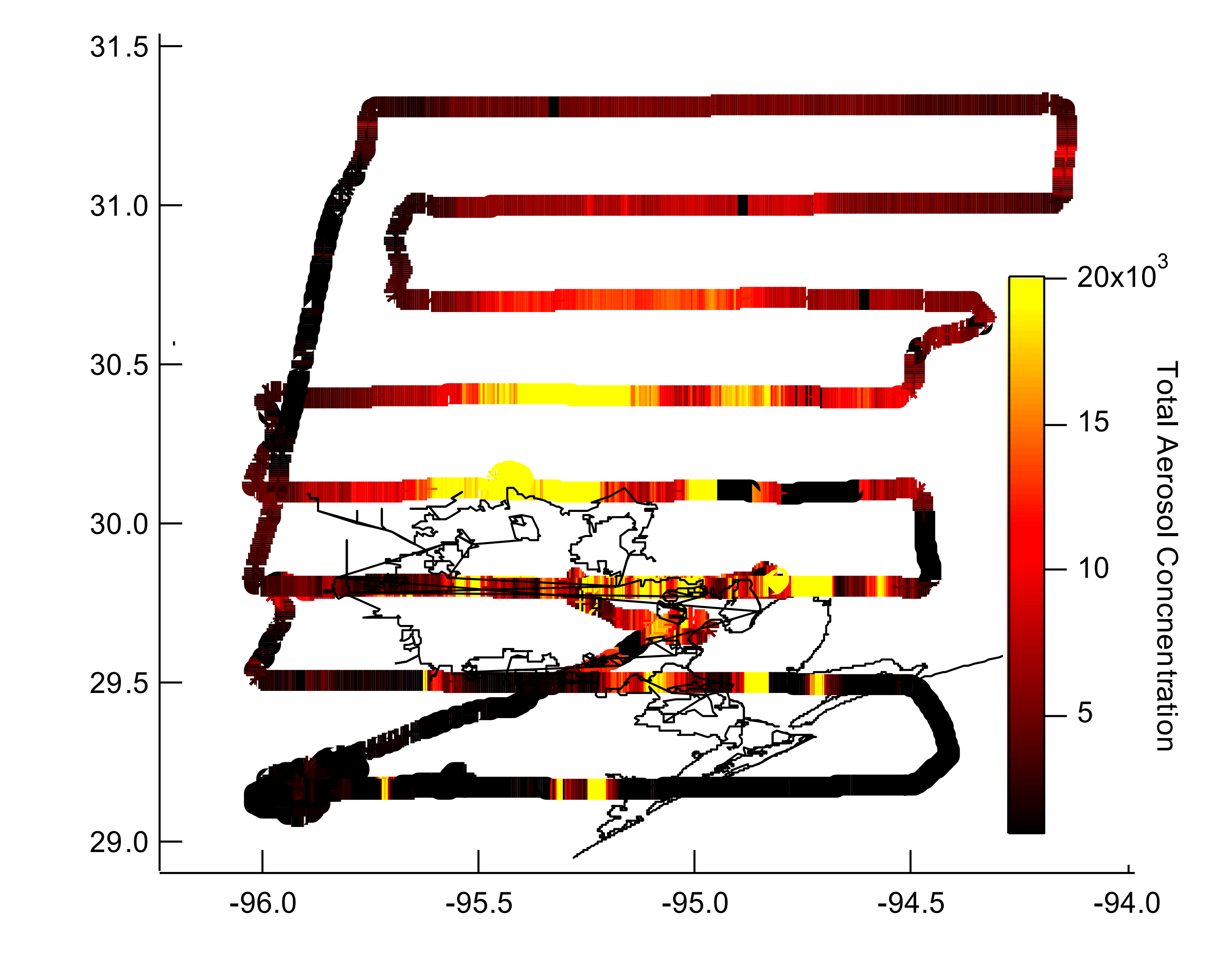
Introducing compreshensive composition into CCN calculation often gives very good CCN closure.

Larger-scale CCN variability (ageing)

How important is external mixing to "overall" CCN prediction

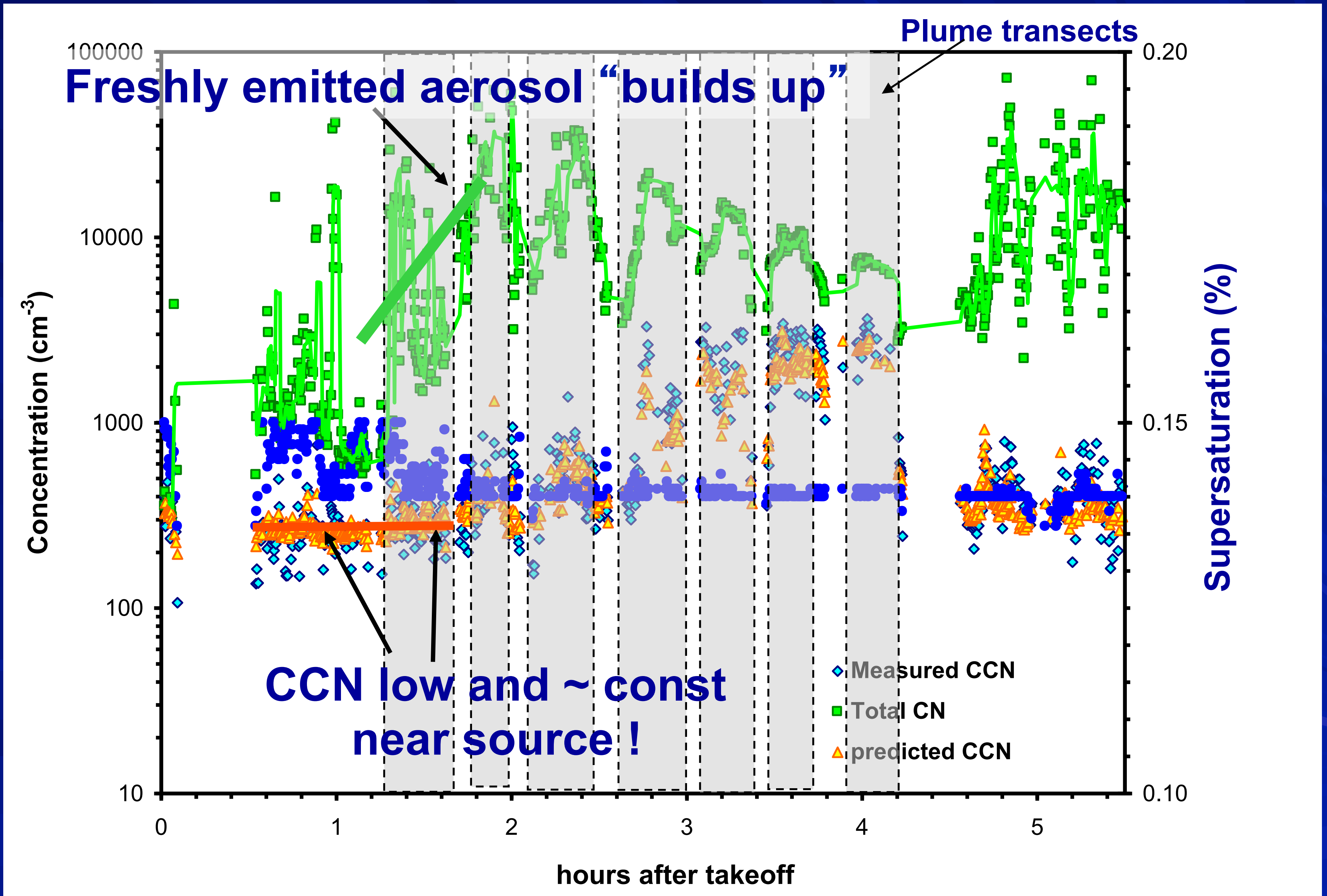
Look at CCN data from GoMACCs (Houston August, 2006) and MILAGRO (Mexico City, March 2006).

September 21: Houston Urban Plume Ageing

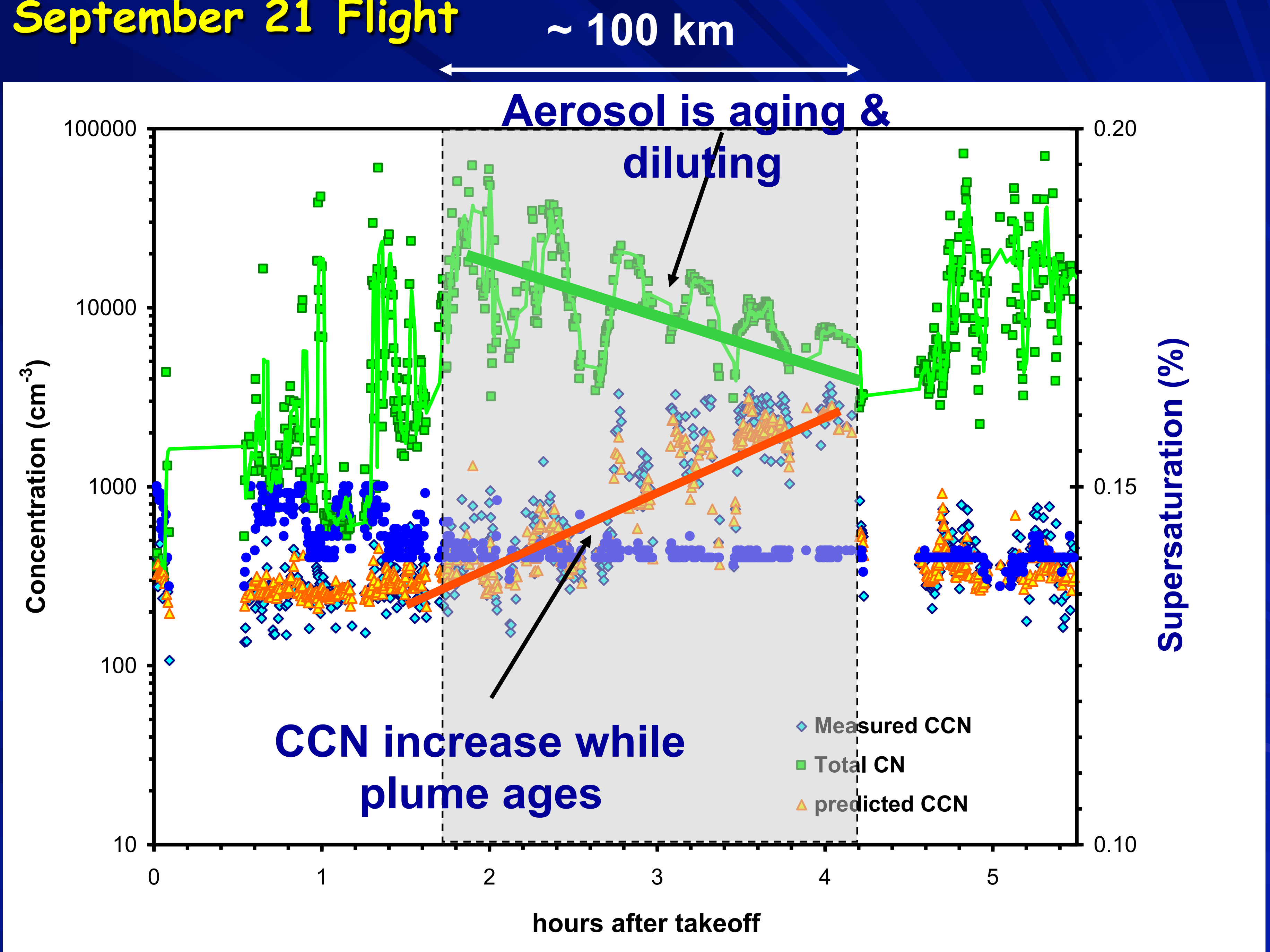


Wind, Flight progression

September 21 Flight



September 21 Flight



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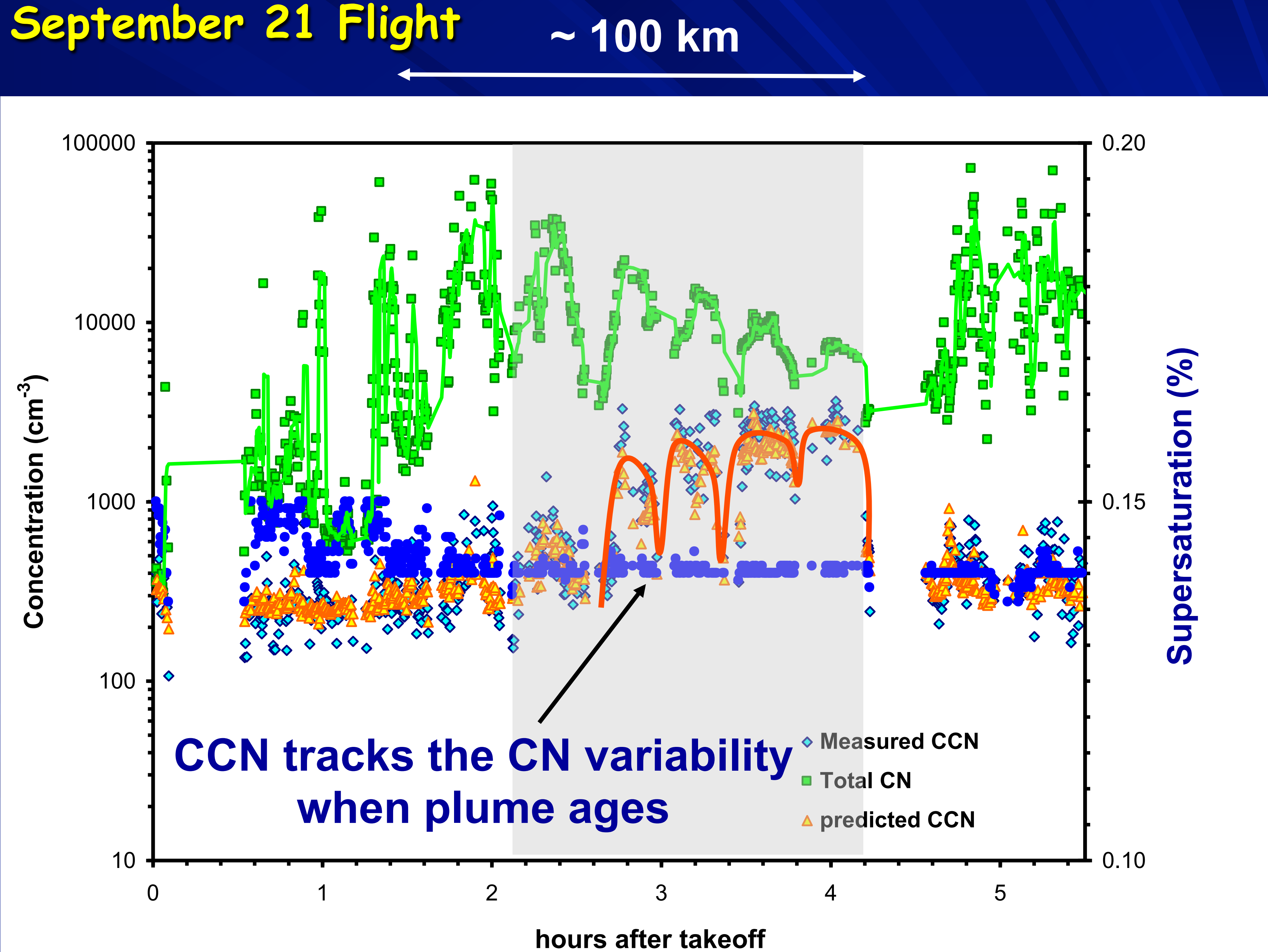
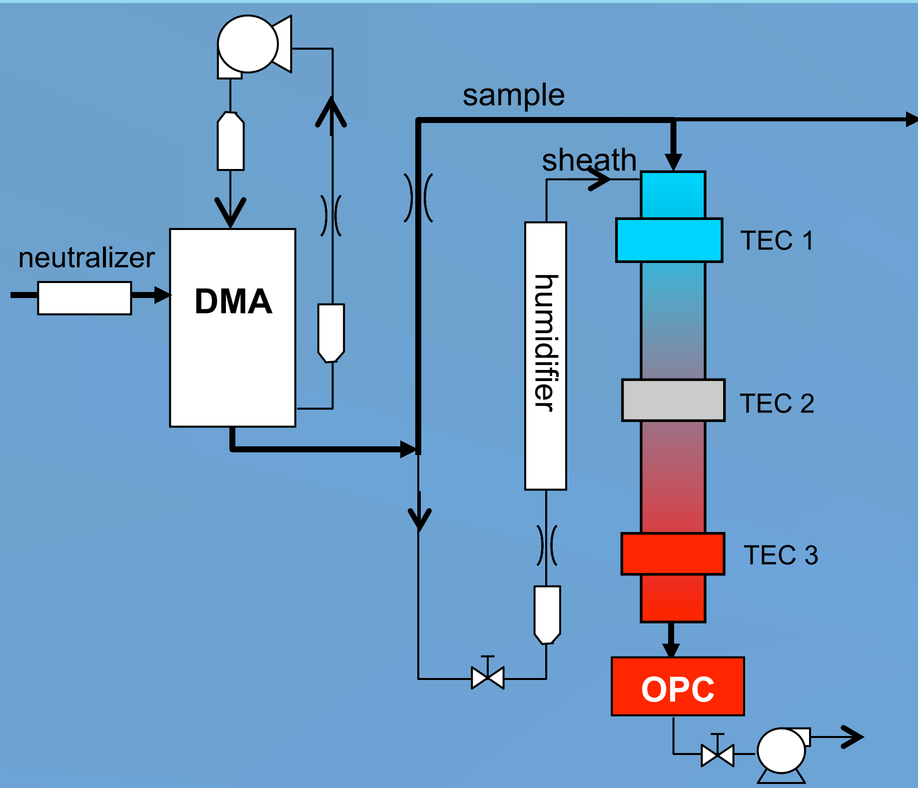


Some "take home" points

- CCN theory is adequate. Closure errors are from lack of information (size-resolved composition, mixing state).
- Aerosol variability close to source regions often does not correlate with CCN variability. CCN levels are often controlled by "background" (or "aged") concentrations.
- As plumes age, CCN increase and covary with total CN. This happens on a typical GCM grid size.
- … so external mixing considerations may be required only for GCM grid cells with large point sources of CCN (like megacities). Encouraging for large-scale models.
- Potential problem: Megacities are increasing in number (primarily in Asia), so the importance of external mixing (i.e. # of GCM cells) may be important in the future.

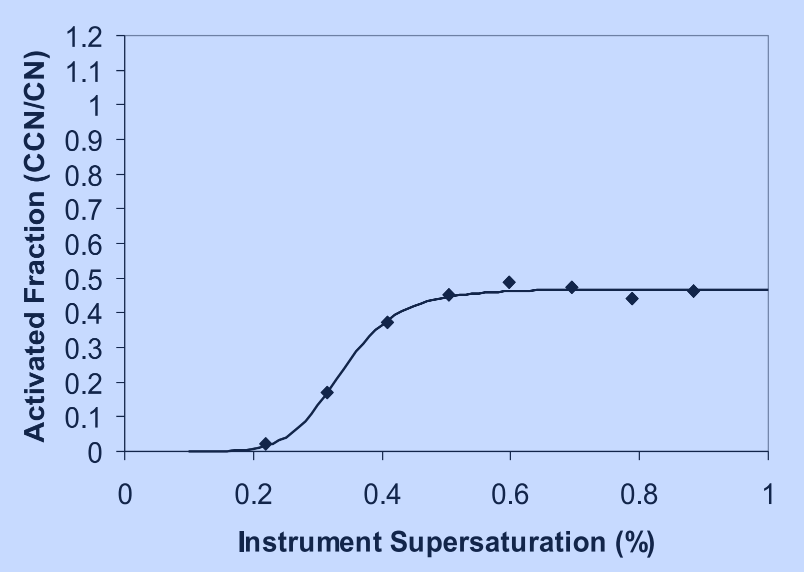
Get more out of CCN instrumentation: Size-resolved measurements

Measure CCN activity of aerosol with known diameter

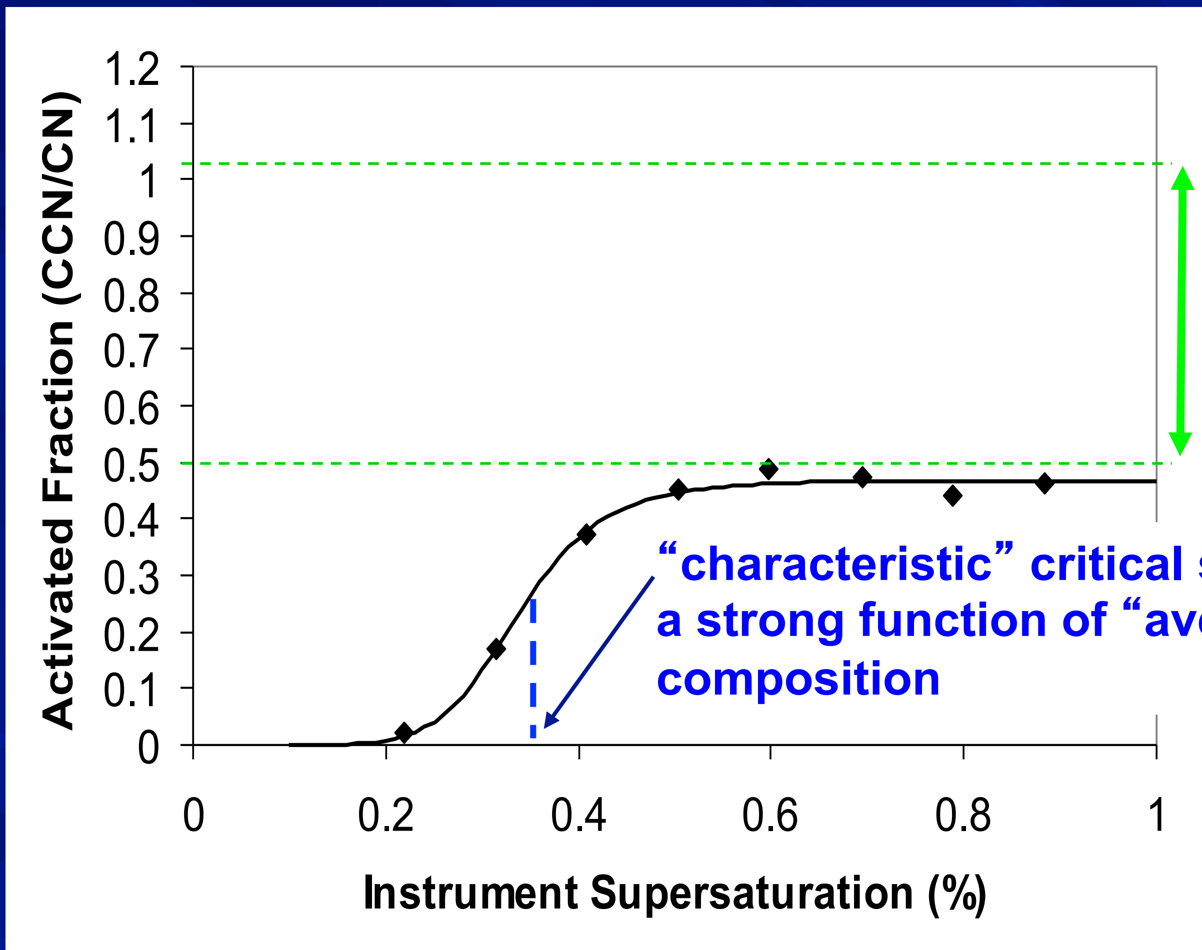


$$\text{Activated Fraction} = \frac{\text{CCN}}{\text{CN}}$$

Results: “activation curves”



What the Activation Curves tell us

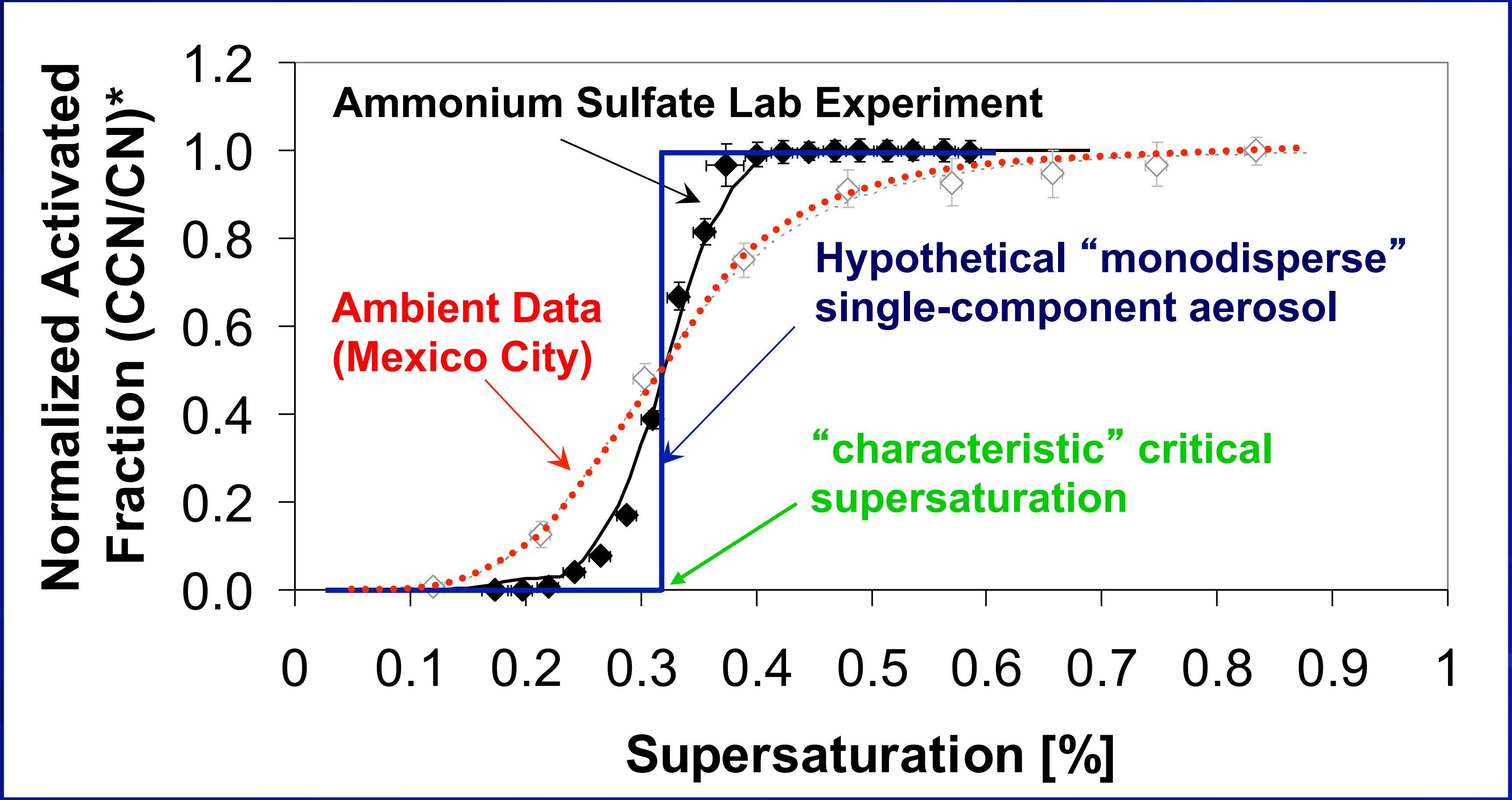


Fraction of non-hygroscopic externally mixed particles

“characteristic” critical supersaturation: a strong function of “average” aerosol composition

What the Activation Curves tell us

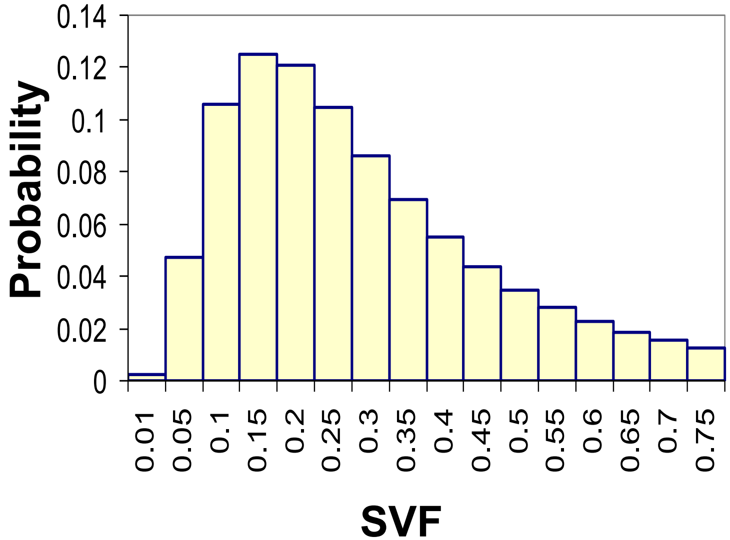
Slope has a wealth of information as well.



Ambient Data Is Much Broader Because Chemistry varies alot

First Approach; slope is from chemistry alone

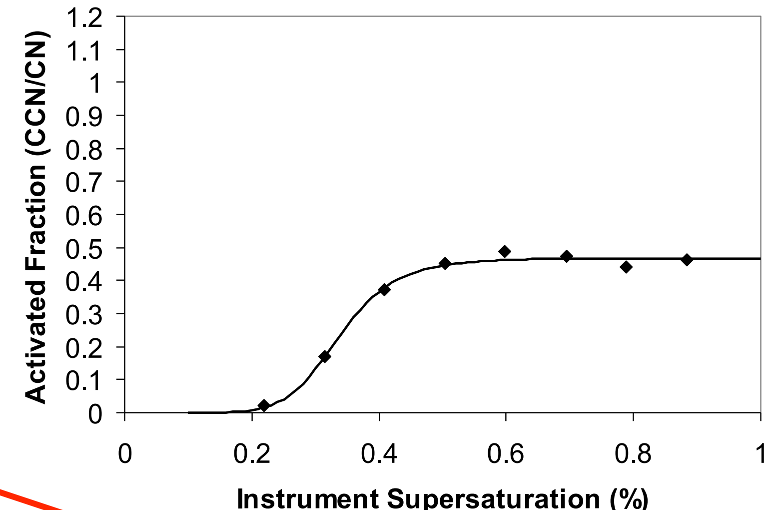
SVF = **S**oluble **V**olume **F**raction



Köhler Theory

$$S_c = f(SVF, d_p)$$

Sigmoidal fit



$$\frac{CCN}{CN} = \left(\frac{E^*}{1 + \left(S / S^* \right)^C} \right)$$

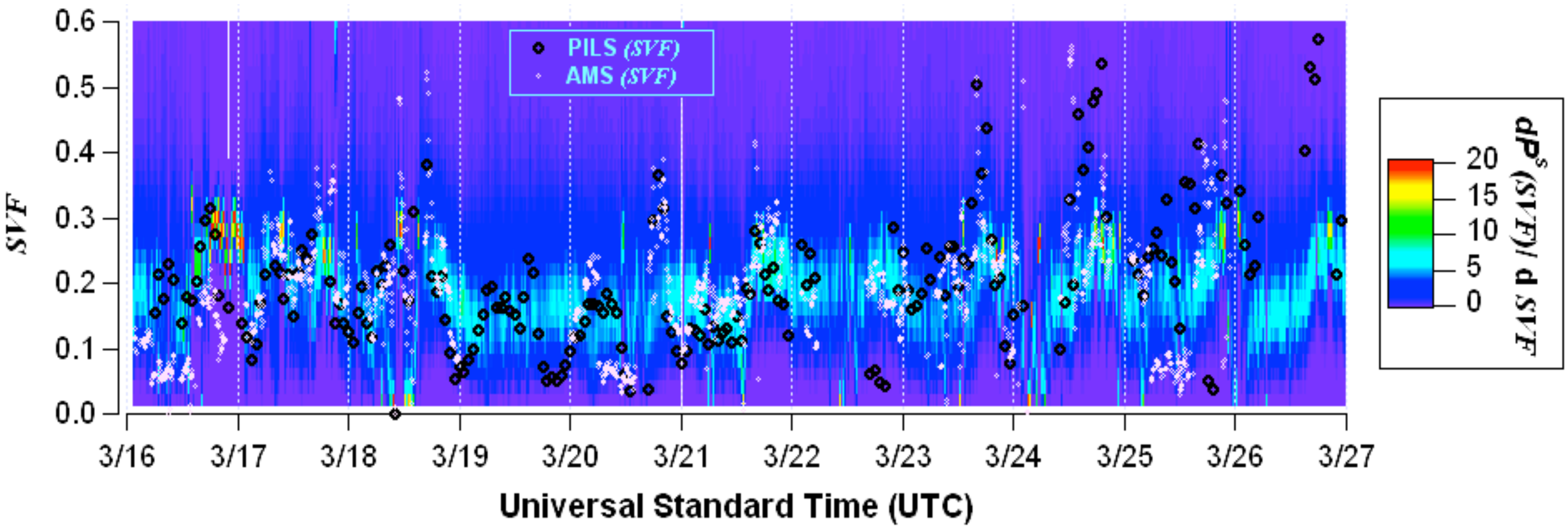
The slope of the activation curve directly translates to the width of the chemical distribution

What we get: PDF of composition as a function of particle size every few minutes.

This is a complete characterization of CCN 'mixing state'.

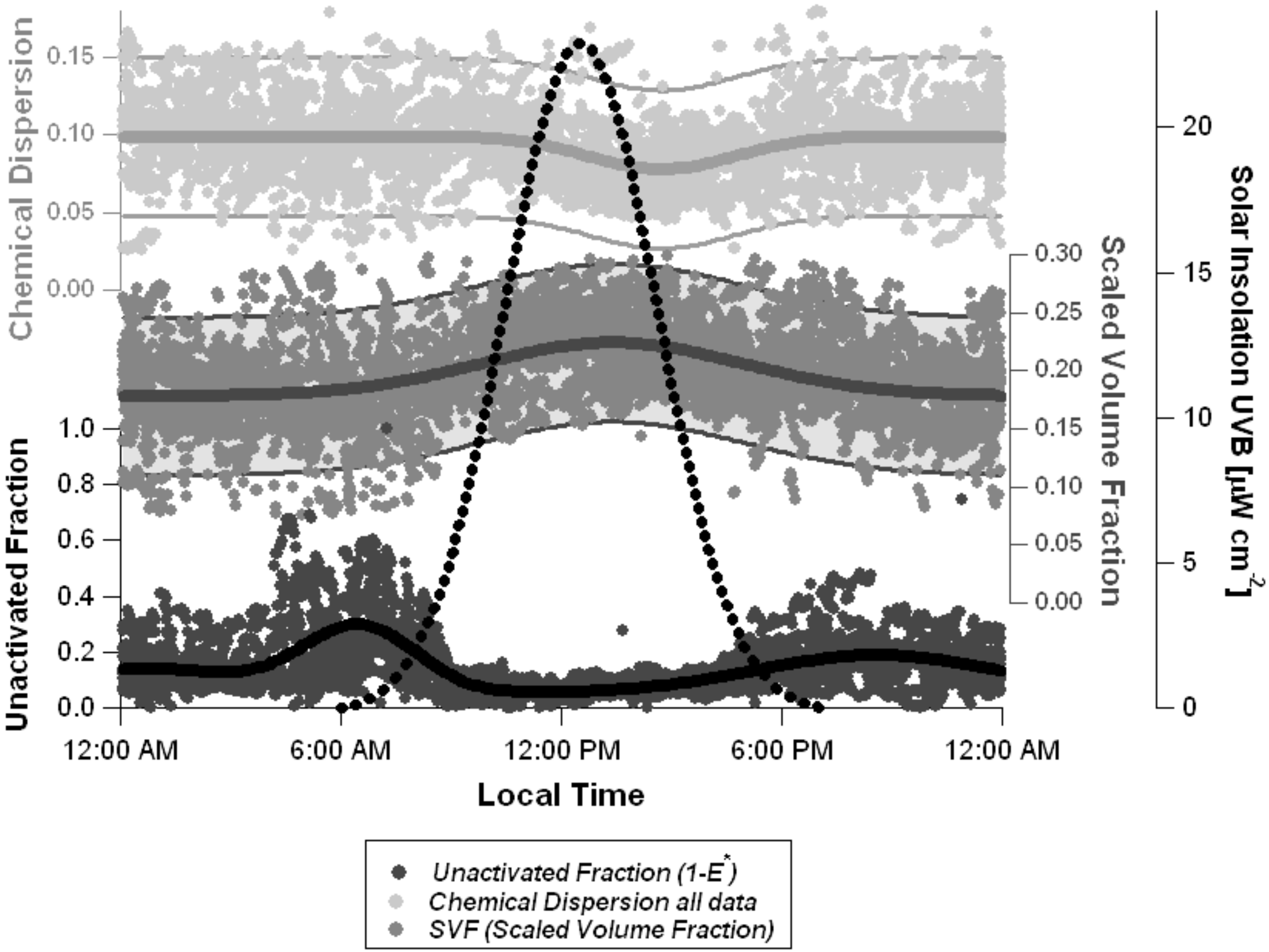
“Chemical Closure”
inferred vs measured soluble fraction

Look at CCN data from MILAGRO (Mexico City, March 2006).
Compare against “bulk” composition measurements



The average distribution of soluble fraction agrees very well with measurements. Our measurements give what’s “important” for CCN mixing state.

Diurnal Variability in CCN mixing state (Mexico City)



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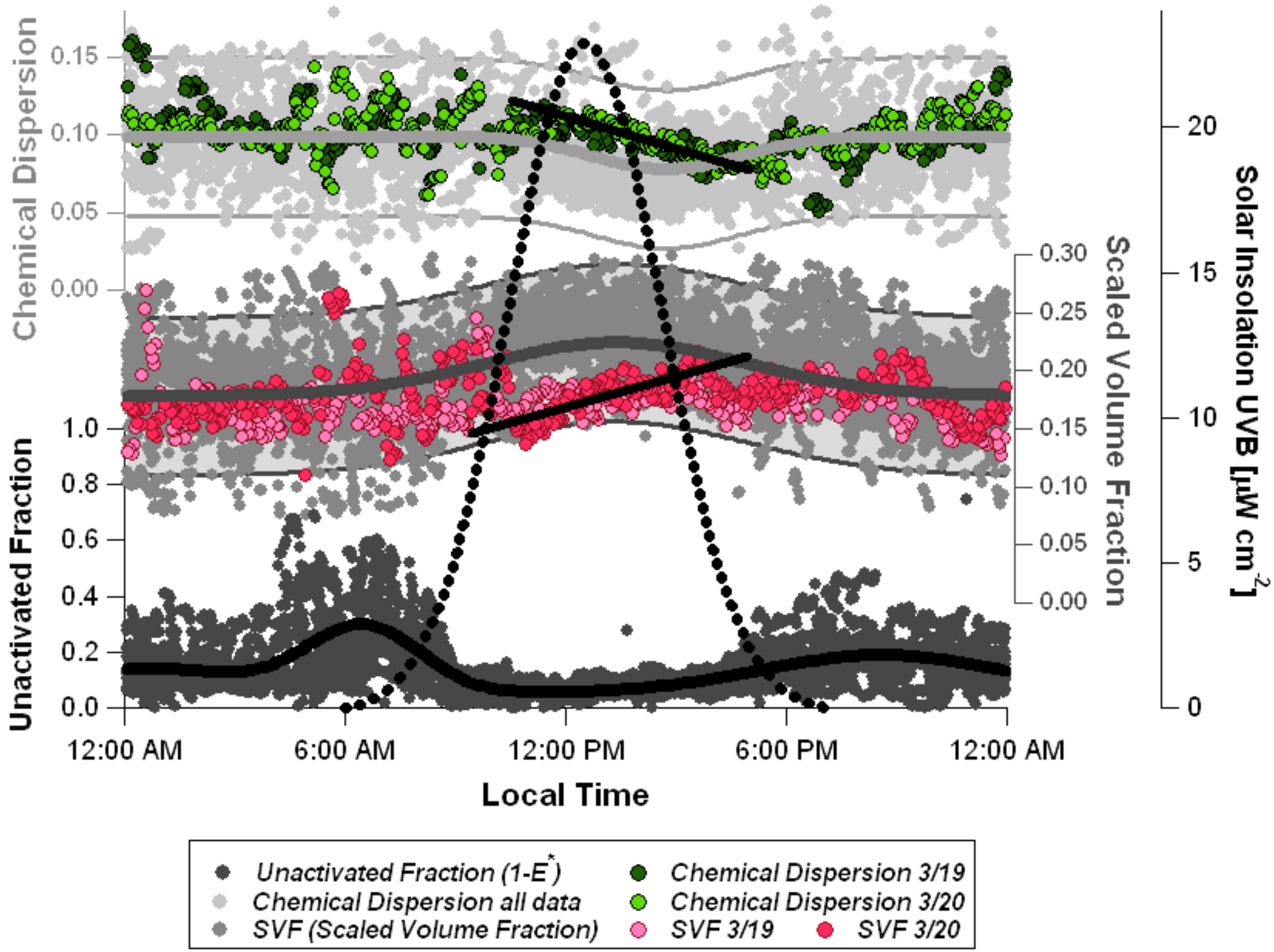


Some "take home" points

- Local signatures of aerosol sources on CCN mixing state largely disappear when the sun comes up (Photochemistry? Boundary layer mixing? New particle formation and growth? Cloud processing?).

- … so external mixing considerations may be required only for GCM grid cells with large point sources of CCN (like megacities). Encouraging for large-scale models, which really need simple but effective ways to predict CCN.

- Potential problem: Megacities are increasing in number (primarily in Asia), so the importance of external mixing (i.e. # of GCM cells) may be important in the future.

CCN: Looking at what's important

- The problem of CCN prediction in global models is not "hopeless". Good news.
- Size distribution plus assuming a uniform mixture of sulfate + insoluble captures most of the CCN variability (on average, to within 20-25% but often larger than that).
- Scatter and error is because we do not consider mixing state and impact of organics on CCN activity.

How important is this kind of uncertainty?

The term "good closure" is often used, but how "good" is it really?

Due to time limitations… let's get to the point.

CCN predictions: "take home" points

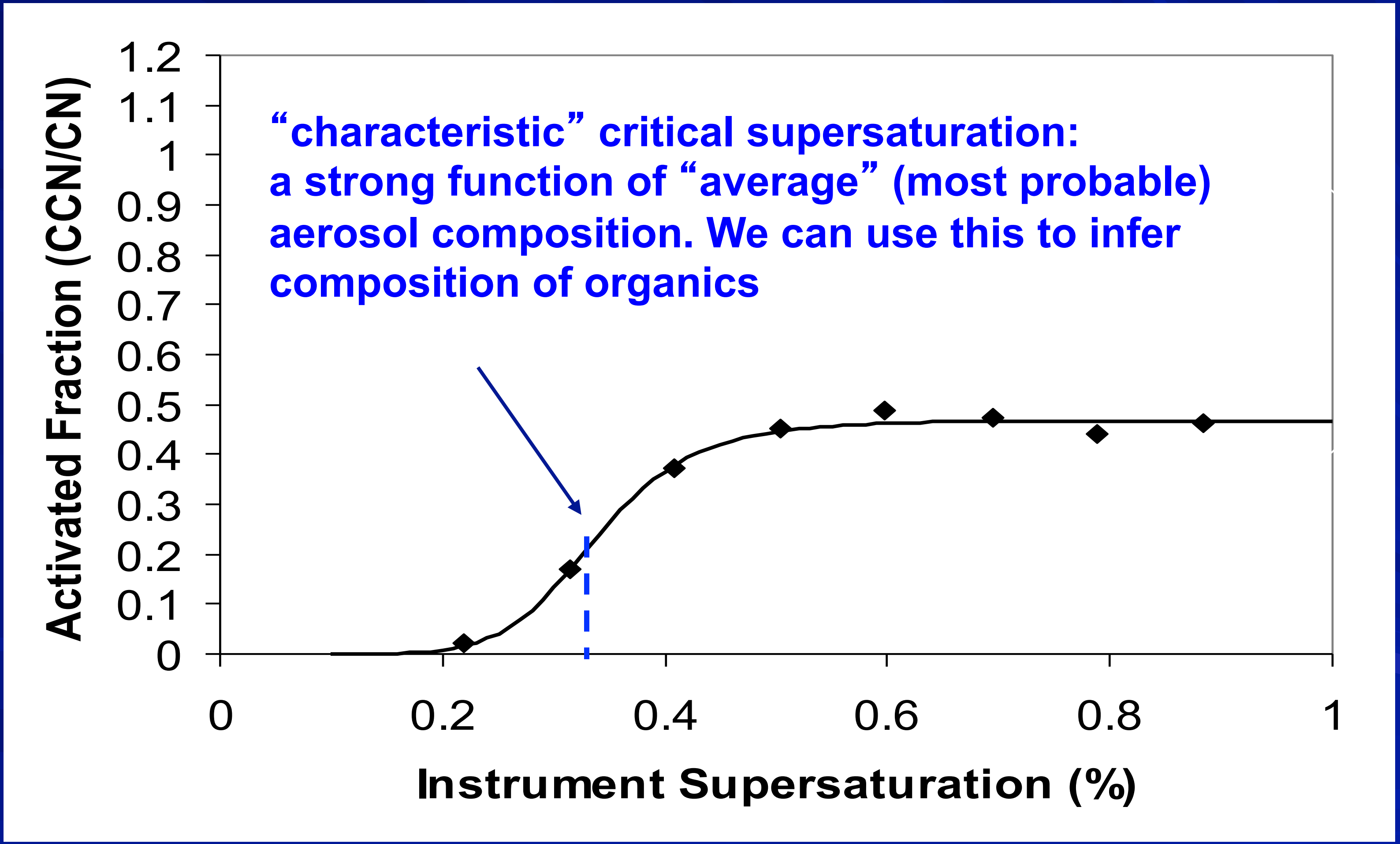
- Resolving size distribution and a uniform mixture of sulfate + insoluble can translate to 50% uncertainty in indirect forcing.
- The effect on precipitation can be equally large (or even larger because of nonlinearities).
- Even if size distribution were perfectly simulated, more **simplistic treatments of aerosol composition** imply even larger uncertainties in indirect effect. Size is **not** the only thing that matters in CCN calculations.
- Scatter and error is because we do not consider mixing state and impact of organics on CCN activity.

Organics & CCN: The Challenges

- Since CCN theory works well, we can use it to infer the impact of organics on droplet formation.
- We want "key" properties that can easily be considered in current parameterizations
- Desired information:
  - Average molar properties (molar volume, solubility)
  - Droplet growth kinetics
  - Chemical heterogeneity (mixing state) of aerosol
  - Surface tension depression

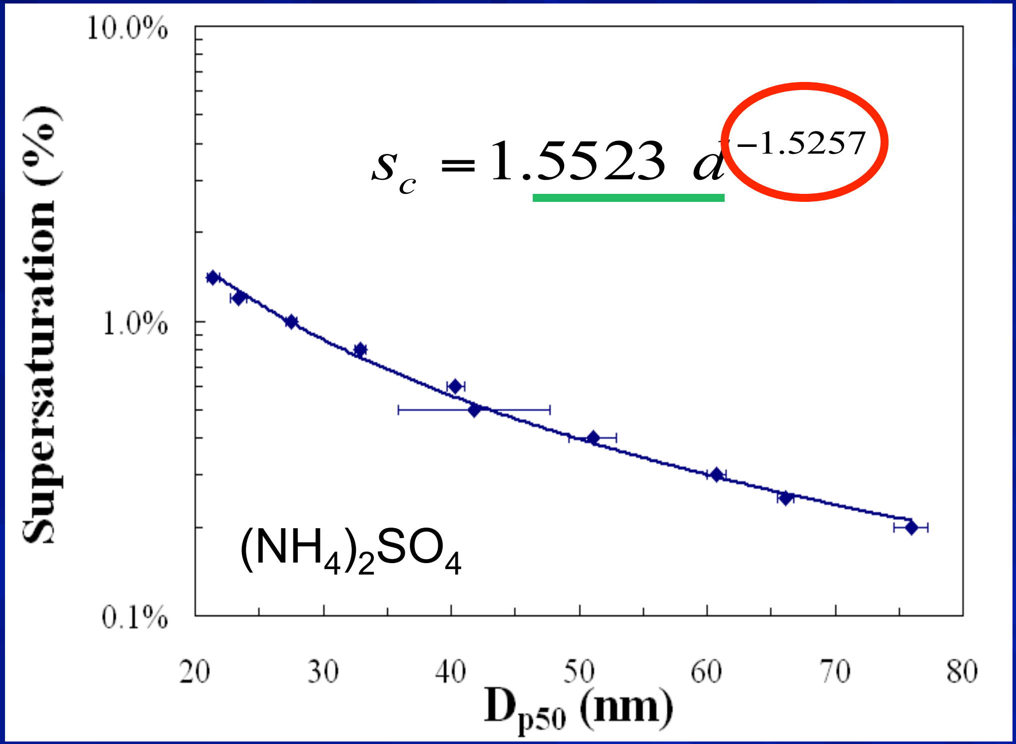
Once more, size-resolved CCN measurements can provide a key source of data

Go back to the Activation Curves



Inferring Molar Volume from CCN activity: Köhler Theory Analysis (KTA)

- Plot characteristic supersaturation as a function of dry particle size.
- Fit the measurements to a power law expression.
- Relate fitted coefficients to aerosol properties (e.g. molecular weight, solubility) by using Köhler theory:



$$S_c = \left(\frac{256 M_w \sigma}{27 RT \rho_w}\right)^{1/2} \left[\sum_i \left(\frac{\rho_w}{M_w}\right)\left(\frac{M_i}{\rho_i}\right)\frac{1}{\varepsilon_i \upsilon_i}\right]^{1/2} d^{-3/2} = \omega d^{-3/2}$$

Padró *et al.*, ACPD; Asa-Awuku *et al.*, ACPD

If we know the inorganic composition, we can "infer" the organic properties

$$\frac{M_{organic}}{\rho_{organic}} = \frac{\varepsilon_{organic}\upsilon_{organic}}{\frac{256}{27}\left(\frac{M_w}{\rho_w}\right)^2\left(\frac{1}{RT}\right)^3 \sigma^{-3}\omega^{-2} - \sum_{i\neq organic}\frac{\rho_i}{M_i}\varepsilon_i\upsilon_i}$$

Constants

Molar Volume
This is what you need to know about organics for CCN activity

Measured surface tension and CCN data

$$\varepsilon_i = \frac{m_i/\rho_i}{\sum_i m_i/\rho_i}$$

From IC/WSOC measurement

Method shown to work well for laboratory-generated aerosol (Padró *et al.*, ACPD) and SOA generated from ozonolysis of biogenic VOC (Asa-Awuku *et al.*, ACPD), even marine organic matter!

KTA: Major findings on soluble organics

Many "aged" soluble organics from a wide variety of sources have a 200-250 g mol^{-1} . For example:

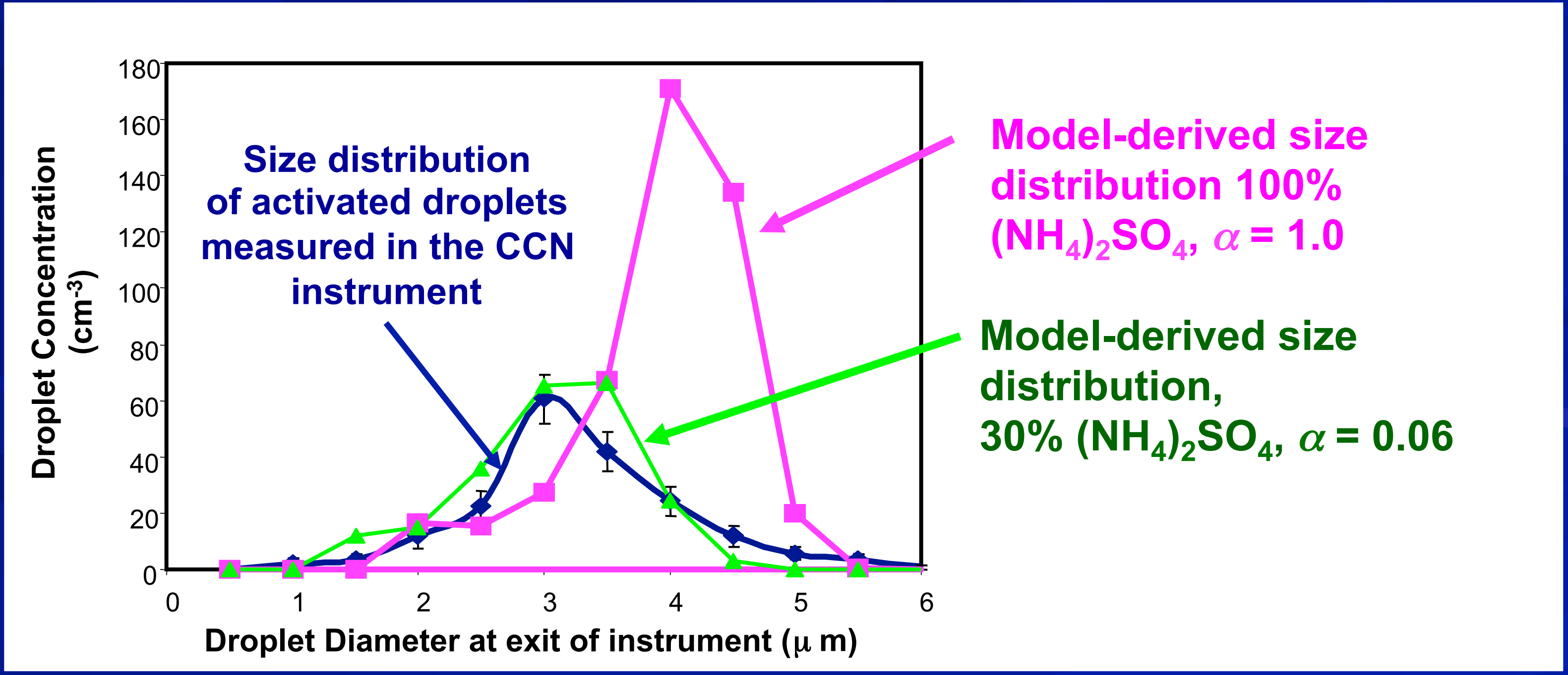
- Aged Mexico City aerosol from MILAGRO.
- Secondary Organic Aerosol from
  a-pinene and monoterpene oxidation (Engelhart et al., ACPD).
  Ozonolysis of Alkenes (Asa-Awuku et al., ACPD)
  Oleic Acid oxidation (Shilling et al., 2007)
- In-situ cloudwater samples collected aboard the CIRPAS Twin Otter during the MASE, GoMACCs field campaigns
- … and the list continues (e.g., hygroscopicity data from the work of Petters and Kreidenweis).

What varies mostly is not the "thermodynamic" properties of the complex organic "soup" but the fraction of soluble material… Complexity sometimes simplifies things for us.

Growth kinetics from CCN measurements

Size of activated droplets measured in the instrument.
The impact of composition on growth kinetics can be inferred:

- compare against the growth of $(NH_4)_2SO_4$ (thus giving a sense for the relative growth rates), and,
- use a model of the CCN instrument (Nenes et al., 2001), to parameterize measured growth kinetics in terms of the water vapor uptake coefficient, α.

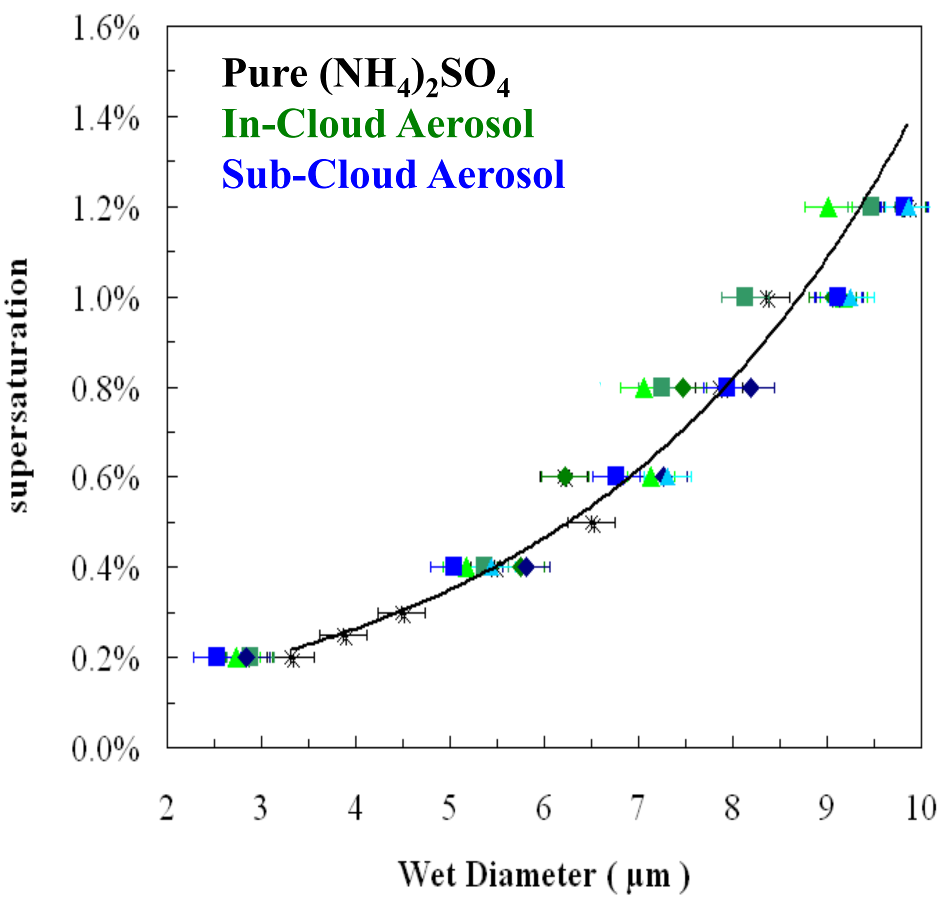


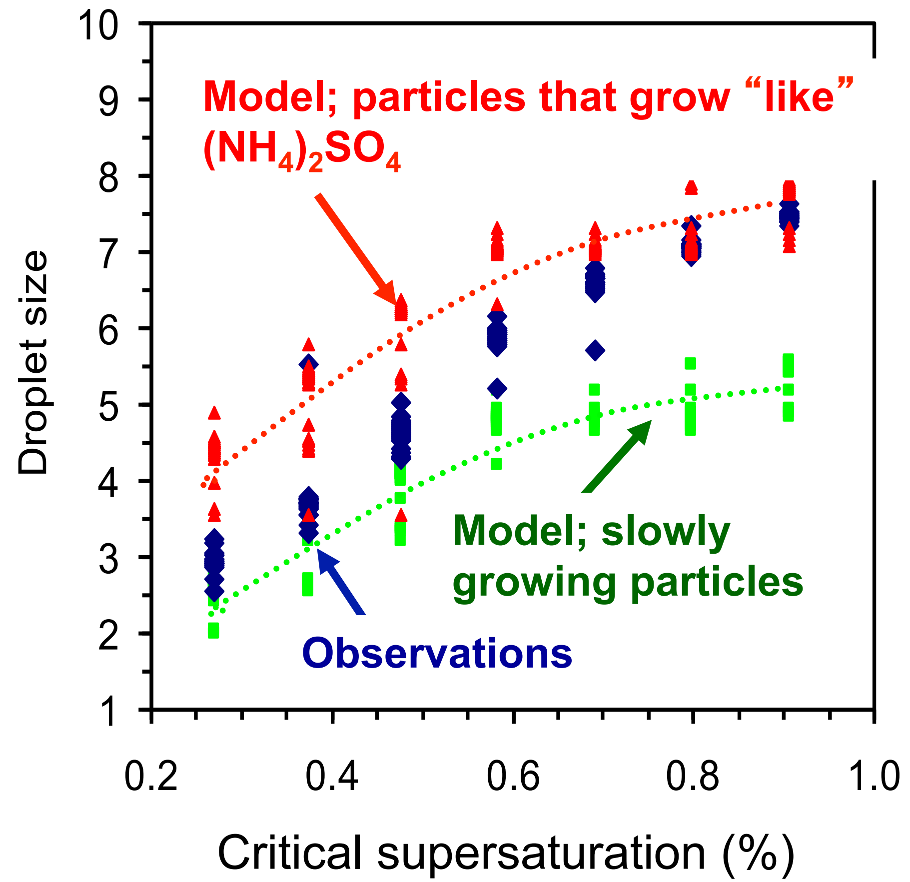
Roberts and Nenes (2005); Lance et al. (2006)

Do growth kinetics of CCN vary?

Measurements of droplet size in the CCN instrument

Marine Stratocumulus (MASE) All CCN grow alike	Urban (Mexico City, MILAGRO) CCN don't grow alike.

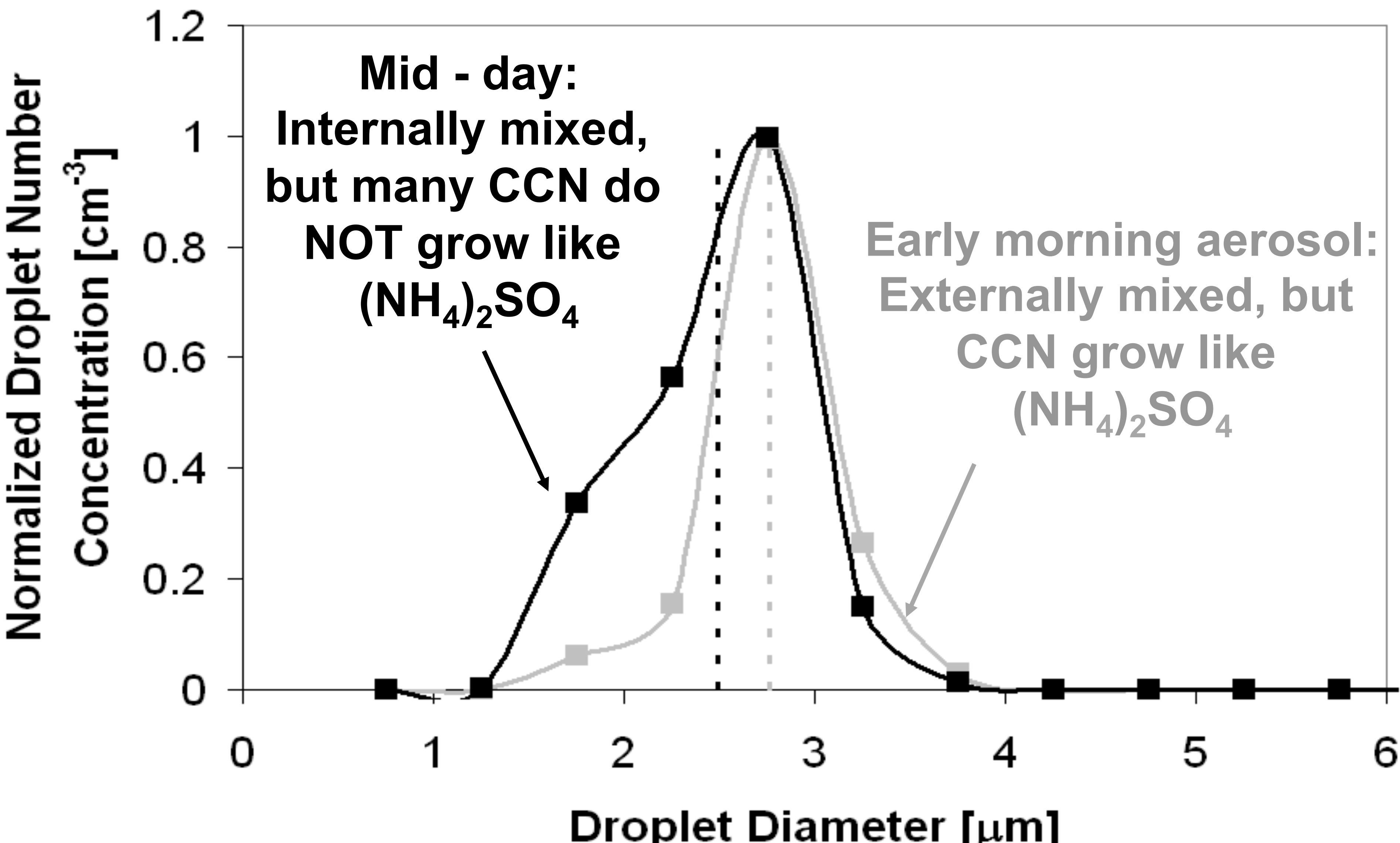




~ 250 g mol^{-1}, no surfactants

Strong surfactants present

Mexico City droplet growth kinetics



Does *growth kinetics* change?

Measurements of droplet size in the CCN instrument

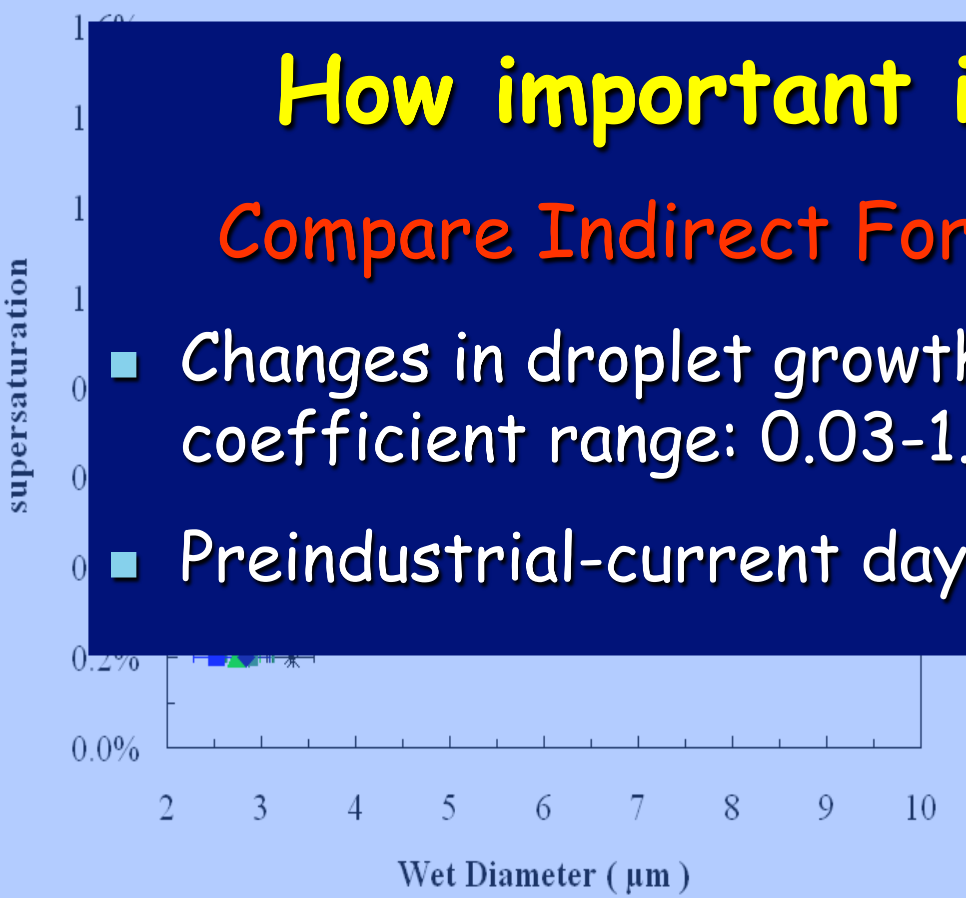
Marine Stratocumulus (MASE)
All CCN grow alike

Urban (Mexico City, MIRAGE)
CCN don't grow alike.

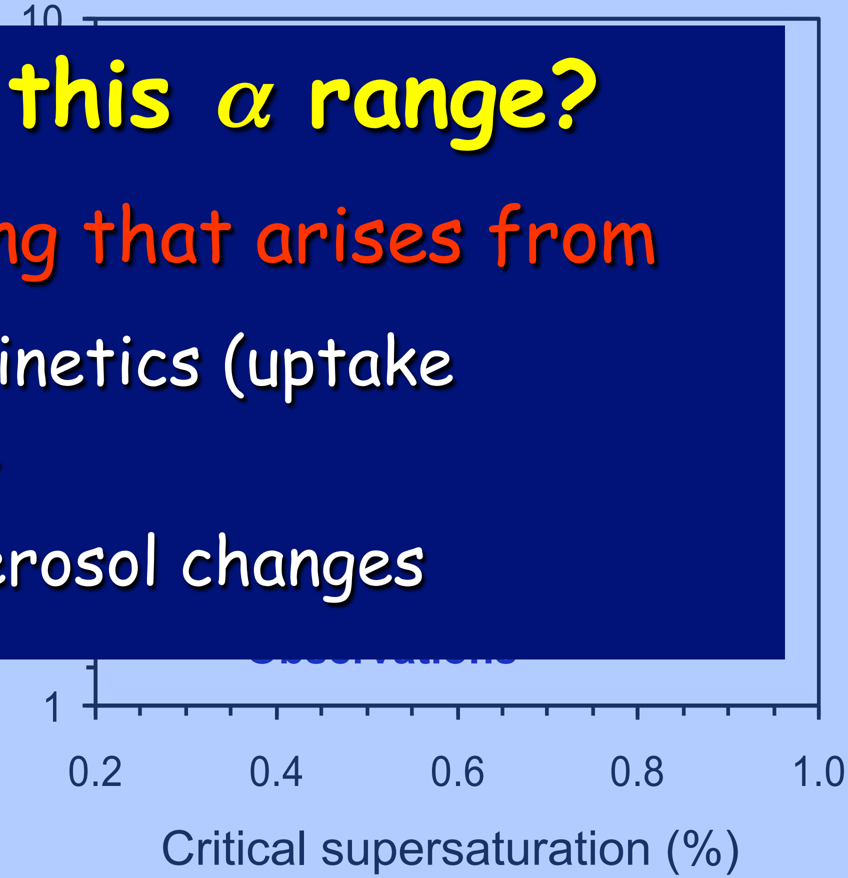
How important is this α range?

Compare Indirect Forcing that arises from

- Changes in droplet growth kinetics (uptake coefficient range: 0.03-1.0).
- Preindustrial-current day aerosol changes



~ 250 g mol^{-1}, no surfactants



Strong surfactants present

Global Modeling Framework Used

General Circulation Model

- NASA GISS II’ GCM
- 4’×5’ horizontal resolution
- 9 vertical layers (27-959 mbar)

Aerosol Microphysics

- The TwO-Moment Aerosol Sectional (TOMAS) microphysics model (Adams and Seinfeld, *JGR*, 2002) is applied in the simulations.

- Model includes 30 size bins from 10 nm to 10 μm.
- For each size bin, model tracks: Aerosol number, Sulfate mass, Sea-salt mass

- Bulk microphysics version is also available (for coupled feedback runs).

Global Modeling Framework Used

Emissions

Current day, preindustrial

Cloud droplet number calculation

Nenes and Seinfeld (2003); Fountoukis and Nenes (2005) cloud droplet formation parameterizations.

- ✓ Sectional and lognormal aerosol formulations.
- ✓ Can treat very complex internal/external aerosol, and effects of organic films on droplet growth kinetics.

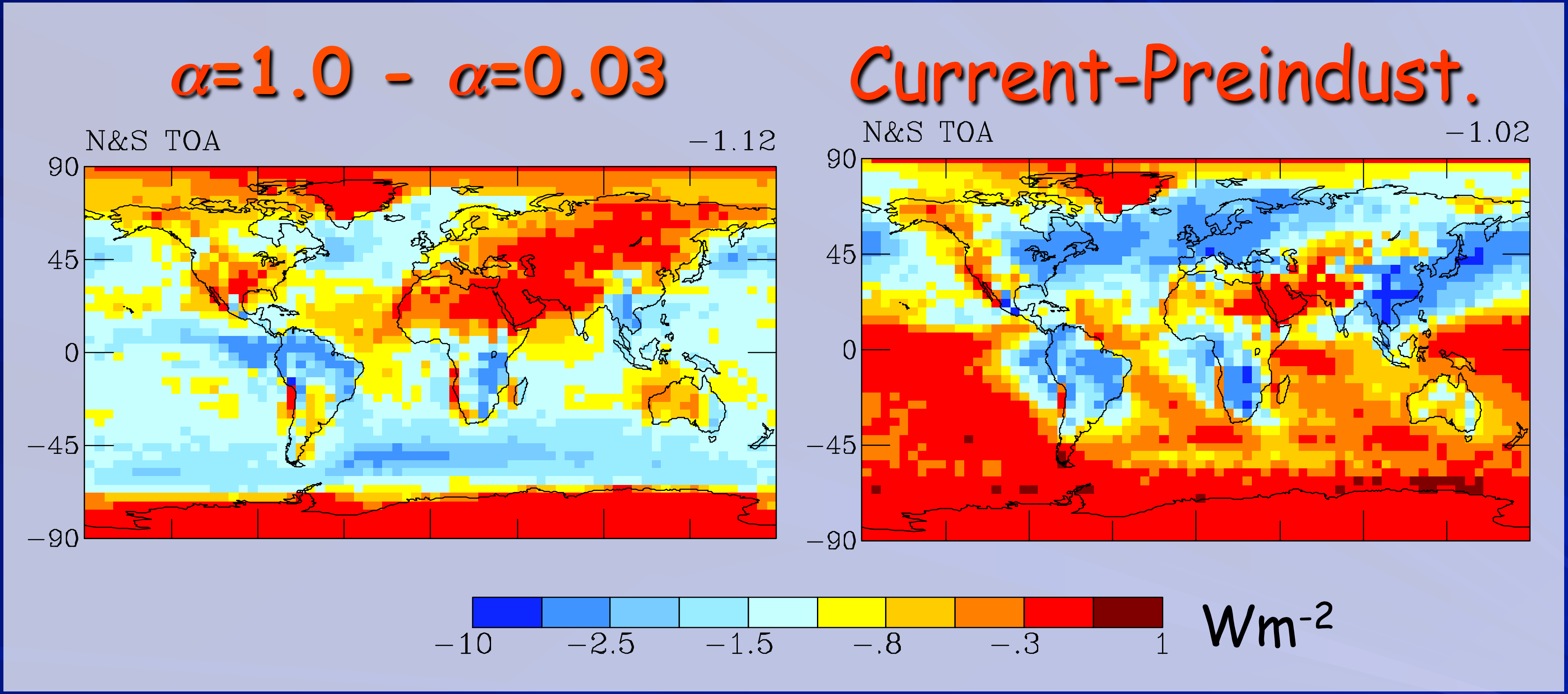
Autoconversion

Khairoutdinov & Kogan (2000), Rotstayn (1997)

In-cloud updraft velocity (for N&S only)

- Prescribed (marine: 0.25-0.5 ms^{-1};continental: 0.5-1 ms^{-1}).
- Diagnosed from large-scale TKE resolved in the GCM.

Sensitivity of indirect forcing to the water vapor uptake coefficient



Forcing from range in growth kinetics (-1.12 W m^{-2}) is as large as Present-Preindustrial change (-1.02 W m^{-2}) !

Spatial patterns are very different.

Nenes et al., *in preparation*

Overall Summary

- CCN theory is adequate for describing cloud droplet formation.
- Simplified treatments of aerosol composition get “most” of the CCN number right, but the associated uncertainty in CCN and indirect forcing can still be large.
- Size-resolved measurements of CCN activity can be used to infer the effects of organics in a simple way. It seems that simple descriptions are possible and as a community we need to systematically delve into this.
- The impact of organics on droplet growth kinetics is a important issue that remains unconstrained to date. This is a “chemical effect” that has not been appreciated enough…

Overall Summary

- Droplet formation parameterizations are at the point where they can explicitly consider all the chemical "complexities" of CCN calculations and droplet growth kinetics.
- Observations should provide the "constraints" of organic properties – classified with respect to age and source.
- They can even begin incorporating effects of dynamics (entrainment, variable updraft).
- People developing aerosol-cloud parameterizations need to work very hard at linking them at the cloud-scale, starting off from idealized "conceptual" feedback models.
- A lot of work to do… but it's exciting and challenging (not impossible).

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