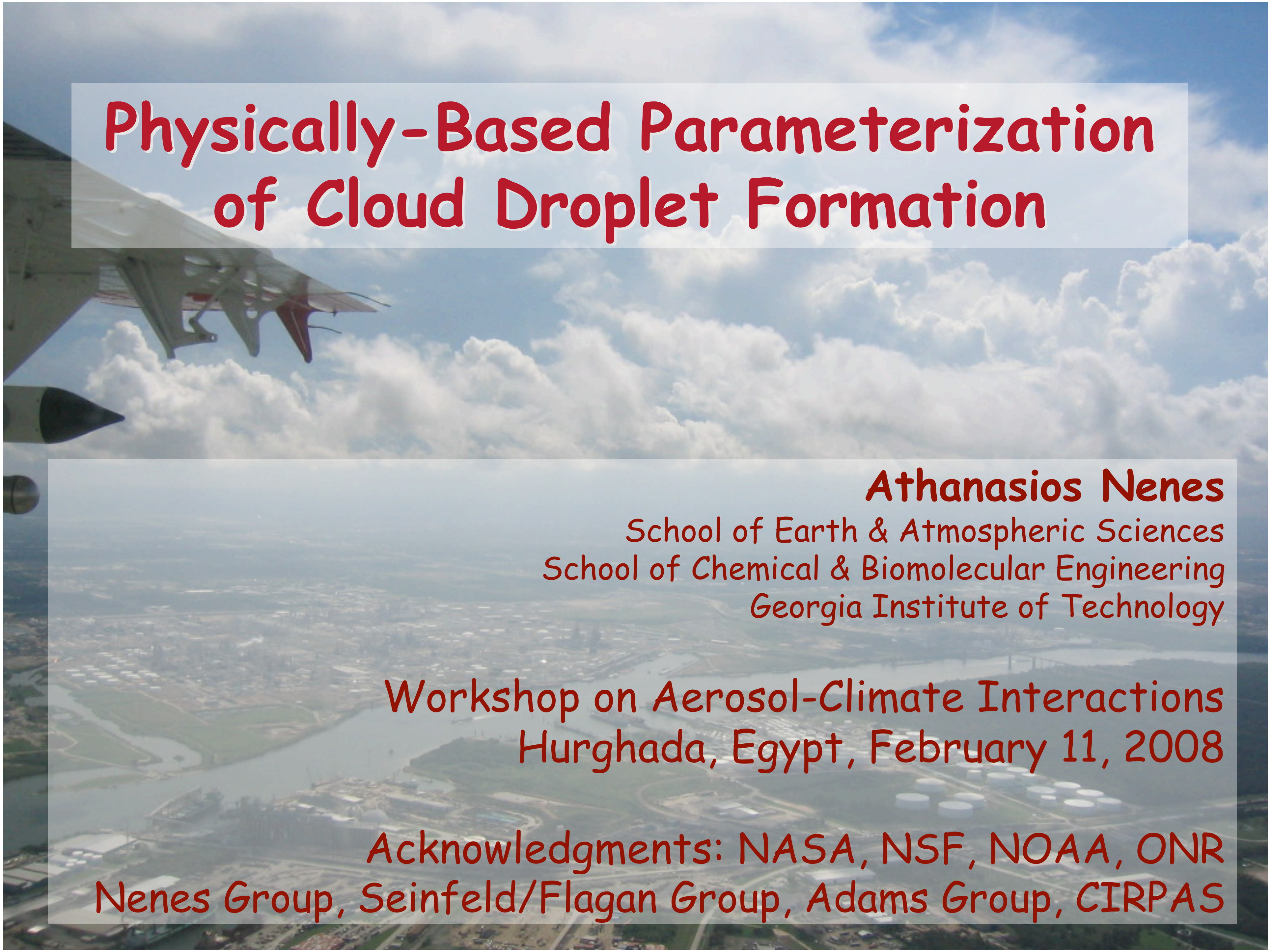

Physically-Based Parameterization of Cloud Droplet Formation

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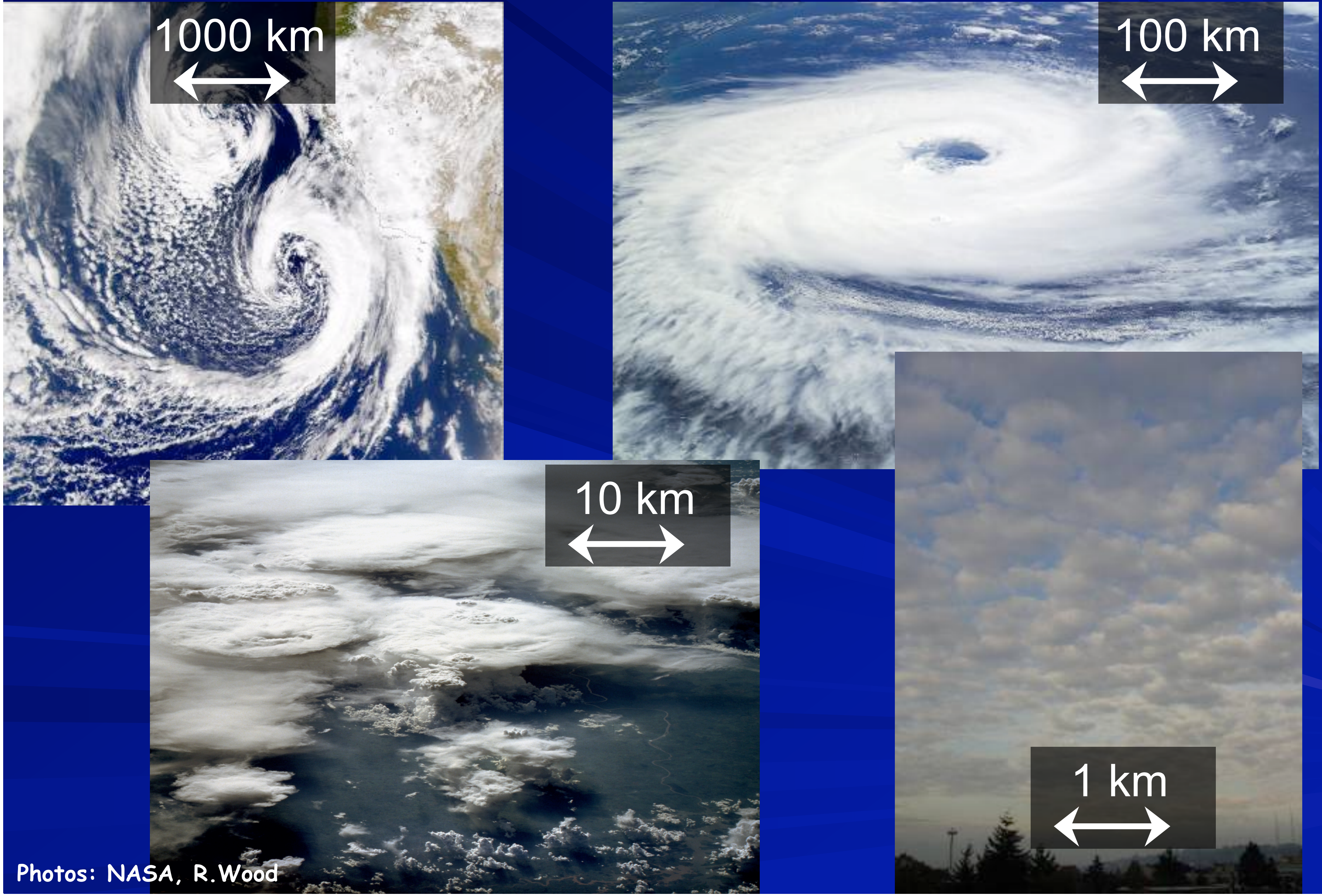
Workshop on Aerosol-Climate Interactions
Hurghada, Egypt, February 11, 2008

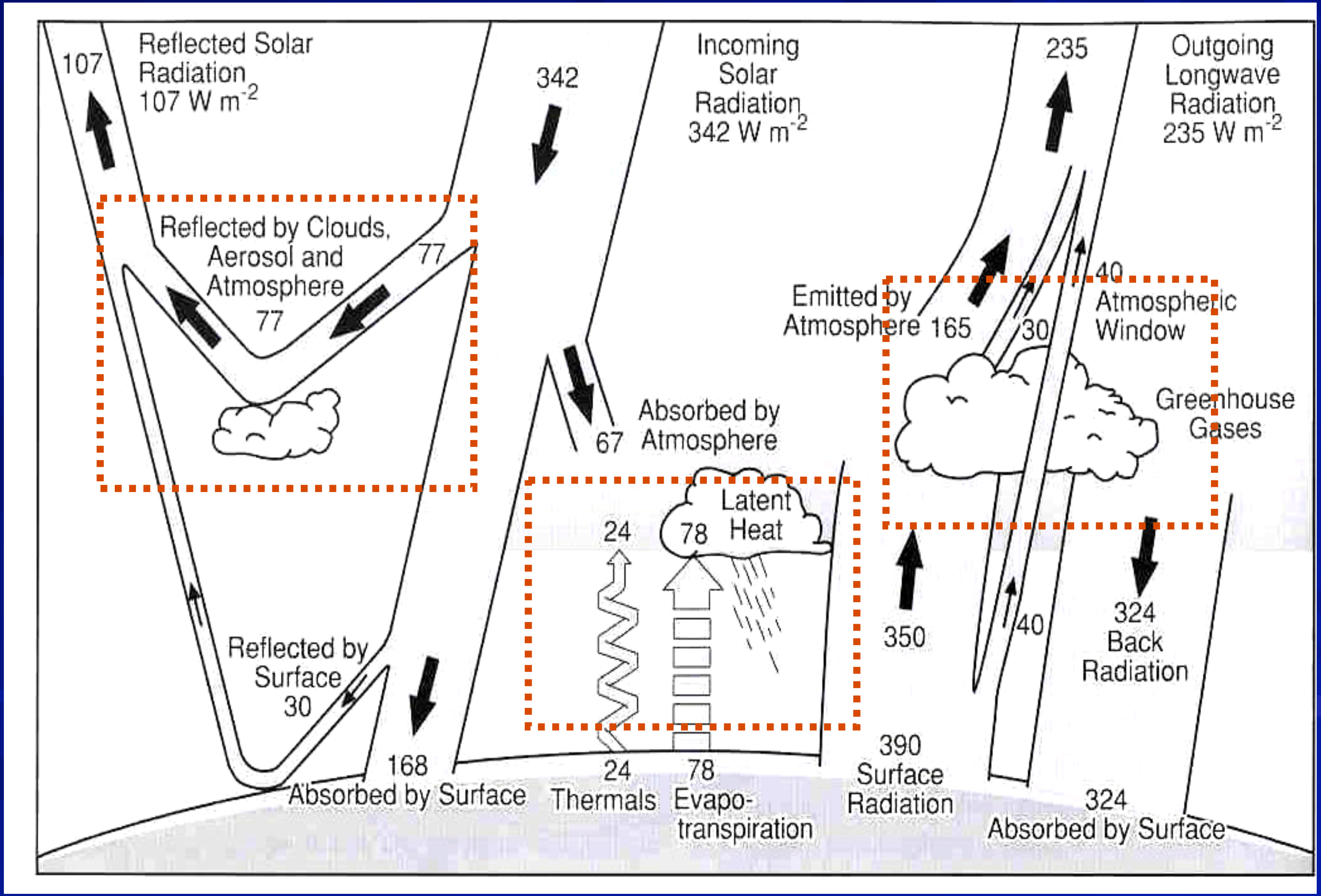
Acknowledgments: NASA, NSF, NOAA, ONR
Nenes Group, Seinfeld/Flagan Group, Adams Group, CIRPAS

Clouds are everywhere and found at all scales…



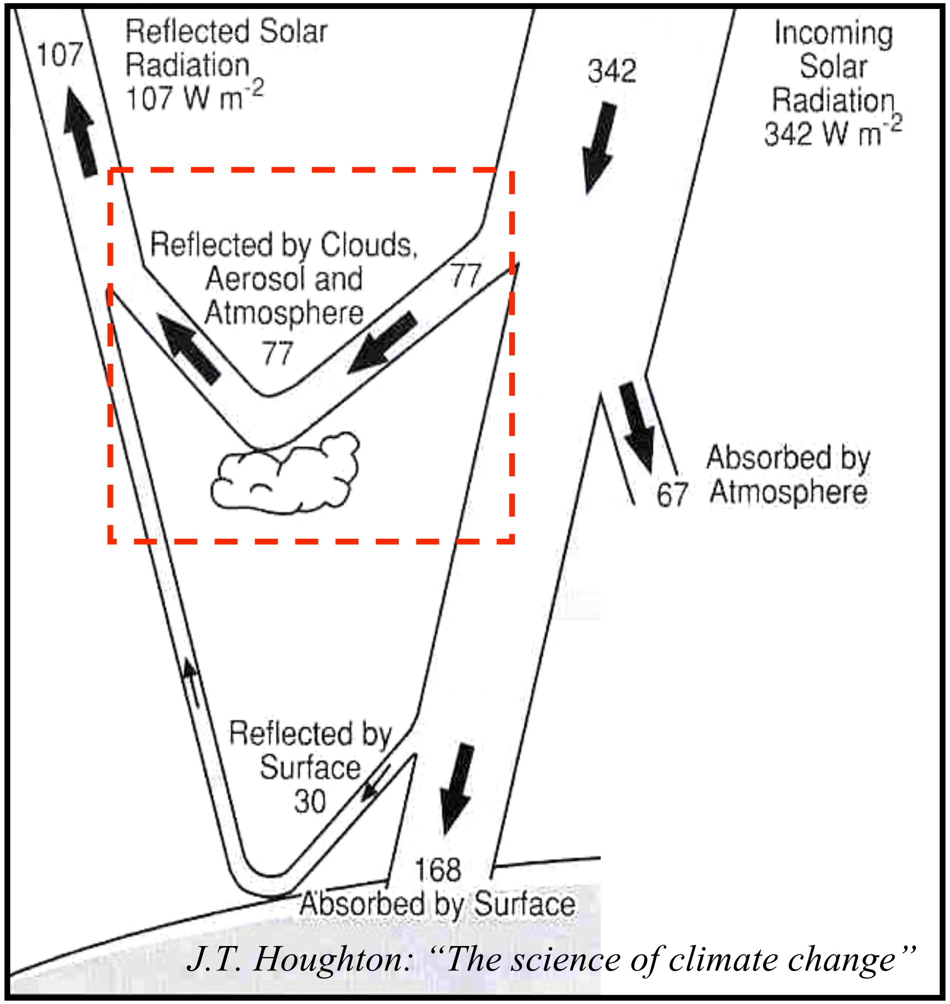
Photos: NASA, R.Wood

Clouds play a central role in the climate system



J.T. Houghton: "The science of climate change"

Clouds: major contributor to SW global albedo



J.T. Houghton: "The science of climate change"

Facts:

- Clouds account for ~50% of planetary reflectivity (albedo).
- Small changes in clouds yield large changes in global energy balance.
- **1%** increase in global cloud cover can counteract warming from greenhouse gases.

Consequence:

Understanding cloud formation is required for assessments of climate change.

Clouds are VERY dynamic (difficult to simulate).

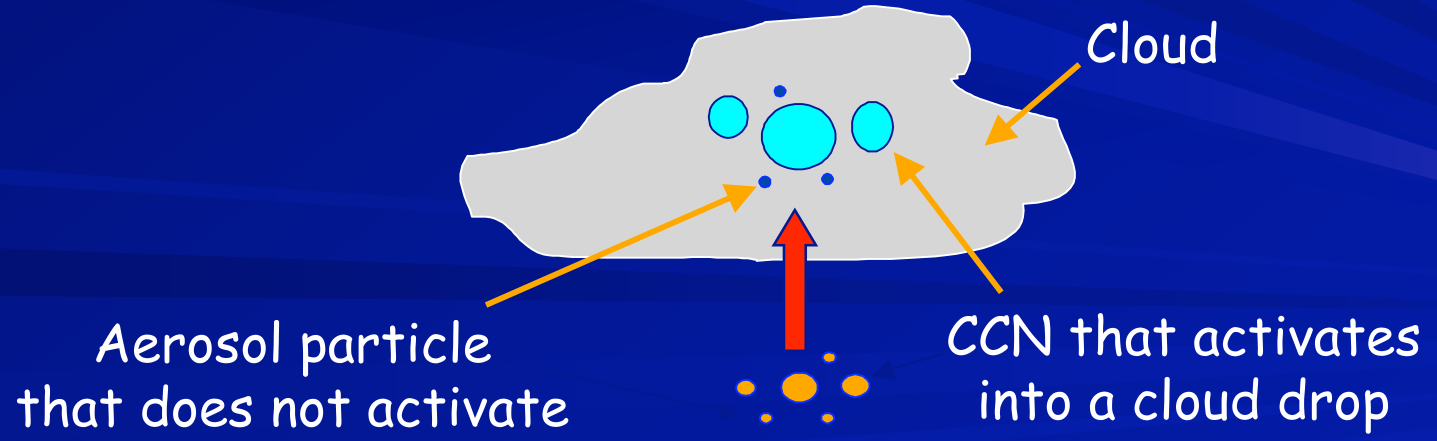
How do (liquid water) clouds form?

Clouds form in regions of the atmosphere where there is too much water vapor (it is "supersaturated").

This happens when air is cooled (primarily through expansion in updraft regions and radiative cooling).

Cloud droplets nucleate on pre-existing particles found in the atmosphere (aerosols). This process is known as activation.

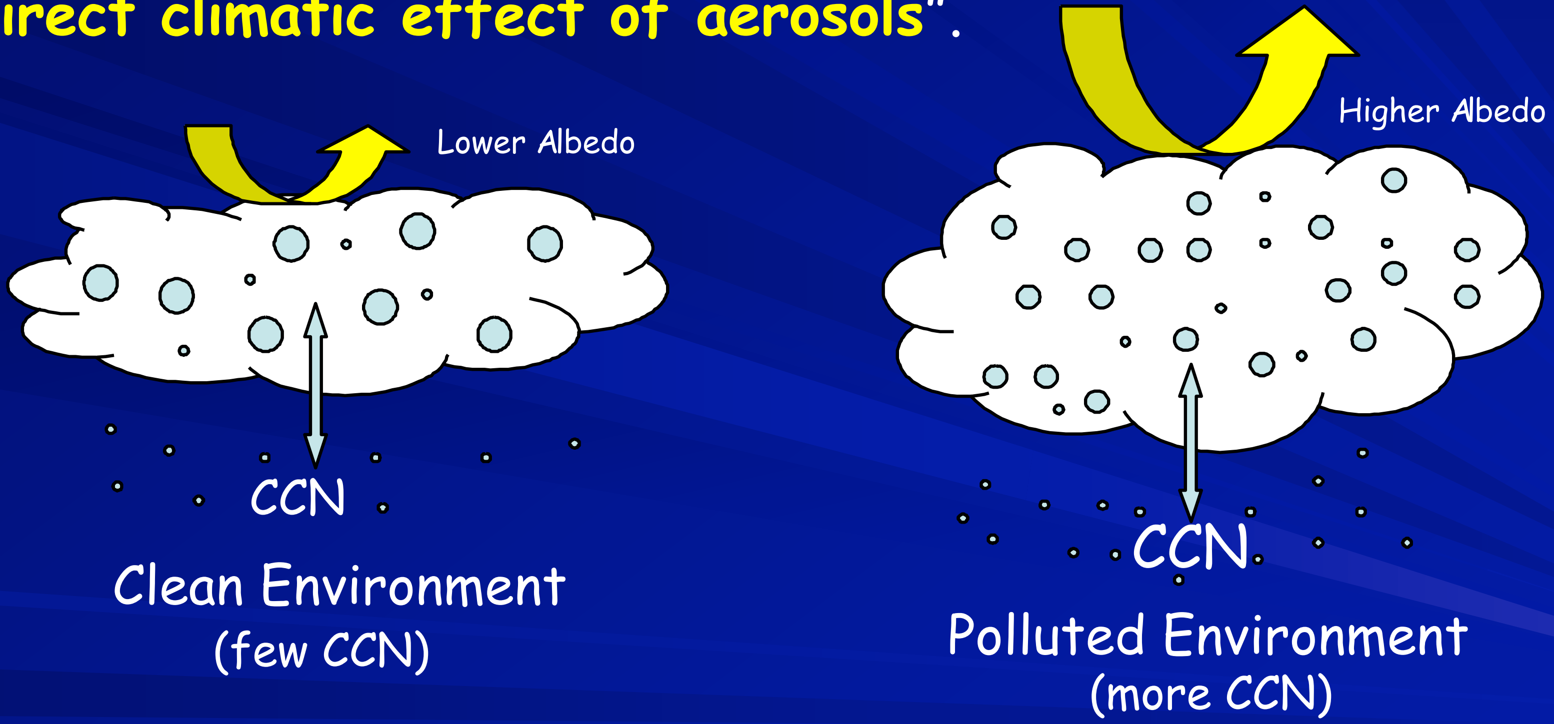
Aerosols that can become droplets are called cloud condensation nuclei (CCN).



Can humans affect clouds and the hydrological cycle?

Yes! By changing global CCN concentrations.

Result: Clouds that are "whiter", precipitate less (persist longer) and potentially cover larger areas of the globe. This is thought to yield a net cooling on climate and is termed as the "**indirect climatic effect of aerosols**".

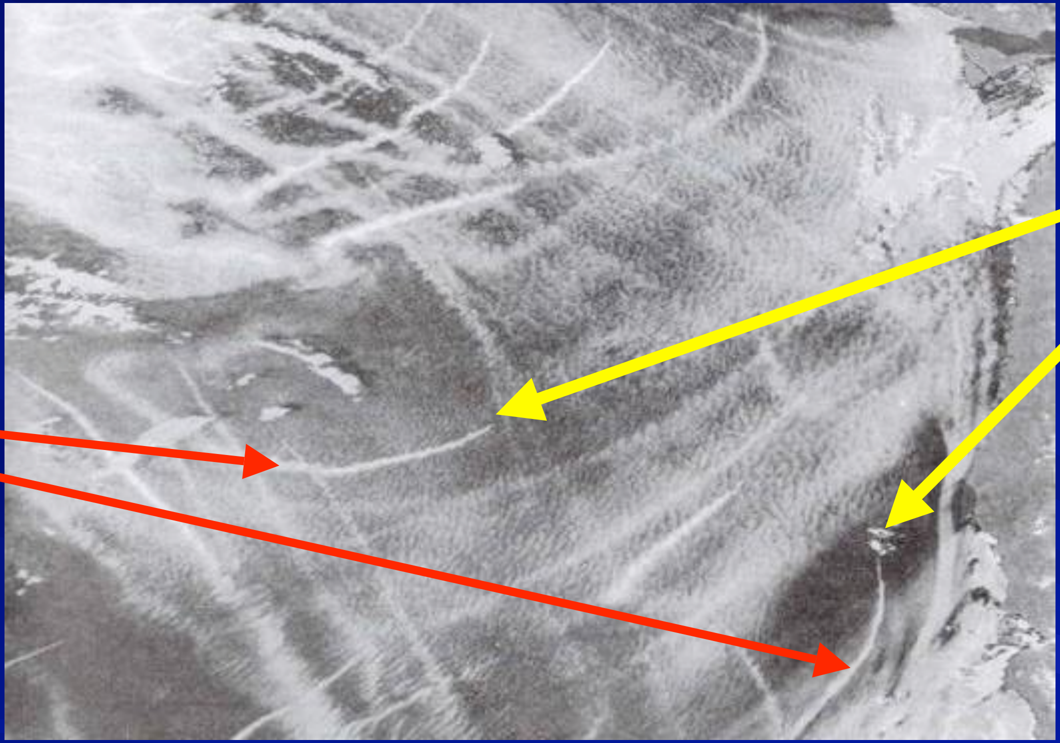


Increasing particles tends to cool climate (potentially alot).

Quantitative assessments done with climate models.

Observational evidence of indirect effect

"Ship tracks": features of high cloud reflectivity embedded in marine stratus. A result of ship plumes affecting clouds above.



"Ship Track"

Ship plume incorporated into cloud

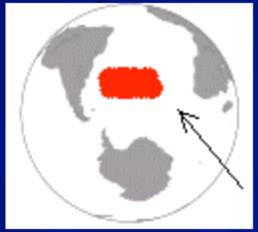
Clouds are whiter and last longer

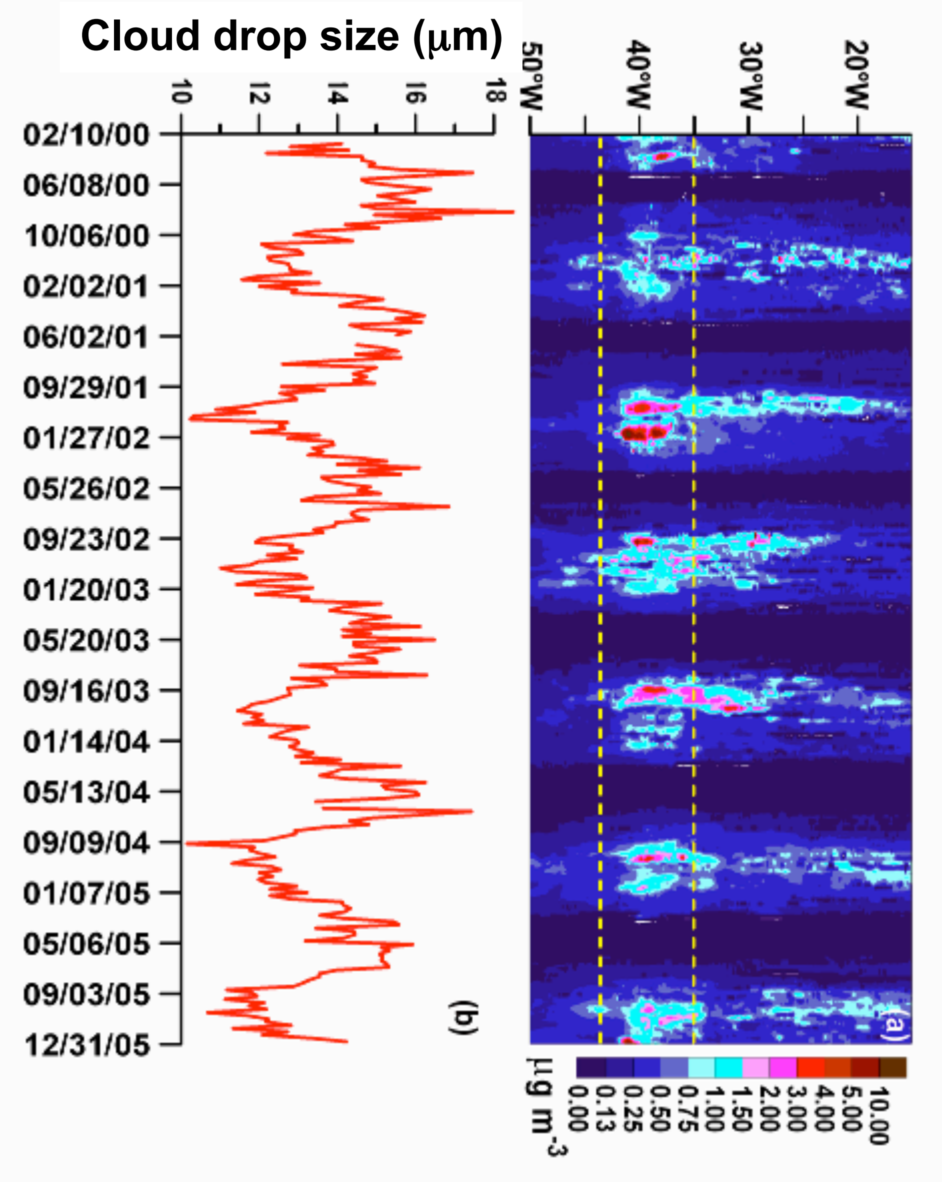
Pollution ↑ ⇒ Droplet number ↑ ⇒ Droplet size ↓

Droplet size ↓ ⇒ Cloud reflectivity ↓ AND Precip ↓

Phytoplankton affect clouds too…

Location: East of Patagonia (South America)





Low chlorophyll,
cloud have large drops

High Chlorophyll,
Clouds have small drops

Phytoplankton increase CCN.

Their activity strongly impact clouds.

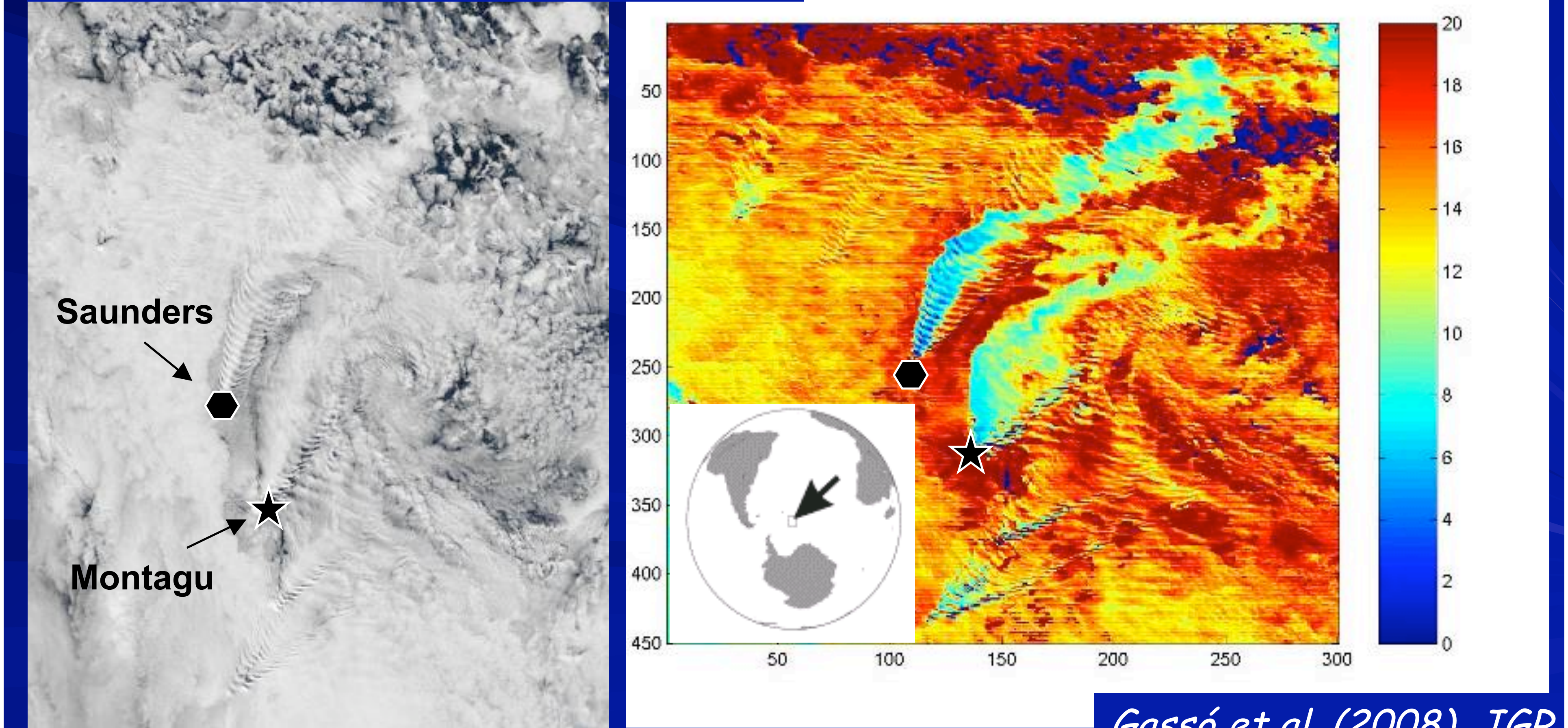
The changes are comparable to contrasts between polluted and clean environments (forcing ~ -15 W m^{-2}).

Meskhidze and Nenes, Science, 2006

So do volcanoes (even when “sleeping”) …

Volcanoes continuously emit SO_2 which becomes sulfate aerosol. The aerosol can substantially increase CCN in volcanic plumes. Cloud in the plume are much more reflective than outside.

Location: Sandwich Islands , ~55S,~30W



Gassó et al. (2008), JGR

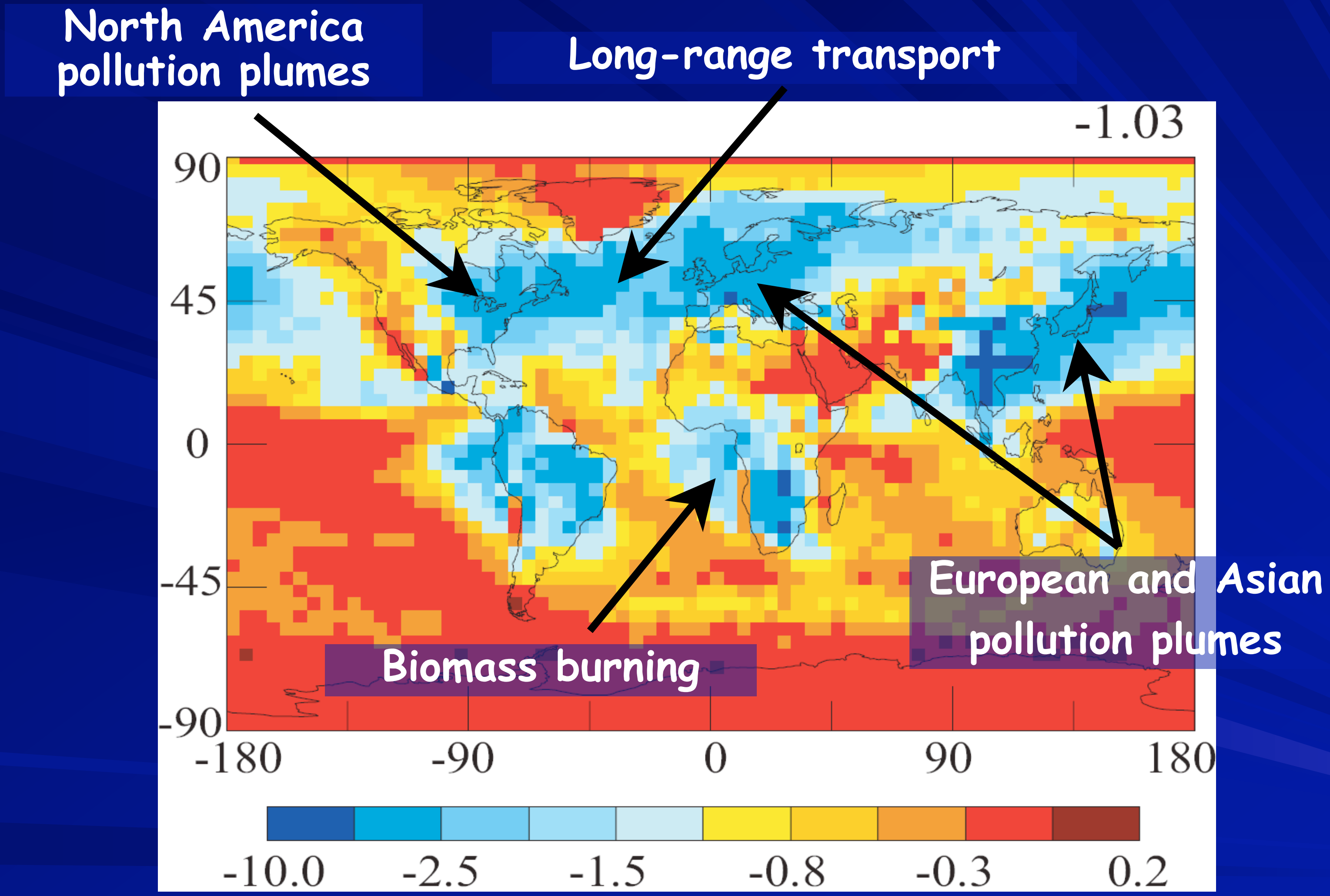
Anthropogenic Indirect Effect: How do we estimate its global impact?

We use a global climate model (GCM)

- simulation with “current day” emissions
- simulation without anthropogenic emissions (“preindustrial” emissions)

- compute the change in energy (radiation) between two simulations (“indirect forcing”)
- compare annual average forcing to greenhouse gas warming (~ 2.5 W m^{-2})
- Net forcing (greenhouse + indirect) can be used as an index for climate change.

Indirect Forcing calculation (W m^{-2})

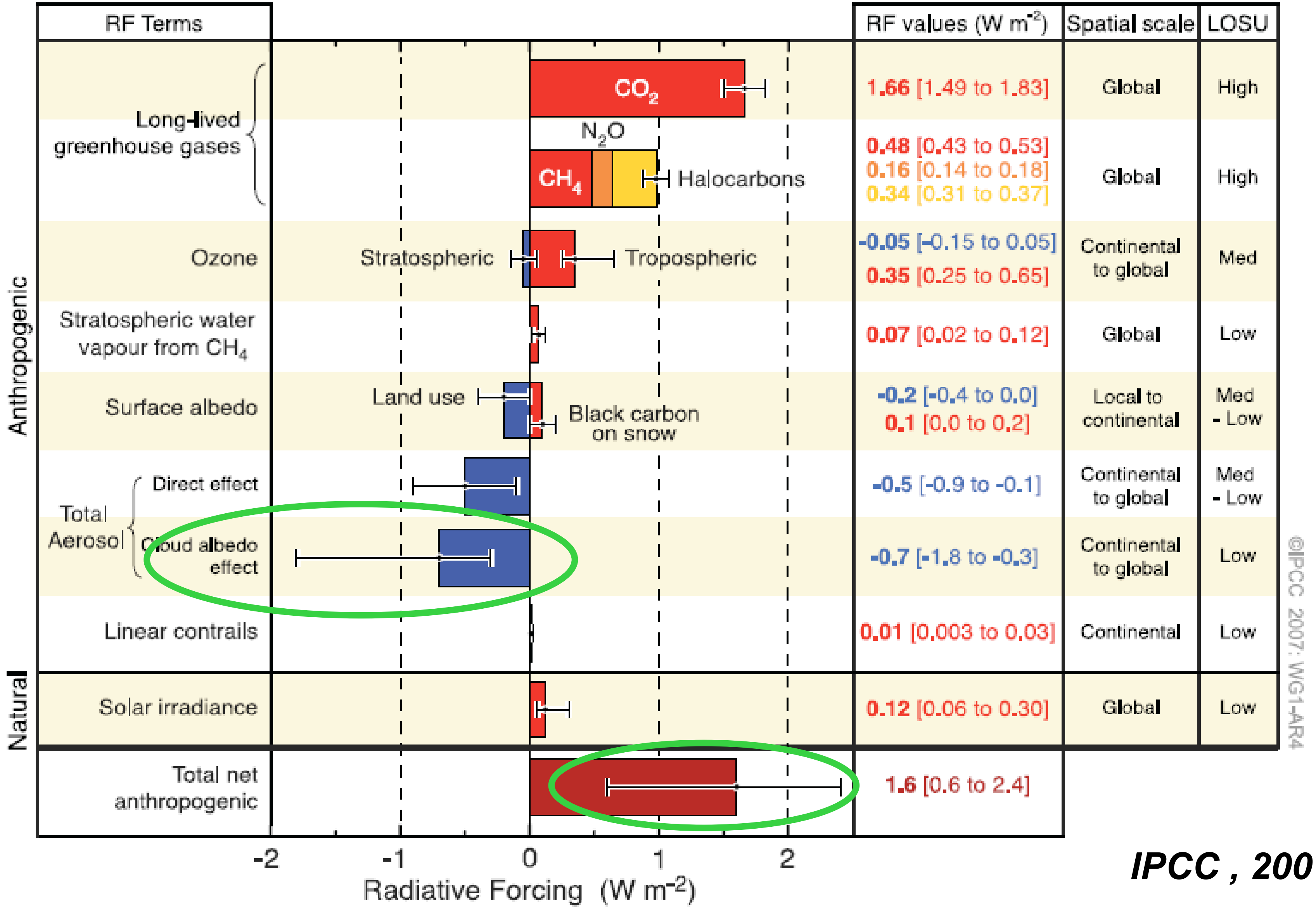


Sotiropoulou et al., in preparation

Spatial pattern of IF follows that of aerosol variations

Anthropogenic Climate Forcing

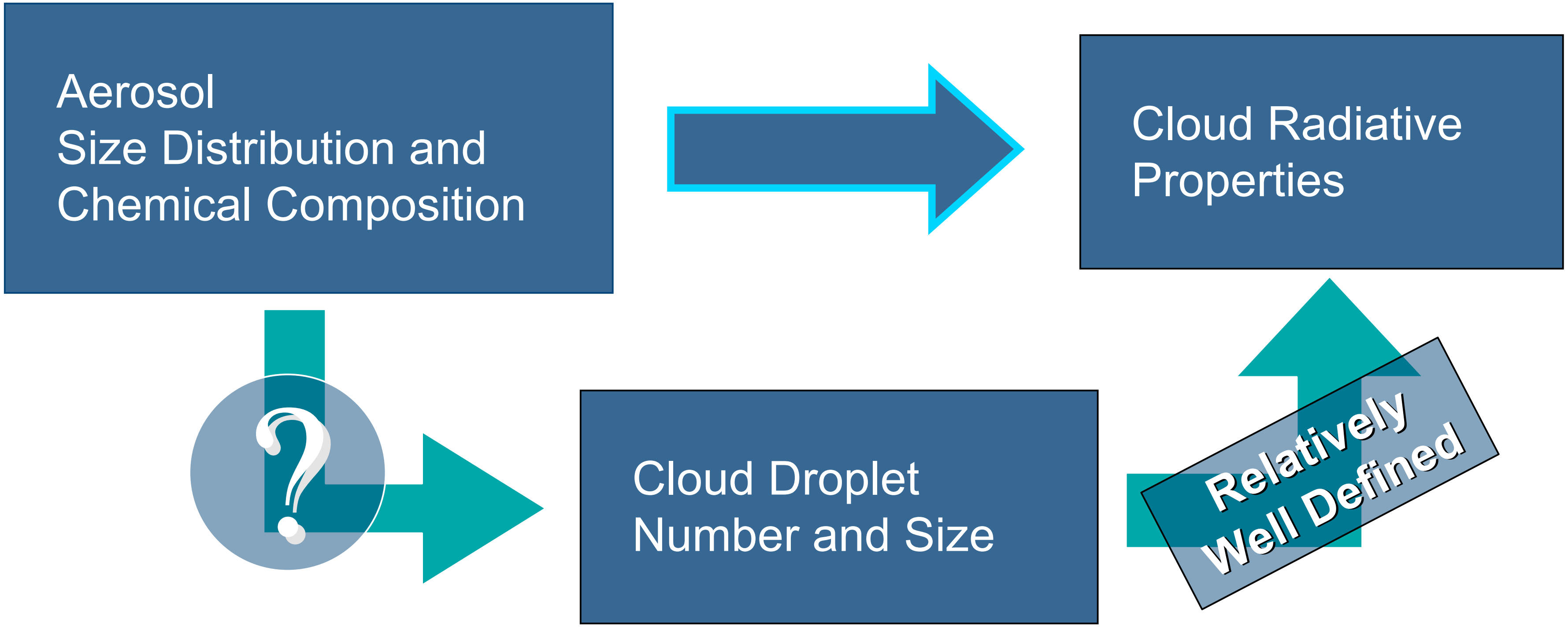
RADIATIVE FORCING COMPONENTS



		RF Terms	RF values (W m^{-2})	Spatial scale	LOSU
Anthropogenic	Long-lived greenhouse gases	CO$_2$	1.66 [1.49 to 1.83]	Global	High
		CH$_4$ N$_2$O Halocarbons	0.48 [0.43 to 0.53] 0.16 [0.14 to 0.18] 0.34 [0.31 to 0.37]	Global	High
	Ozone	Stratospheric Tropospheric	-0.05 [-0.15 to 0.05] 0.35 [0.25 to 0.65]	Continental to global	Med
	Stratospheric water vapour from CH$_4$		0.07 [0.02 to 0.12]	Global	Low
	Surface albedo	Land use Black carbon on snow	-0.2 [-0.4 to 0.0] 0.1 [0.0 to 0.2]	Local to continental	Med - Low
	Total Aerosol	Direct effect	-0.5 [-0.9 to -0.1]	Continental to global	Med - Low
		Cloud albedo effect	-0.7 [-1.8 to -0.3]	Continental to global	Low
	Linear contrails		0.01 [0.003 to 0.03]	Continental	Low
Natural	Solar irradiance		0.12 [0.06 to 0.30]	Global	Low
	Total net anthropogenic		1.6 [0.6 to 2.4]		

IPCC , 2007

Quantification of the Indirect Effect in Global Models



Aerosol
Size Distribution and
Chemical Composition

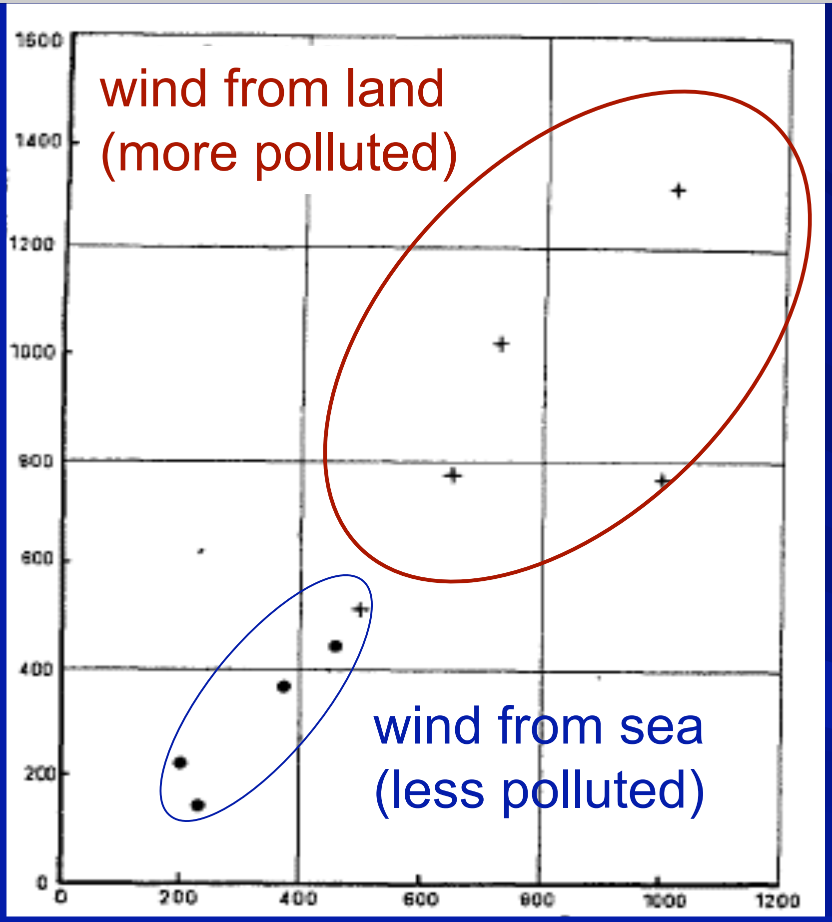
Cloud Radiative
Properties

Cloud Droplet
Number and Size

This problem has historically been reduced to finding the relationship between aerosol number concentration and cloud droplet number concentration. **Empirical** relationships are often used.

Aerosol-cloud microphysical observations

….first measurements at one site (1967)



wind from land
(more polluted)

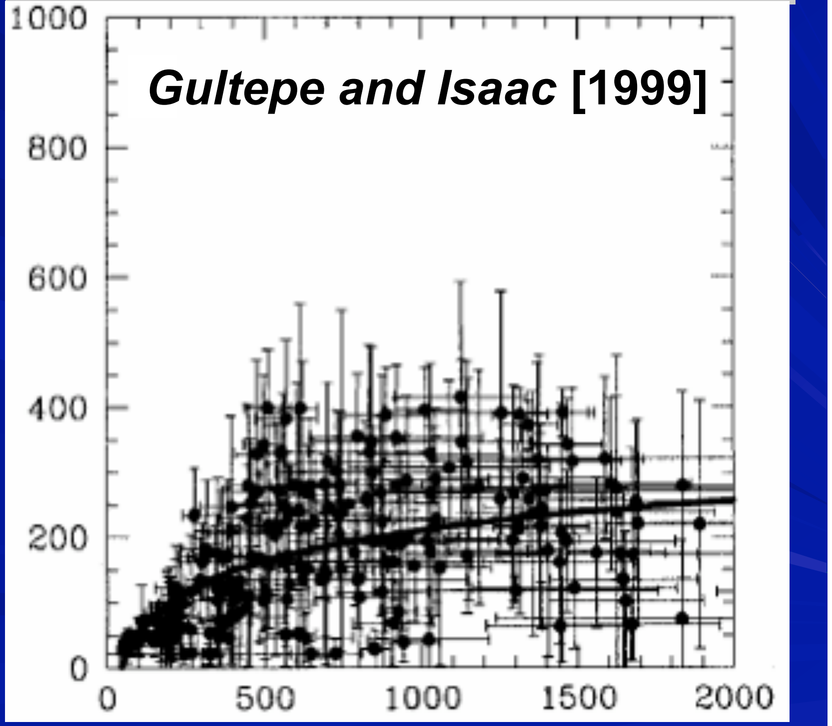
wind from sea
(less polluted)

Observed cloud droplet concentration [cm^{-3}]

Predicted droplet concentration from aerosol spectrum [cm^{-3}]

Twomey and Warner (1967)

….recent measurements (1999)

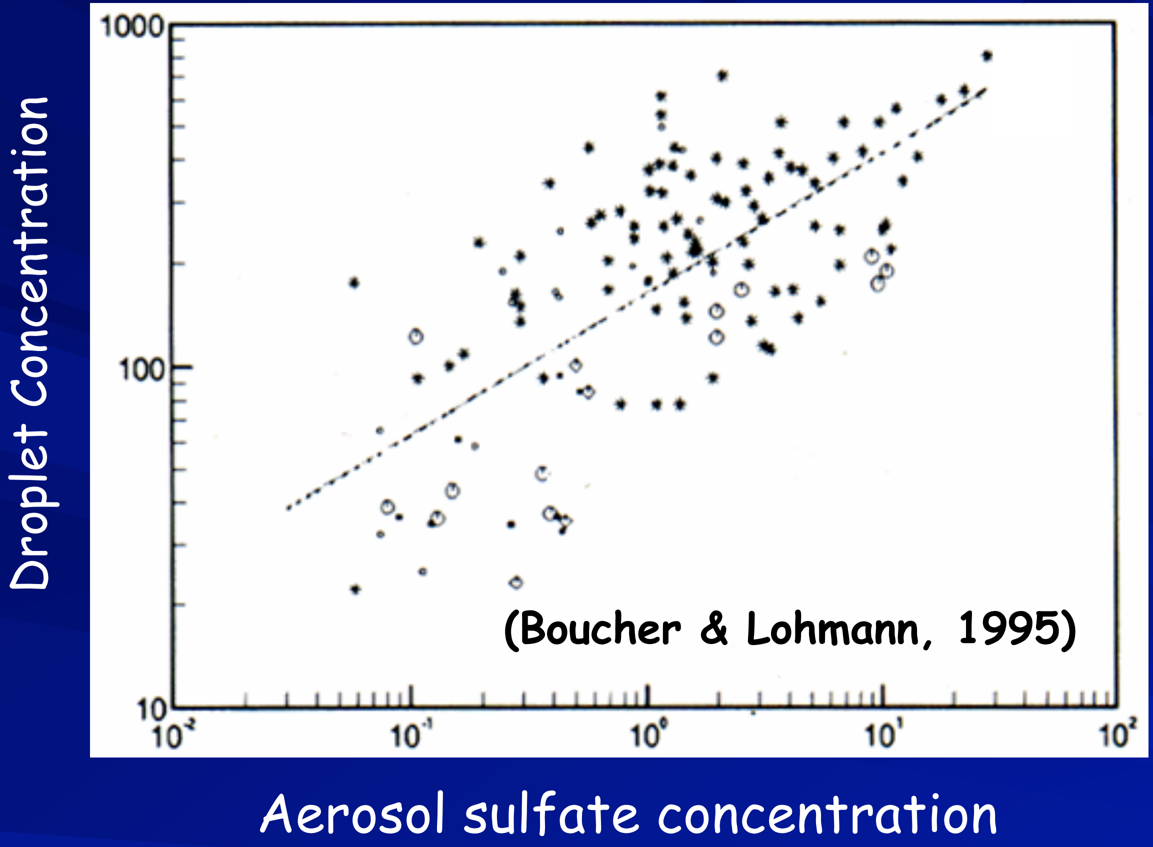


***Gultepe and Isaac* [1999]**

Droplet Number [cm^{-3}]

Aerosol Concentration [cm^{-3}]

Approach for aerosol-N_d: empirical



(Boucher & Lohmann, 1995)

Large variability.

Why?

Unaccounted:

- Meteorology
- Cloud microphysics
- Composition
- etc…

Many studies still utilize this type of approach.

Large predictive uncertainty, without “chances” of improving.

Current Direction: Use simplified but physically based approach for important processes

Dynamics

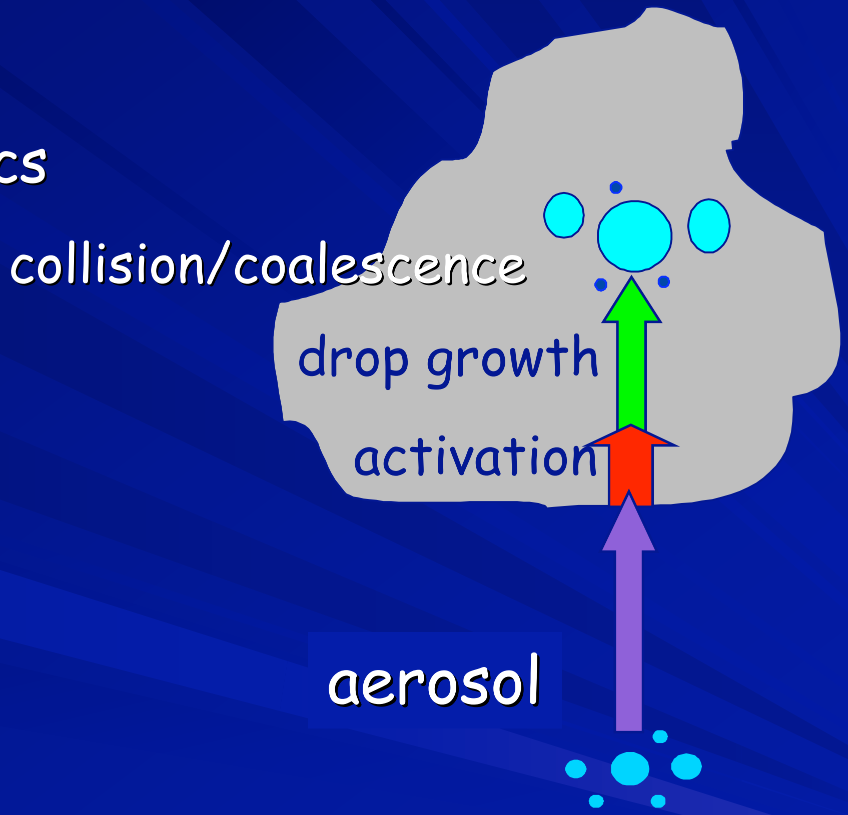
- Updraft Velocity
- Large Scale Thermodynamics

Particle characteristics

- Size
- Concentration
- Chemical Composition

Cloud Processes

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



collision/coalescence

drop growth

activation

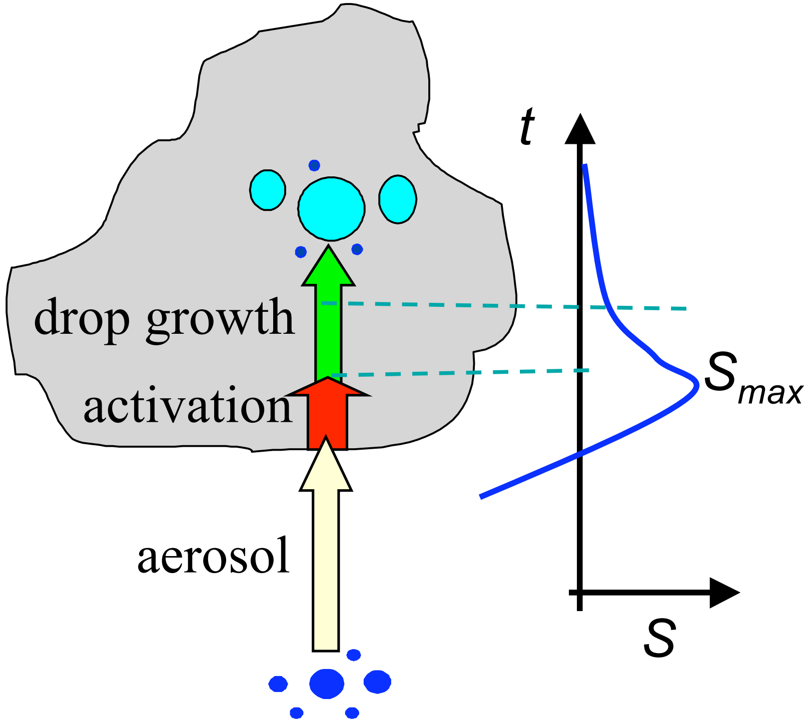
aerosol

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

Including explicit physics in GCMs is possible…

Tempting: use the "simple story of droplet formation"

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

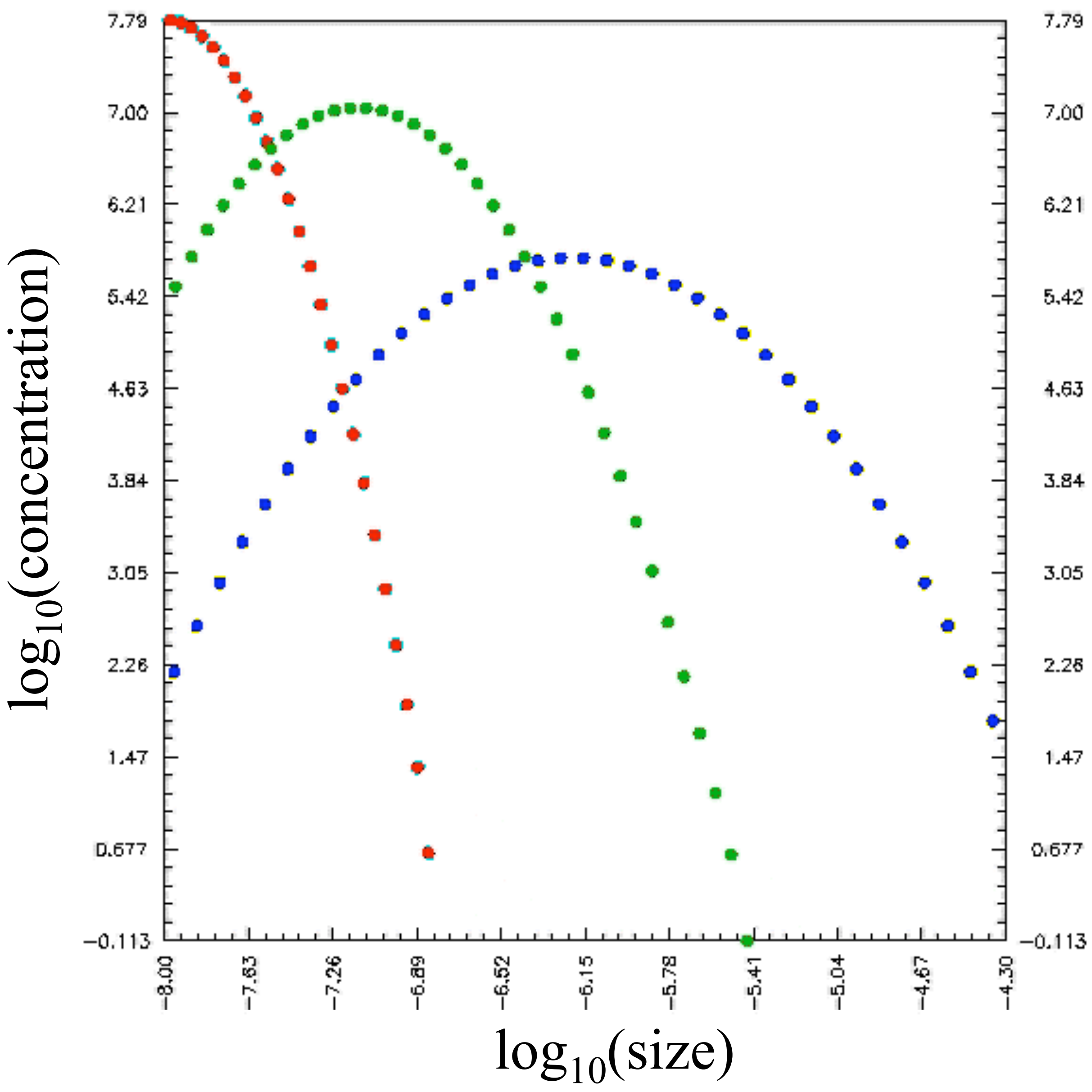


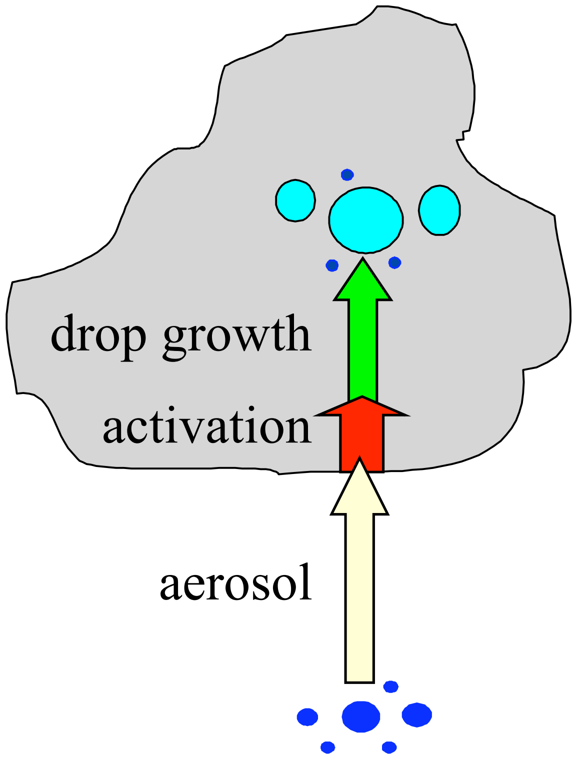
Steps are:

- 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

Cloud droplet formation in updrafts



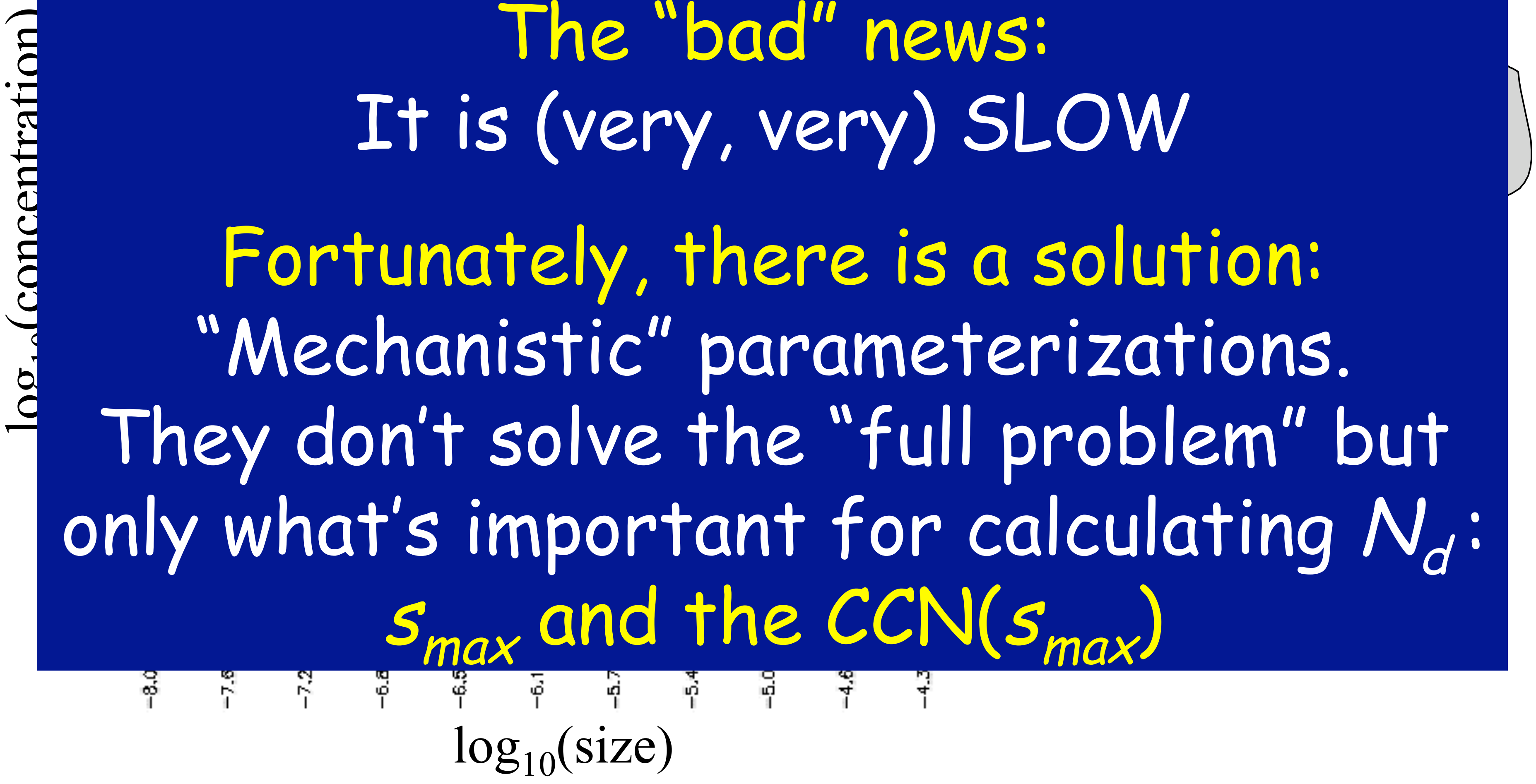


drop growth

activation

aerosol

Cloud droplet formation in updrafts



The “good” news:

The theory is established

The “bad” news:

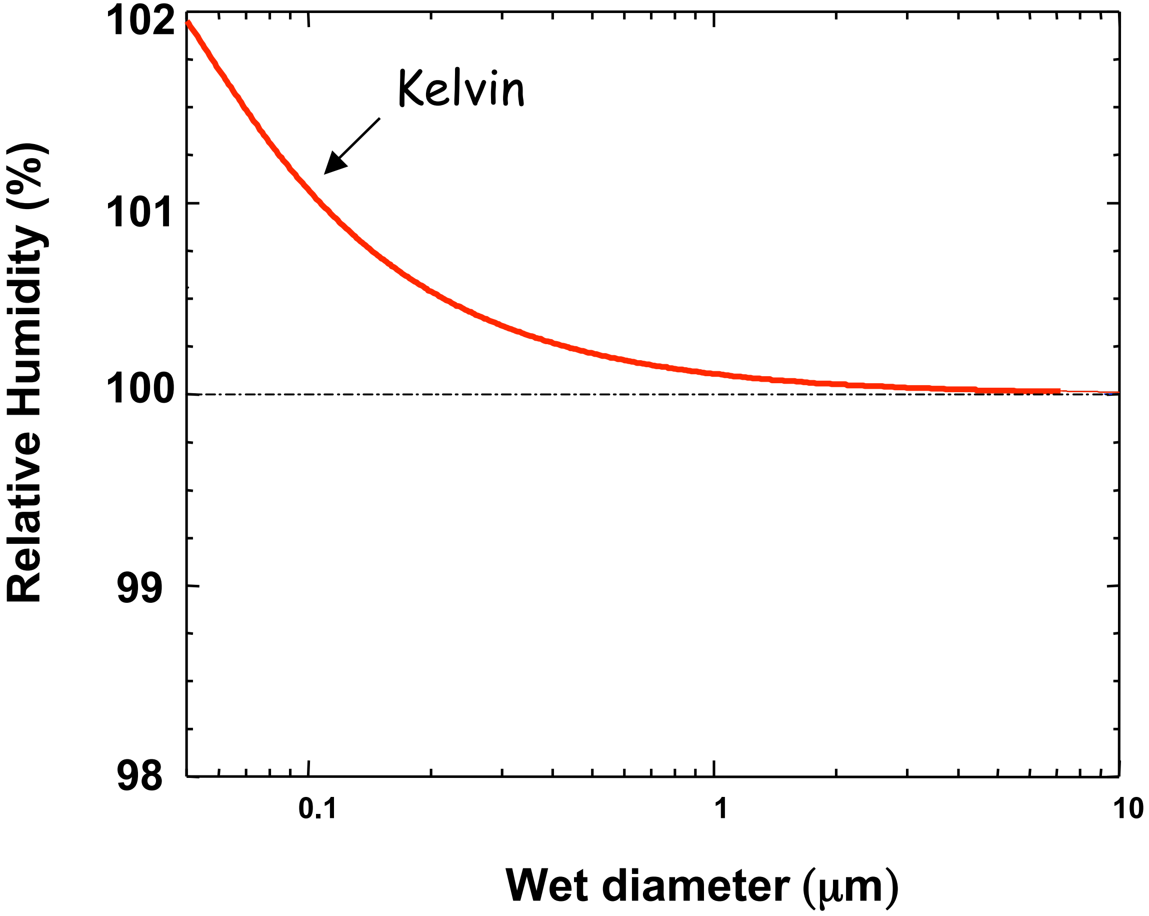
It is (very, very) SLOW

Fortunately, there is a solution:

“Mechanistic” parameterizations. They don’t solve the “full problem” but only what’s important for calculating N_d : s_{max} and the CCN(s_{max})

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

A particle acts as a CCN when it cannot be in *stable* equilibrium with water vapor.



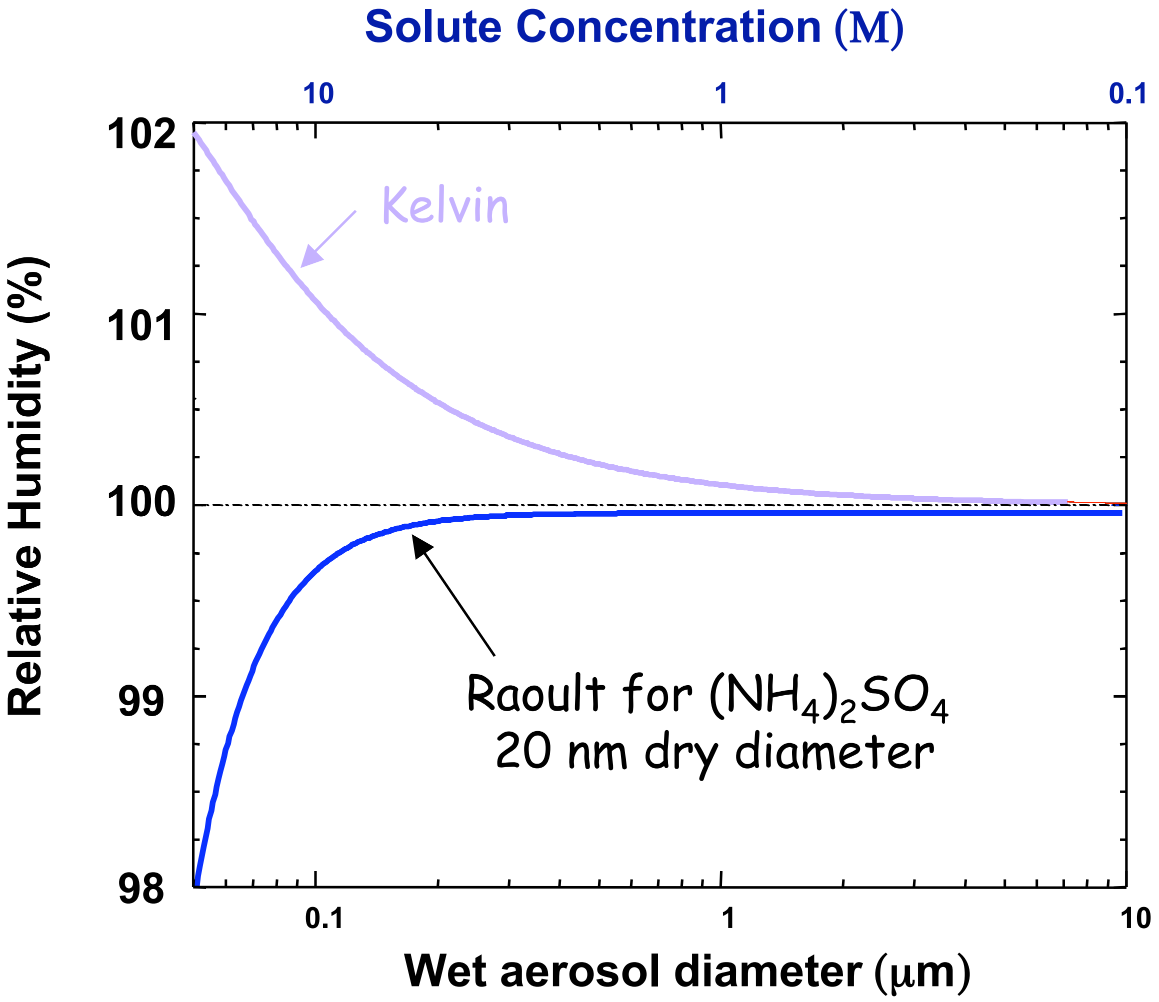
Take a pure H_2O drop

Droplet curvature enhances vapor pressure (Kelvin effect).

Drop is always in unstable equilibrium with water vapor (they either evaporate or grow uncontrollably).

When does an aerosol particle act as a CCN ?

A particle acts as a CCN when it cannot be in *stable* equilibrium with water vapor.



Now add some solute; e.g., $(NH_4)_2SO_4$

Dissolved material in the droplet tends to decrease the vapor pressure (Raoult effect).

When does an aerosol particle act as a CCN ?

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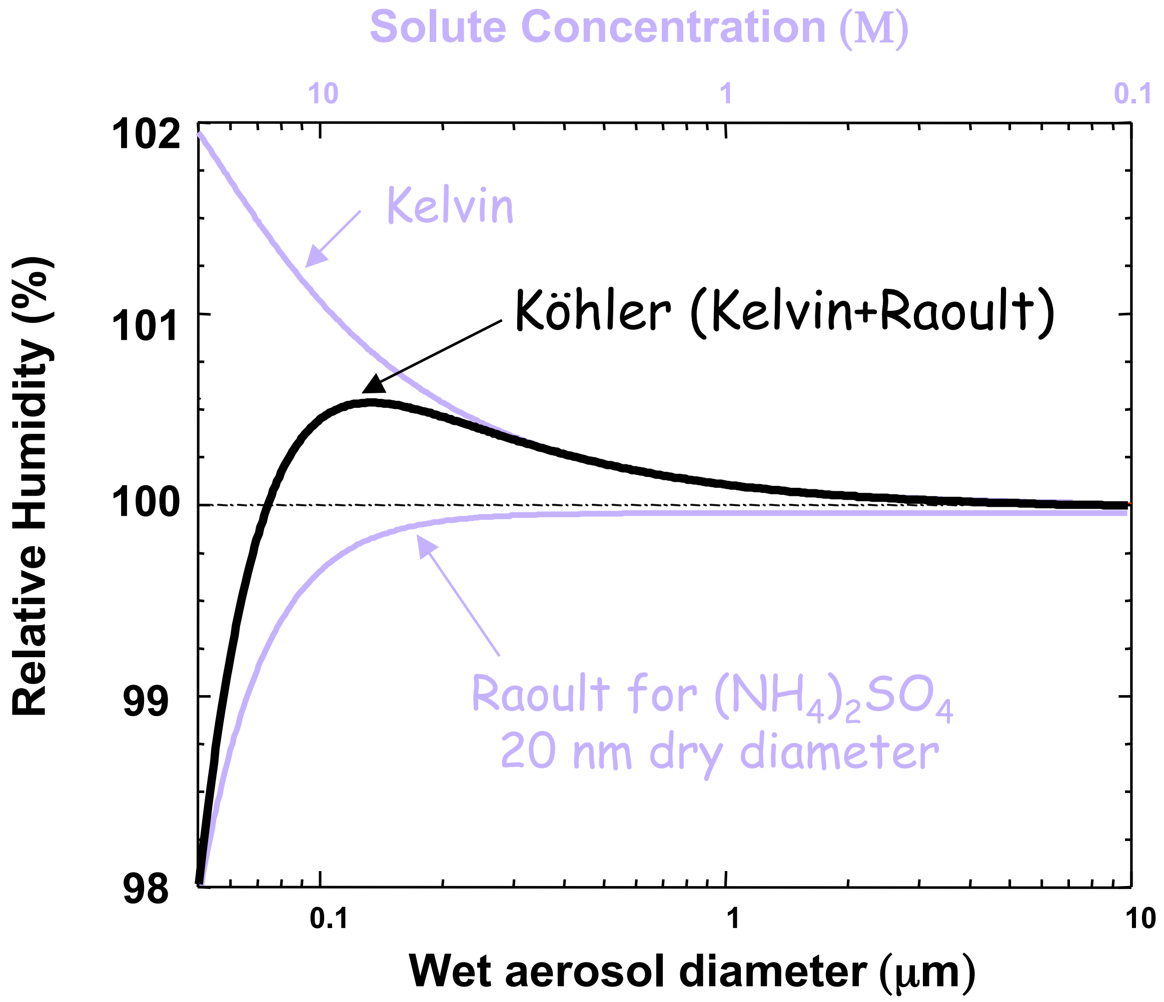


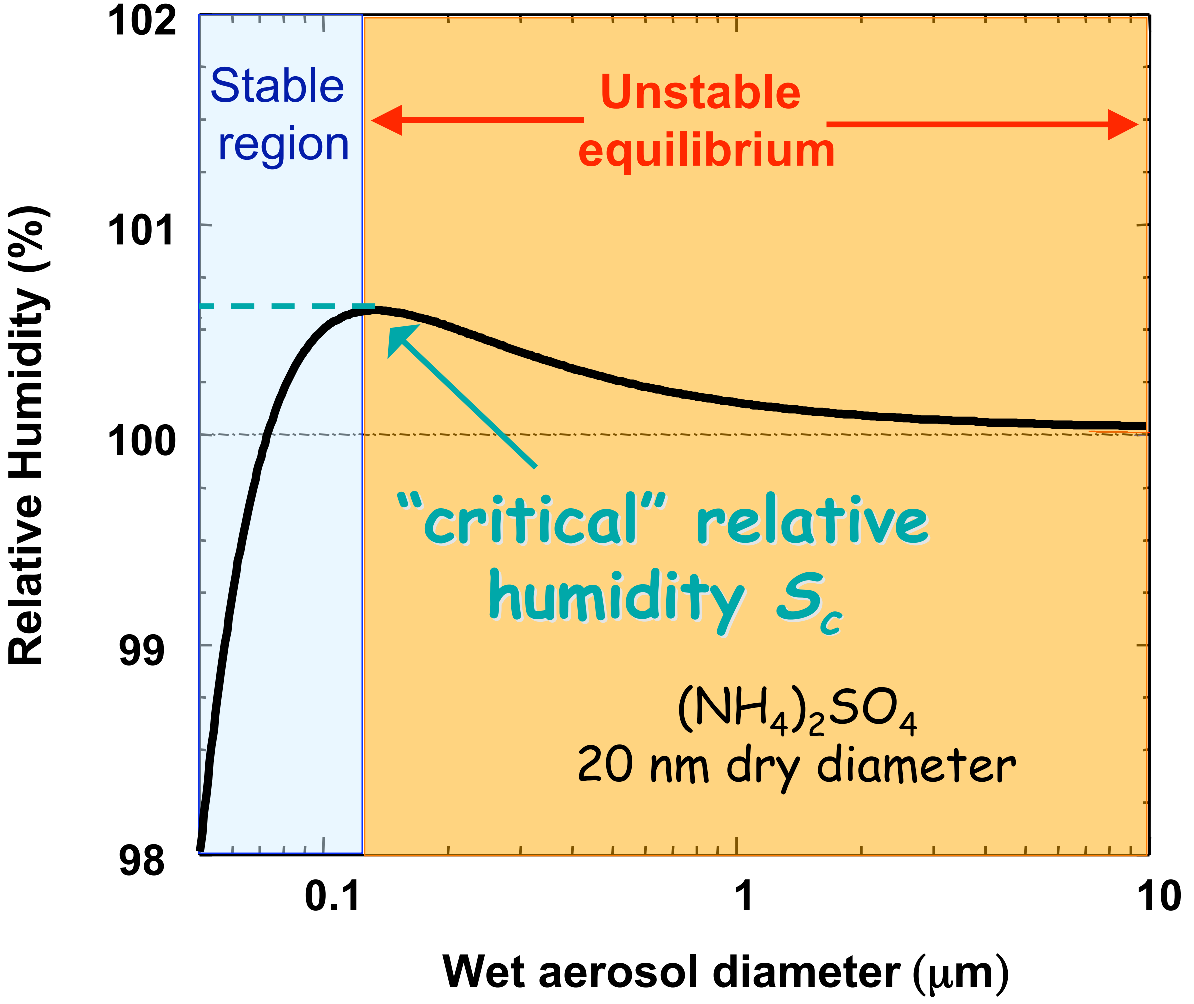
Now add some solute; e.g., $(NH_4)_2SO_4$

Dissolved material in the droplet tends to decrease the vapor pressure (Raoult effect).

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

When does an aerosol particle act as a CCN ?

Aerosol can be in stable equilibrium with supersaturated water.
If in "unstable" region, it acts as a CCN and nucleates a drop.

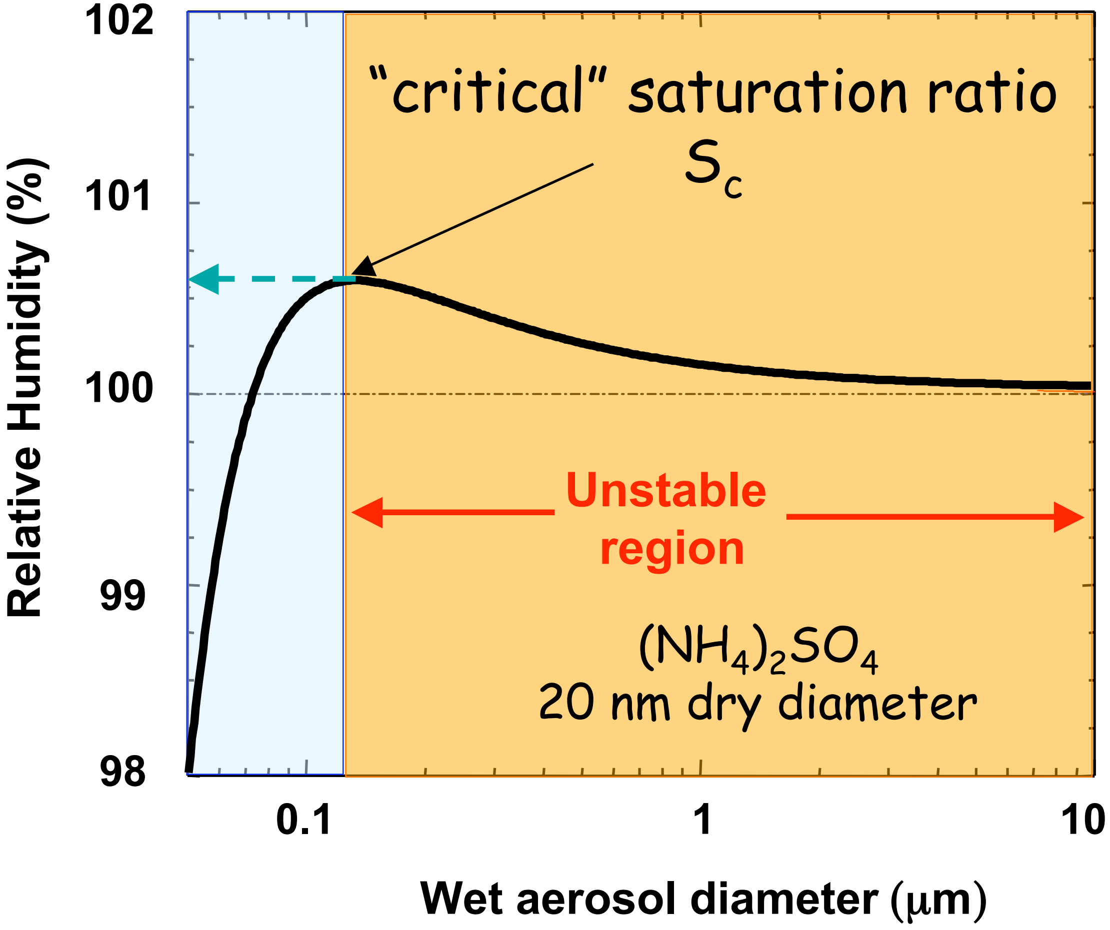


Every particle has a characteristic S_c

S_c depends on dry size, chemical composition and surface tension.

When does an aerosol particle act as a CCN ?

When **ambient** saturation is above S_c **the particle becomes unstable** and forms a droplet. Only then it can be a CCN.



S_c depends on dry size, chemical composition and surface tension.

$$\boxed{\frac{S_c - 100}{100}} = \left(\frac{4A^3}{27B}\right)^{1/2}$$

S_c, critical supersaturation

A: "surface tension" parameter

B: "solute" parameter

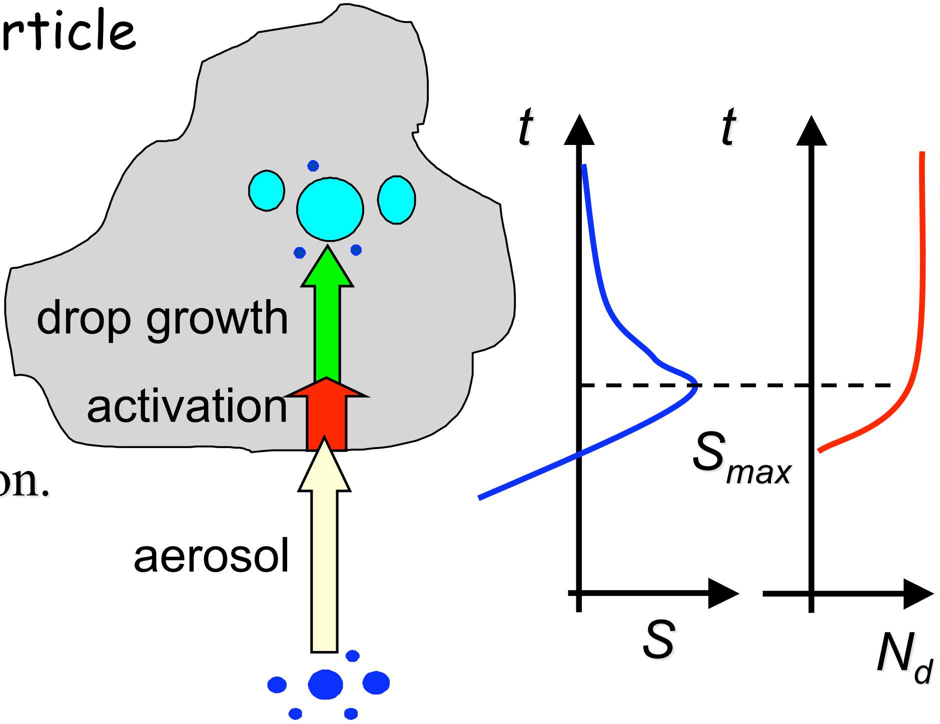
Steps for building a “mechanistic” parameterization

Input: P,T, vertical velocity, particle characteristics.
Output: Cloud droplet number

How:

Determine CCN (s)

- Use a aerosol model to predict size distribution and chemical composition.
- Köhler theory for computing CCN properties.



Determine s_{max}

- Derive expression for the condensational growth of CCN; include within the supersaturation balance for the parcel, and solve for the maximum.

  Challenge: to derive an expression of the condensation rate at S_{max}.
  Our solution: “Population splitting” (Nenes and Seinfeld, JGR, 2003)

$N_d = CCN(s_{max})$

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).

Water vapor condensation from kinetically "limited" CCN

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

Water vapor condensation from CCN that "instantaneously" activate

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

"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).

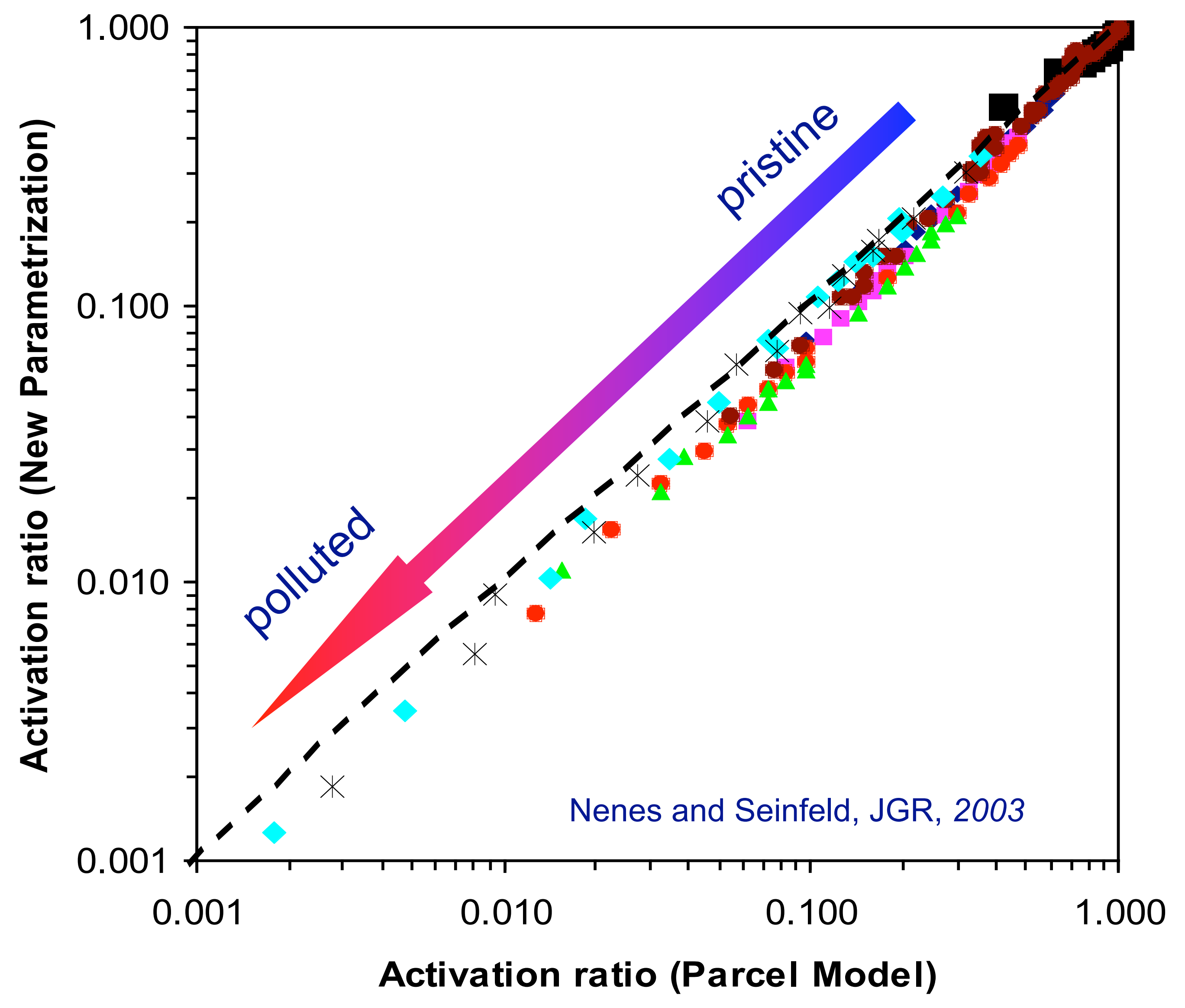
Examples:

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)

Evaluation against detailed cloud model



In-situ data evaluation : the “real” test

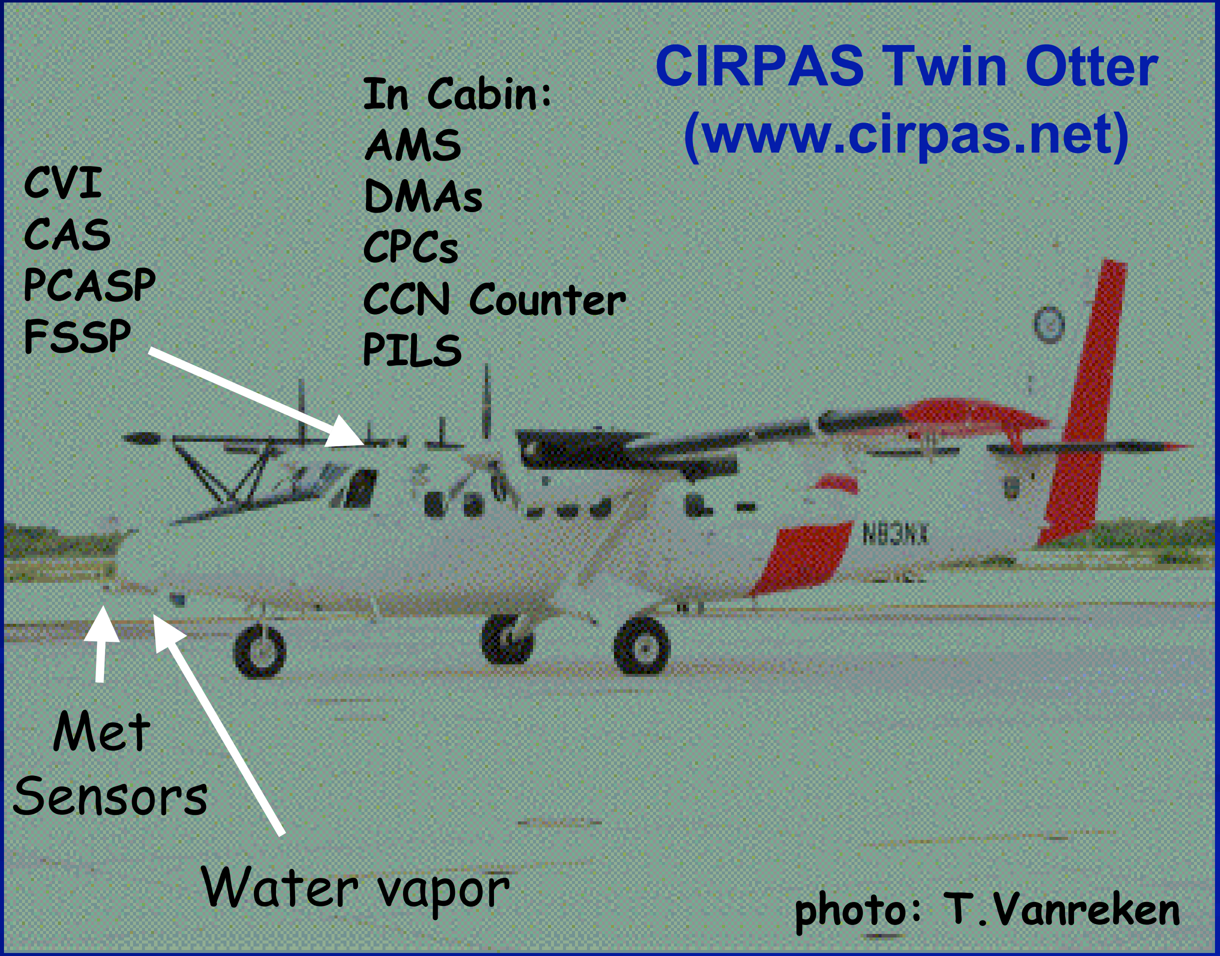
Airborne platforms are a major “workhorse” for producing the aerosol-cloud datasets we need for parameterization evaluation and development.

CIRPAS Twin Otter

NOAA P3

Evaluate parameterizations with in-situ aerosol/cloud microphysical measurements.

(Are they "good enough" for real clouds?)



Cloud droplet concentration

FSSP, CAS

Aerosol number concentration

CPC

Aerosol size distribution

DMA, PCASP, APS

Aerosol composition

AMS, PILS

Updraft velocity

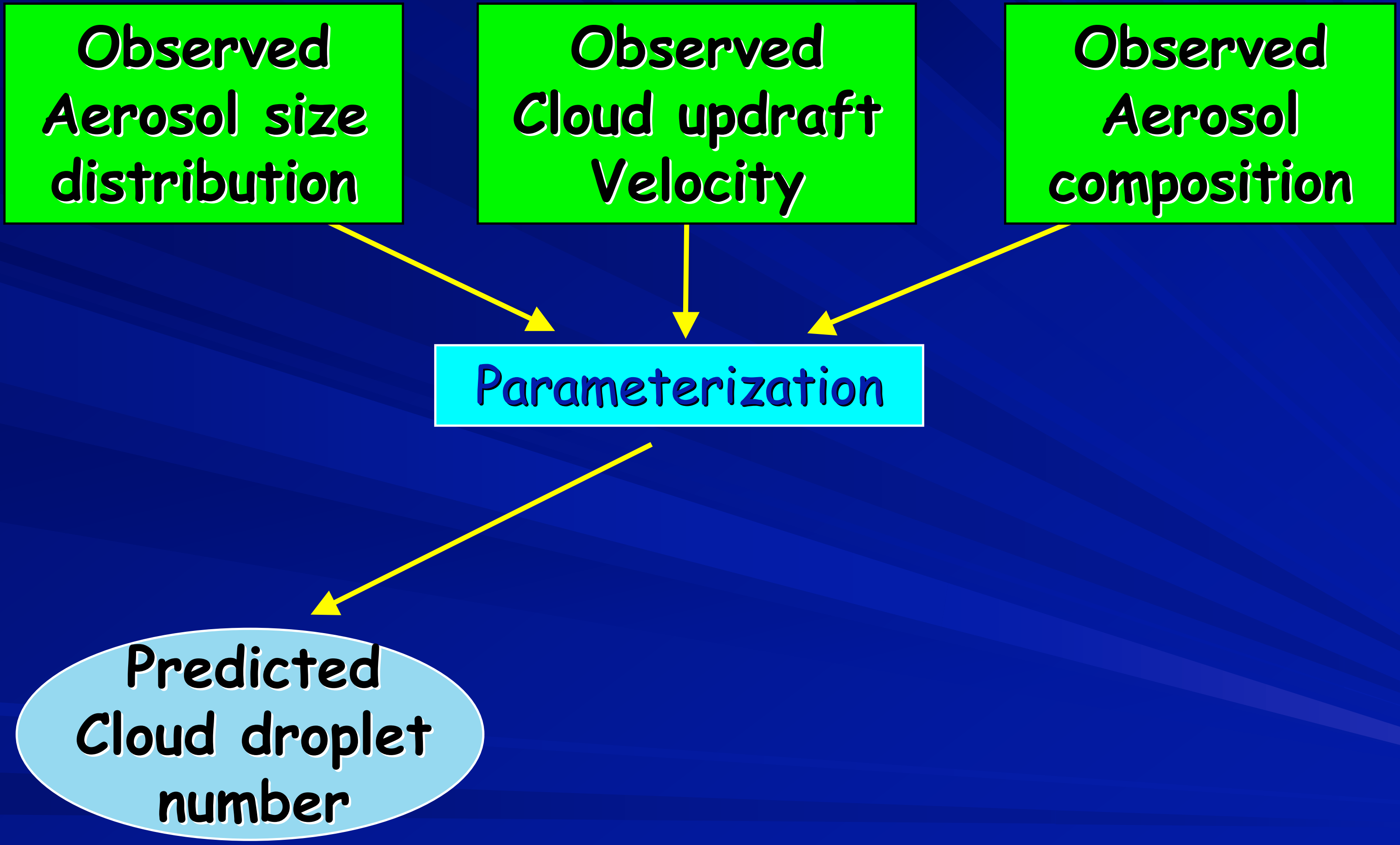
Parameterization Evaluation
CDNC "closure"

Observed
Aerosol size
distribution

Observed
Cloud updraft
Velocity

Observed
Aerosol
composition

Parameterization Evaluation
CDNC "closure"



Observed Aerosol size distribution

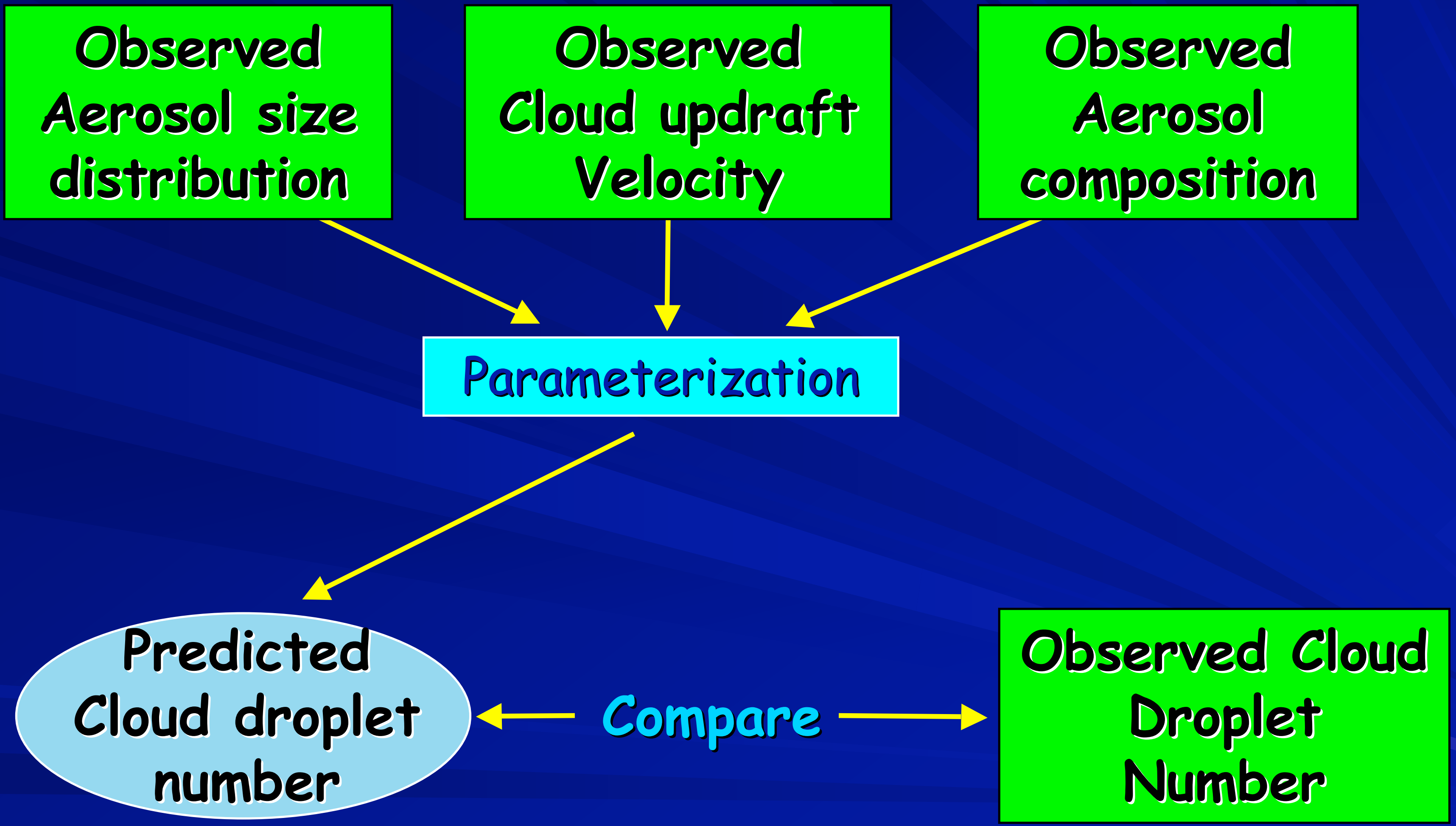
Observed Cloud updraft Velocity

Observed Aerosol composition

Parameterization

Predicted Cloud droplet number

Parameterization Evaluation
CDNC “closure”



Observed Aerosol size distribution

Observed Cloud updraft Velocity

Observed Aerosol composition

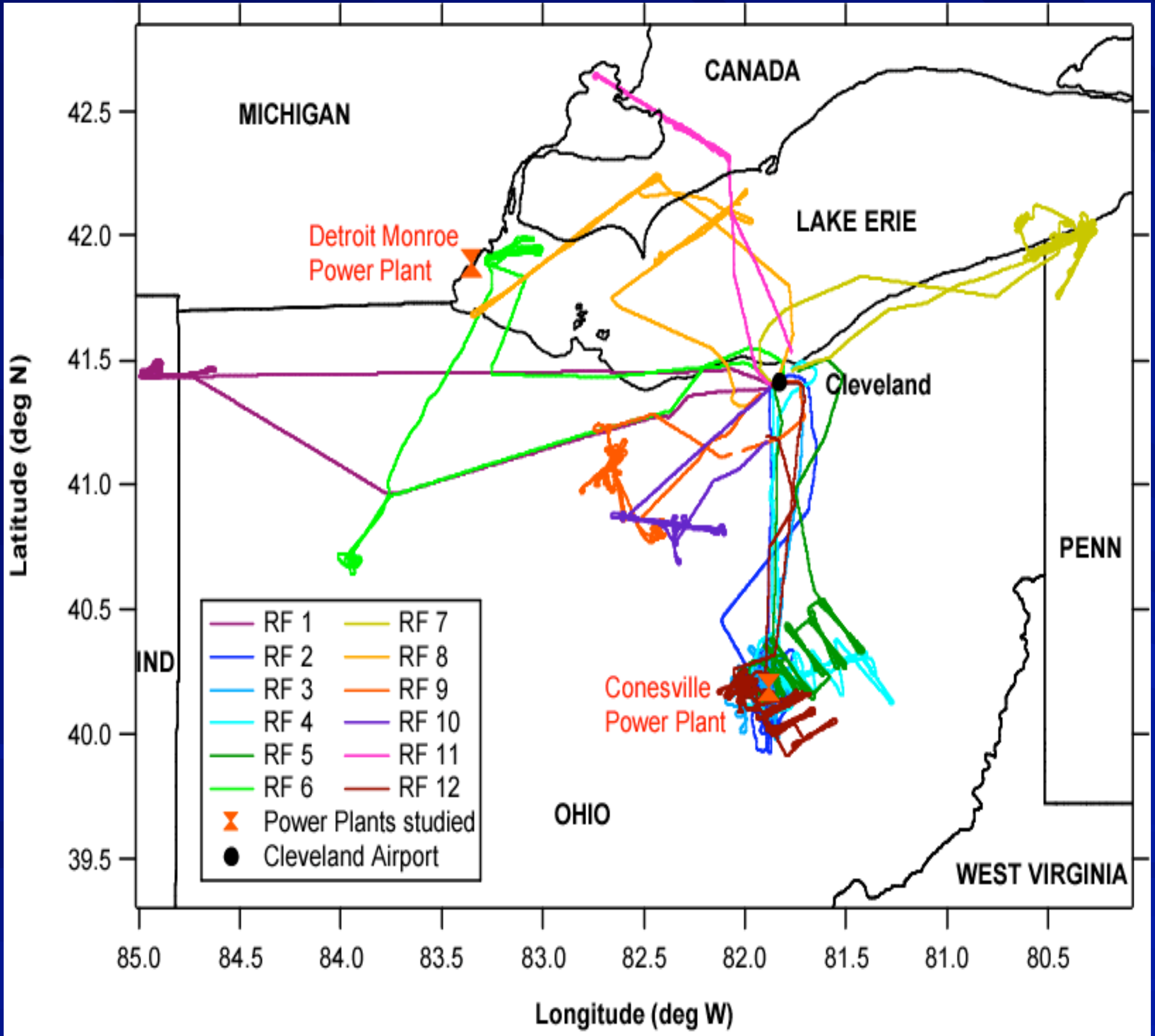
Parameterization

Predicted Cloud droplet number

Compare

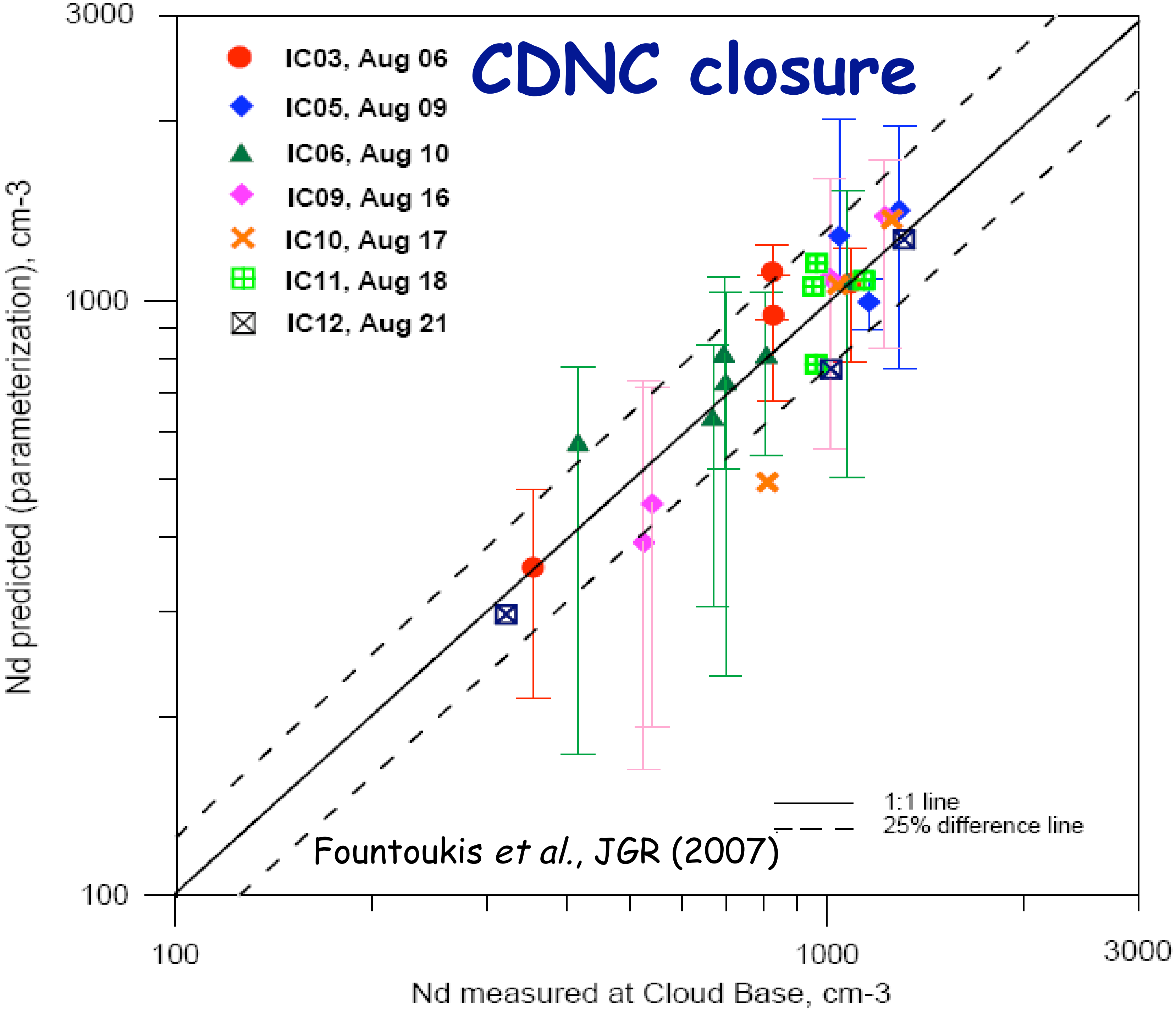
Observed Cloud Droplet Number

CDNC closure during ICARTT (Aug.2004)



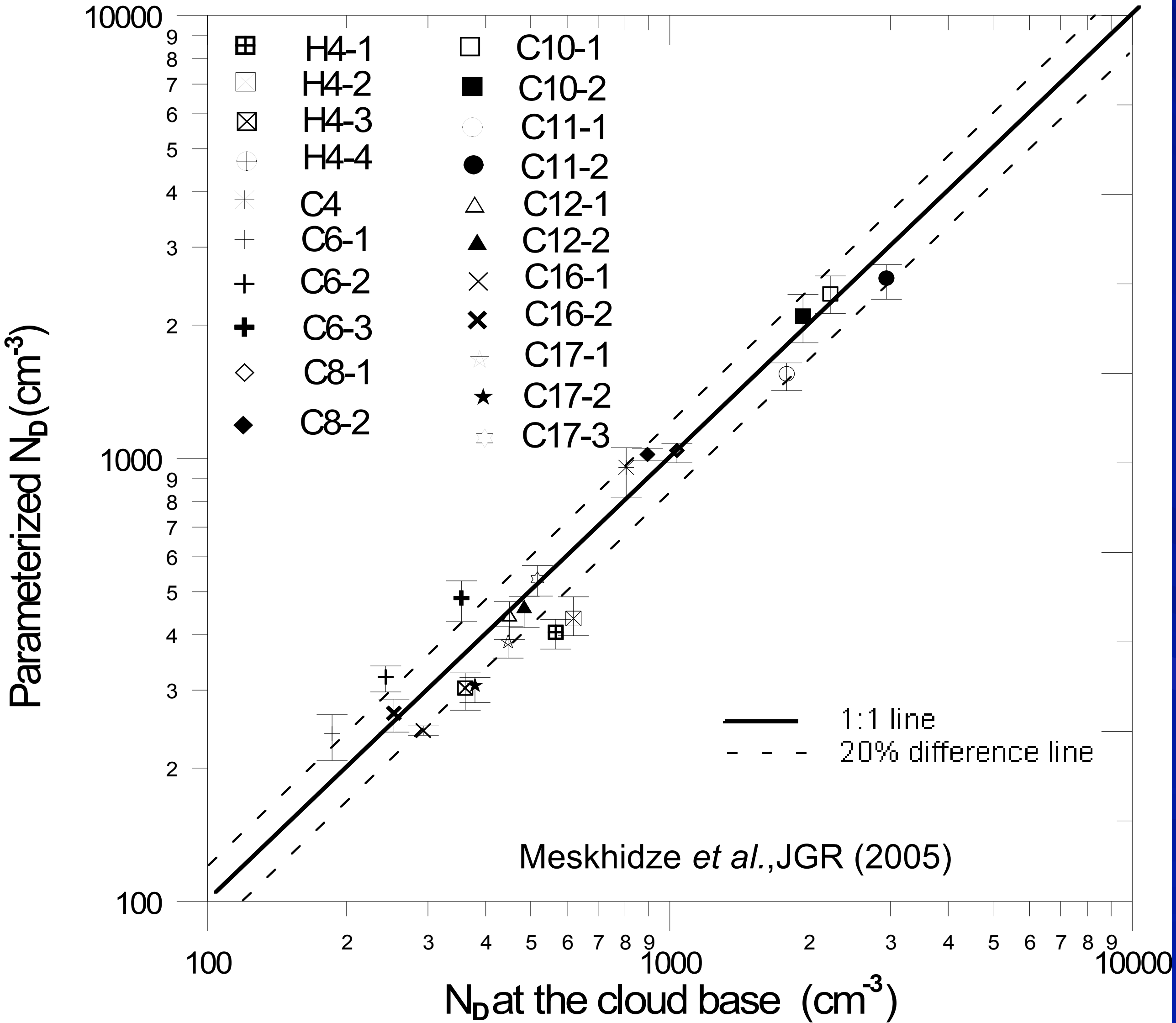
- Cumuliform and Stratiform clouds sampled
- Investigate the effect of power plant plumes on clouds

CDNC closure



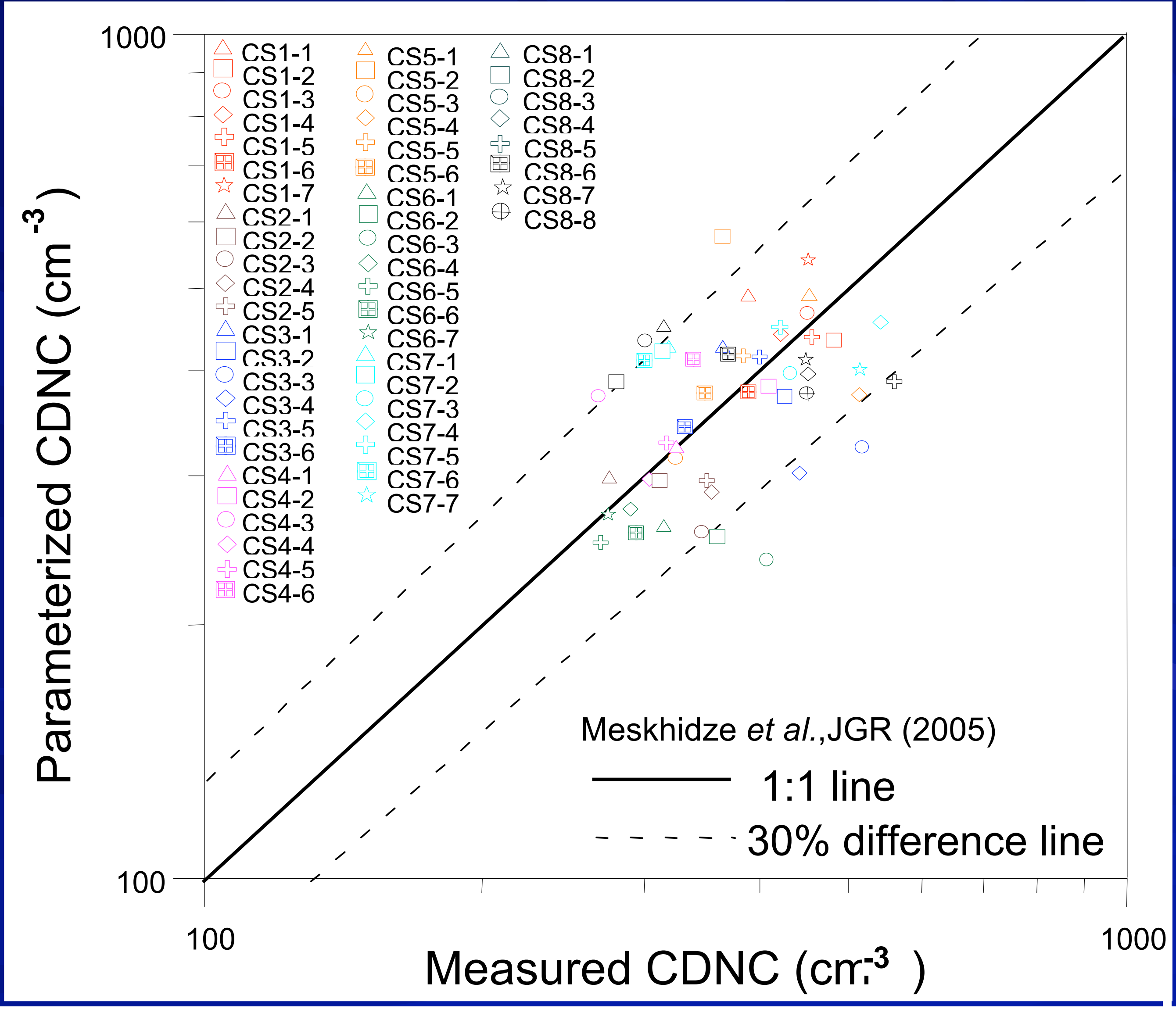
Fountoukis *et al.*, JGR (2007)

CRYSTAL-FACE (2002)
Cumulus clouds



Meskhidze *et al.*,JGR (2005)

CSTRIPE (2003)
Coastal Stratocumulus

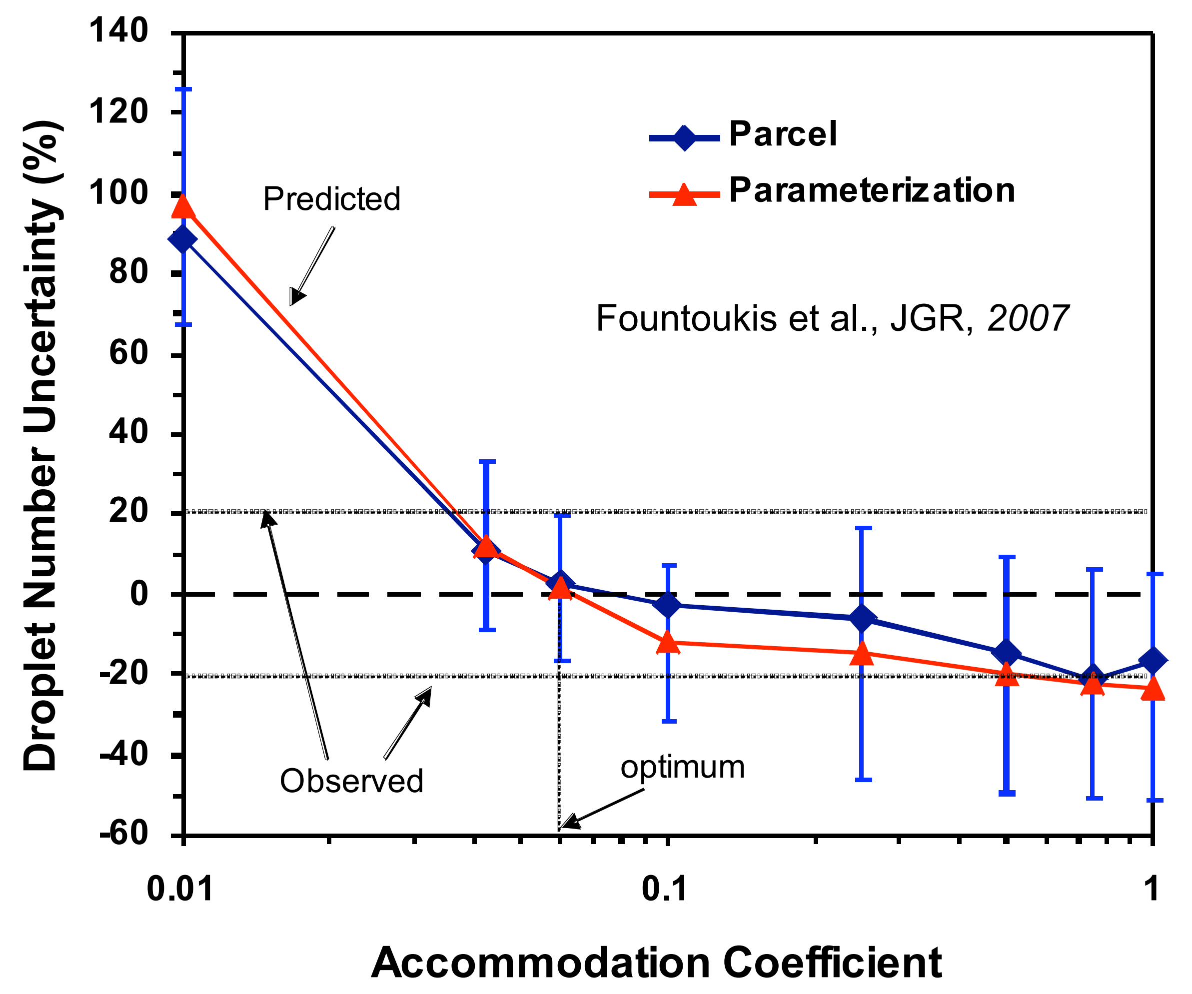


What we have learned from CDNC closure studies

- "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").

Sensitivity of droplet closure to kinetic parameters (*a*)

CIRPAS Twin Otter



Fountoukis et al., JGR, *2007*

ICARTT (2004)

Optimum closure obtained for α between
0.03 – 1.0

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

Parameterization and parcel model exhibit the same sensitivity.

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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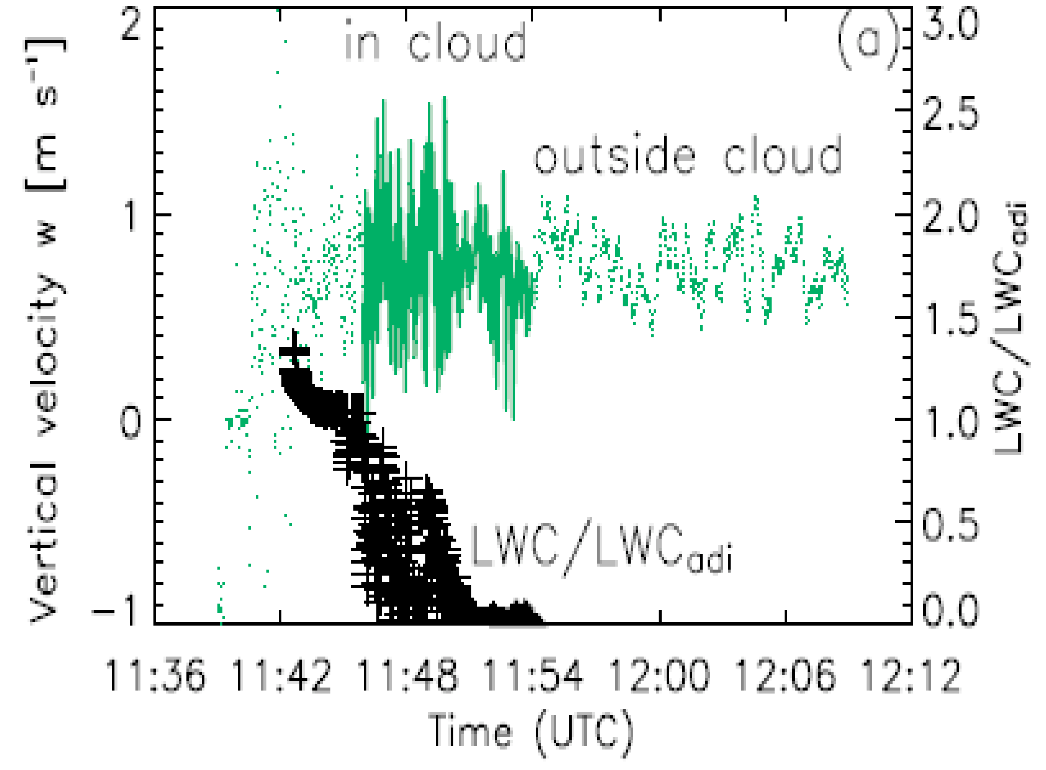
- Very difficult to address… but not impossible.

Real Clouds are not Adiabatic



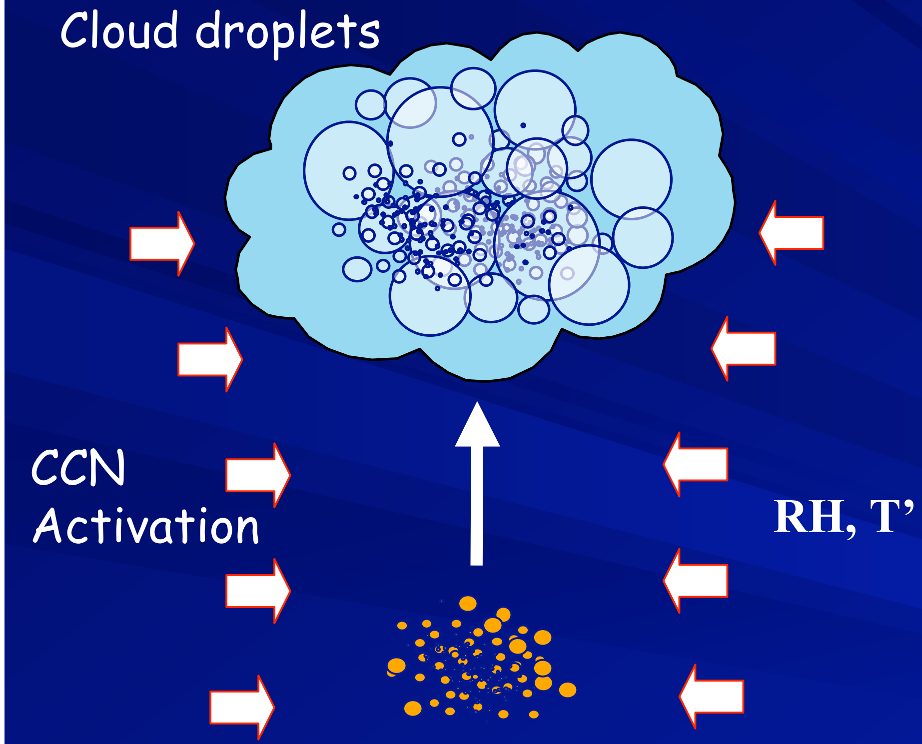
Peng, Y. et al. (2005). J. Geophys. Res., 110, D21213

- 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

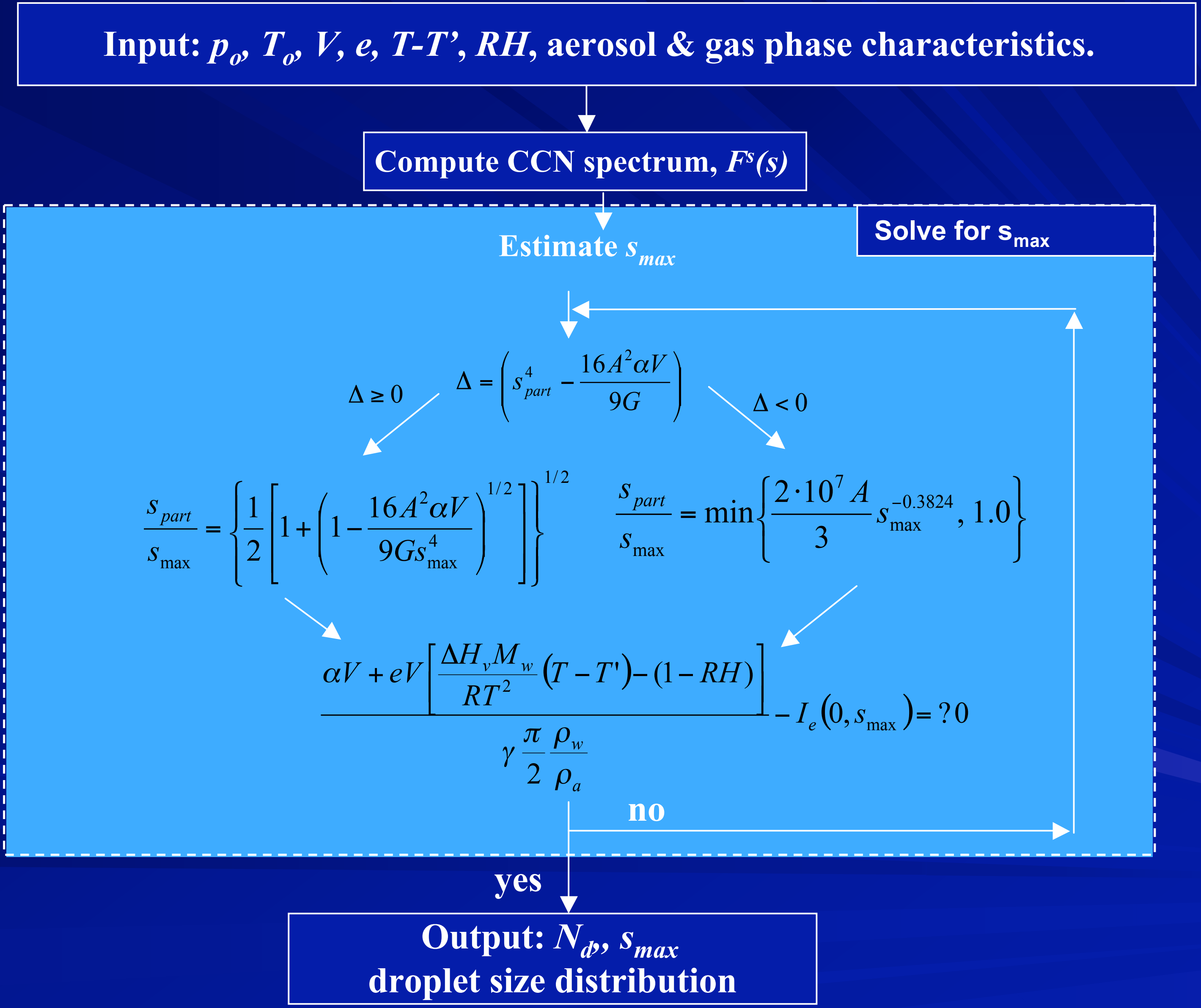
Barahona and Nenes (2007)
Droplet formation in entraining clouds



- 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.

Using the parameterization



Input: p_o, T_o, V, e, $T\text{-}T'$, RH, **aerosol & gas phase characteristics.**

Compute CCN spectrum, $F^s(s)$

Solve for s_{max}

Estimate s_{max}

$$\Delta = \left(s_{part}^4 - \frac{16A^2\alpha V}{9G}\right)$$

$\Delta \geq 0$

$$\frac{s_{part}}{s_{\max}} = \left\{\frac{1}{2}\left[1 + \left(1 - \frac{16A^2\alpha V}{9Gs_{\max}^4}\right)^{1/2}\right]\right\}^{1/2}$$

$\Delta < 0$

$$\frac{s_{part}}{s_{\max}} = \min\left\{\frac{2\cdot 10^7 A}{3} s_{\max}^{-0.3824},\ 1.0\right\}$$

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

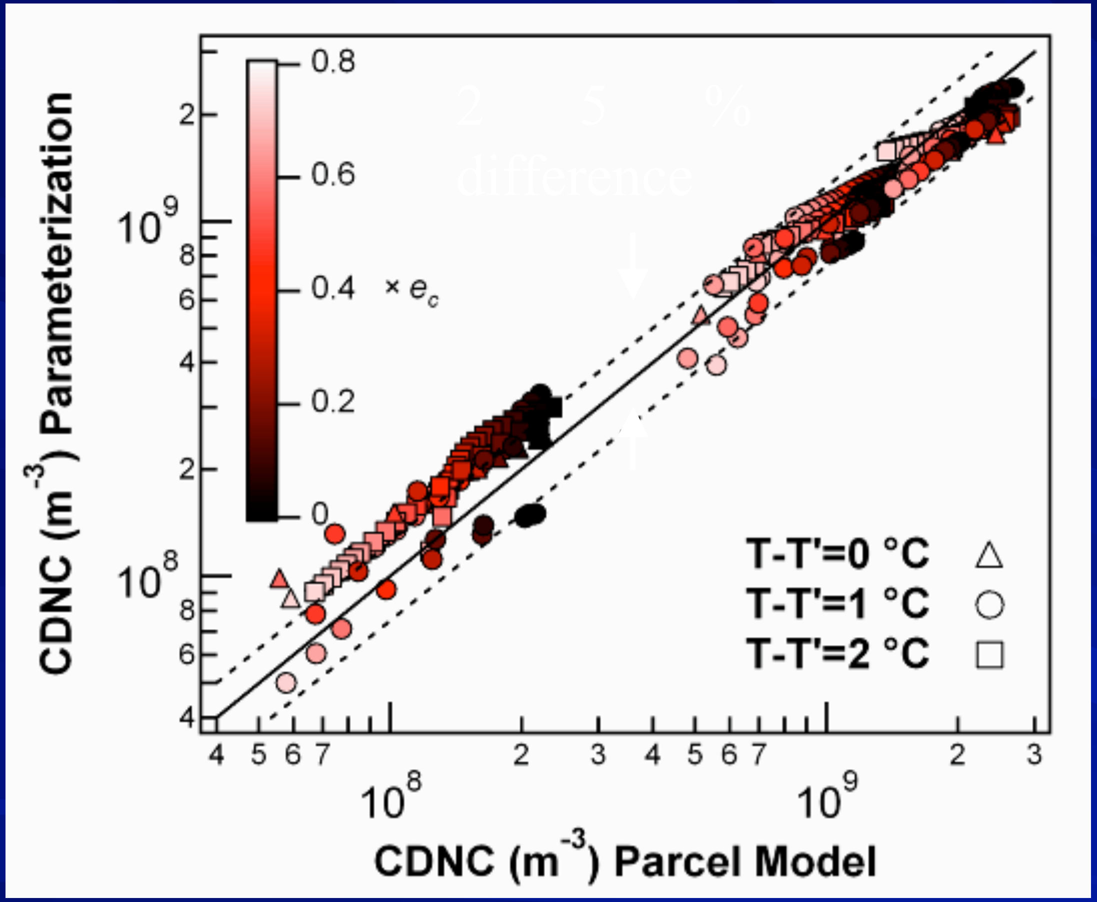
no

yes

Output: N_d, s_{max}
droplet size distribution

Barahona and Nenes, (2007)

Entraining Parameterization vs. 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.

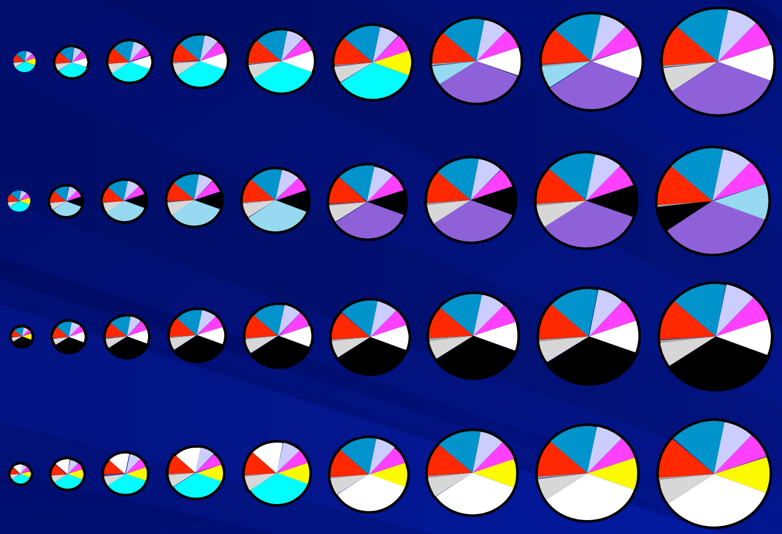
- Comparison with detailed numerical model.
- Parameterization closely follows the parcel model
- Mean relative error ~3%.
- > 10^4 times faster than numerical parcel model.

Barahona and Nenes, (2007)

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, complete 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.

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

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… but we'll talk about it on Wednesday.

Overall Summary (for now)

- Droplet formation parameterizations are at the point where they can explicitly consider all the chemical "complexities" of CCN calculations and droplet growth kinetics.
- They can even begin incorporating effects of dynamics (entrainment, variable updraft).
- Observations should provide the "constraints" of organic properties (more on this on Wednesday).
- 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 (and not impossible).

THANK YOU!