



**GCM Assessments of  
Aerosol-Cloud Interactions:  
Ongoing and future work**

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# Goal: assess the uncertainties

Is CCN prediction theory and existing aerosol-cloud parameterizations "good enough"?

How important are organic species for cloud formation?

Do they:

- Provide solute? What is their MW, solubility?
- Affect droplet surface tension? How much?
- **Affect droplet growth rate? When/how much?**

How much "inherent" indirect effect uncertainty is associated with all these? How does it compare with the uncertainty associated with cloud or large-scale dynamics?

**"Observations can provide the constraints needed."**  
But *how?* (The devil is always in the details).

# Global Modeling Framework (#1)

## General Circulation Model

- NASA GISS II' GCM
- 4'x5' 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  $\mu\text{m}$ .
- 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 (#1)

## Cloud droplet number calculation

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

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

## Autoconversion

Khairoutdinov & Kogan (2000), DelGenio (1996)

## Emissions

Current day, preindustrial

## In-cloud updraft velocity

- Prescribed (marine:  $0.25-0.5 \text{ ms}^{-1}$ ; continental:  $0.5-1 \text{ ms}^{-1}$ ).
- Large-scale TKE in a  $4' \times 5'$  grid is too separated from the cloud scale.

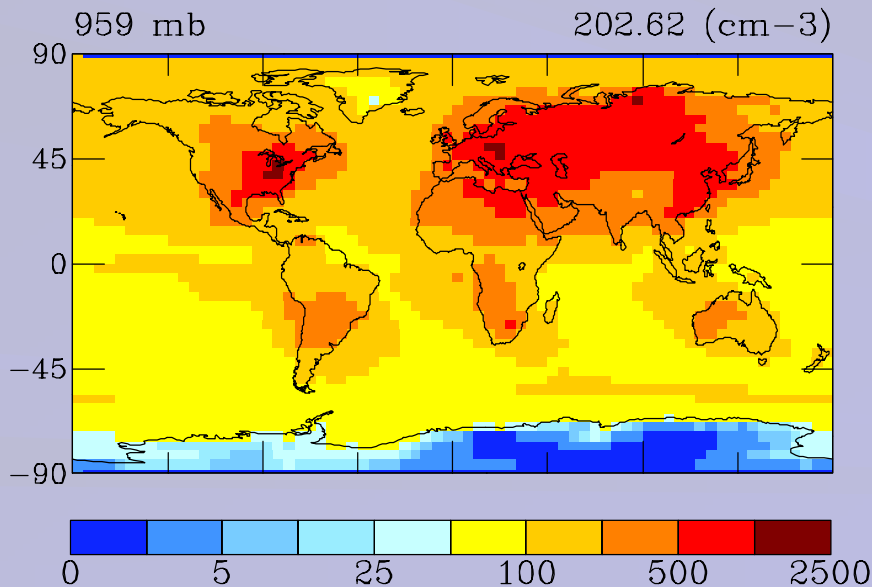
# Current Day Simulation (annual average)

## Conditions for aerosol-cloud linking

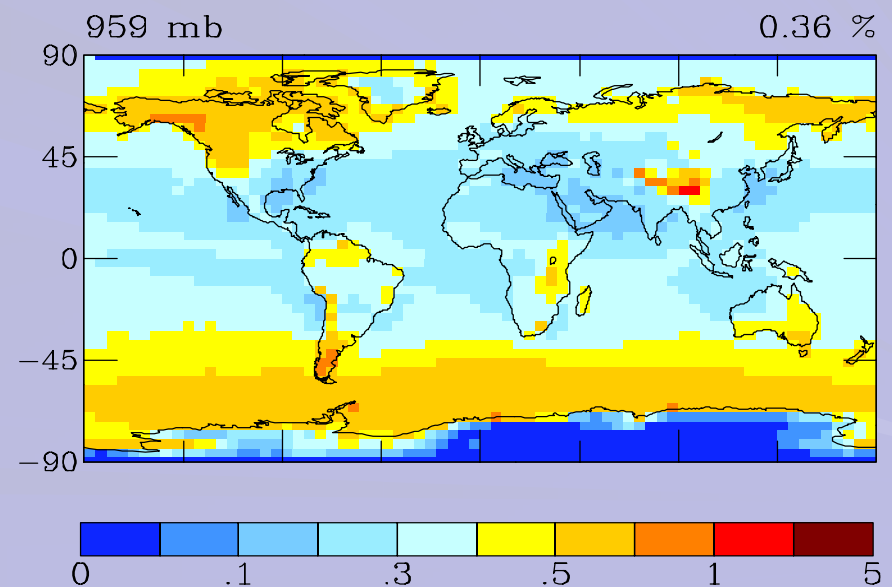
Cloud base updraft ( $\text{m s}^{-1}$ ): 0.25 (marine), 0.5 (continental)

Water vapor uptake coefficient: 0.06

### Cloud droplets ( $\text{cm}^{-3}$ )

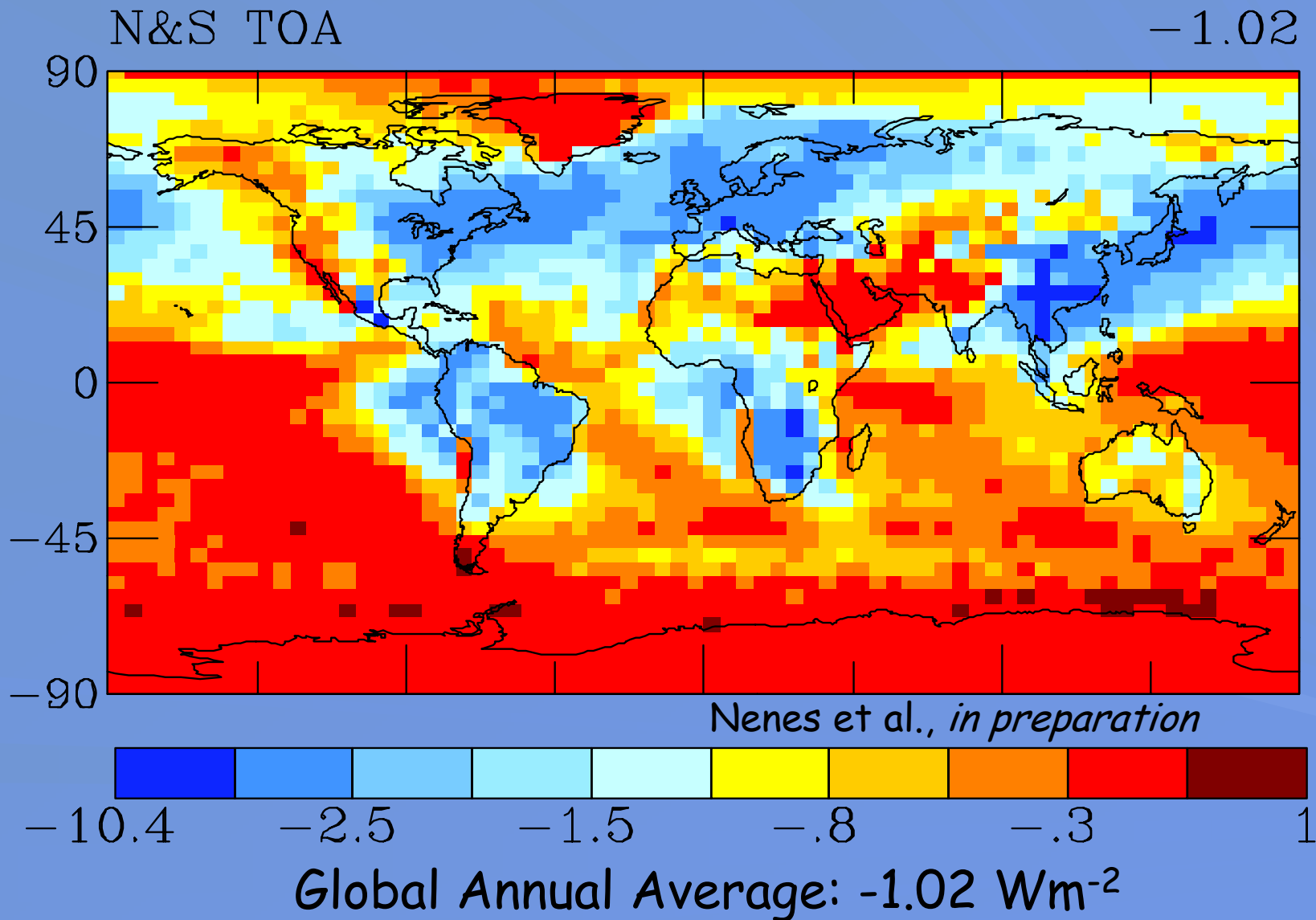


### Cloud $s_{max}$ (%)

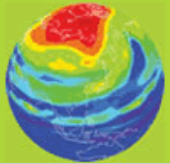


Nenes et al., *in preparation*

# Current day-preindustrial indirect forcing



# Global Modeling Framework (#2)



NASA Global Modeling Initiative (GMI)



GODDARD SPACE FLIGHT CENTER

<http://gmi.gsfc.nasa.gov/gmi.html>

- 3-D chemistry-transport model (CTM)
- Multiple "packages" for e.g., chemistry & aerosol
- Meteorological inputs from GCMs (**GEOS-4 FVGCM** & **GISS-II'**) or data assimilation systems (NASA **DAO**)
- Any vertical resolution; horizontal resolution: **4°x5°**
- Multi-year assessment simulations
- We've put in the aerosol-cloud interactions and radiative calculations.

# Sensitivity of Indirect Forcing to met field ( $\text{W m}^{-2}$ )

Mean -0.98 Max 0 Min -14.7

Mean -0.87

• Depending on the droplet activation parameterization and the met field used, global annual indirect forcing ranges:  
 $-0.75 \text{ W/m}^2$  to  $-1.27 \text{ W/m}^2$

• Different met fields lead up to 30% (Global average) variability in indirect forcing calculations.

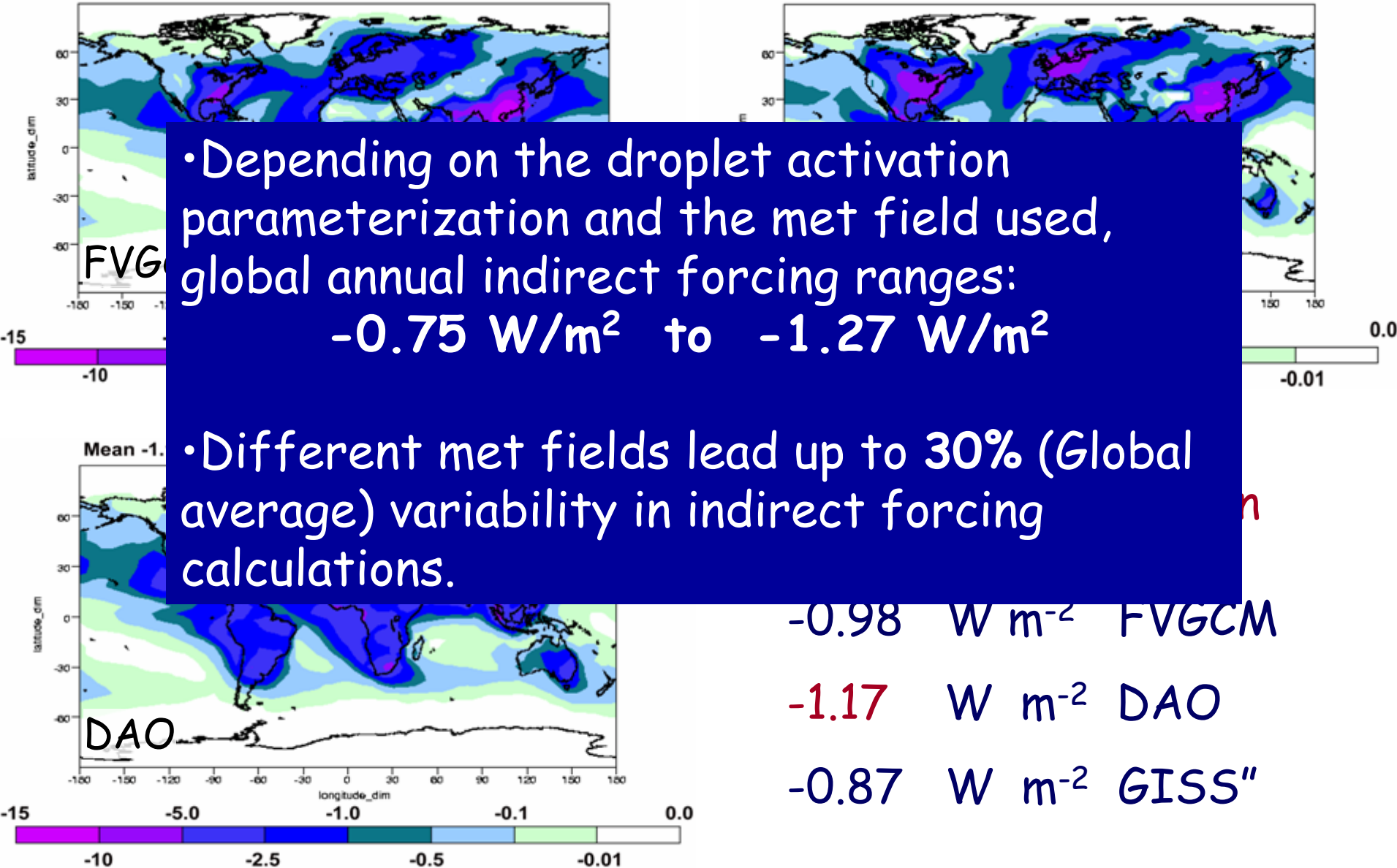
$-0.98 \text{ W m}^{-2}$  FVGCM

$-1.17 \text{ W m}^{-2}$  DAO

$-0.87 \text{ W m}^{-2}$  GISS"

Mean -1.

DAO

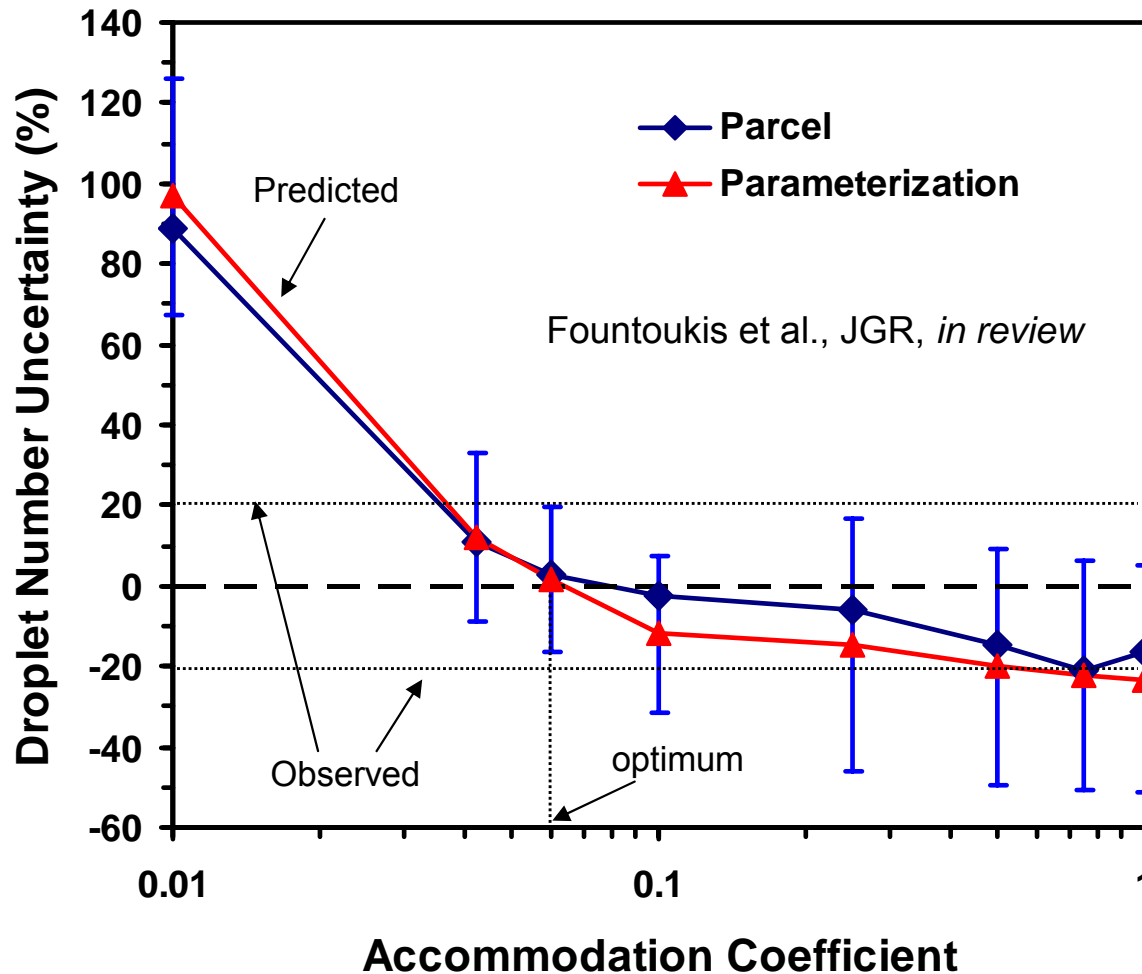




# Constraining the water vapor uptake coefficient



CIRPAS Twin Otter

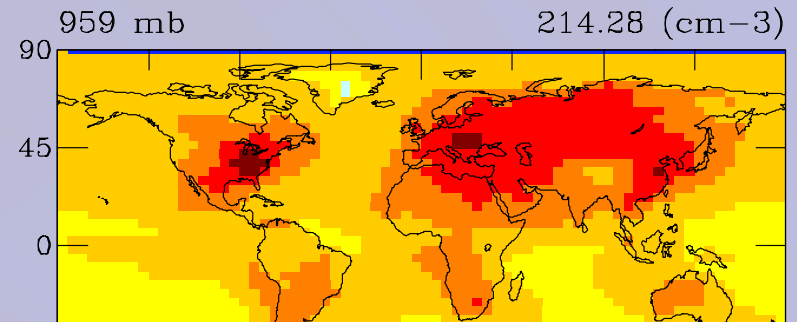
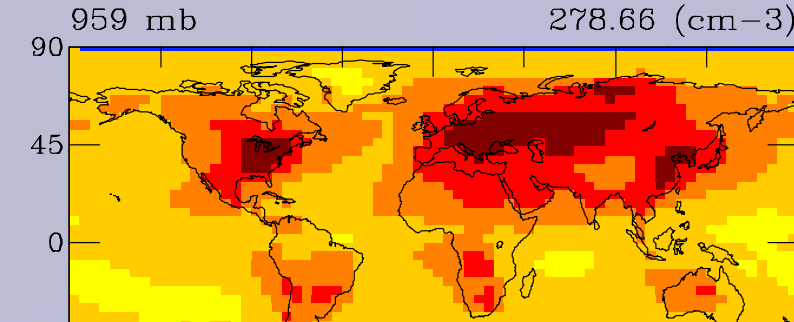


Cloud droplet closure during ICARTT (2004)

Optimum closure obtained for  $\alpha$  between 0.03 - 1.0

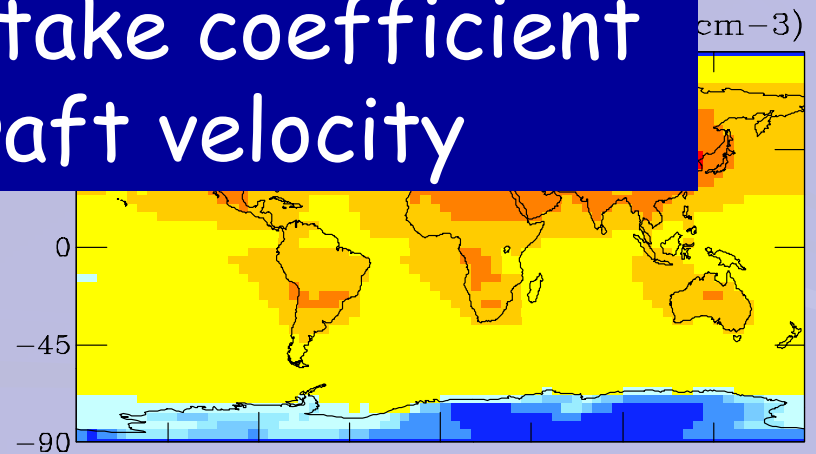
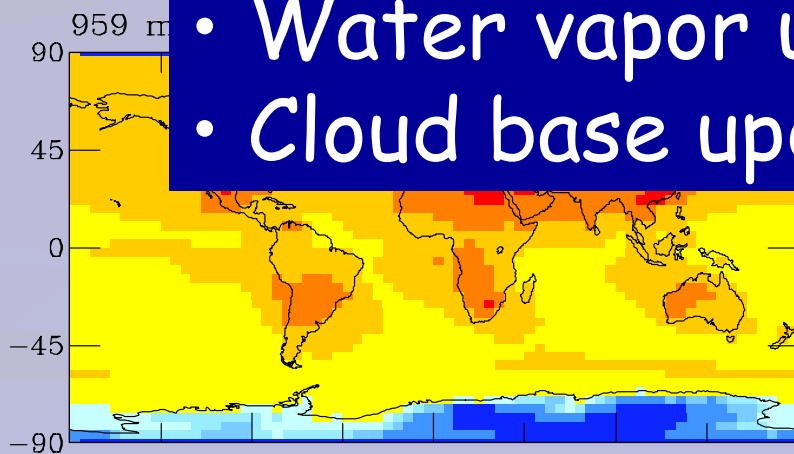
This is the range we use in our GCM simulations

# Current day simulation: Sensitivity of $N_d$ to $w$ and $a$



Droplet number is very sensitive to changes in

- Water vapor uptake coefficient
- Cloud base updraft velocity

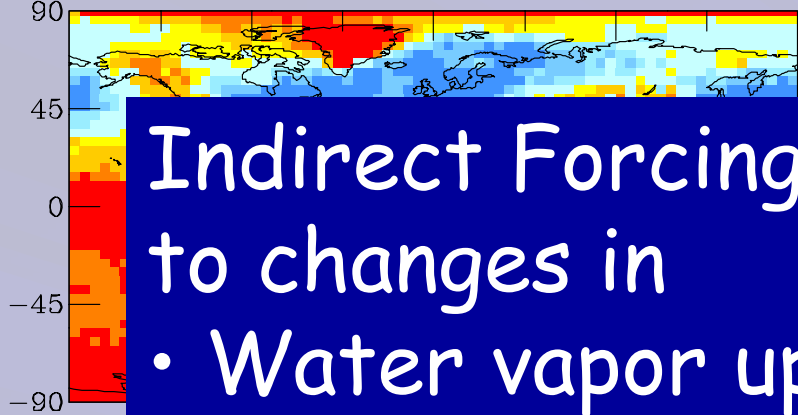


$$w_{\text{ocean}} = 0.25 \text{ ms}^{-1}; w_{\text{land}} = 0.5 \text{ ms}^{-1}$$
$$\alpha = 0.06$$

$$w_{\text{ocean}} = 0.25 \text{ ms}^{-1}; w_{\text{land}} = 0.5 \text{ ms}^{-1}$$
$$\alpha = 1.0$$

# Preindustrial-Current Day forcing: Sensitivity to $w$ and $a$

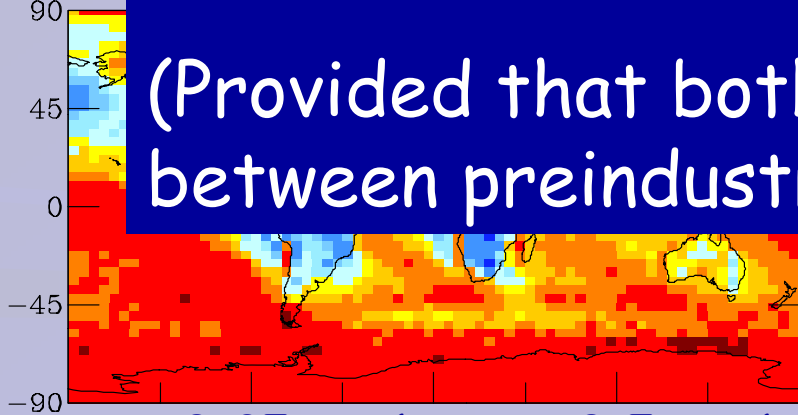
N&S TOA -1.03



$w_{ocean} = 0.25 \text{ ms}^{-1}; w_{land} = 0.5 \text{ ms}^{-1}$

$\alpha = 0.06$

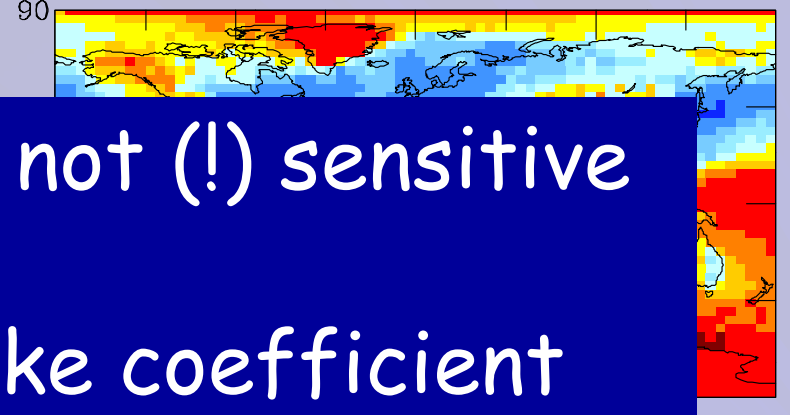
N&S TOA -1.02



$w_{ocean} = 0.25 \text{ ms}^{-1}; w_{land} = 0.5 \text{ ms}^{-1}$

$\alpha = 1.0$

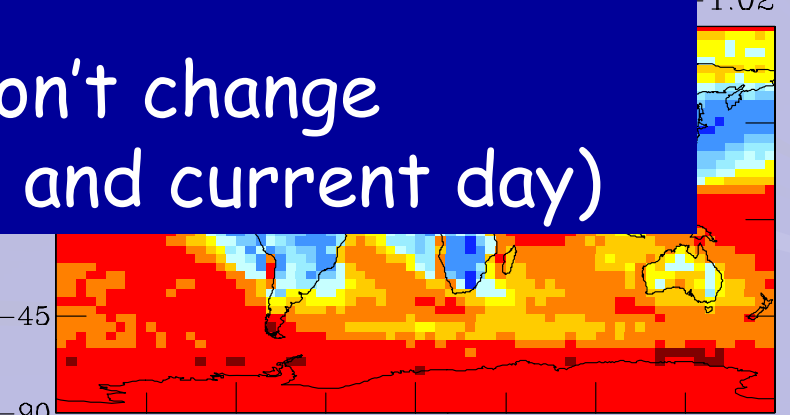
N&S TOA -1.02



$w_{ocean} = 0.25 \text{ ms}^{-1}; w_{land} = 0.5 \text{ ms}^{-1}$

$\alpha = 1.0$

N&S TOA -1.02



$w_{ocean} = 0.25 \text{ ms}^{-1}; w_{land} = 0.5 \text{ ms}^{-1}$

$\alpha = 1.0$

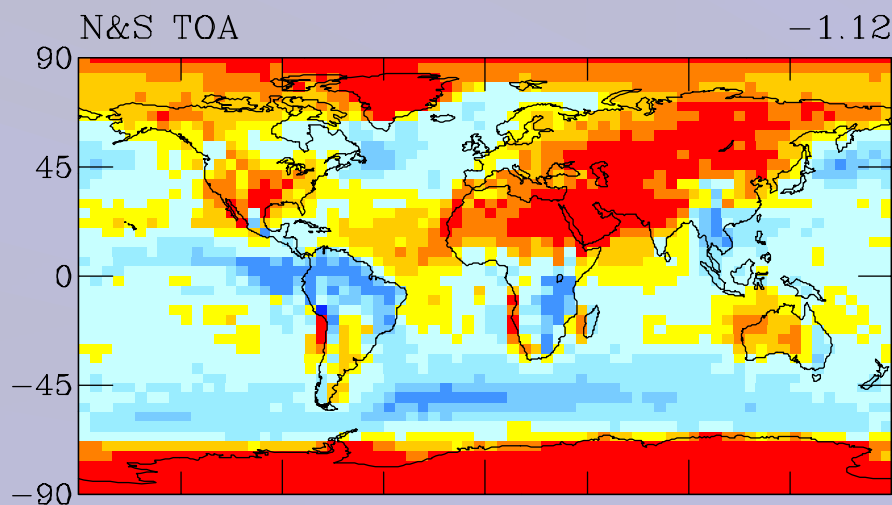
Indirect Forcing is not (!) sensitive to changes in

- Water vapor uptake coefficient
- Cloud base updraft velocity

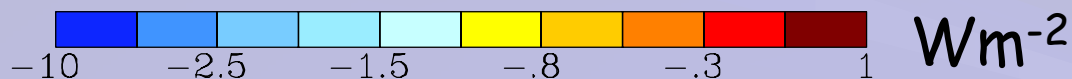
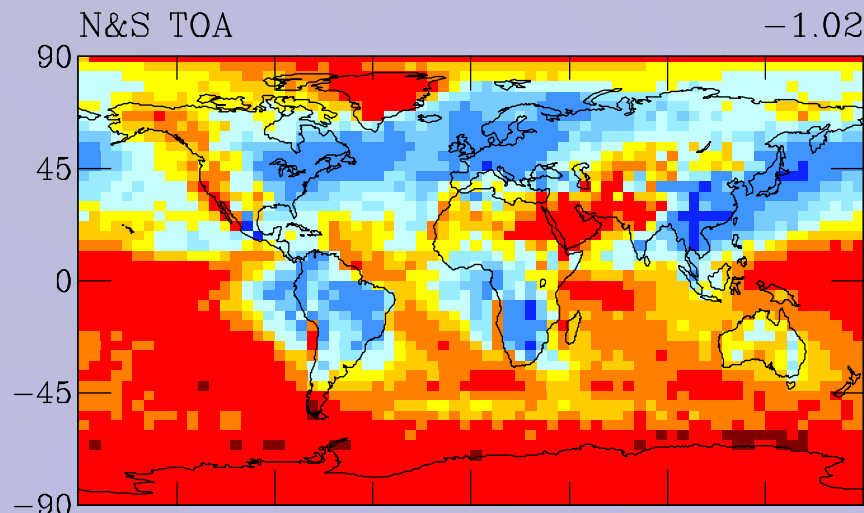
(Provided that both don't change between preindustrial and current day)

# Sensitivity of indirect forcing to the water vapor uptake coefficient

$\alpha=1.0 - \alpha=0.03$



Current-Preindust.

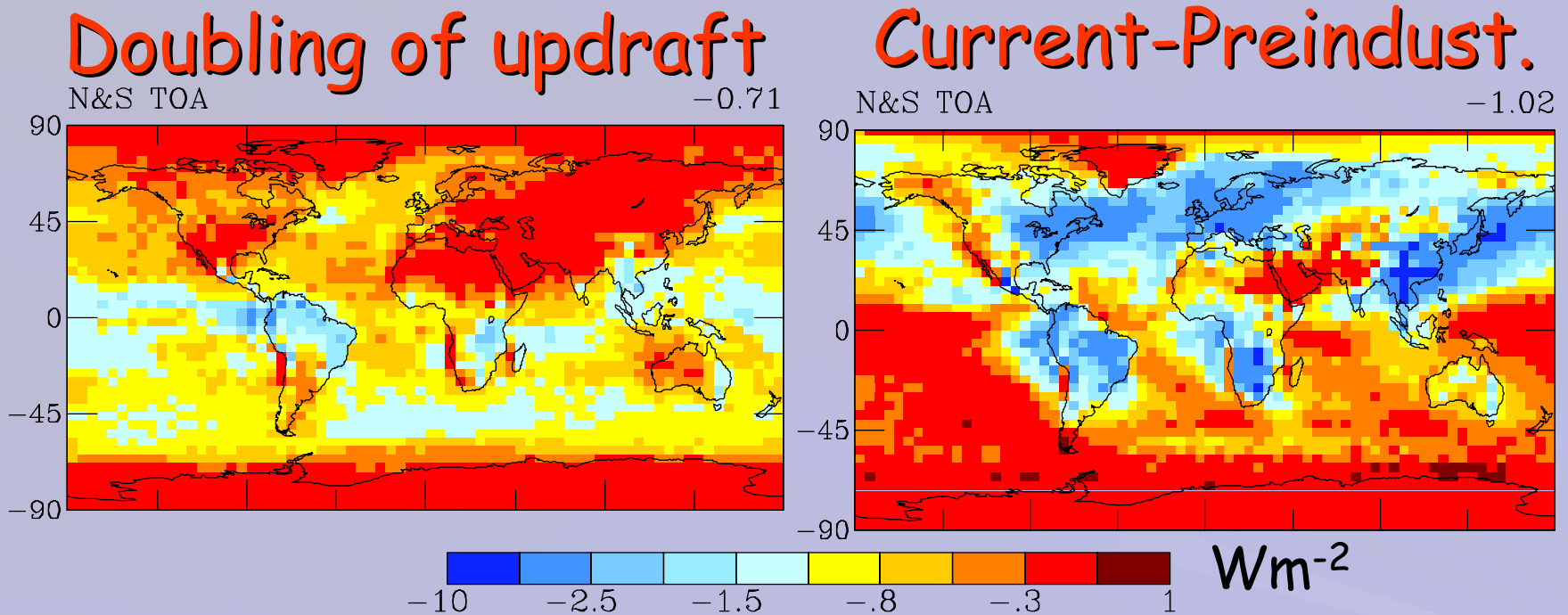


Forcing uncertainty from  $\alpha$  uncertainty ( $-1.12 W m^{-2}$ ) is as large as Present-Preindustrial change ( $-1.02 W m^{-2}$ )!

Spatial patterns are very different.

Nenes et al., *in preparation*

# Sensitivity of indirect forcing to the value of cloud-base updraft velocity



Uncertainty in indirect forcing doubling updraft velocity is a bit smaller than indirect forcing itself.  
Spatial patterns are much different.

# Is CCN prediction theory “good enough”?

## Procedure:

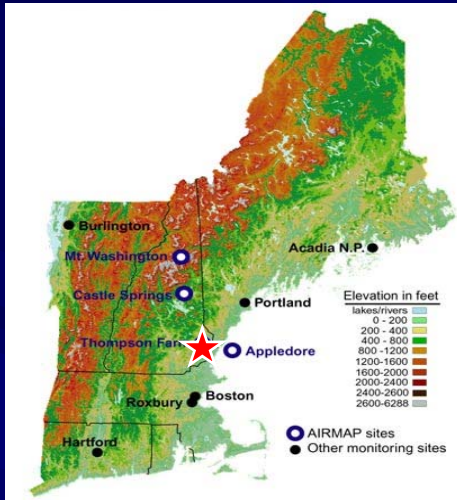
- Use in-situ data and assess CCN closure, for typical assumptions on chemical composition taken in GCMs.
- Quantify CCN prediction error
- Incorporate in global model and assess uncertainty in
  - ✓ Cloud droplet number concentration (CDNC)
  - ✓ Aerosol indirect forcing
  - ✓ Autoconversion of cloudwater to rain

Determine regions where uncertainty is small; define regions where more in-situ constraints are needed.

# In-situ Aerosol/CCN observations

## 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.



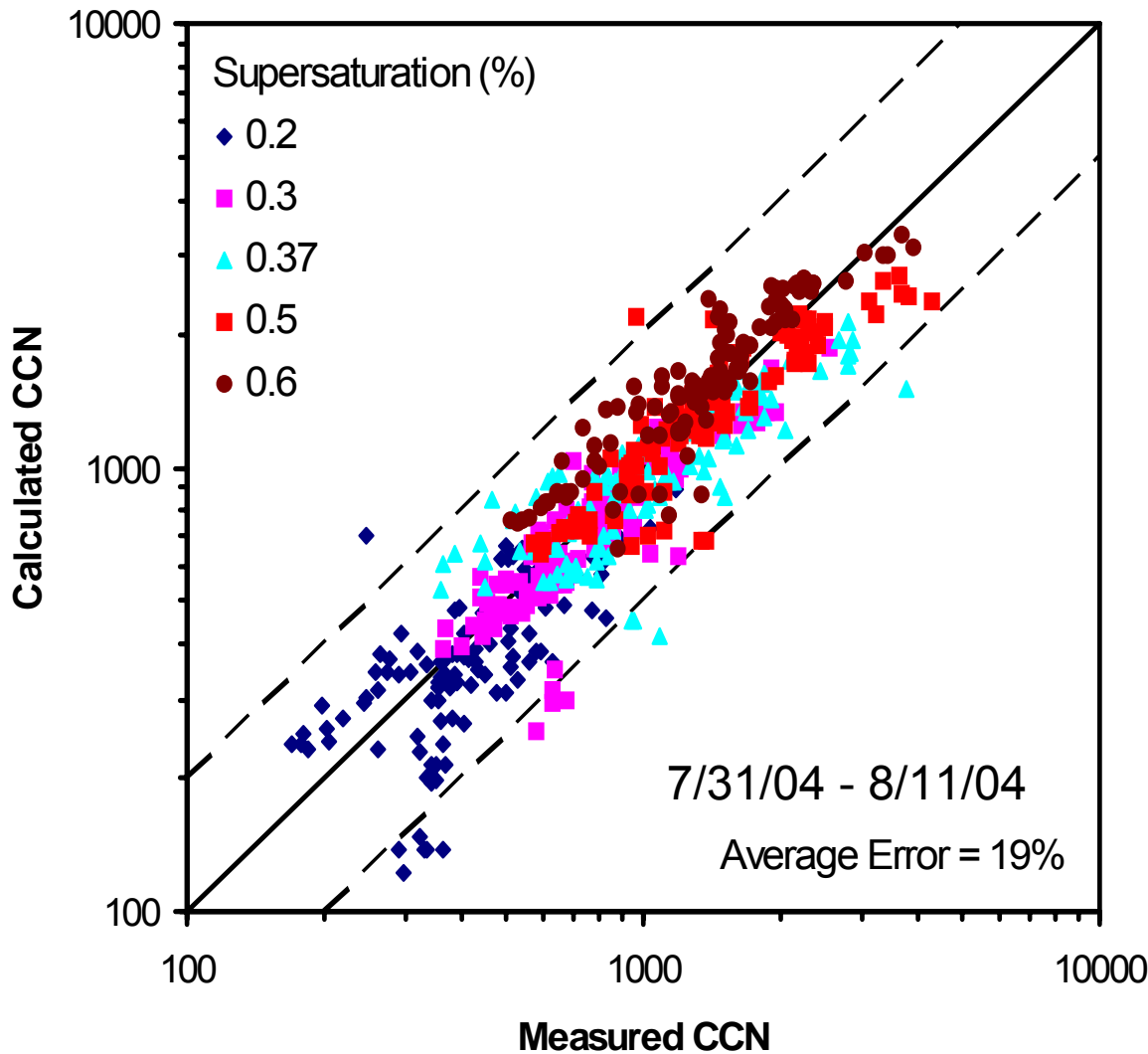
Two DMT CCN counters  
(Roberts and Nenes, AST, 2005;  
Lance et al., AST, 2006)

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



From measured size distribution and chemical composition calculate CCN concentrations.

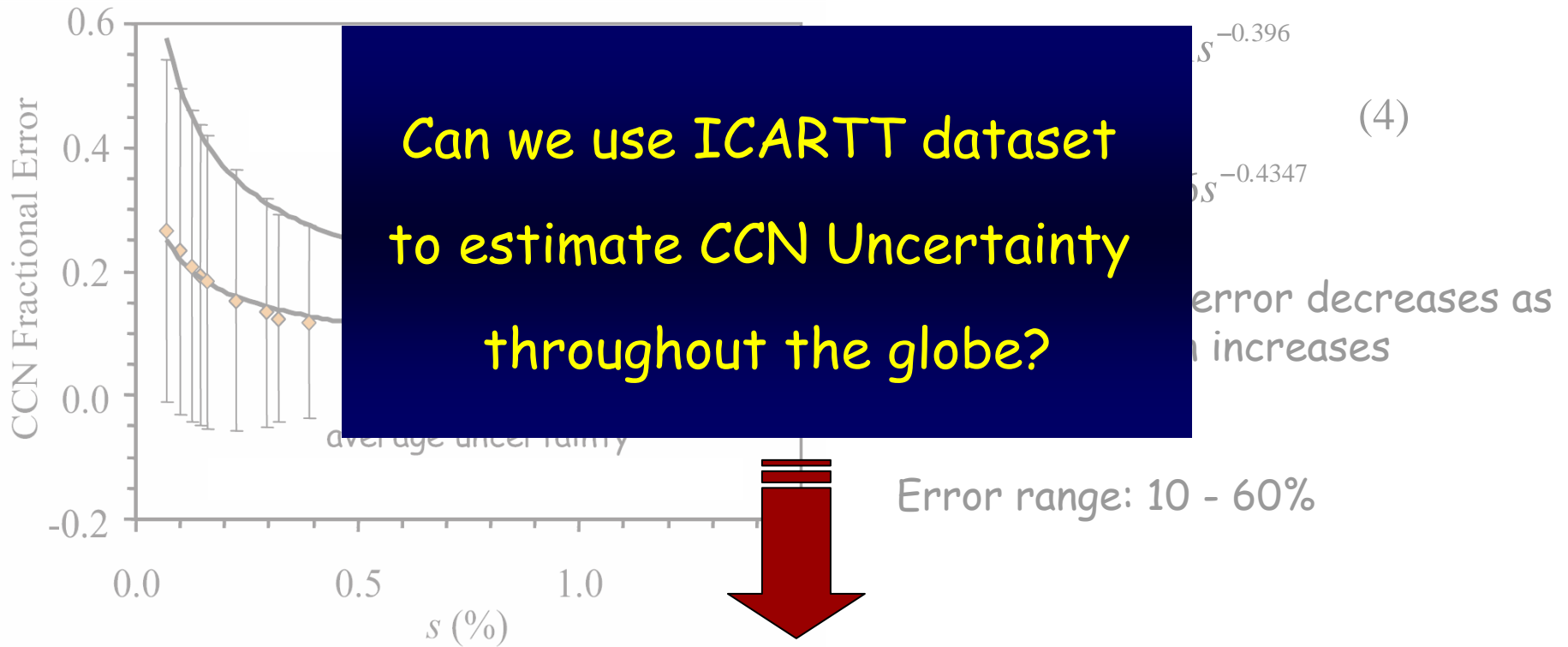
20% overprediction (average).

What does this prediction uncertainty mean for indirect forcing?



# CCN Prediction Uncertainty

ICARTT dataset



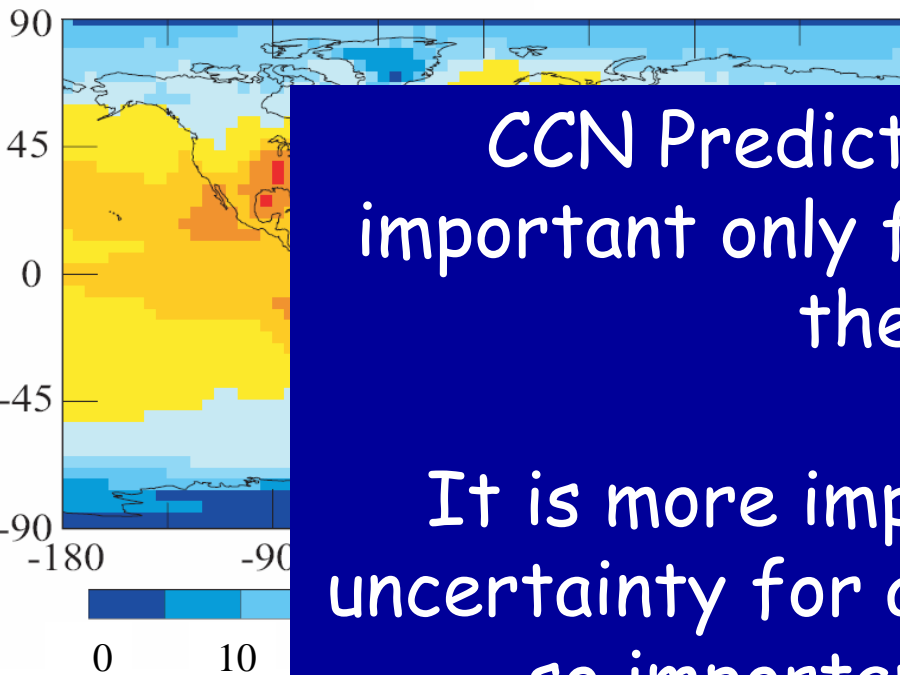
**YES!**

The degree of CCN closure is typical of polluted environments & larger than for pristine ones (Medina et al., in review). Using the dataset will provide an upper limit for CCN prediction and Indirect Forcing Uncertainty

# Prediction Uncertainties

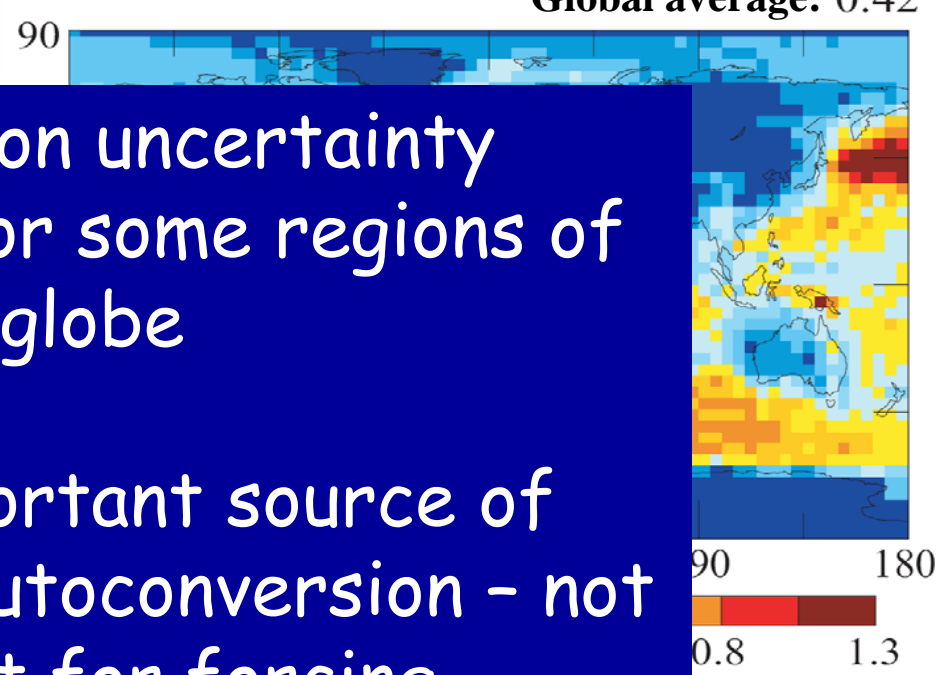
## CCN Prediction

Global average: 28%



## Indirect Forcing ( $W m^{-2}$ )

Global average: 0.42



CCN Prediction uncertainty  
important only for some regions of  
the globe

It is more important source of  
uncertainty for autoconversion - not  
so important for forcing.

Larger CCN prediction uncertainty  
is found in regions where  
in-cloud  $s_{max}$  is low

Larger uncertainty is  
predicted downwind of  
industrialized and biomass  
burning regions

# Parameterizing drizzle: how it's done now

Improved precipitation parameterizations that consider microphysics exist, and are also used,

*liquid water cloud fraction*

*average collection efficiency*

$$\left(\dot{q}_l\right)_P = -C_1 \frac{0.104 g E \rho^{4/3}}{\mu \left(N_d \rho_w\right)^{1/3}} \left(\frac{q_l}{C_1}\right)^{7/3}$$

*droplet number*

(Rotstajn, 1997)

This is a step in the right direction, but the effects of spectral broadening (droplet size distribution) are not explicitly considered.

We seek an explicit link between aerosol, activation and subsequent coalescence at the "updraft" scale.

We are doing this now by predicting droplet size distribution in the updrafts that form clouds online in the GCM.

# Parameterizing drizzle: what we are working on

Two-moment schemes developed for small-scale models can be used instead

(e.g., Cohard and Pinty, 2000; R4 and R6 schemes of Liu & Daum)

$$(\dot{q}_l)_P = - \frac{2.7 \times 10^{-2} \rho q_c \left( \frac{1}{16} \times 10^{-20} \sigma^3 D_v - 0.4 \right)}{\frac{3.7}{\rho q_c} \left( 0.5 \times 10^6 \sigma - 7.5 \right)^{-1}}$$

*spectral dispersion*      *average droplet size*

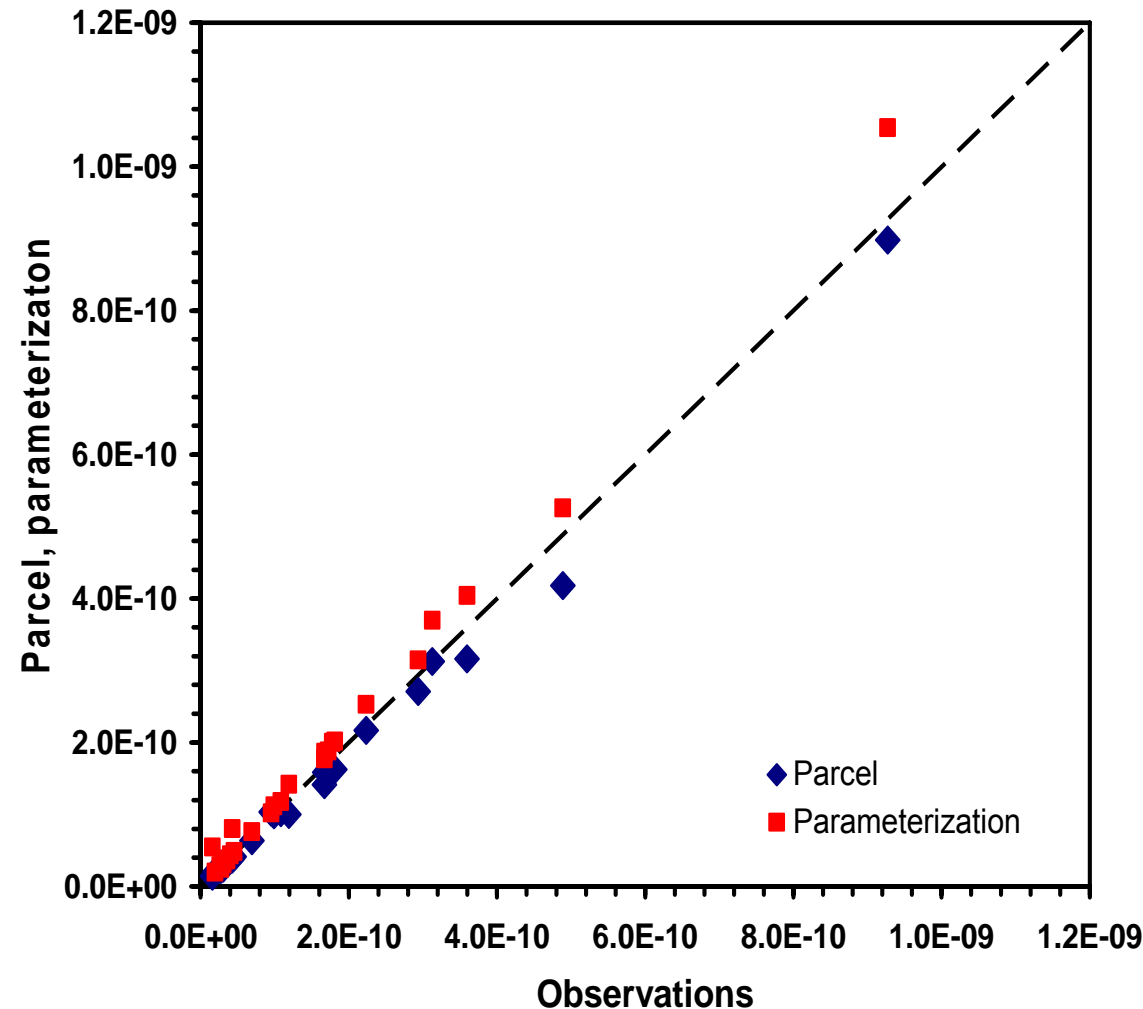
We have all the elements we need (dispersion, droplet size) for a comprehensive treatment of precipitation. Why not include it in the GCM?

**Challenge:** How do we obtain these parameters in the global model?

**Solution:** From the *Nenes and Seinfeld Activation Parameterization*

# Parameterizing drizzle: predicting droplet size in GCMs

## Autoconversion rate ( $\text{kg s}^{-1}$ )

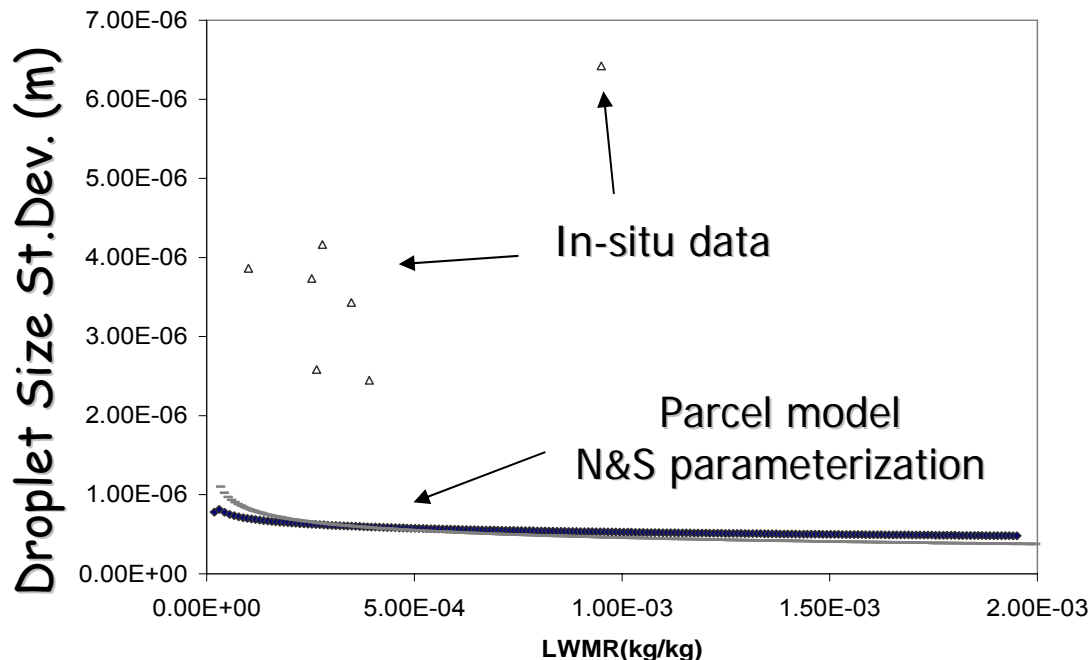


- Predict size distribution with Nenes and Seinfeld parameterization and cloud parcel model for **adiabatic cases** of CRYSTAL-FACE (cumulus) clouds.
- Use droplet number & size distribution to predict autoconversion rate.
- Use in-situ data to calculate autoconversion as well.
- The parameterization (and parcel model) capture the spectral width for adiabatic clouds well.
- Is it always like this? No.
- We can address this problem.

# New cloud droplet formation parameterization (Includes entrainment)

## Why need a new parameterization?

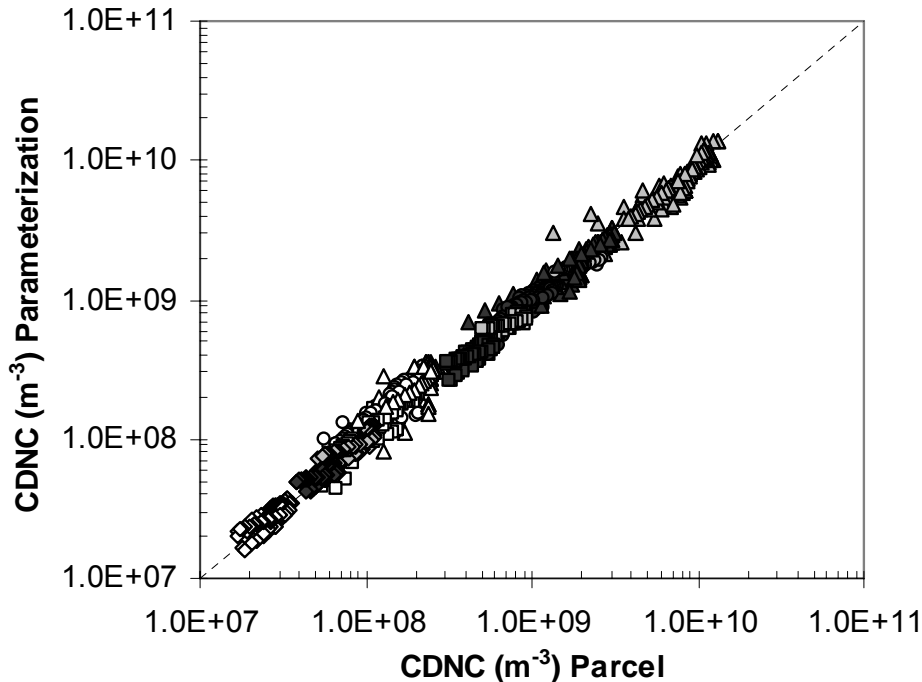
- Current parameterizations are adiabatic. Clouds are generally not.
- Droplet number predictions are good even for slightly diabatic conditions (although  $N_d$  can still be overestimated for strong entrainment).
- Nenes and Seinfeld can predict droplet size distribution, but they are too narrow (adiabatic), so autoconversion calculations would generally be "off".



- Comparison of predicted size distribution "width" vs. liquid water content for non-adiabatic CRYSTAL-FACE (cumulus) clouds.
- Parameterization and cloud parcel model agree great with each other, but not with the data (even though cloud droplet number is captured to within 5%).
- An entraining parameterization would improve this because entrainment broadens the distribution.

# New cloud droplet formation parameterization (Includes entrainment)

- The first parameterization of its kind (Barahona and Nenes, *in prep*).
- Complex organics can be treated, same conceptual framework ("population splitting") as the adiabatic parameterization.
- Mixing is parameterized in terms of an entrainment rate.
- Versions for lognormal and sectional aerosol developed.
- Same CPU requirements as the adiabatic "version".



We've looked at 4000 cases  
Average error:10%

We plan to use CRYSTAL-FACE,  
CSTRIPE, ICARTT, MASE,  
TEXAS-AQS data to constrain  
the entrainment rate.

The predicted in-cloud droplet  
size distribution will be evaluated  
with the same dataset.

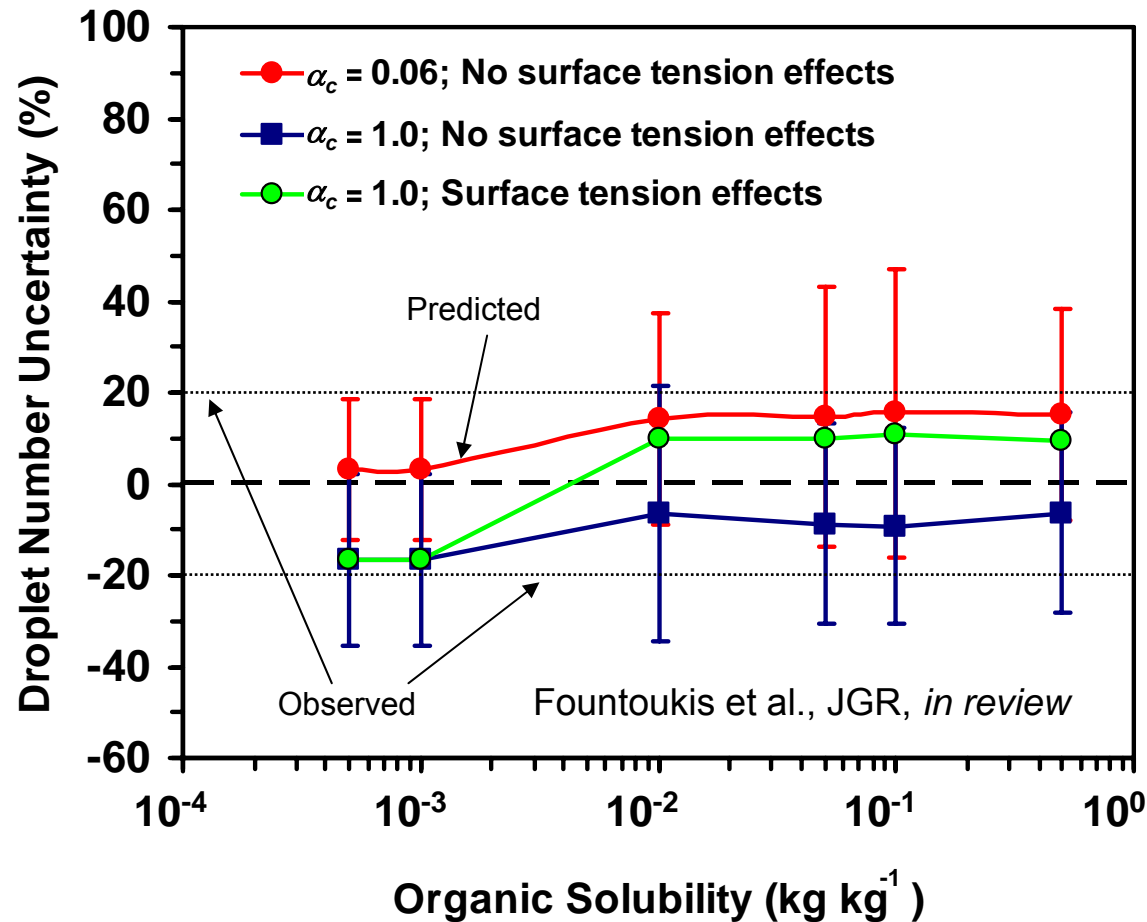
**THANK YOU!**



# ICARTT (2004) Constraining Properties of organics



CIRPAS Twin Otter



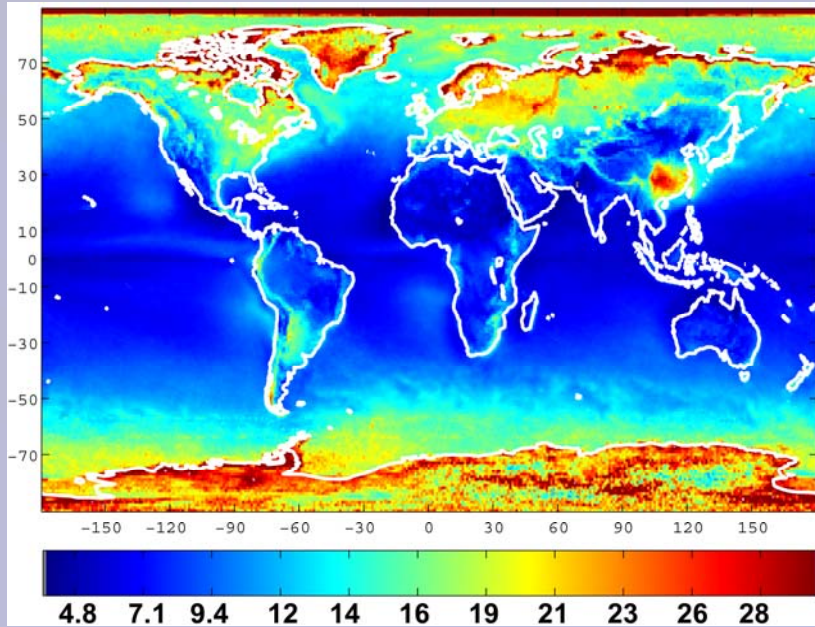
Optimal closure  
if organics  
have a  
solubility less  
than 1 g kg<sup>-1</sup>

This is consistent  
with our CCN  
measurements as  
well.

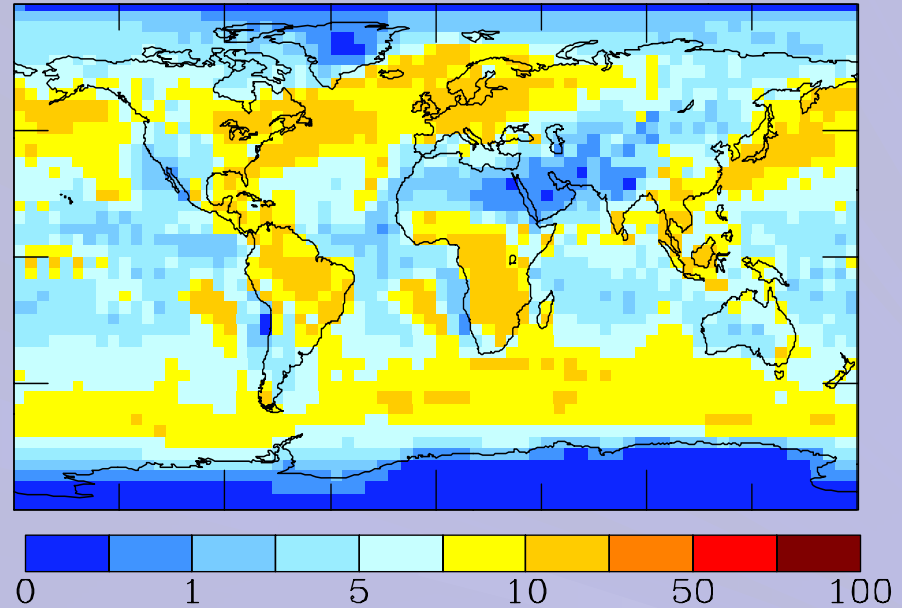
# Current Day Simulation (annual average)

## Cloud optical depths

**MODIS**



**GISS**



Cloud optical depths are quite close.

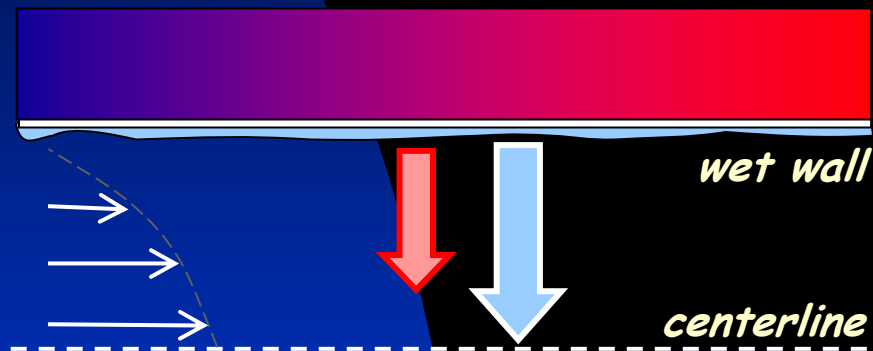
There are local differences (sometimes large).

*Nenes et al., in preparation*

# Measuring CCN: a key source of data

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

**Our Method:** Take a metallic cylinder, wet its walls internally. Cool one end, heat the other, and flow air through it.



Wall saturated with  $H_2O$   
Linear temperature gradient.

$H_2O$  diffuses more quickly than heat and arrives at centerline first.

Roberts and Nenes, *AS&T*, 2005

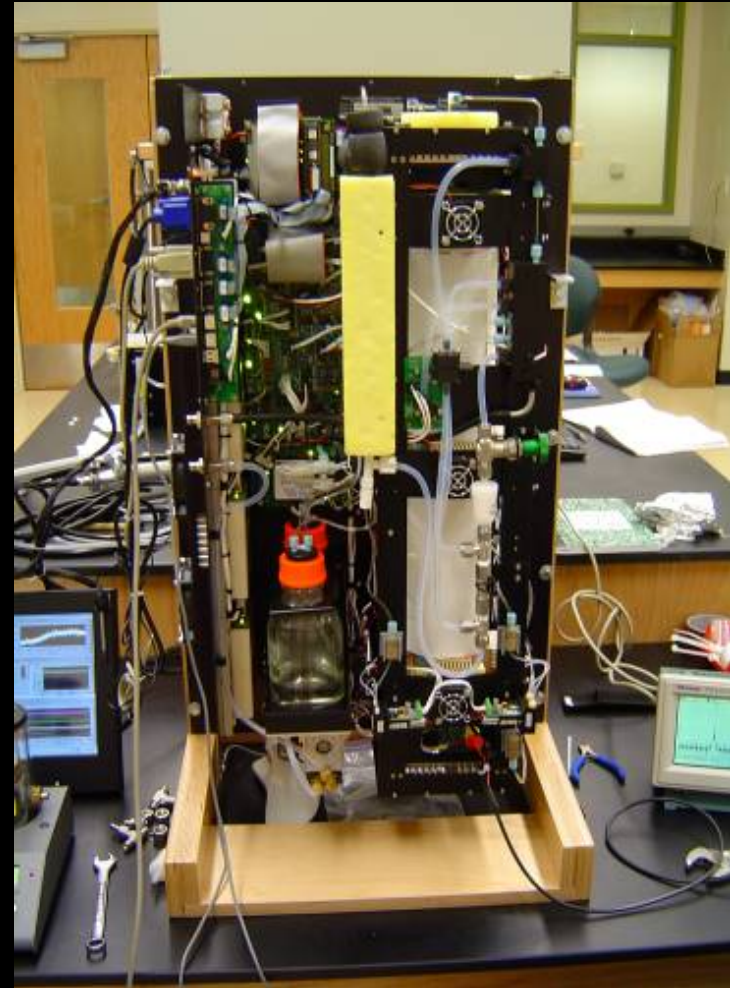
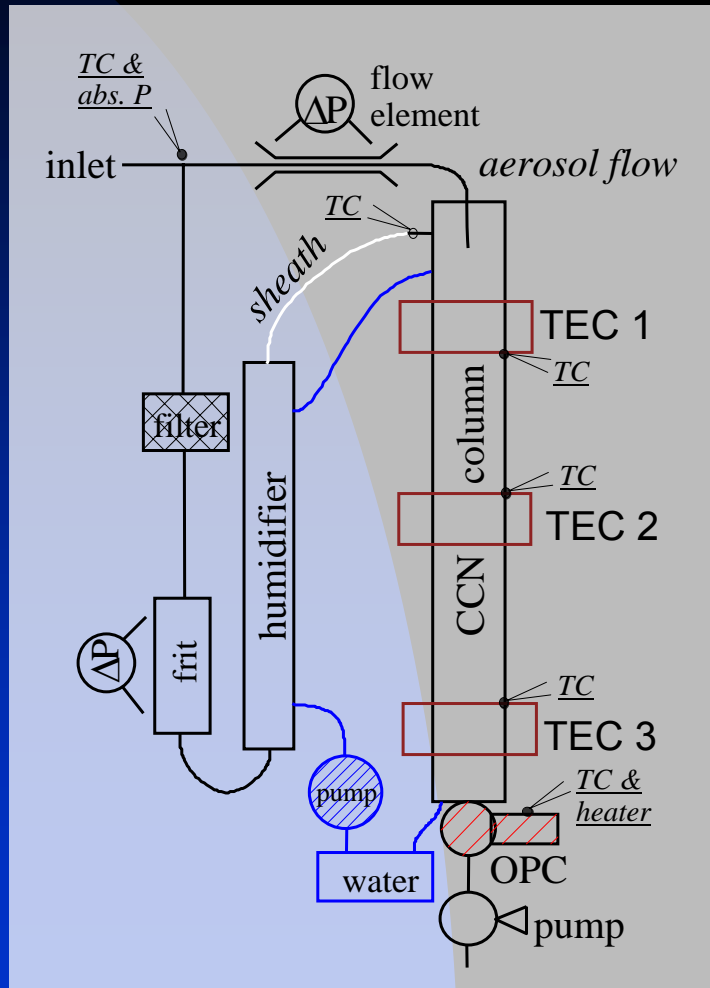
Lance et al., *AS&T* (2006)

The flow is *supersaturated* with water vapor at the centerline.

- Constant supersaturation develops - great for counting CCN!
- Supersaturation is controlled by  $P$ , flow rate,  $T$  gradient
- CCN can be measured over a wide range of supersaturations

# Droplet Measurement Technologies

## CCN Counter



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

# Are GCM parameterizations "good enough"?

Evaluate cloud droplet formation theory and parameterizations with in-situ Aerosol/cloud microphysical measurements.



Cloud droplet concentration

FSSP, CAS

Aerosol number concentration

CPC

Aerosol size distribution

DMA, PCASP, APS

Aerosol composition

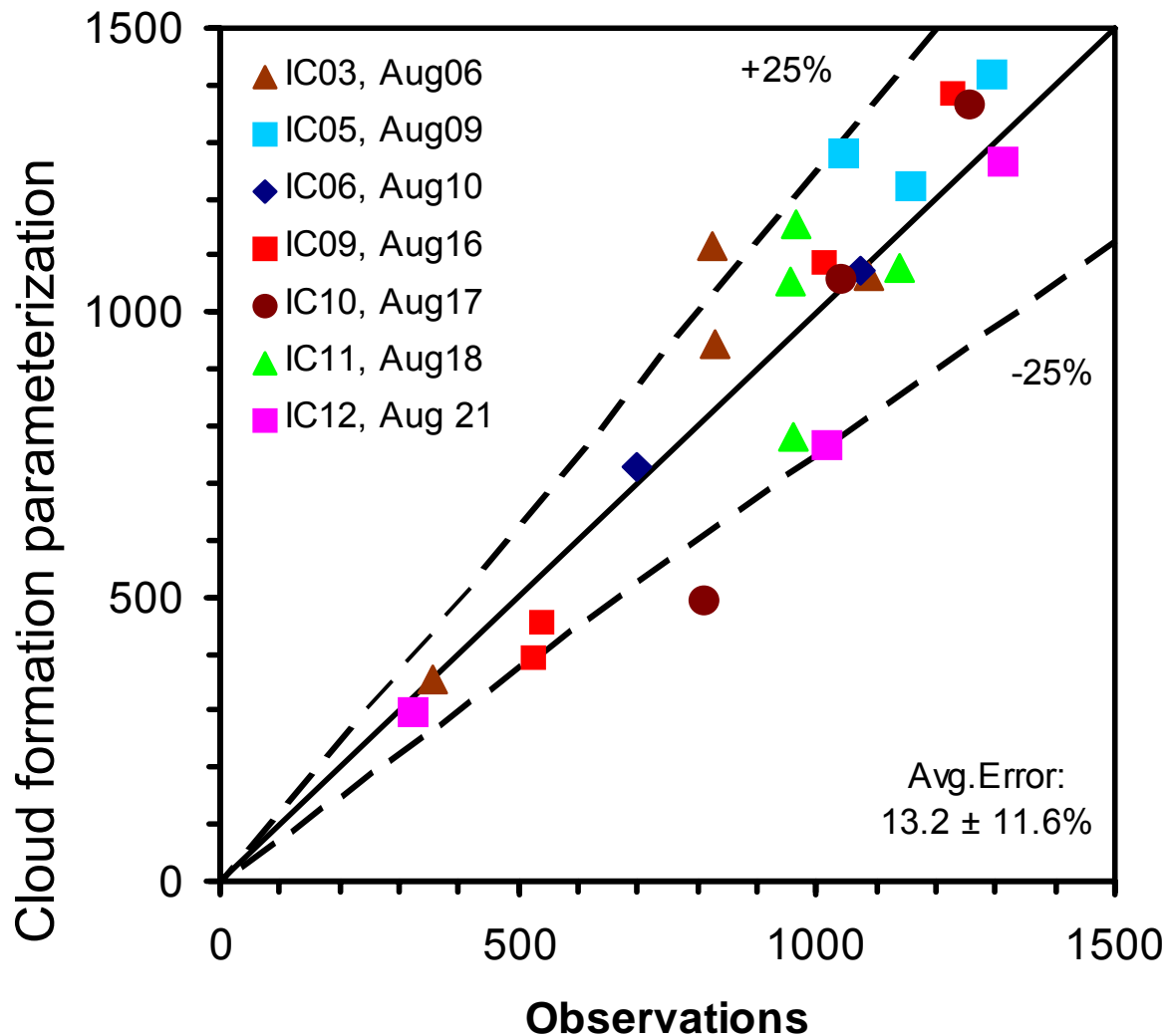
AMS, PILS

Updraft velocity

# ICARTT (2004) Continental Stratus



CIRPAS Twin Otter



Downwind of  
power plants over  
Lake Erie and  
South Ohio.

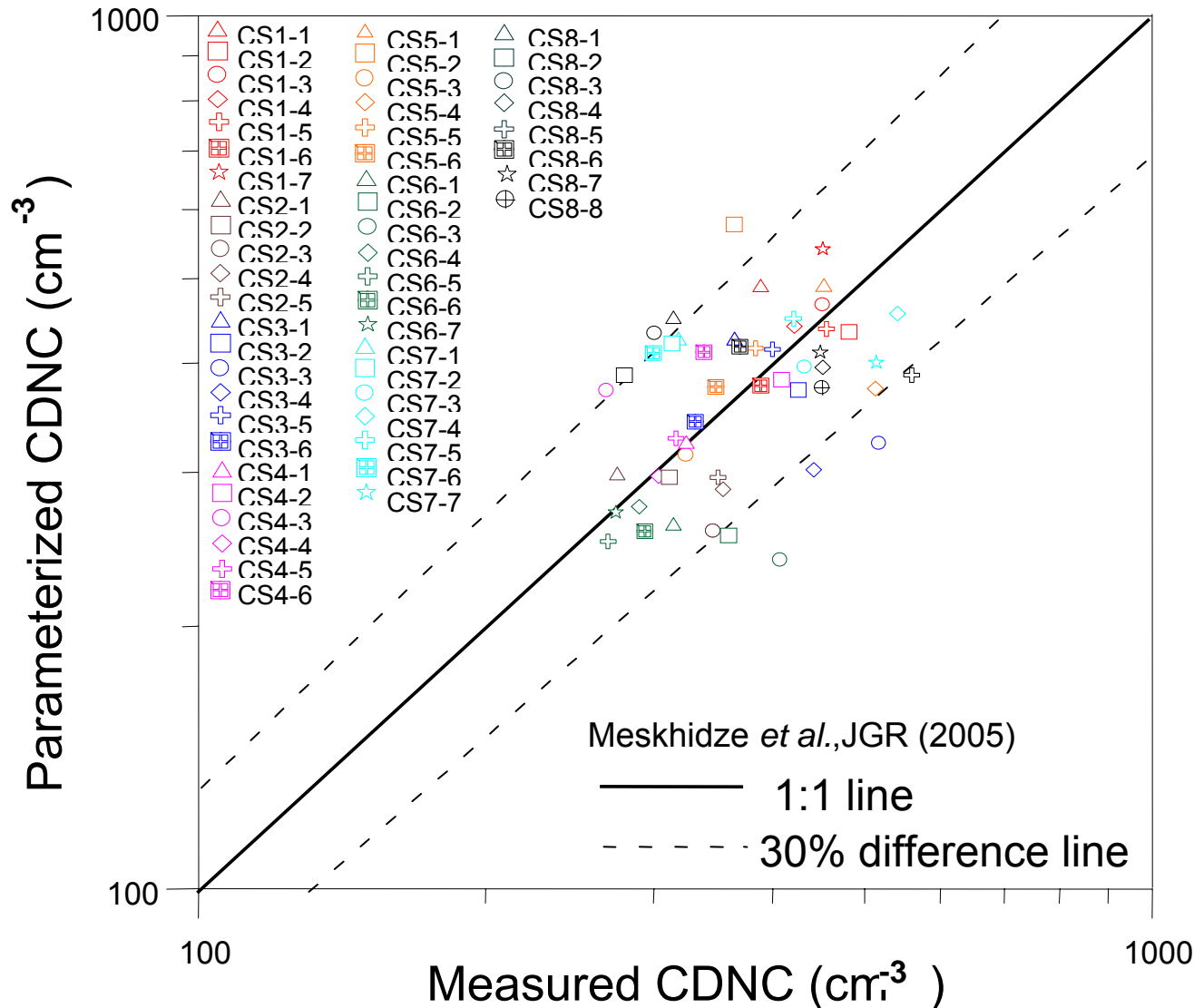
Parameterization  
agrees with  
observed CDNC

Cloud formation  
parameterizations  
do a  
**GOOD JOB**

# CSTRIPE (2003) Coastal Stratocumulus



CIRPAS Twin Otter



Parameterization  
agrees with  
observed CDNC

Gaussian PDF of  
updraft velocity  
is sufficient to  
capture CDNC

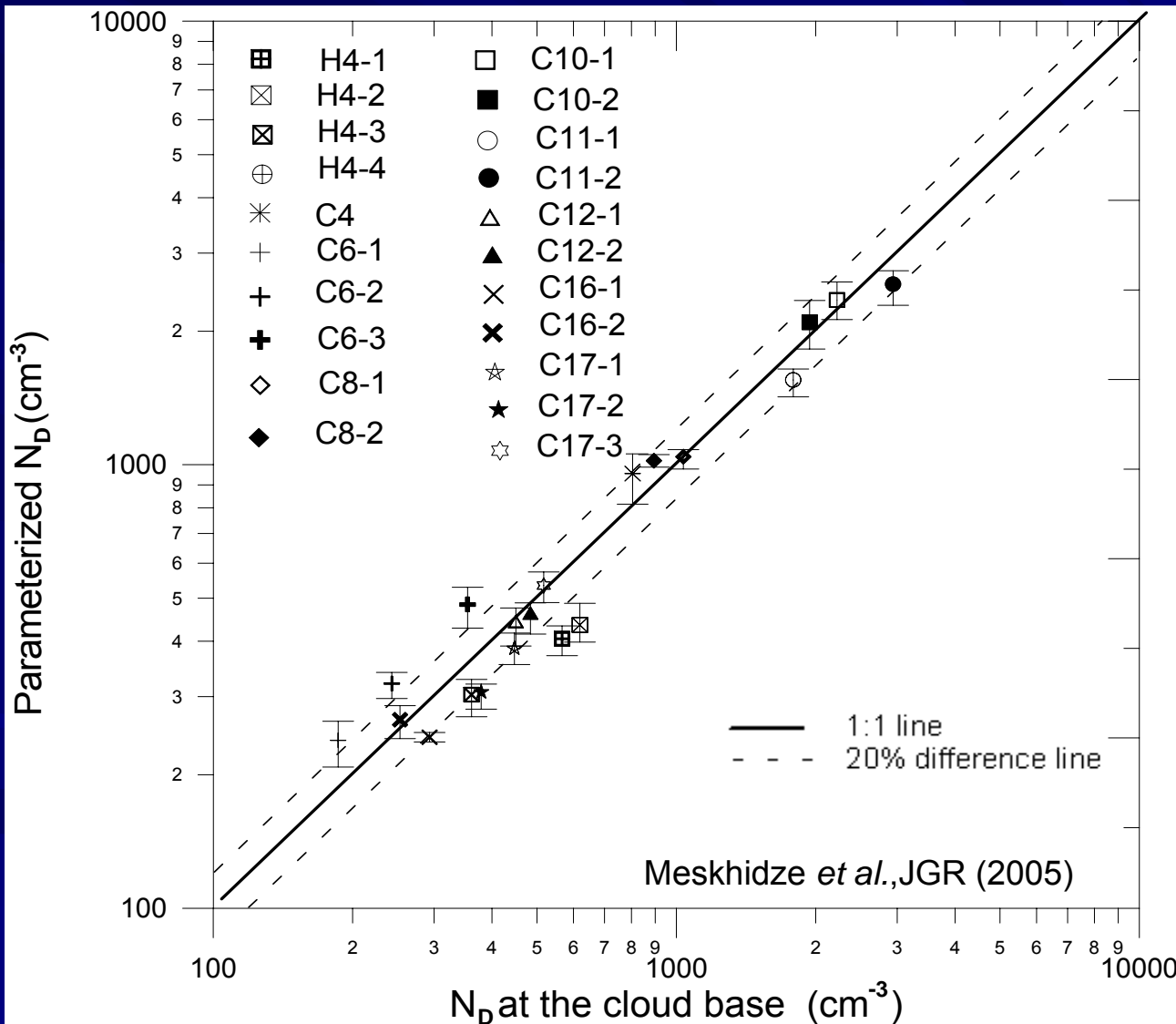
Average updraft  
captures equally  
well.

$\alpha \sim 0.06$

# CRYSTAL-FACE (2002) Cumulus clouds



CIRPAS Twin Otter



Parameterization  
agrees with  
observed CDNC

Single updraft  
sufficient to  
describe CDNC

$\alpha \sim 0.03 - 0.08$   
within updraft  
uncertainty



# Current Day Simulation (annual average)

CCN @0.2% ( $\text{cm}^{-3}$ )

Global average:  $190 \text{ cm}^{-3}$

