

From Emissions to Impacts: Insights from and Advances in Atmospheric Chemistry

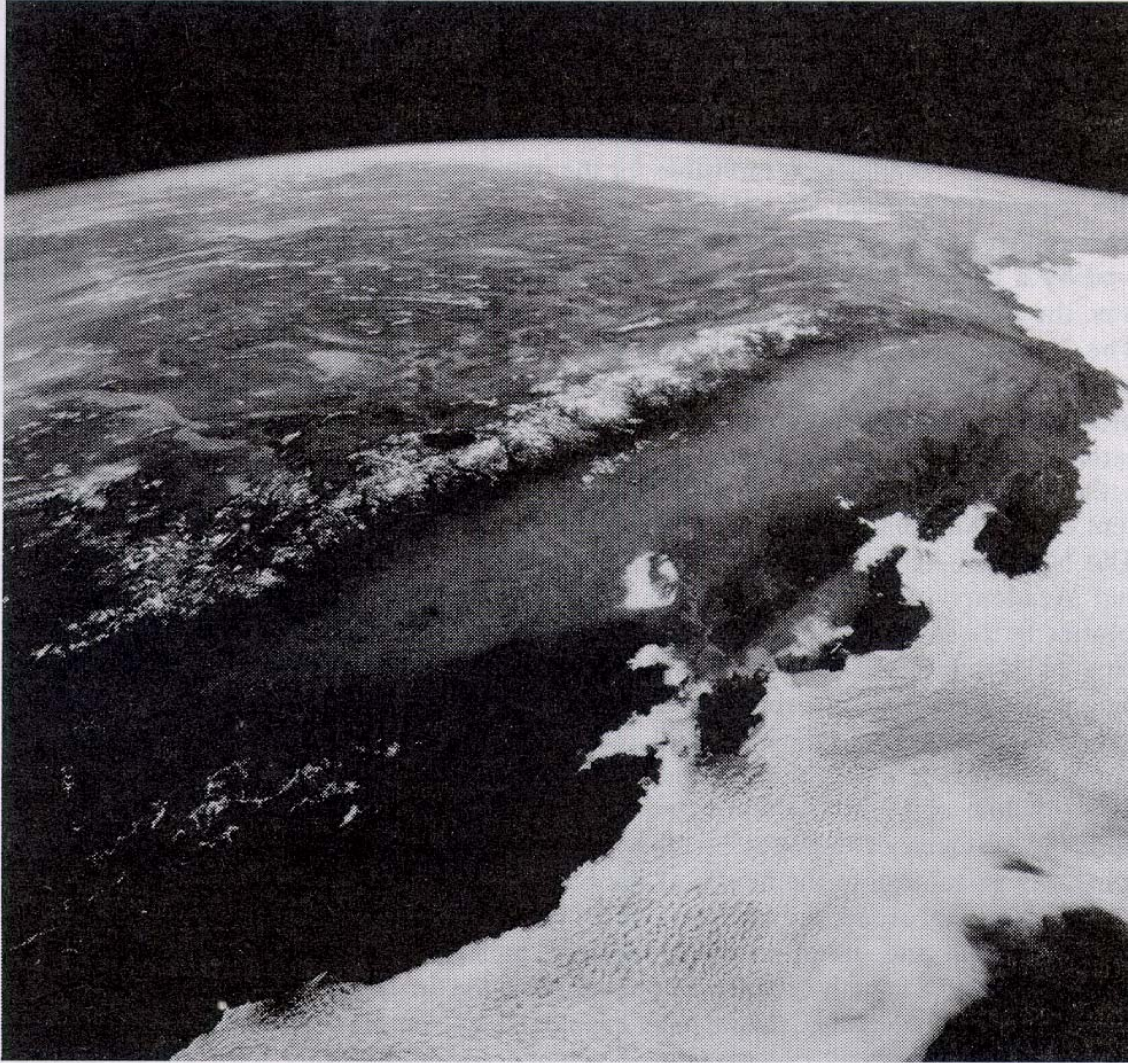
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AAAS Annual Meeting
Boston, MA
16 February 2008

**Acknowledgments: NASA, NSF, NOAA, ONR, CIRPAS
Nenes, Seinfeld/Flagan, and Adams Groups**

Earth's brightness controls climate



Light colors:

reflect sunlight
and keep planet
cool

Dark colors: absorb
and warm planet

Haze and clouds
lighten and cool
the planet.

They act as the
planet's "t-shirt"

The impact of chemistry on climate

Gas phase atmospheric compounds:

They are the greenhouse gases - emissions and their chemical transformations controls their warming potential.

Examples: O_3 , CO_2 , H_2O , CFC's, N_2O , CH_4

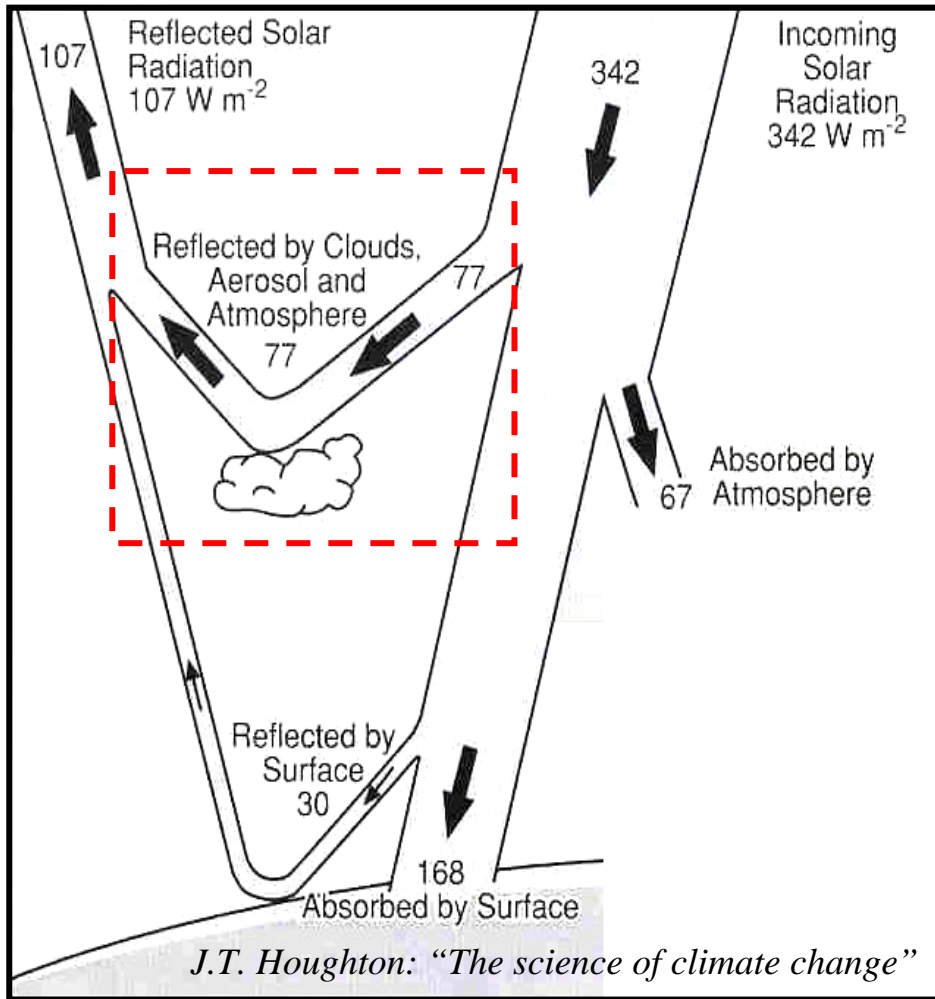
Solid/liquid phase atmospheric particles:

Some components can absorb radiation but some can strongly cool the planet by scattering (reflecting) incoming solar radiation.

Absorbers: Black carbon, liquid water/ice (IR)

Scatterers: Most aerosol, liquid water (Visible)

Clouds: major contributor to shortwave planetary albedo



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).

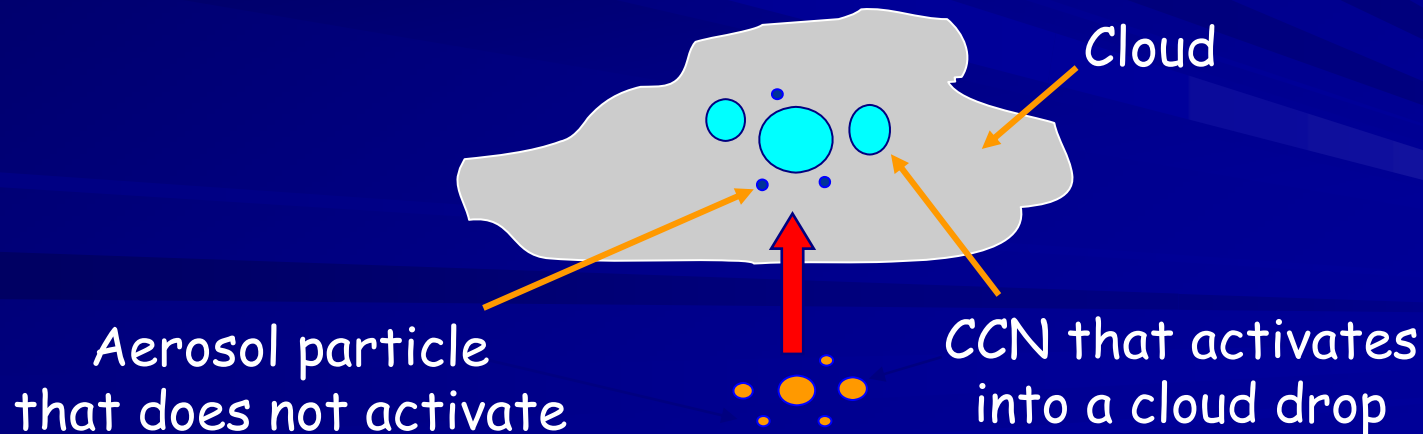
How do (liquid water) clouds form?

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.

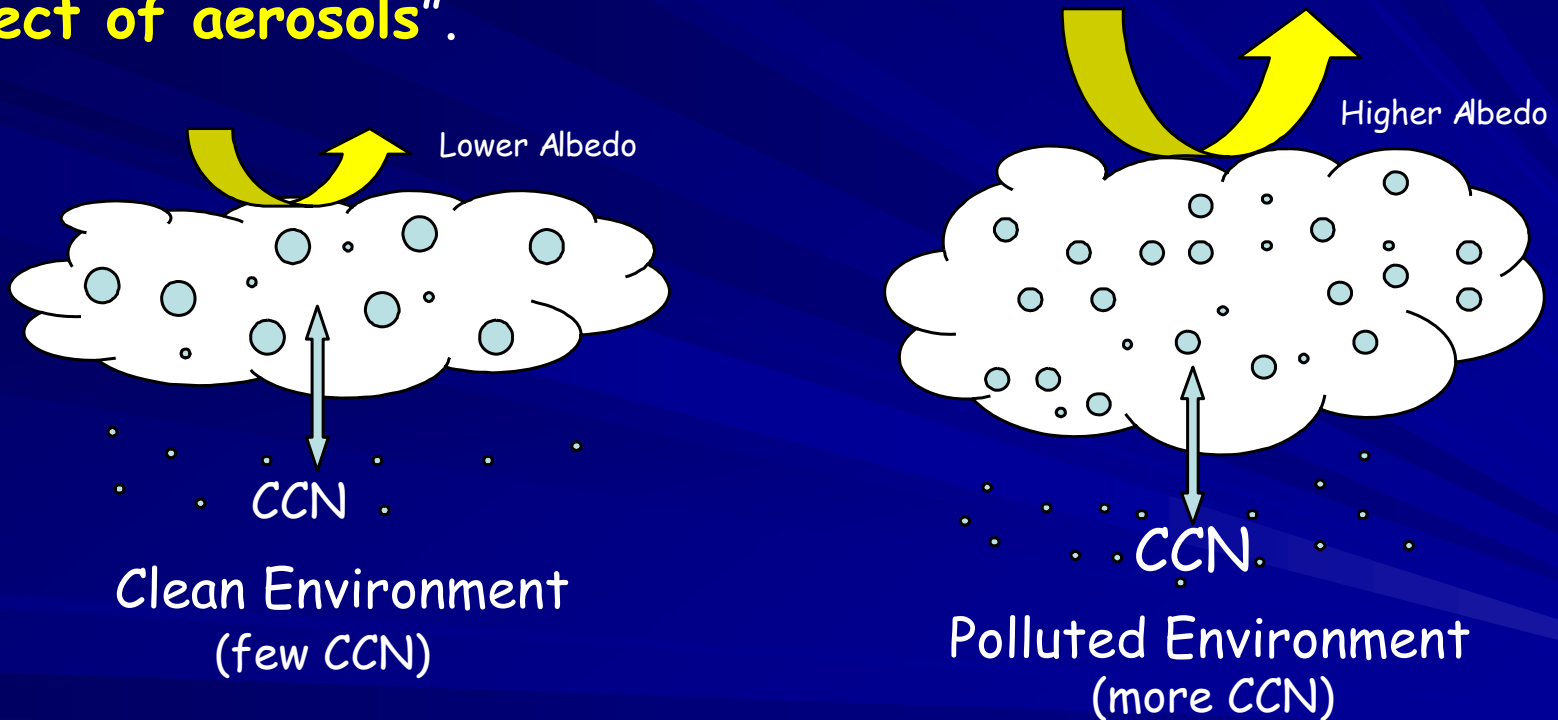
Aerosols that can become droplets are called cloud condensation nuclei (CCN).



Can humans affect clouds and the hydrological cycle?

Yes! By changing global CCN concentrations (air pollution).

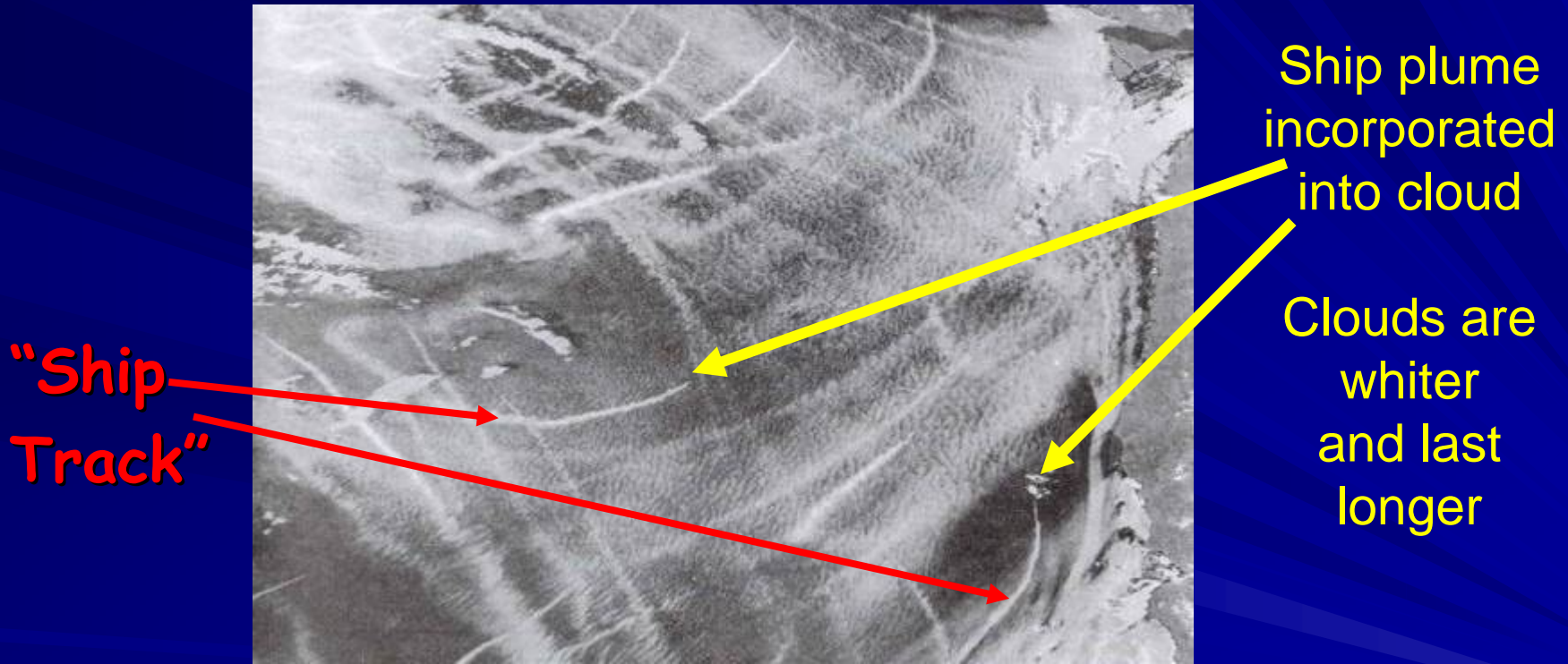
Result. Clouds are "whiter", precipitate less (persist longer) and potentially cover larger areas of the globe. This yields a net cooling on climate and is called the "**indirect climatic effect of aerosols**".



Increasing particles tends to cool climate (potentially a lot).
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.

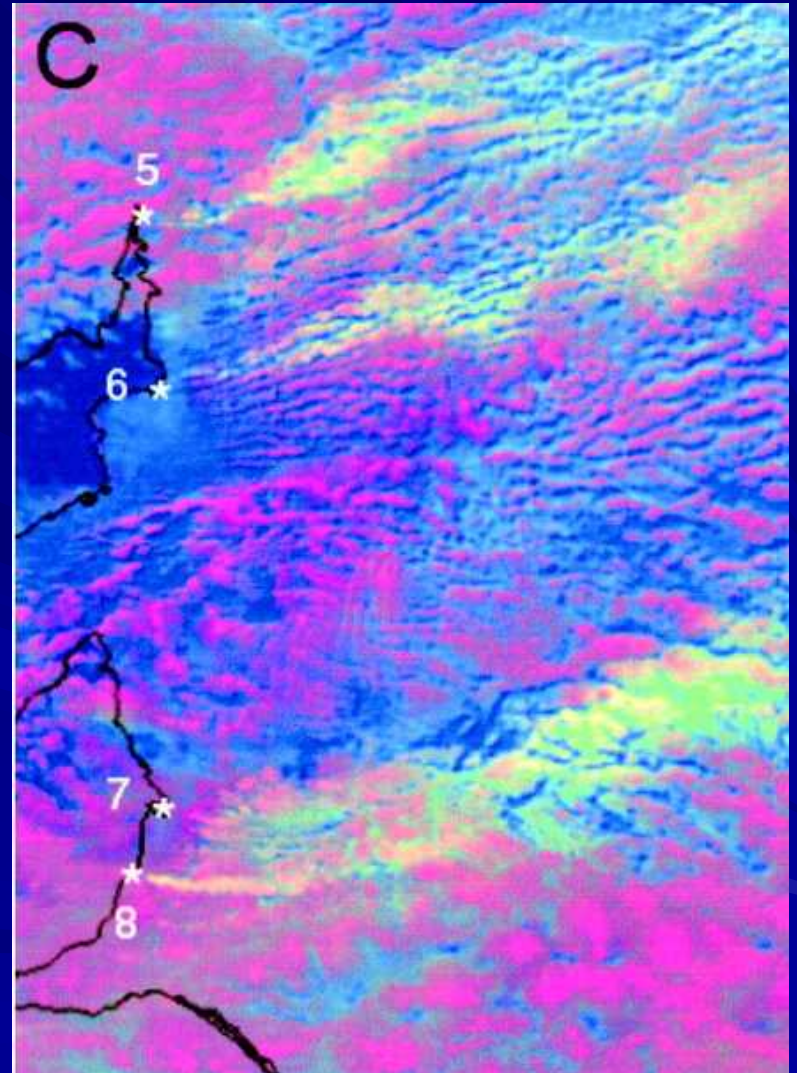


Pollution \uparrow \Rightarrow Droplet number \uparrow \Rightarrow Droplet size \downarrow
Droplet size \downarrow \Rightarrow Cloud reflectivity \downarrow **AND** Precip \downarrow

Observational evidence of indirect effect

Satellite observation of clouds in the Black Sea.

Red: Clouds with low reflectivity.
White: Clouds that reflect a lot.
Blue: Clear sky.



Observational evidence of indirect effect

Air pollution can affect clouds

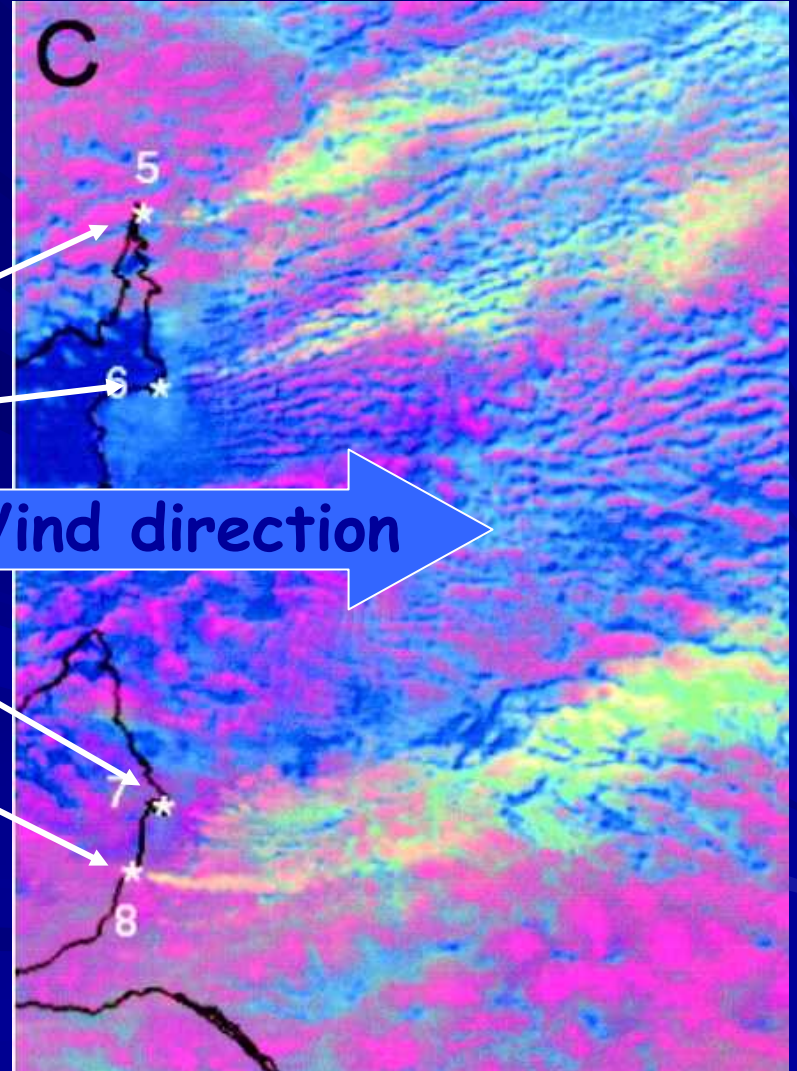
Satellite observation of clouds in the Black Sea.

Power plant

Lead smelter

Port

Oil refineries



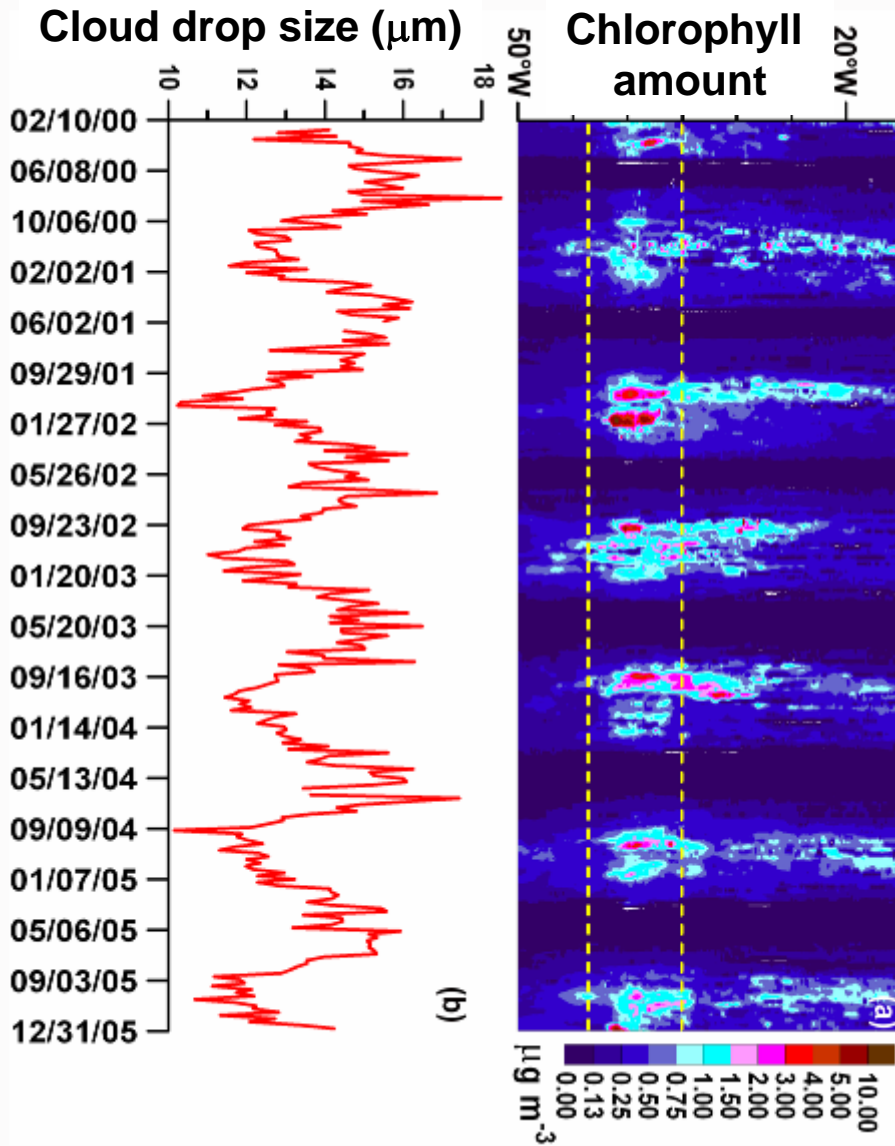
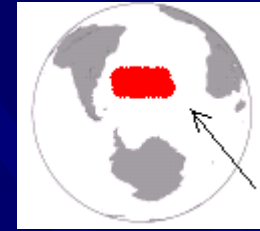
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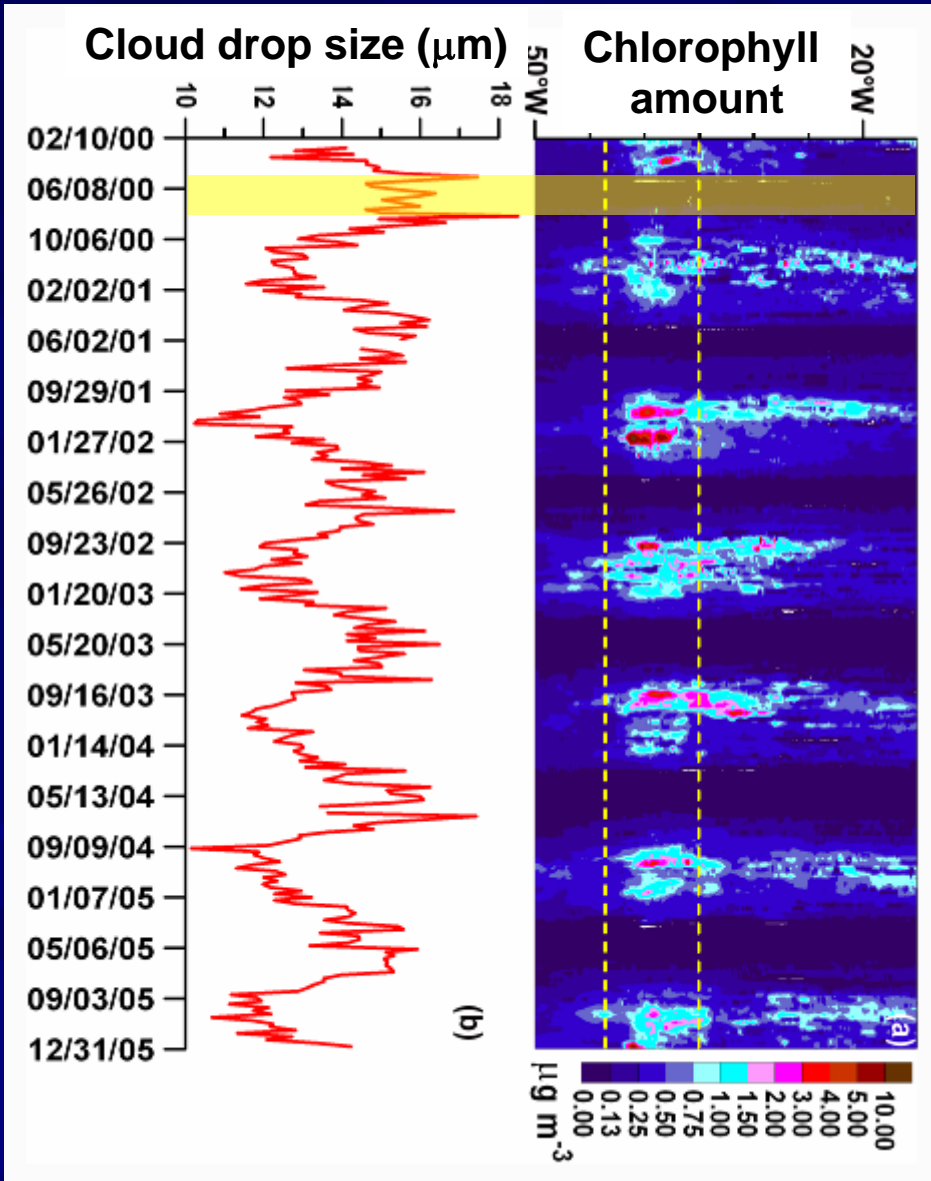
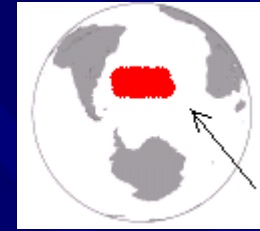
Phytoplankton affect clouds too...

Location: East of Patagonia (South America)



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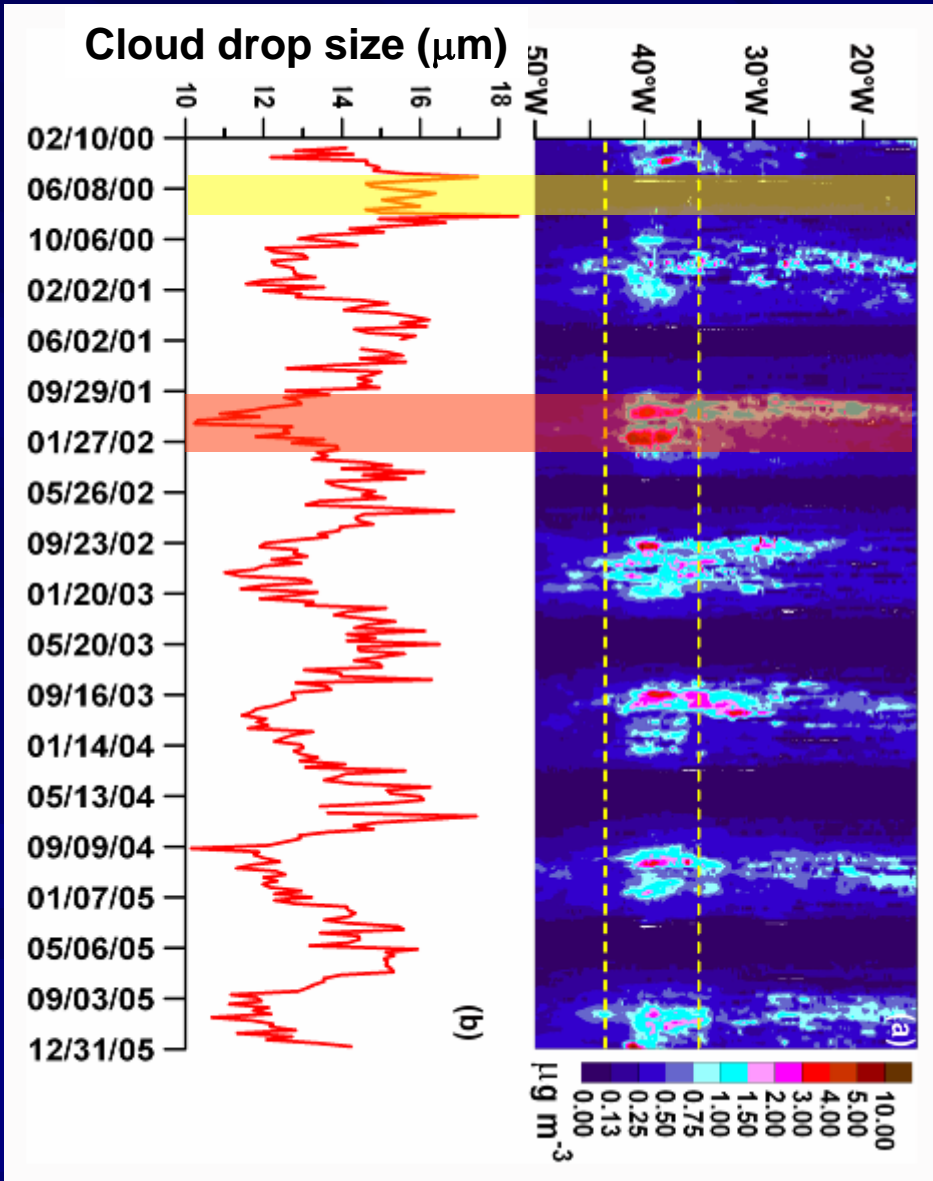
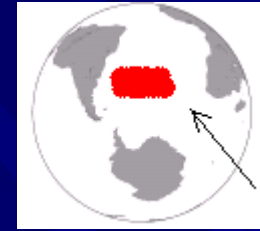
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Low chlorophyll period,
clouds have large drops
(not very reflective)

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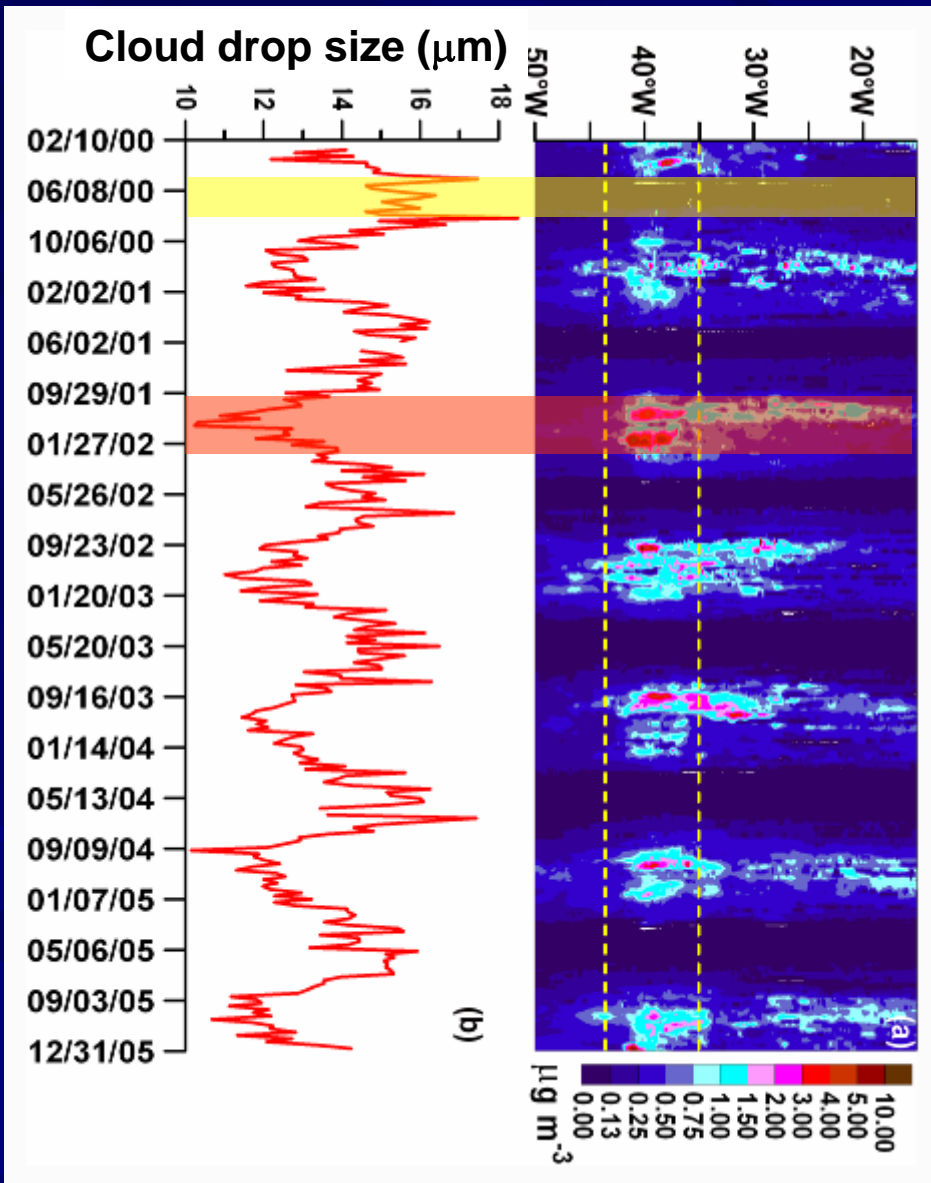
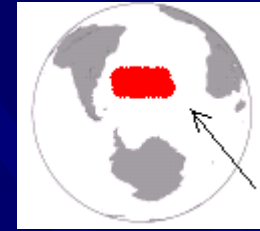
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- ← Low chlorophyll period,
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- ← High Chlorophyll period,
Clouds have small drops
(very reflective)

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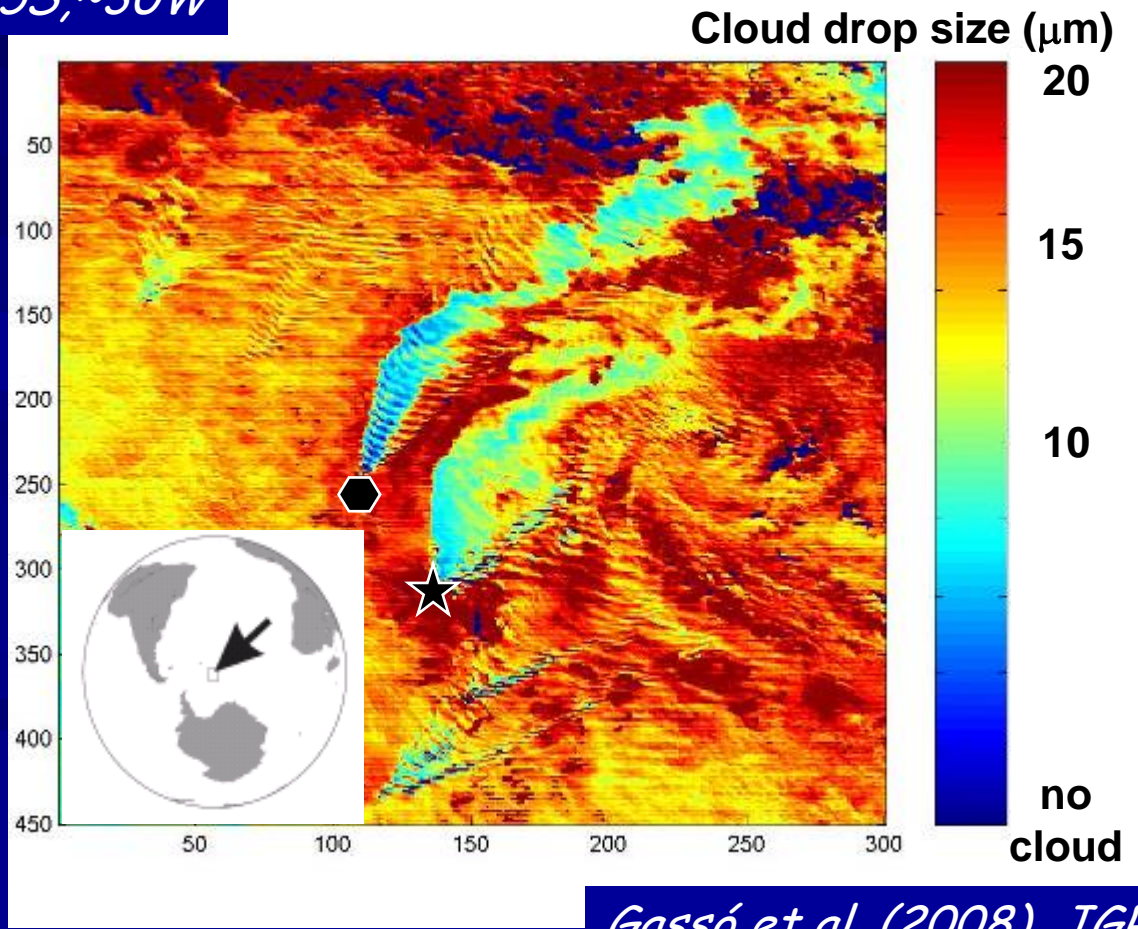
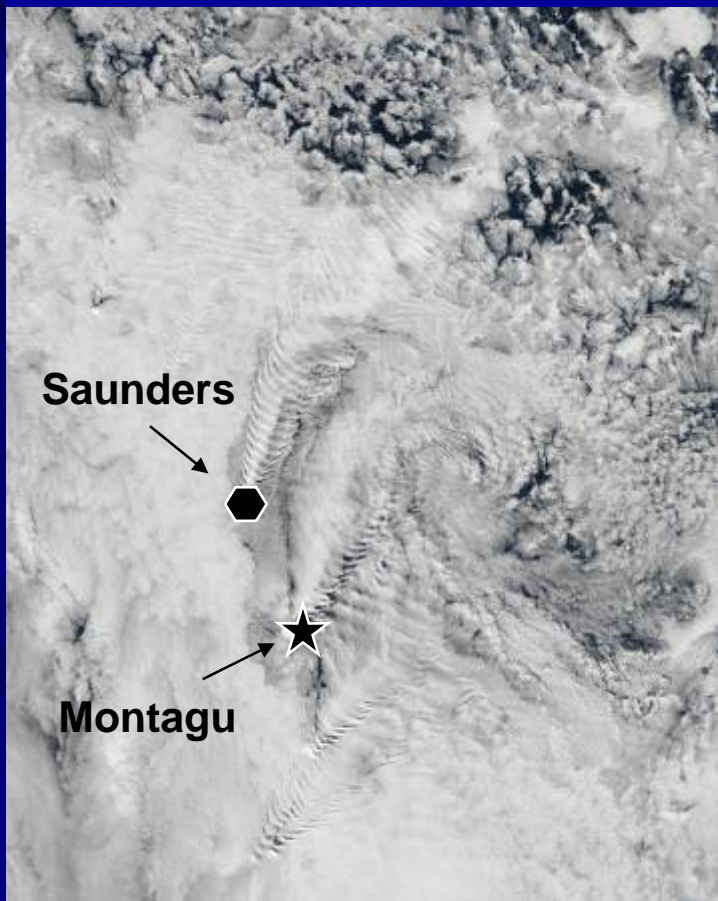
Phytoplankton emissions
increase particle loads, and
strongly impact clouds.

Changes are comparable to
contrasts between polluted
and clean environments
(forcing $\sim -15 \text{ W m}^{-2}$).

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, $\sim 55\text{S}, \sim 30\text{W}$



Gassó et al. (2008), JGR

Anthropogenic Indirect Effect: How do we estimate its magnitude?

We use a global climate model (GCM)

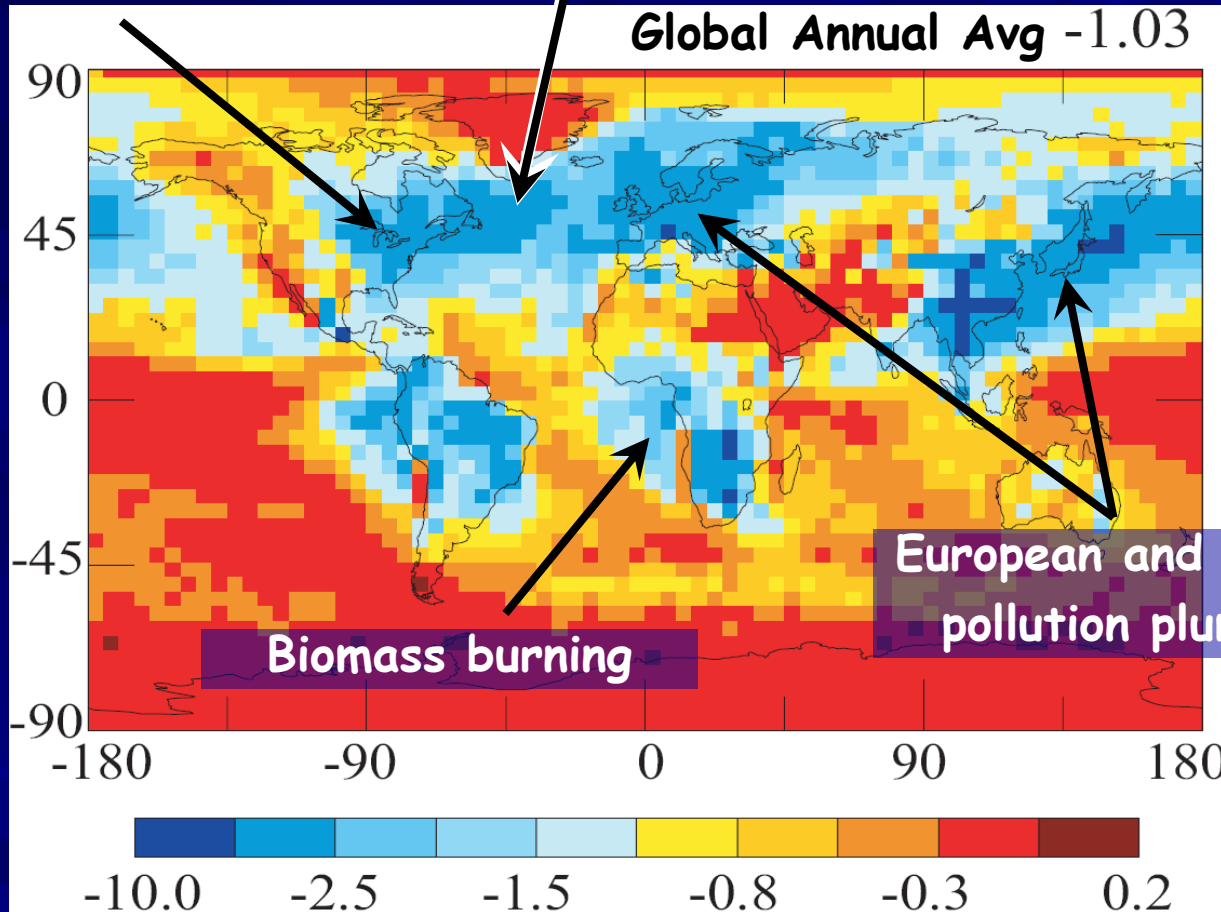
- simulation with "current day" emissions
- simulation without anthropogenic emissions ("preindustrial" emissions)
- compute the change in the planetary energy balance between the simulations ("indirect forcing")
- compare annual average forcing to greenhouse gas warming ($\sim 2.5 \text{ W m}^{-2}$)
- Net forcing (greenhouse + indirect) can be used as an index for climate change.

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

North America
pollution plumes

Long-range transport

Global Annual Avg -1.03



European and Asian
pollution plumes

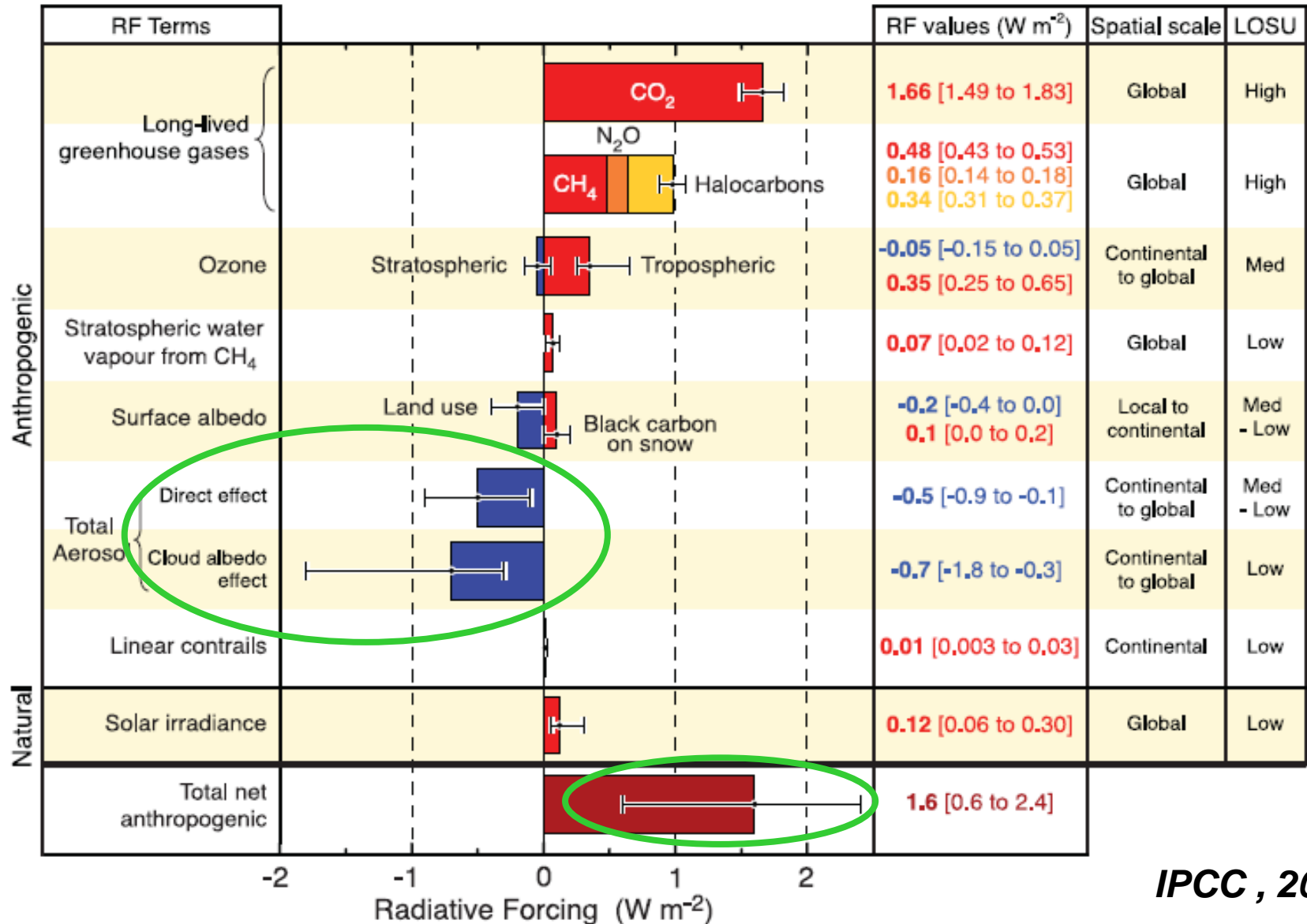
Biomass burning

Sotiropoulou et al., in review

Spatial pattern of IF follows that of aerosol variations

Anthropogenic Climate Forcing

RADIATIVE FORCING COMPONENTS



©IPCC 2007: WG1-AR4

The importance of reducing IE uncertainty

The indirect effect diminishes the impact of global warming. We don't know exactly by how much.

- If IE is low ($\sim -0.3 \text{ Wm}^{-2}$) then we are "feeling" most of the warming ($2.3 - 0.3 = 2.0 \text{ Wm}^{-2}$) from greenhouse gases.
- If IE is high ($\sim -2.0 \text{ Wm}^{-2}$) then we are "feeling" very little of the warming ($2.3 - 2.0 = 0.3 \text{ Wm}^{-2}$) from greenhouse gases.

In both cases, we will experience global warming, but the difference in climate sensitivity between both states is *huge*.

If we are closer to the latter, improving air quality will lead to accelerated global warming.

The importance of reducing IE uncertainty

The regional impacts of indirect effect can be much larger than global warming on a regional scale.

- Changes in warming/heating patterns \Rightarrow large changes in circulation and local climate (e.g., onset of the Indian Monsoon Season).
- Changes in ground-level solar radiation \Rightarrow reduction of photosynthetic activity (crop reduction).
- Changes in redistribution of water/precipitation with large impacts on water resources.

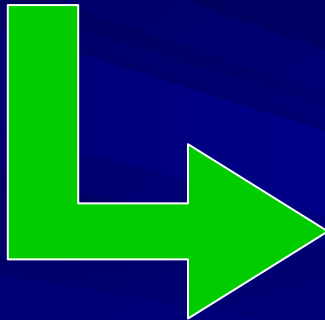
The indirect effect is thought to have a deleterious impact on climatic sensitive areas (US midwest, subsaharan Africa). The impacts will only become larger because of the development of Asian Nations.

Quantification of the Indirect Effect in Global Models

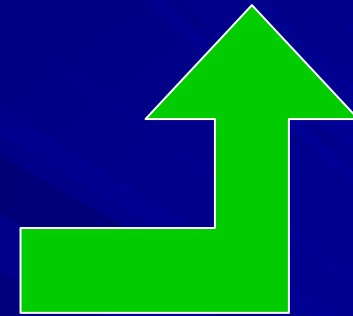
Aerosol
Size Distribution and
Chemical Composition



Cloud Radiative
Properties

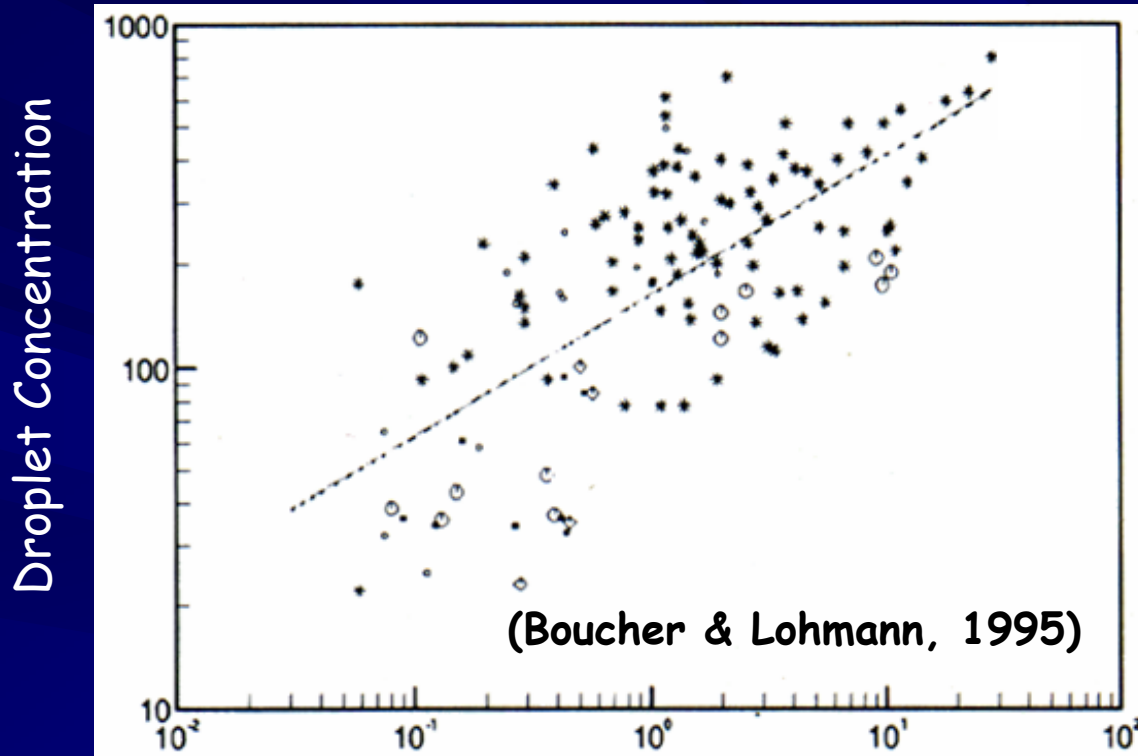


Cloud Droplet
Number and Size



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

Approach for aerosol- N_d ; empirical



Aerosol sulfate concentration
(proxy for pollution)

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 approaches for cloud processes

Dynamics

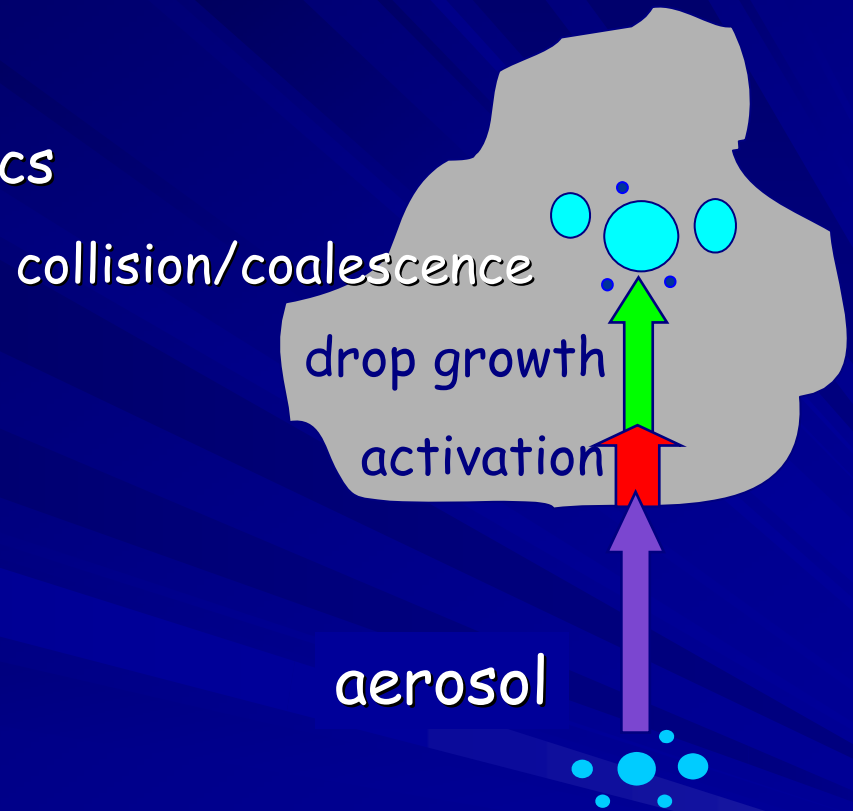
- Updraft Velocity
- Large Scale Thermodynamics

Particle characteristics

- Size
- Concentration
- Chemical Composition

Cloud Processes

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

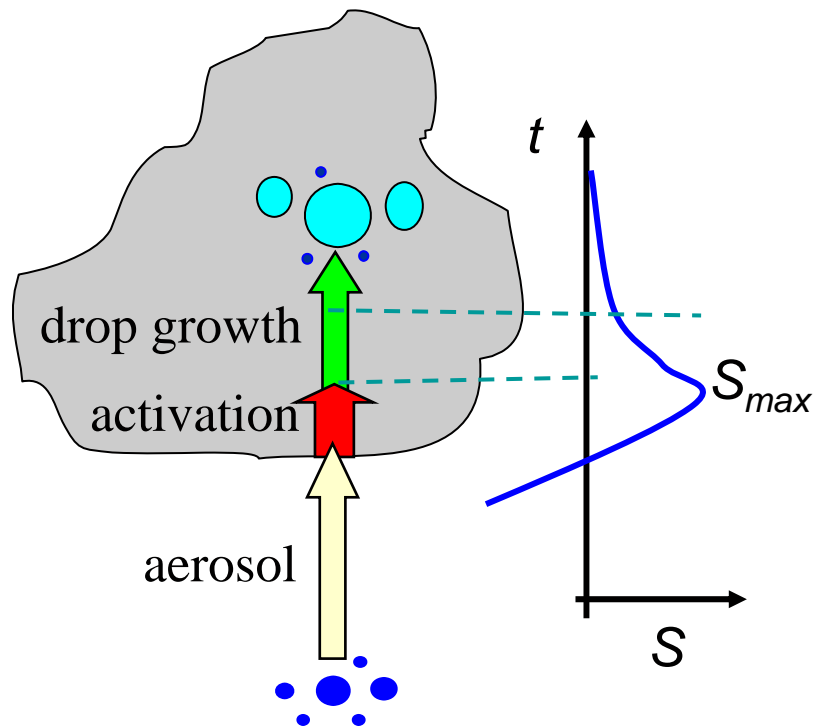


All the links need to be incorporated in global models
The links need to be **COMPUTATIONALLY** feasible.

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.

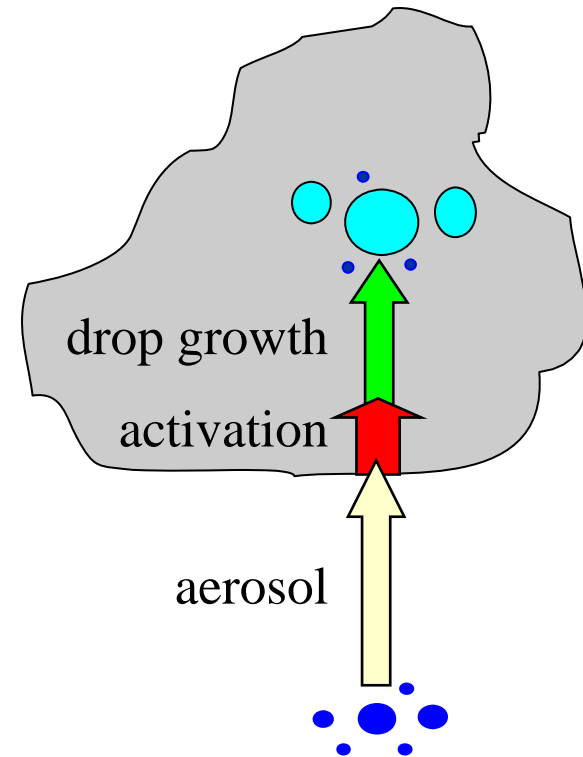
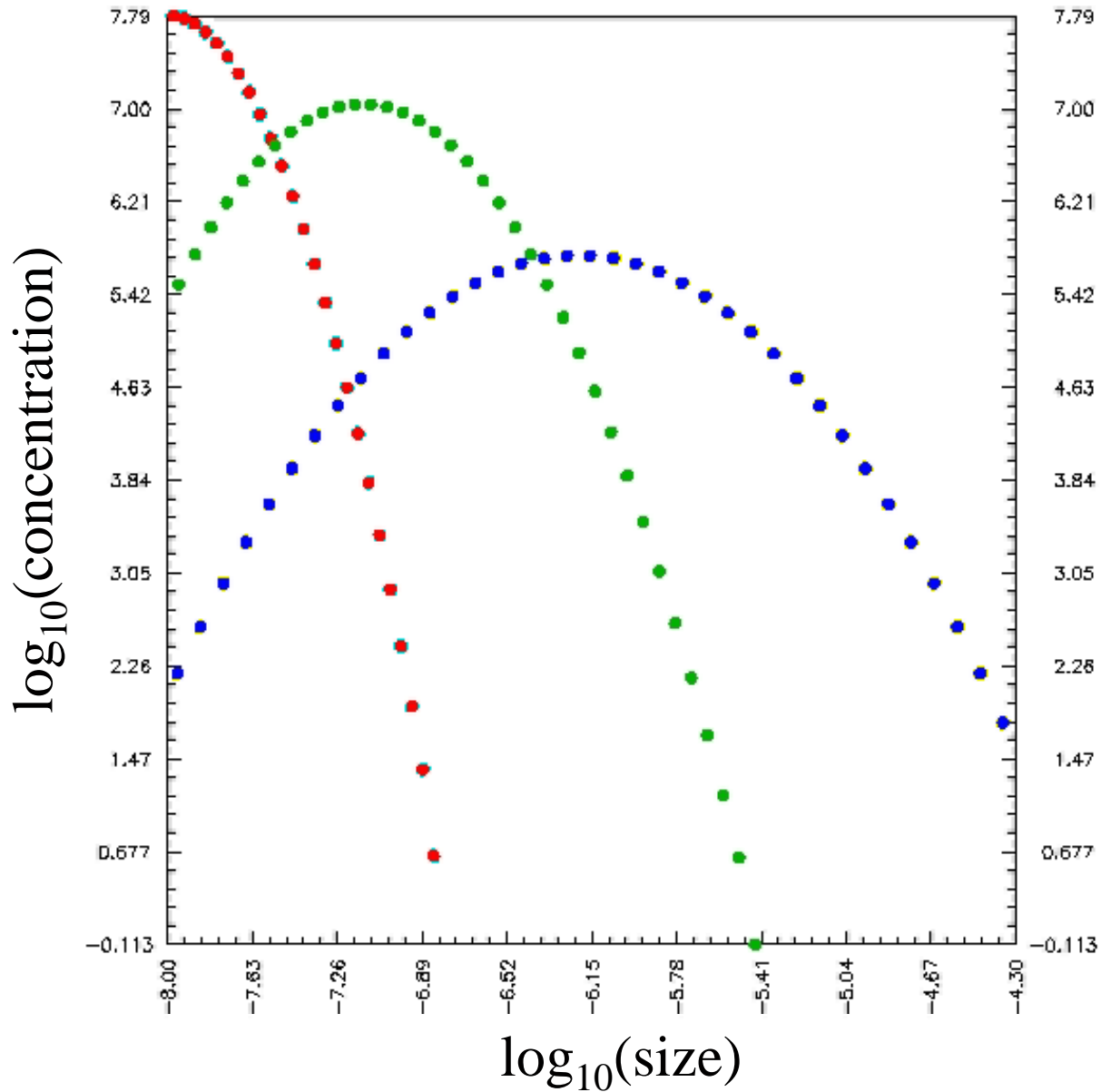


Steps are:

- 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



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

$\log_{10}(\text{concentration})$

$\log_{10}(\text{size})$

"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)

Are the parameterizations "good enough"?

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



CIRPAS Twin Otter



NOAA P3

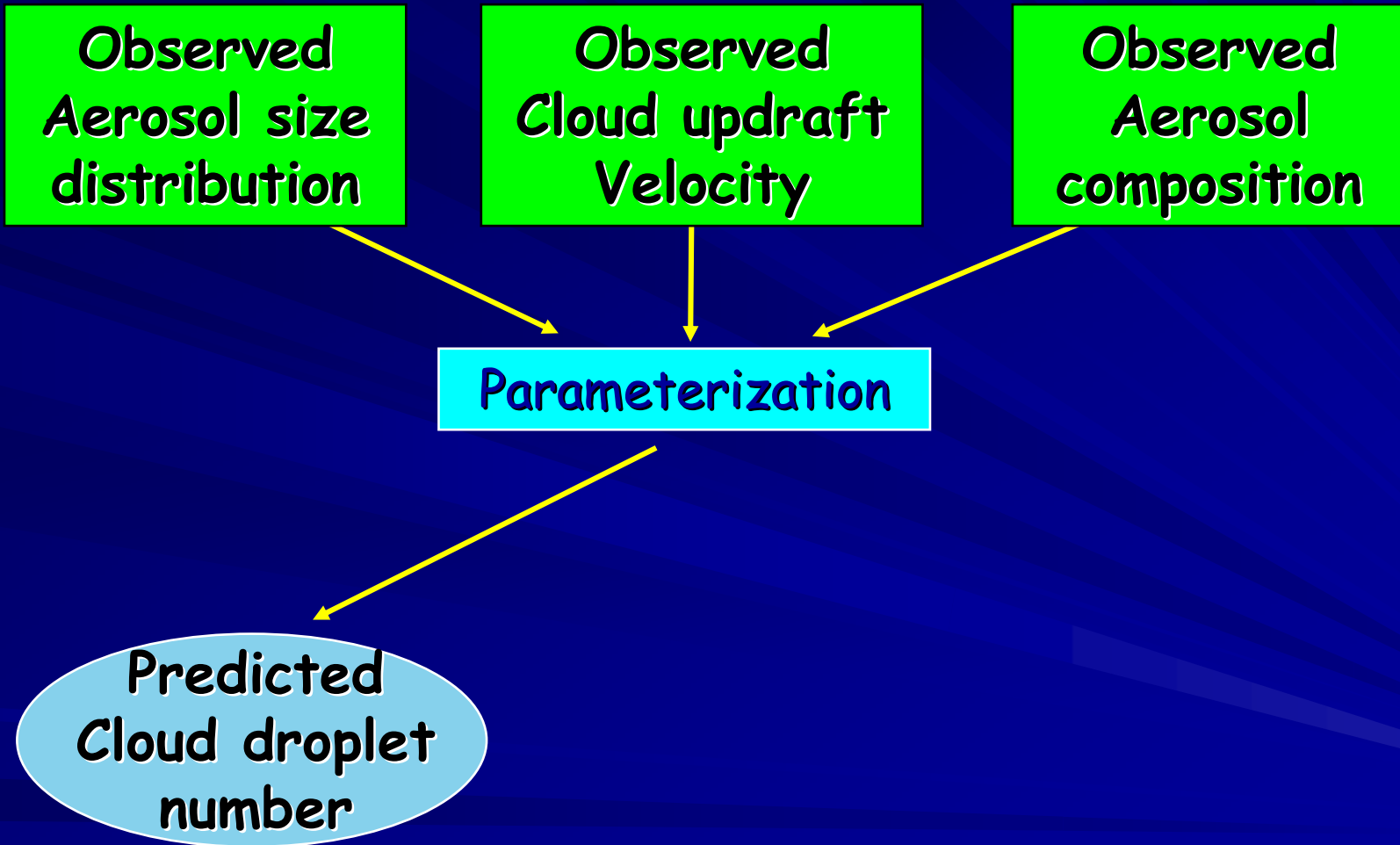
Parameterization Evaluation CDNC "closure"

Observed
Aerosol size
distribution

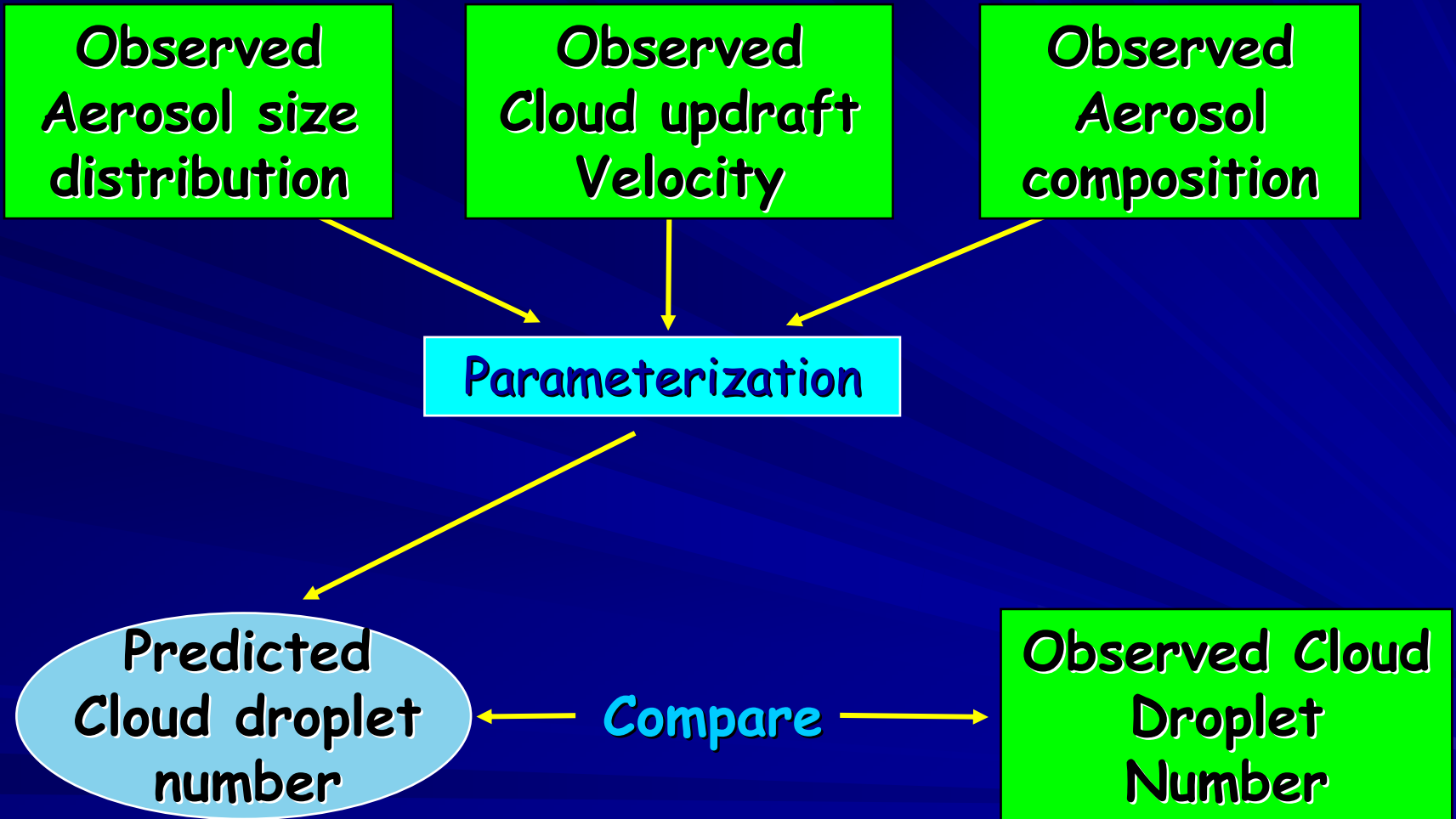
Observed
Cloud updraft
Velocity

Observed
Aerosol
composition

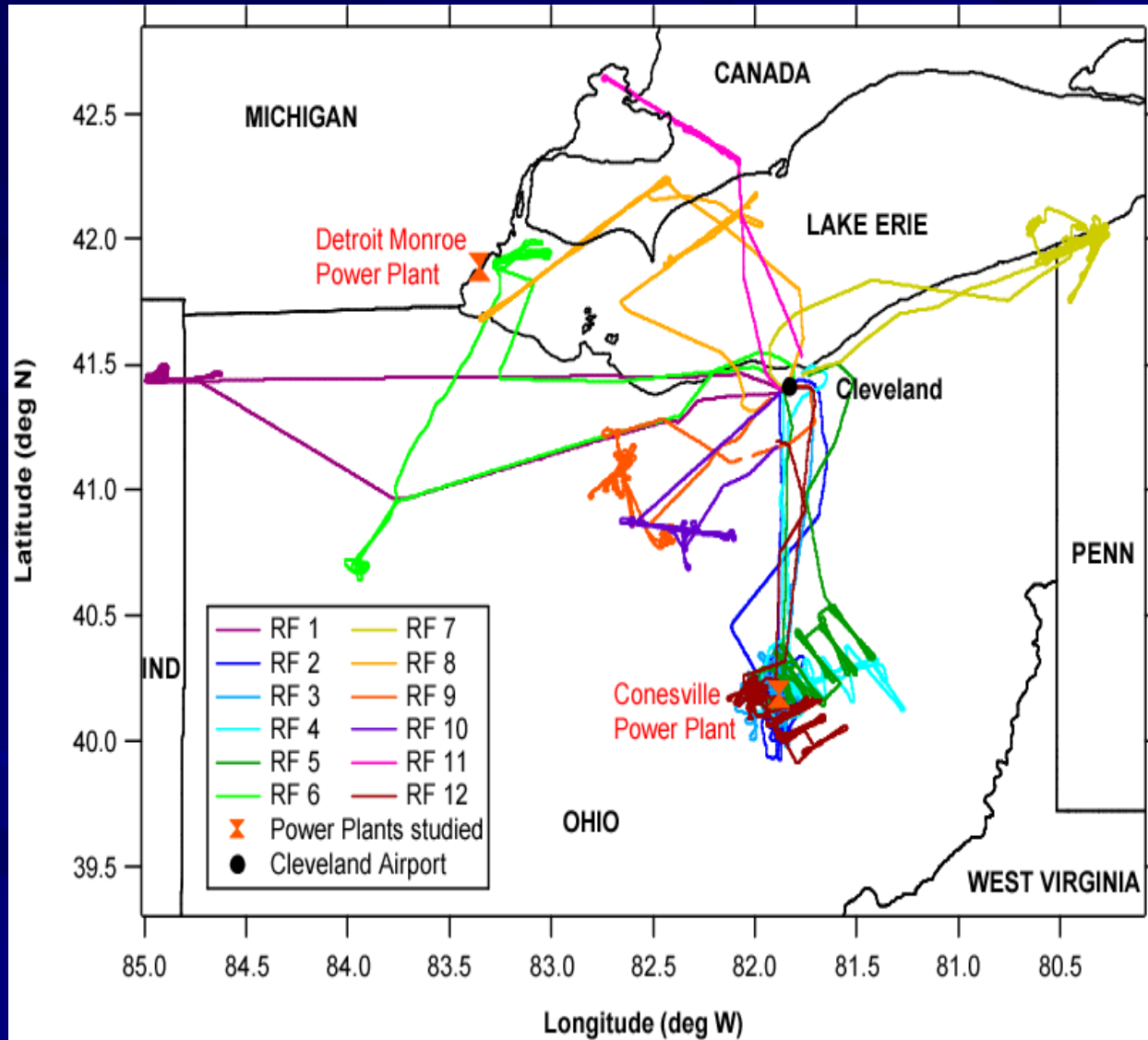
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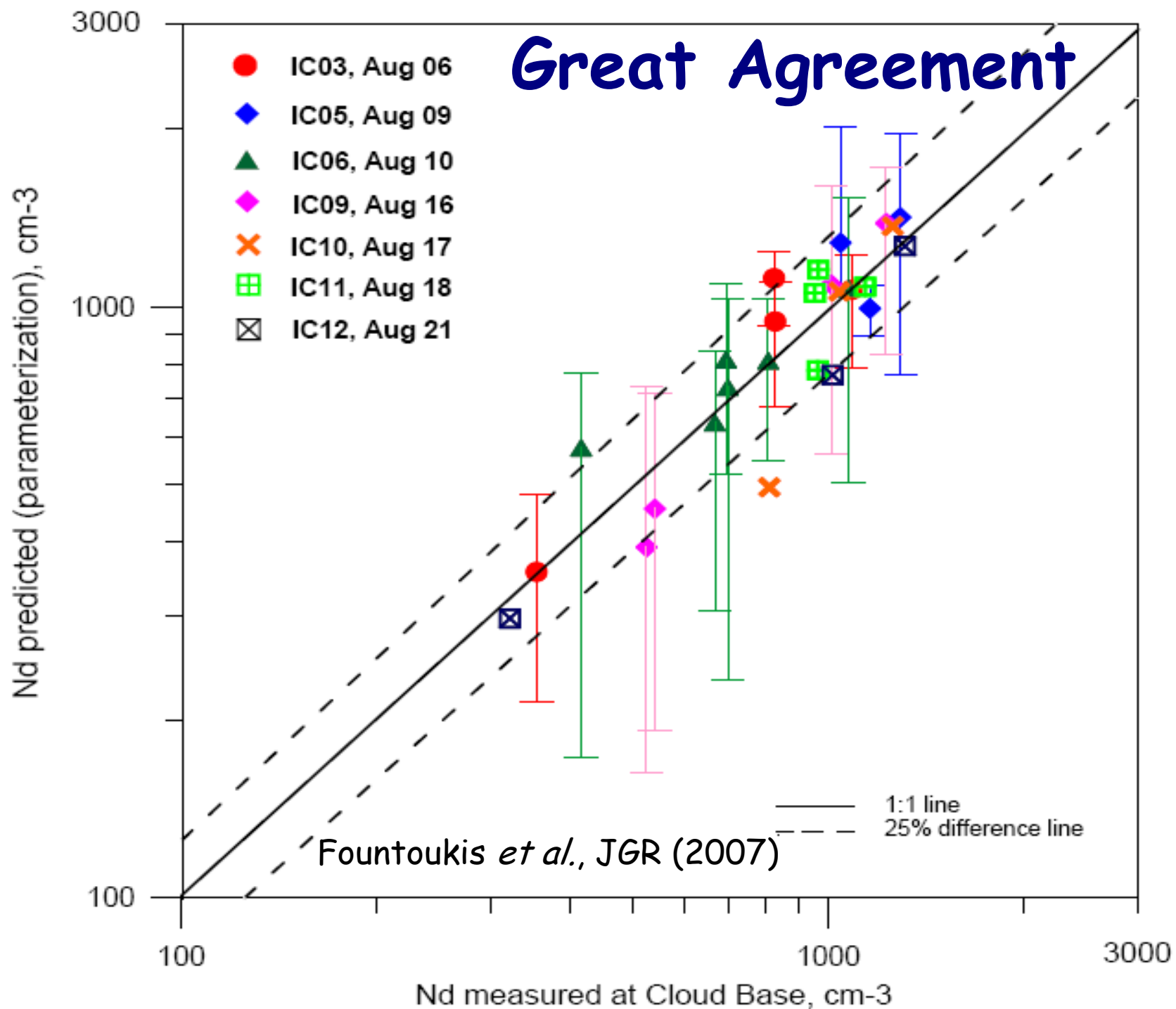
CDNC closure during ICARTT (Aug. 2004)



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



Great Agreement



Summary

- Impacts of atmospheric chemistry on global climate are very important.
- The impacts of aerosol on clouds appreciably diminish warming from greenhouse gases.
- The sensitivity of climate to CO_2 increases are very uncertain, ranging from modest to very large. This is because the indirect effect is very uncertain.
- The indirect effect can regionally be dominant. These impacts will only become larger because of the development of Asian Nations.

Summary

- Climate models are beginning to include physically-based descriptions of aerosol-cloud interactions.
- Observations should provide the “constraints” and “tests” for evaluation of the improved physics developed for climate models.
- A lot of work to do... but we are really seeing the improvements. This could can only be accomplished through the coordinated effort of the scientific community and the support from the funding agencies.

THANK YOU!



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

Barahona and Nenes (2007) parameterization.

- Water vapor uptake coefficient: 0.06
- Entrainment rate varied between $0.0-0.6 \times e_c$
- "Out-of-cloud" RH varied between 65 and 90%
- "Out-of-cloud" T was between 0.1 and 1 K below T_{cloud}

Autoconversion

Khairoutdinov and Kogan (2000)

In-cloud updraft velocity

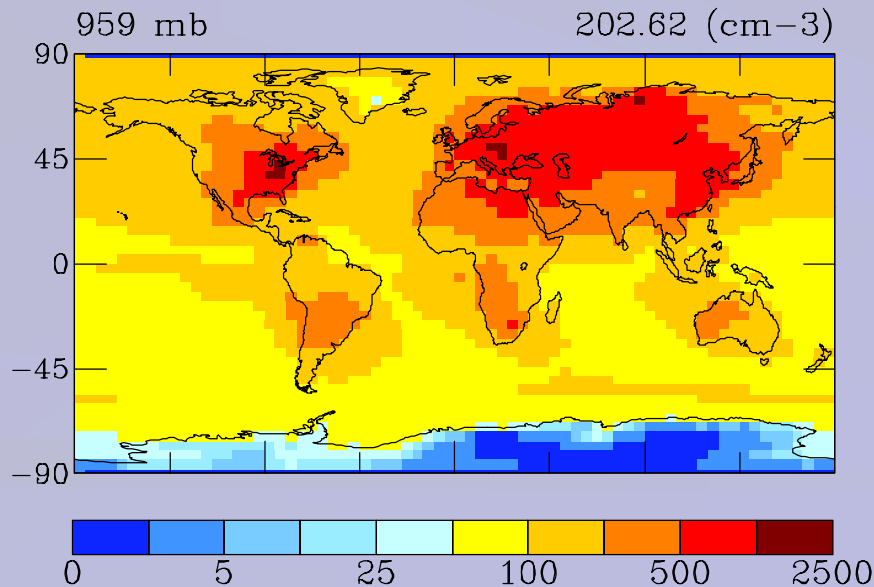
- Prescribed (marine: $0.25-0.5 \text{ ms}^{-1}$; continental: $0.5-1 \text{ ms}^{-1}$).
- Large-scale TKE in a $4' \times 5'$ grid is too separated from the cloud scale.

Current Day Simulation (adiabatic)

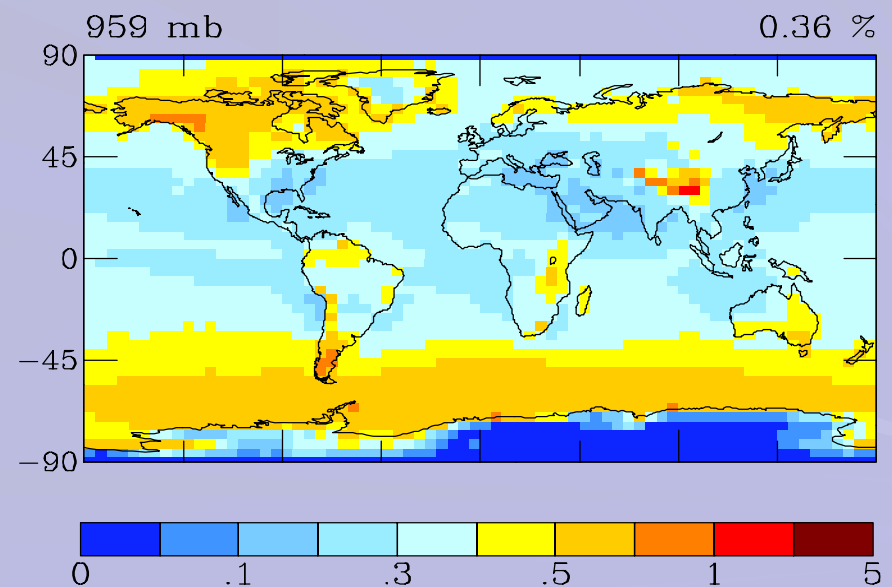
Entrainment rate: $0.0 e_c$

Annual Average Values

Cloud droplets (cm^{-3})

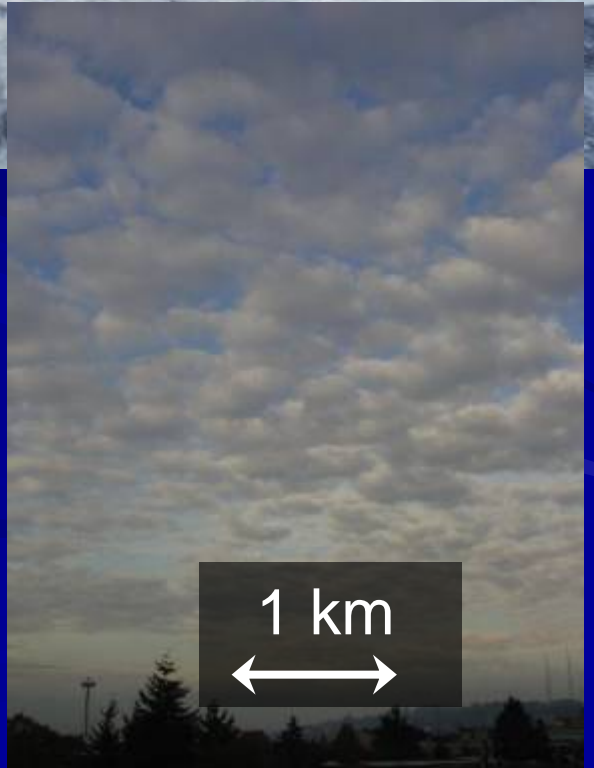
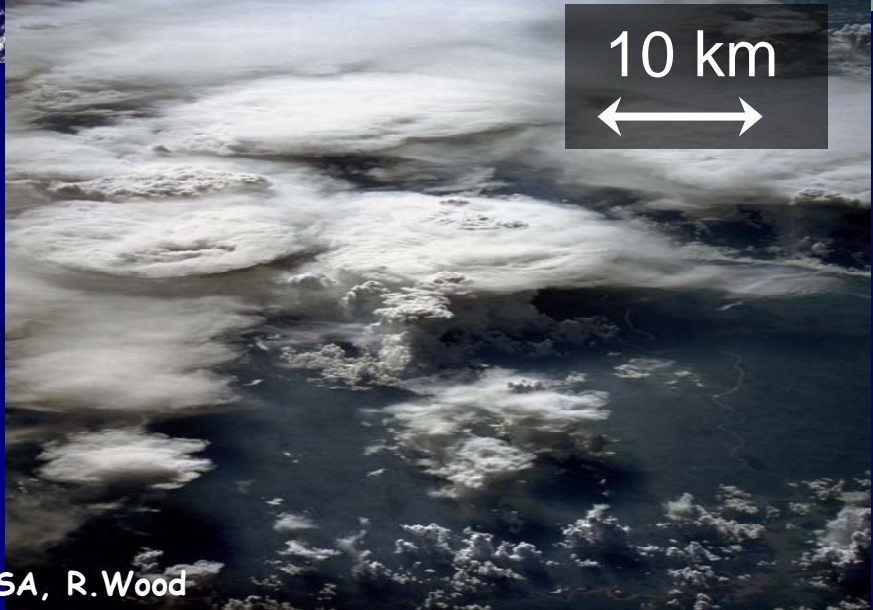
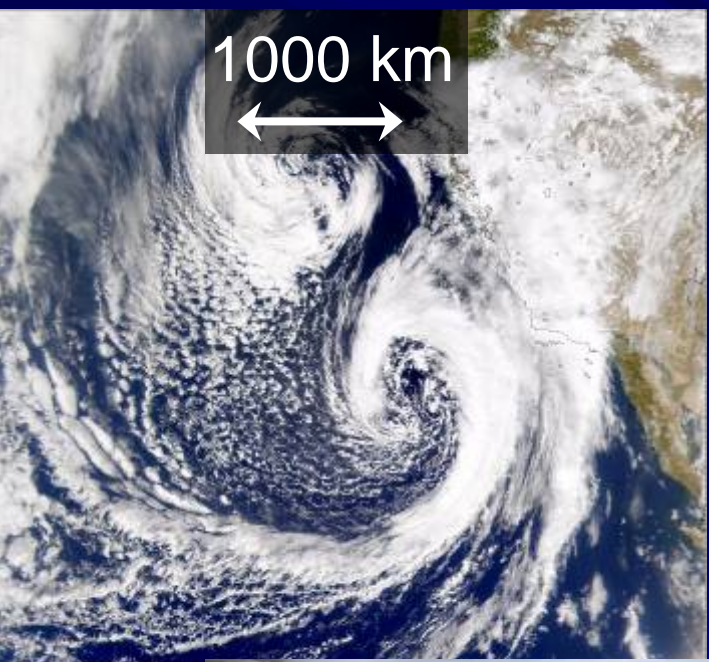


Cloud s_{max} (%)

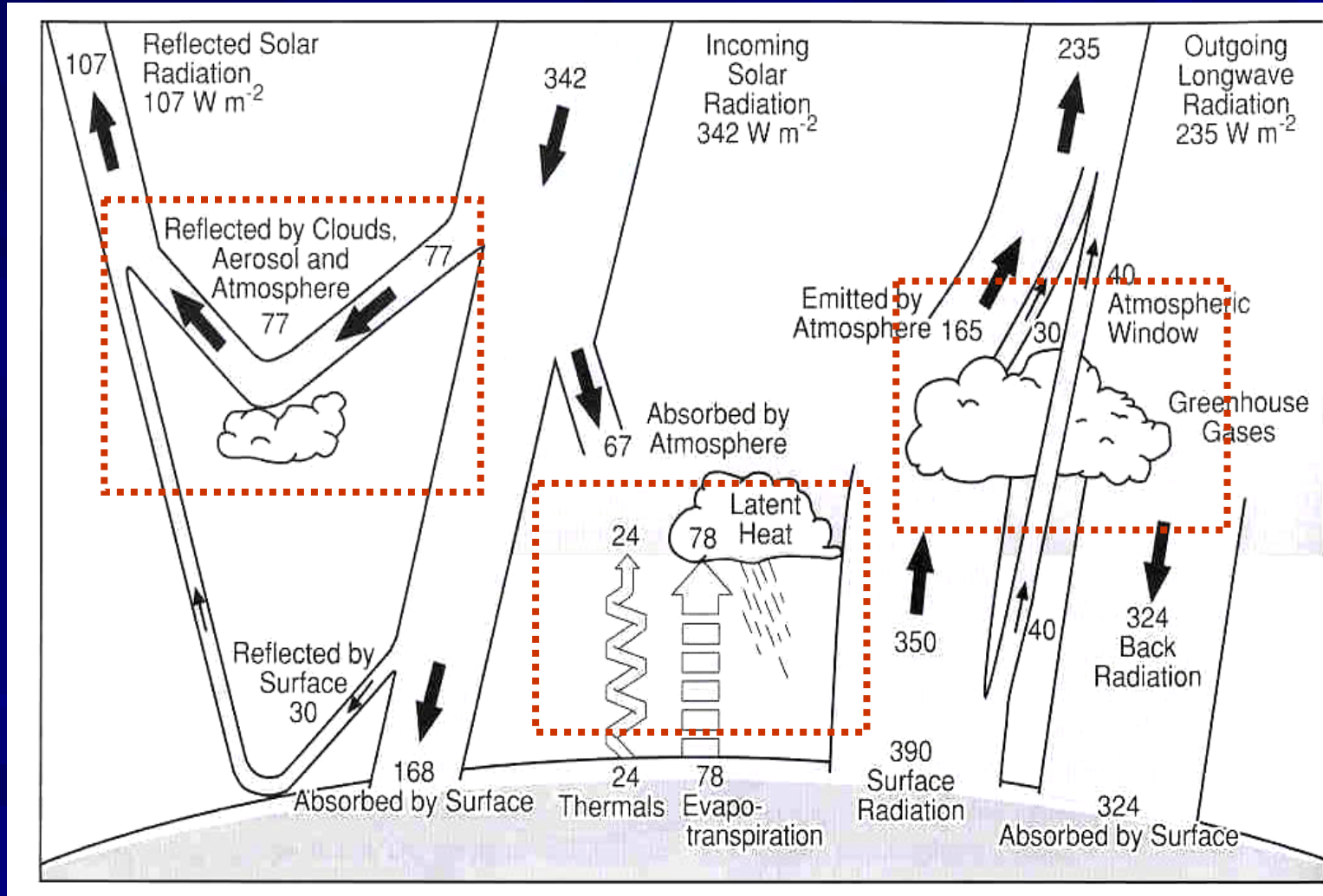


Nenes et al., *in preparation*

Clouds are everywhere and found at all scales...

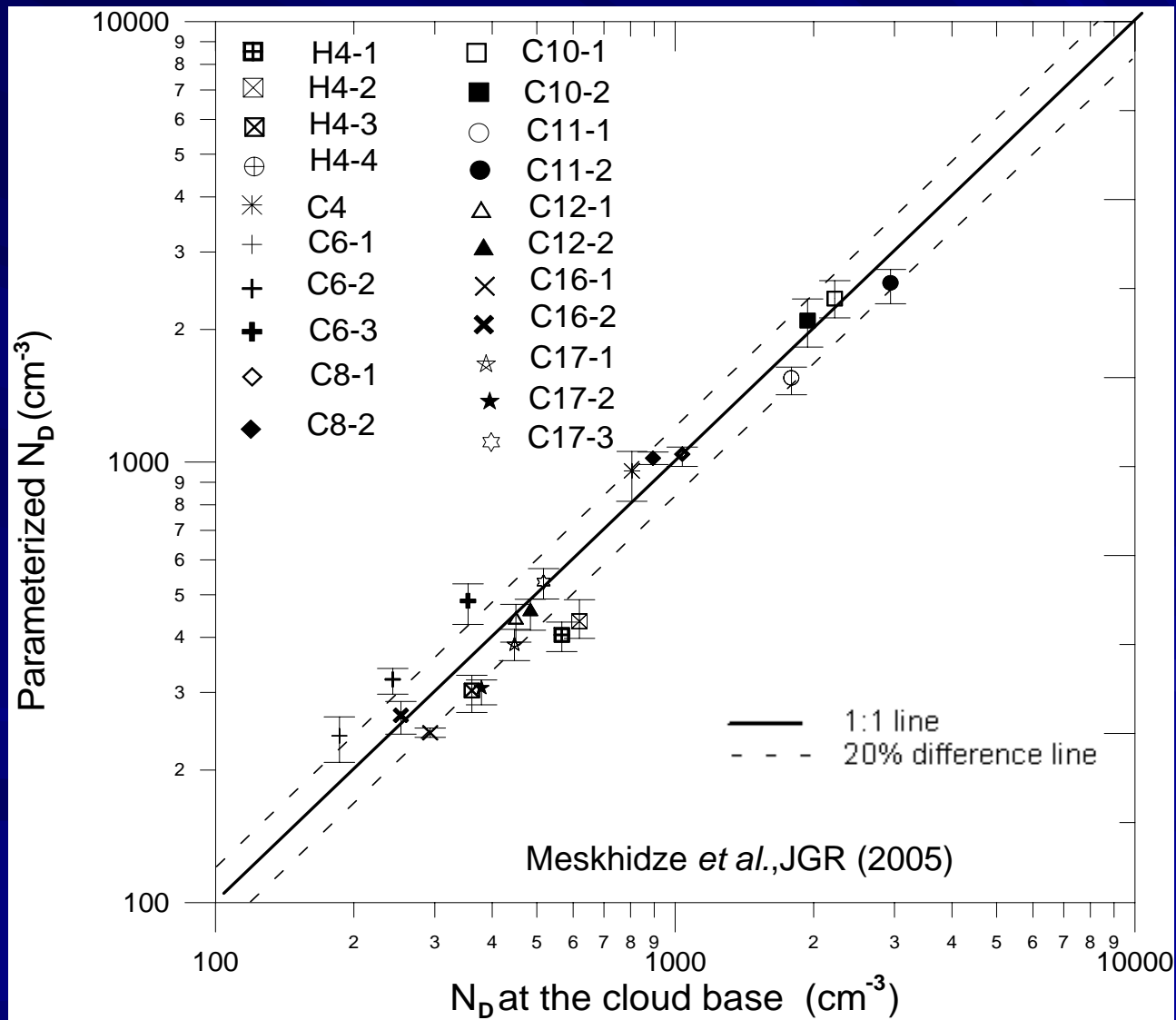


Clouds play a central role in the climate system

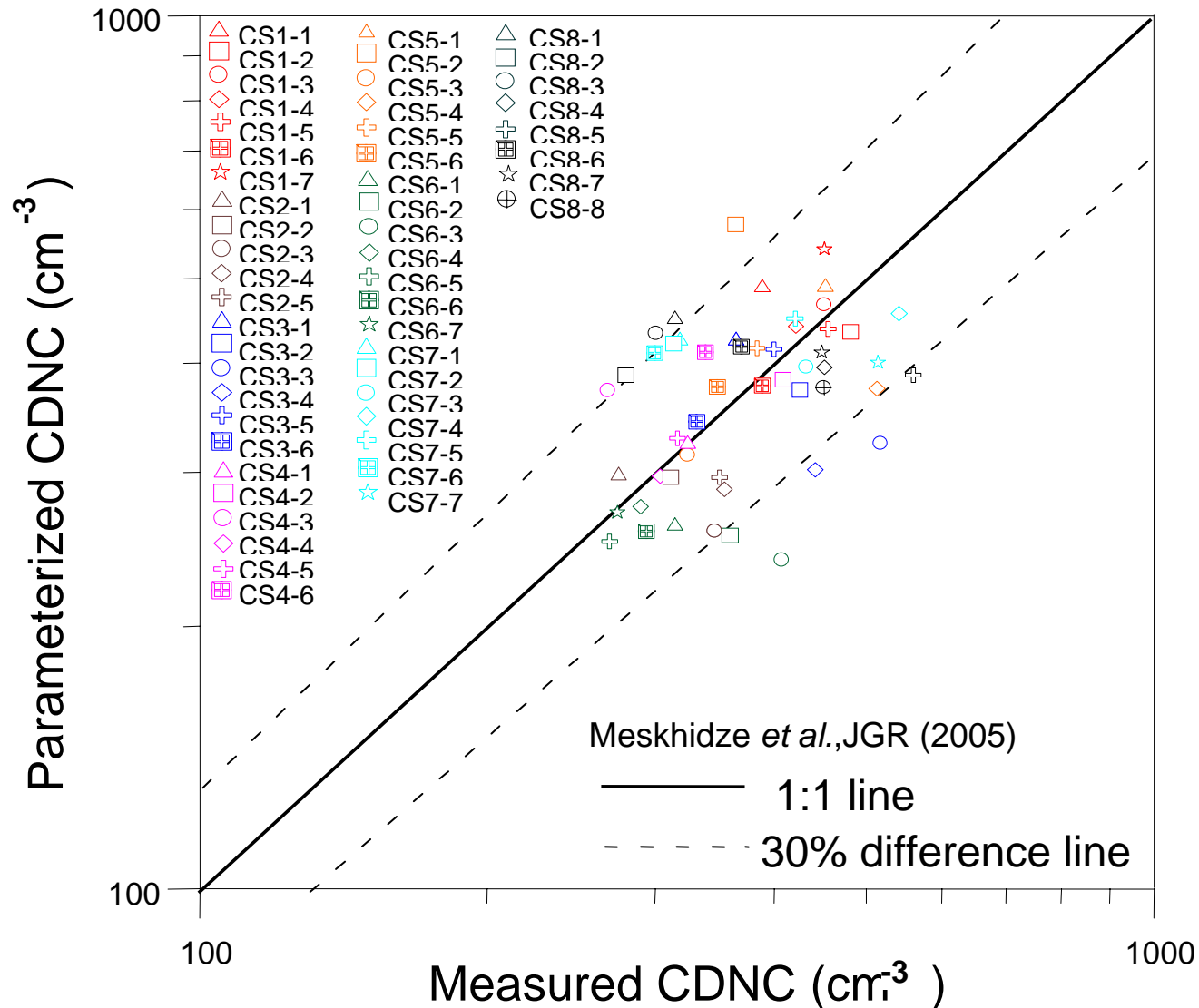




CRYSTAL-FACE (2002) Cumulus clouds



CSTRIPE (2003) Coastal Stratocumulus



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