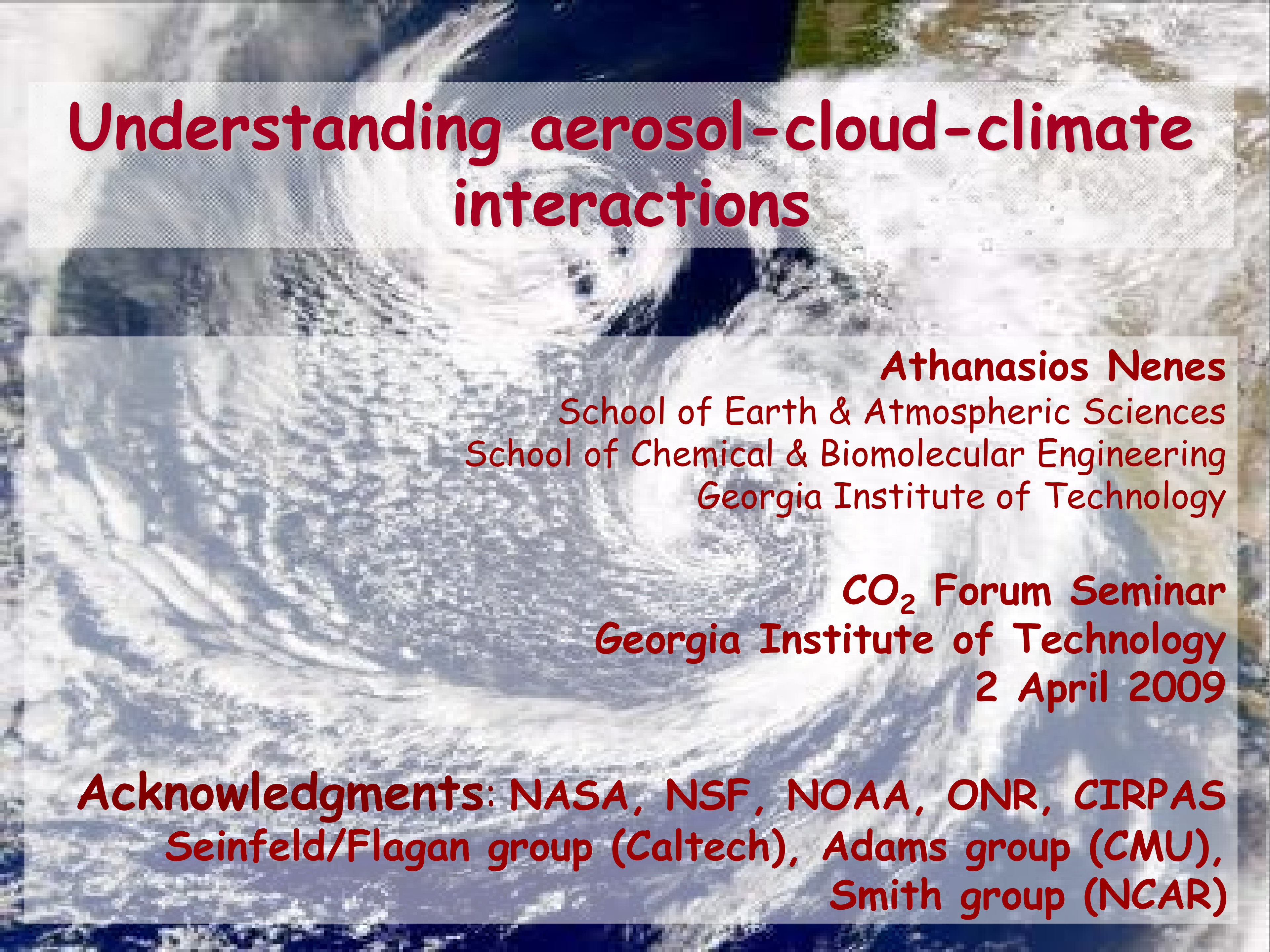

Understanding aerosol-cloud-climate interactions

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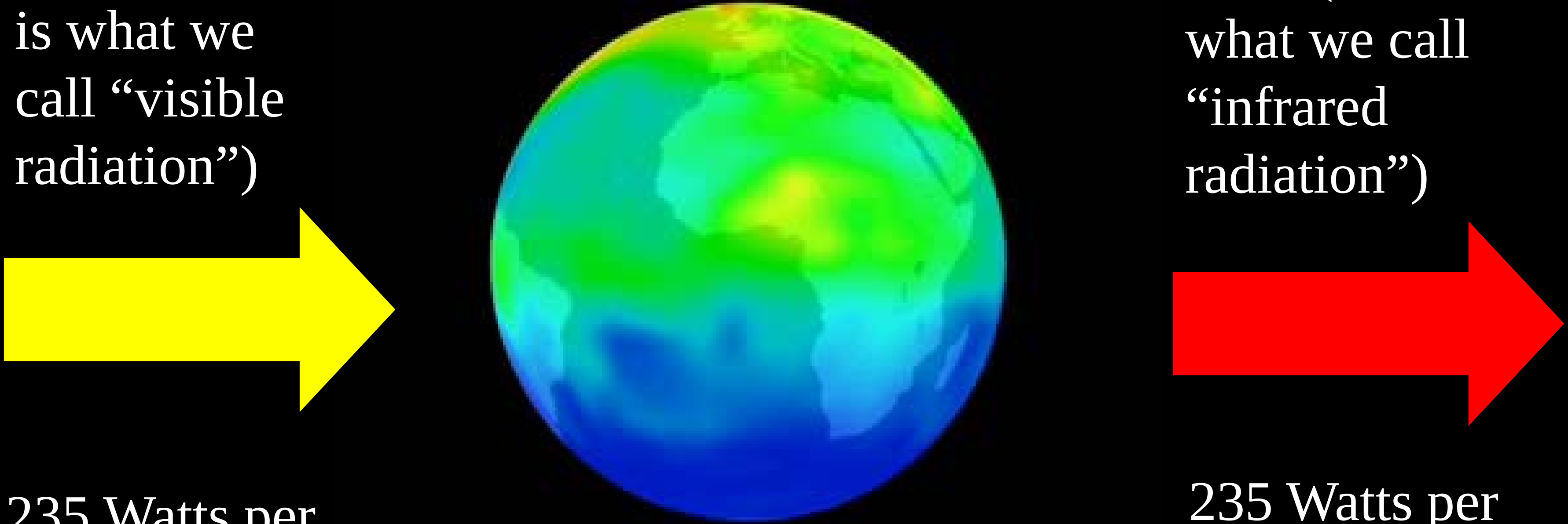
Georgia Institute of Technology

CO_2 Forum Seminar
Georgia Institute of Technology
2 April 2009

Acknowledgments: NASA, NSF, NOAA, ONR, CIRPAS
Seinfeld/Flagan group (Caltech), Adams group (CMU),
Smith group (NCAR)

Earth’s Energy Balance



Sunlight (this is what we call “visible radiation”)

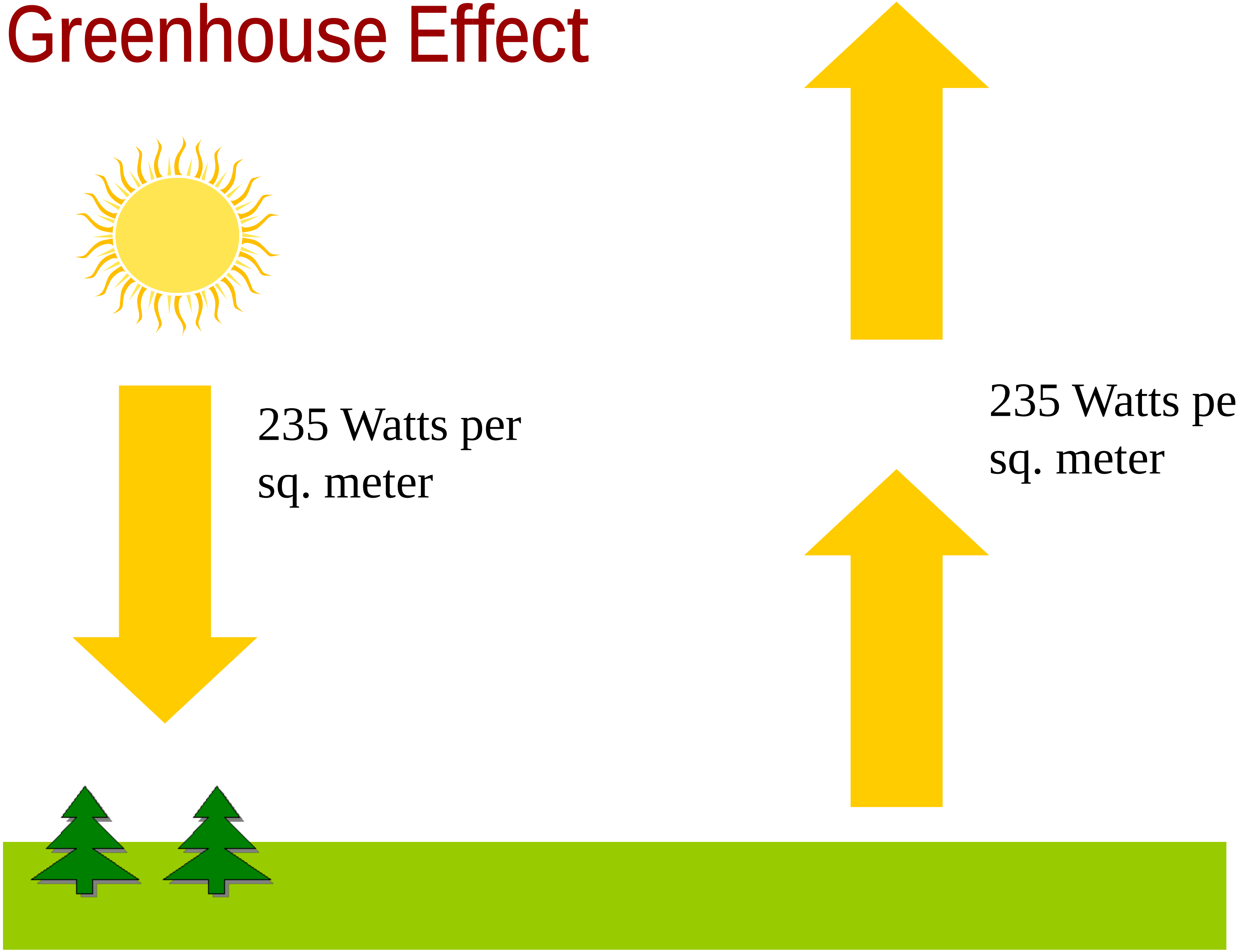
235 Watts per square meter (Wm^{-2})

Heat (this is what we call “infrared radiation”)

235 Watts per square meter (Wm^{-2})

When energy IN = energy OUT, climate is “in balance” (i.e., steady state)

Greenhouse Effect

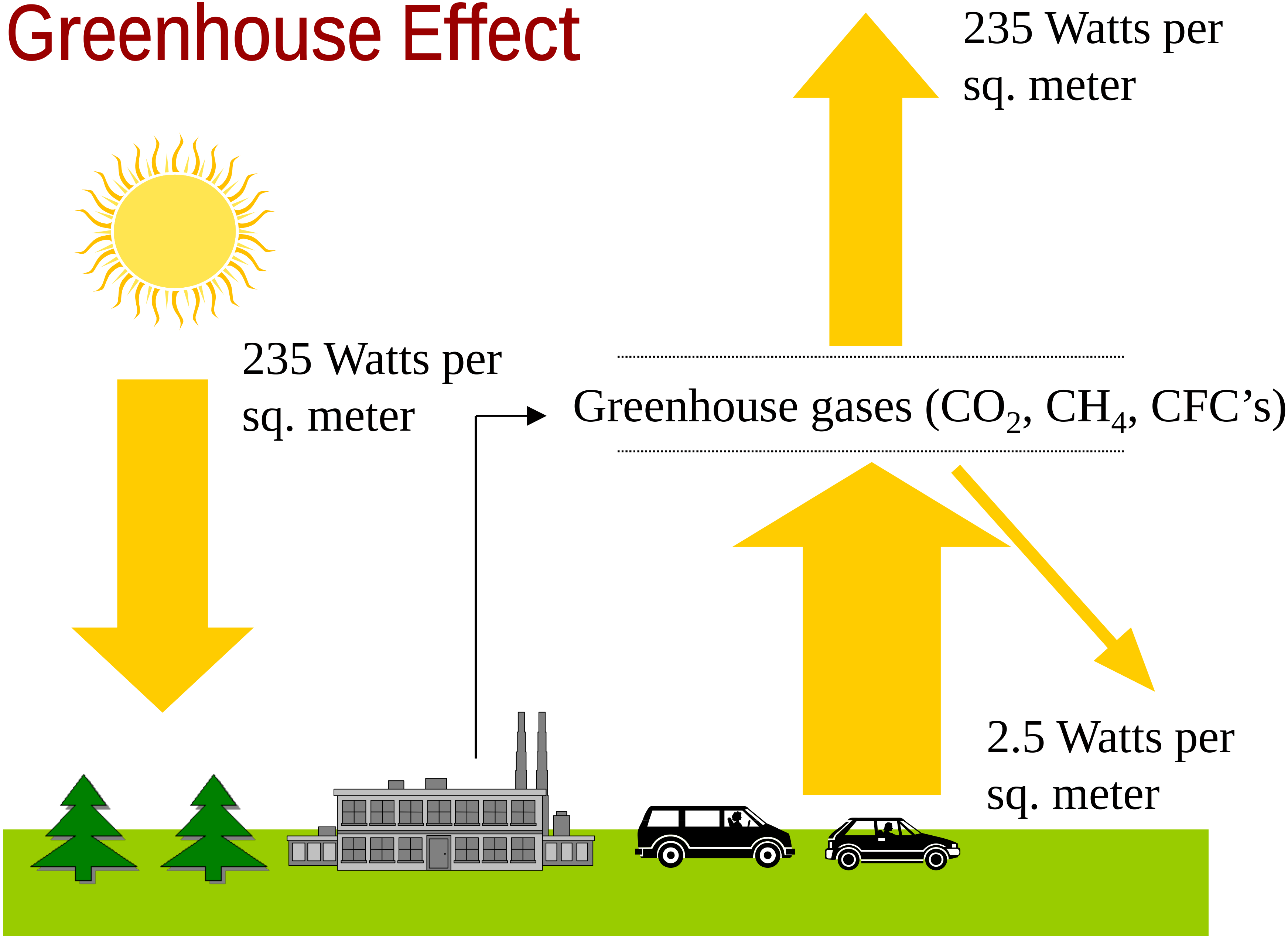


235 Watts per
sq. meter

235 Watts per
sq. meter

“Natural”, “Preindustrial” state of climate (before late 1700’s)

Greenhouse Effect



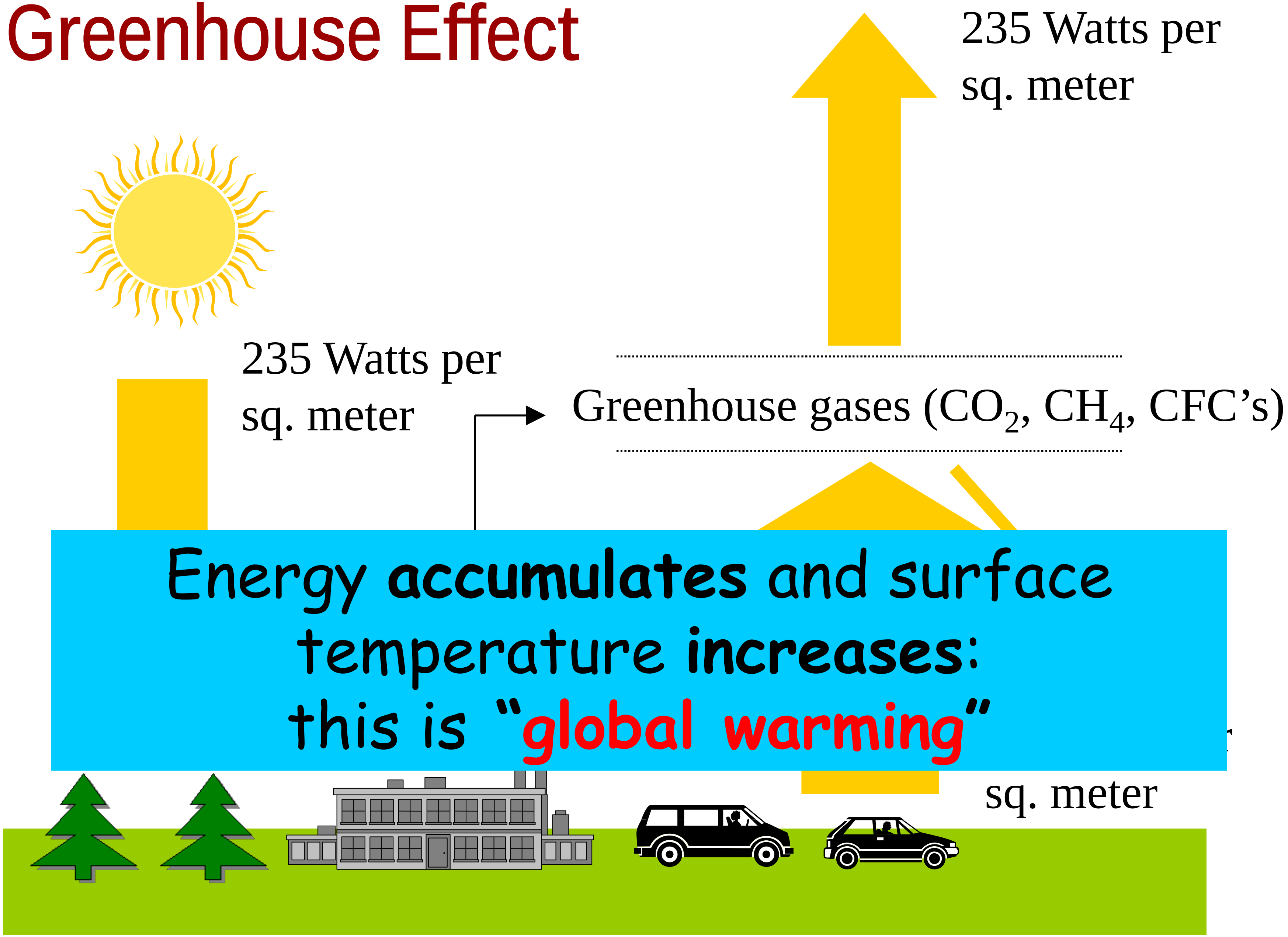
235 Watts per sq. meter

235 Watts per sq. meter

Greenhouse gases (CO_2, CH_4, CFC’s)

2.5 Watts per sq. meter

Greenhouse Effect



235 Watts per sq. meter

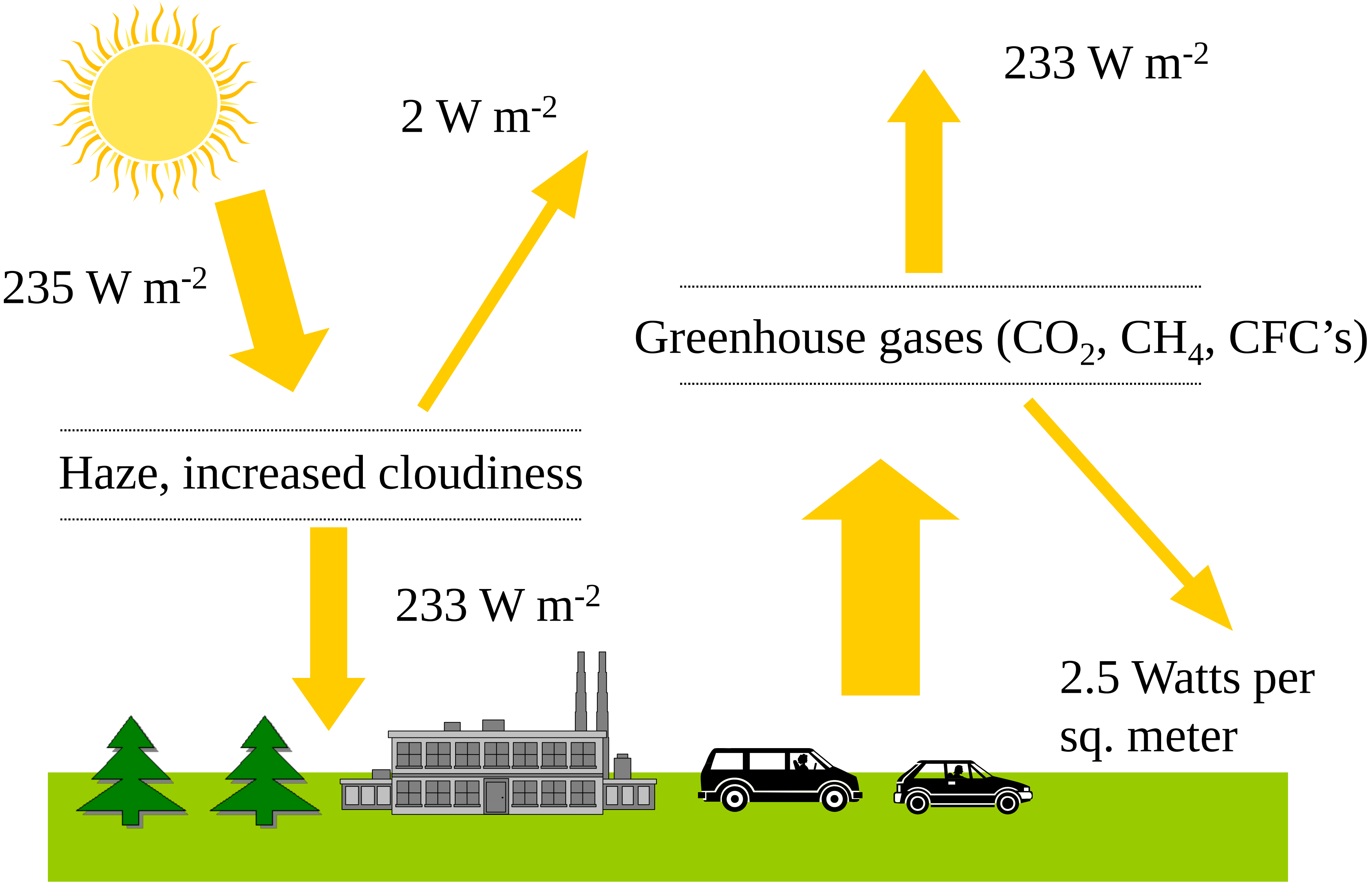
235 Watts per sq. meter

Greenhouse gases (CO_2, CH_4, CFC’s)

sq. meter

Energy **accumulates** and surface temperature **increases**:
this is “global warming”

“Whitehouse” Effect



2 W m^{-2}

233 W m^{-2}

235 W m^{-2}

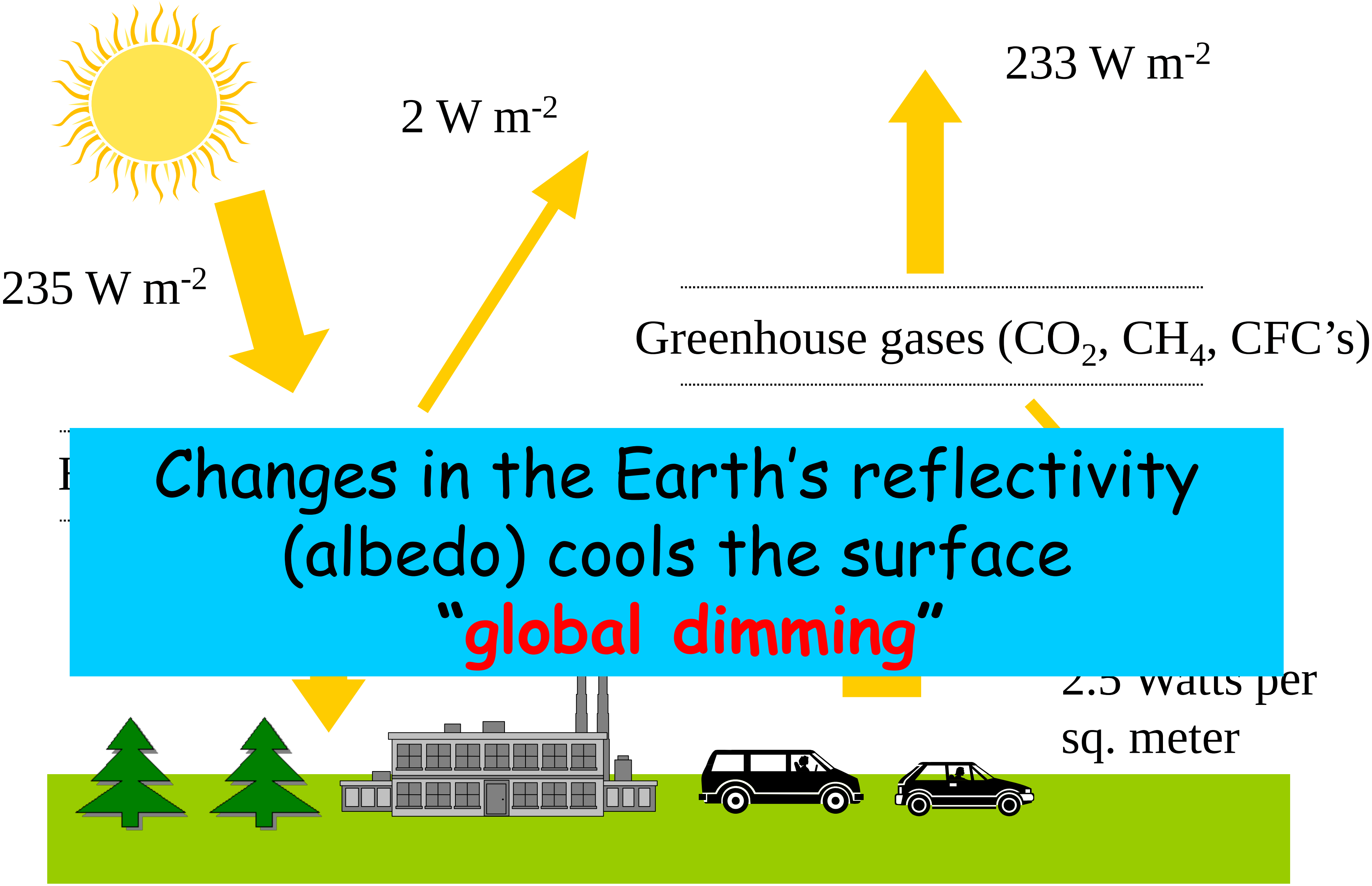
Greenhouse gases (CO_2, CH_4, CFC’s)

Haze, increased cloudiness

233 W m^{-2}

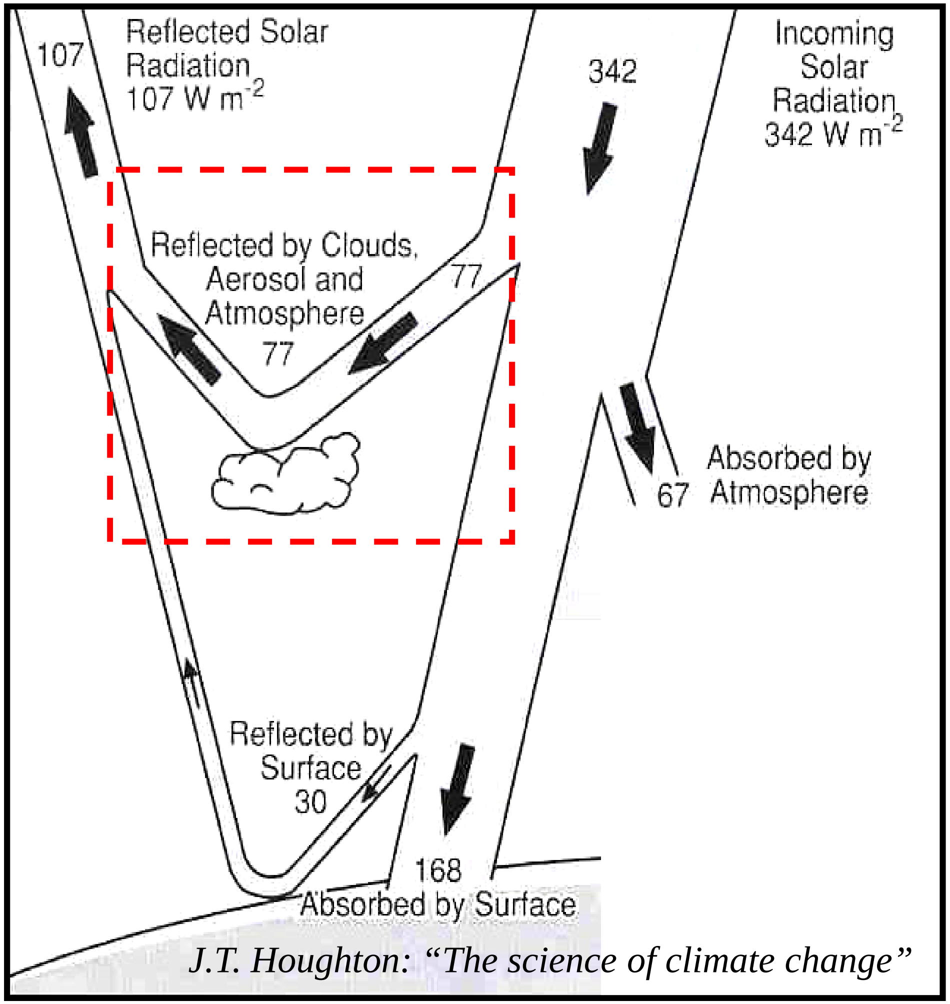
2.5 Watts per
sq. meter

“Whitehouse” Effect



Changes in the Earth’s reflectivity
(albedo) cools the surface
“global dimming”

Earth's Albedo: controlling factor of "global dimming"



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.
- **A few %** increase in global cloud cover can counteract warming from greenhouse gases.

Consequence:

Understanding cloud formation is required for assessments of anthropogenic climate change.

Clouds are VERY dynamic (difficult to simulate).

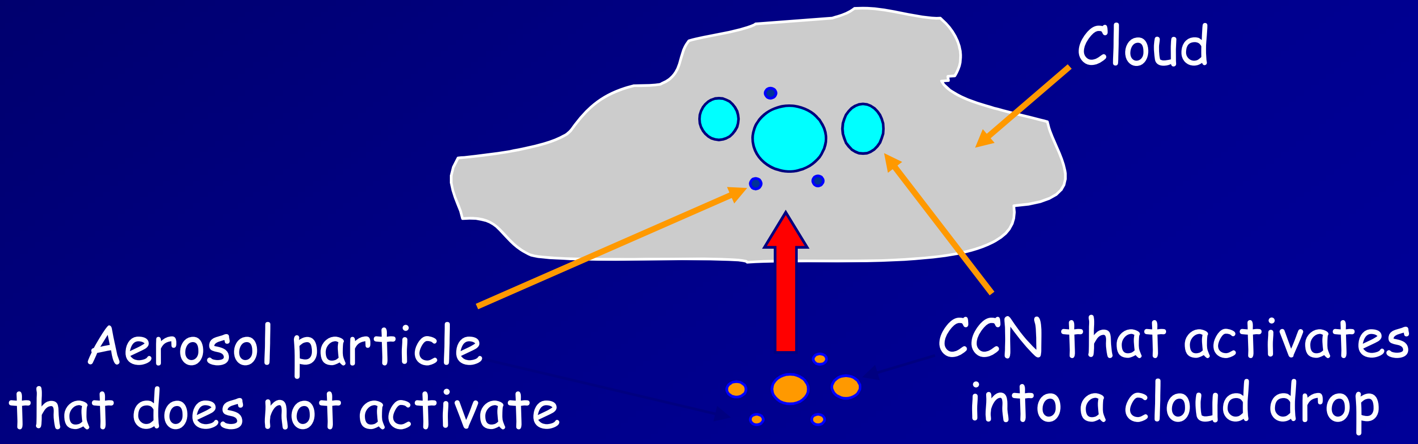
How do (liquid water) clouds form?

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.

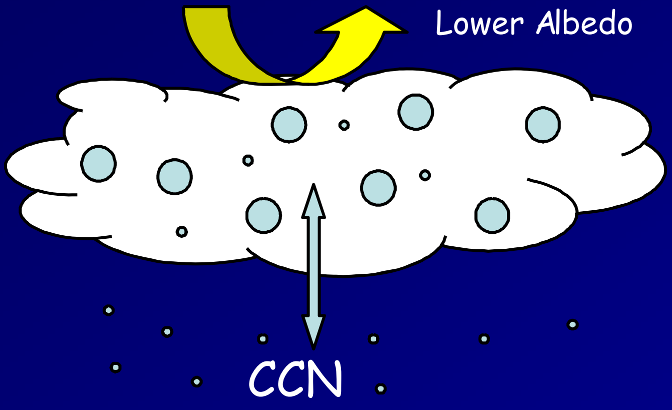
Aerosols that can become droplets are called cloud condensation nuclei (CCN).



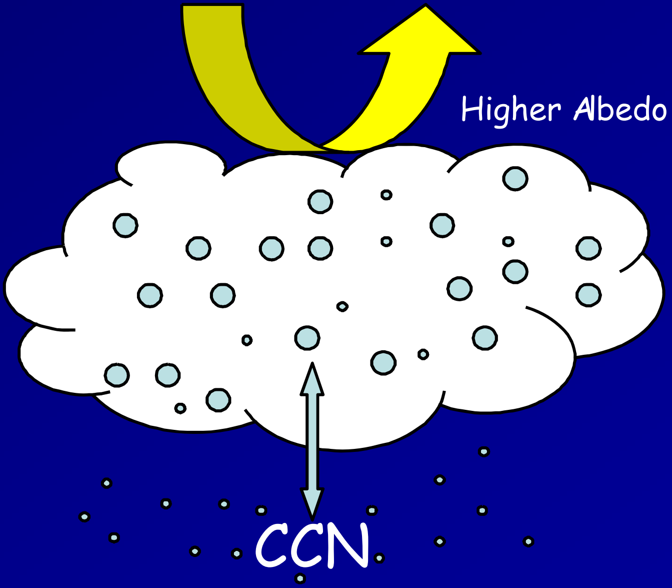
Can humans affect clouds and the hydrological cycle?

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

Result**:** Clouds tend to be "whiter", change in precipitate efficiency. This yields a net cooling on climate and is called the "**indirect climatic effect of aerosols**".



Clean Environment
(few CCN)



Polluted Environment
(more CCN)

Increasing particles tends to cool climate (potentially alot).

Quantitative assessments done with climate models.

Humans have a large impact on global aerosol

Pollution is a global problem. CCN are emitted together with greenhouse gases.

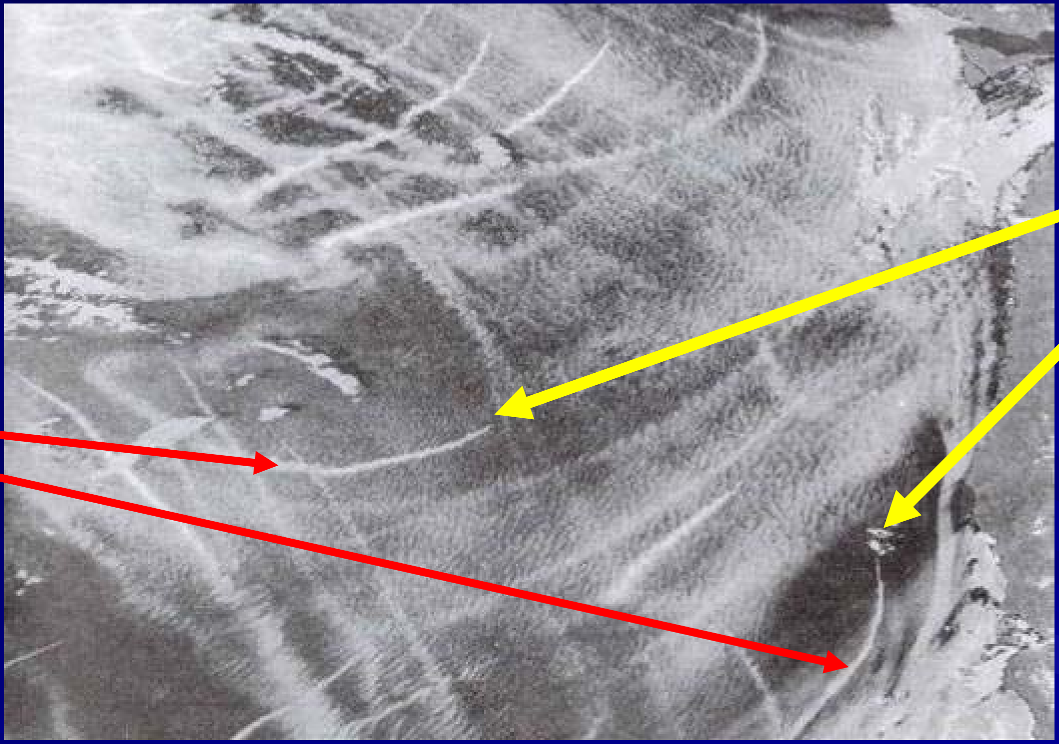
Pollution plumes
(SE Asia, Summer 2002).

Biomass burning
(Mediterranean, Summer 2007).

Photos: NASA

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

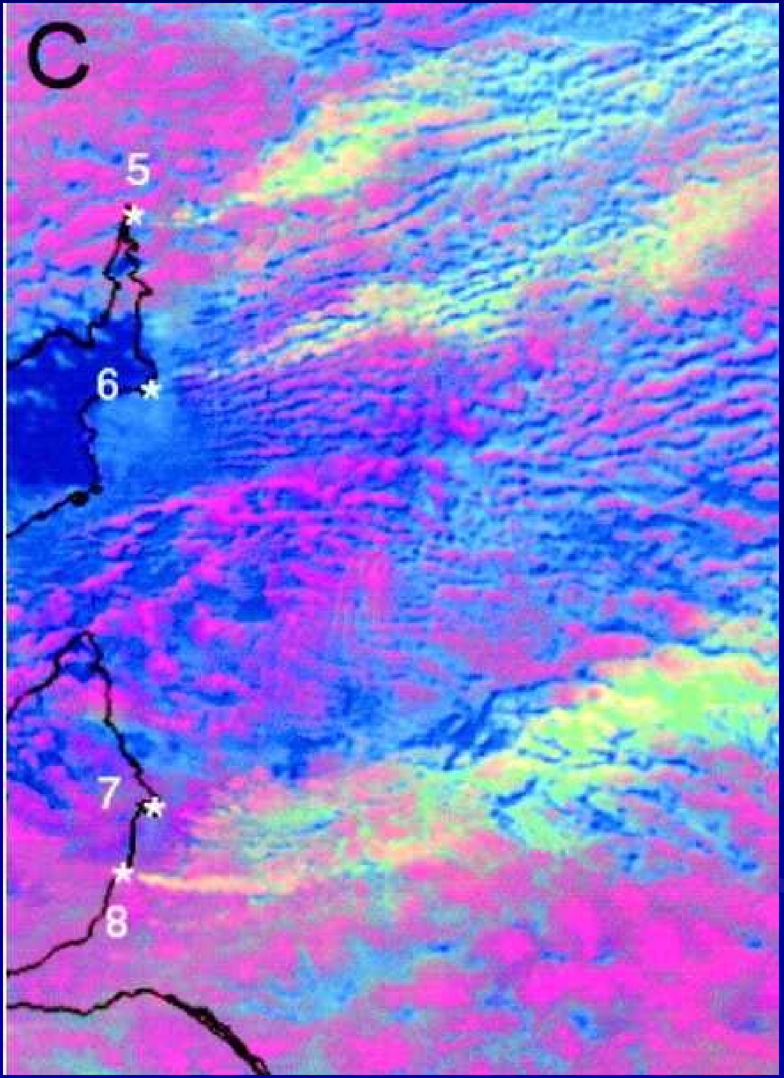
Clouds are whiter and last longer

Pollution ↑ ⇒ Droplet number ↑ ⇒ Droplet size ↓

Droplet size ↓ ⇒ Cloud reflectivity ↑ AND Precip ↓

Observational evidence of indirect effect

Satellite observation of clouds in the Black Sea.



Rosenfeld et al., Science

Red: Clouds with low reflectivity.

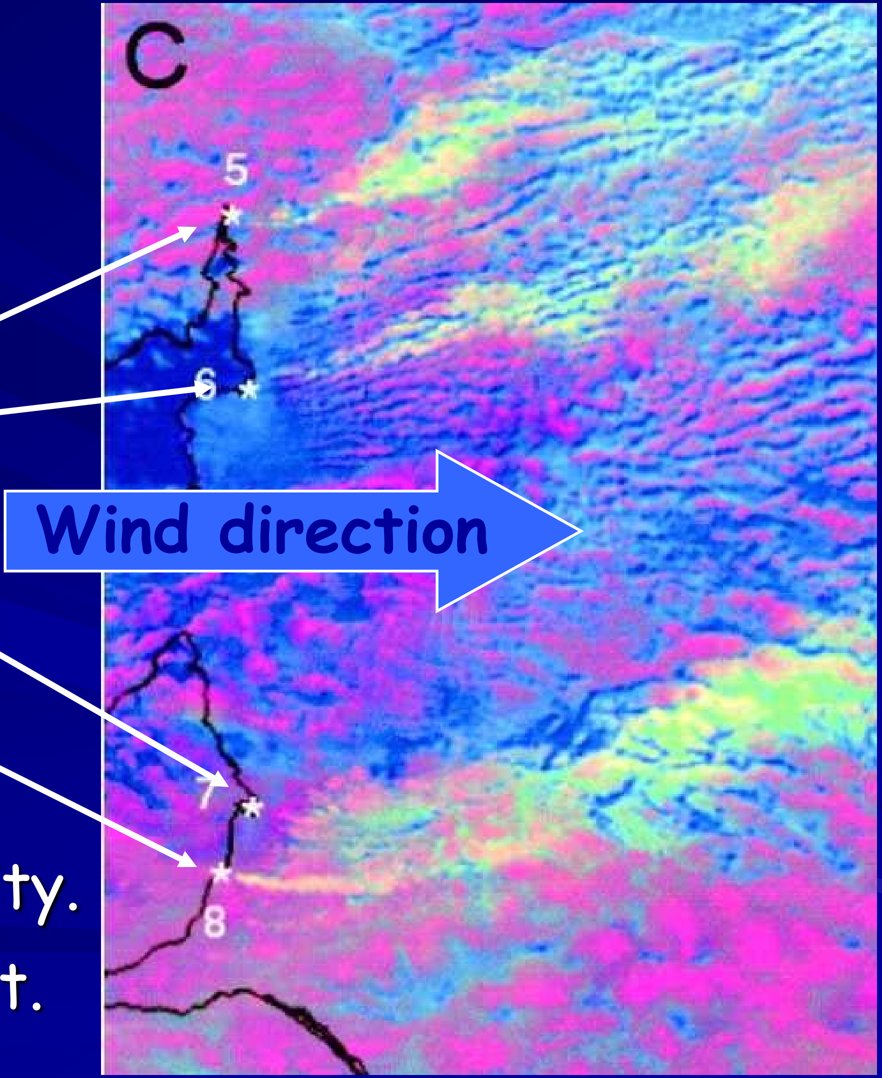
White: Clouds that reflect alot.

Blue: Clear sky.

Observational evidence of indirect effect

Air pollution can affect cloud properties

Satellite observation of clouds in the Black Sea.



Power plant

Lead smelter

Port

Oil refineries

Wind direction

Red: Clouds with low reflectivity.

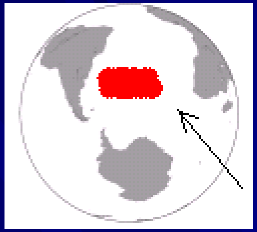
White: Clouds that reflect alot.

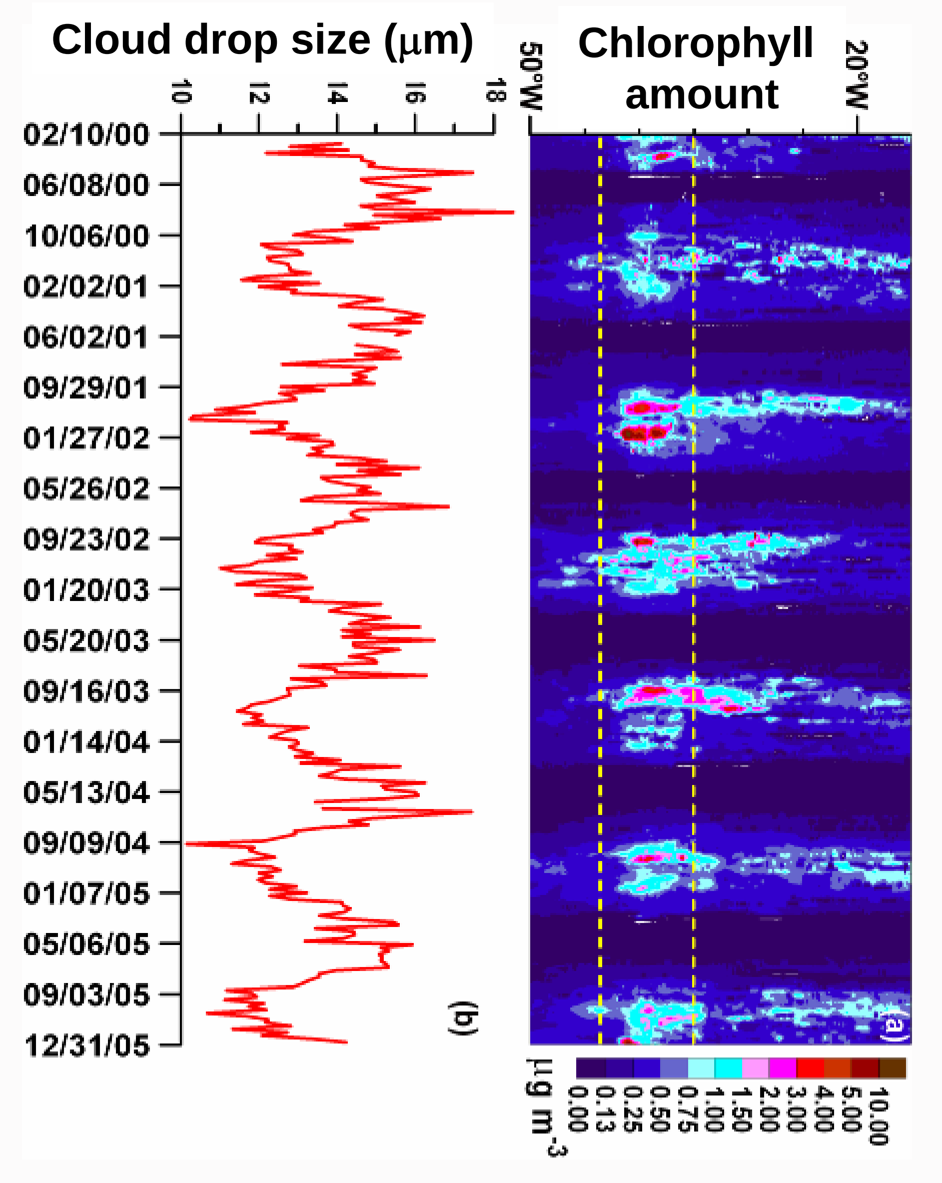
Blue: Clear sky.

Rosenfeld et al., Science

Phytoplankton affect clouds too…

Location: East of Patagonia (South America)





Meskhidze and Nenes, Science, 2006

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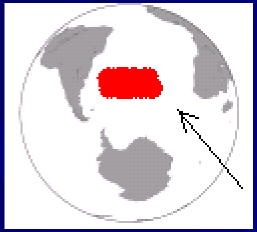

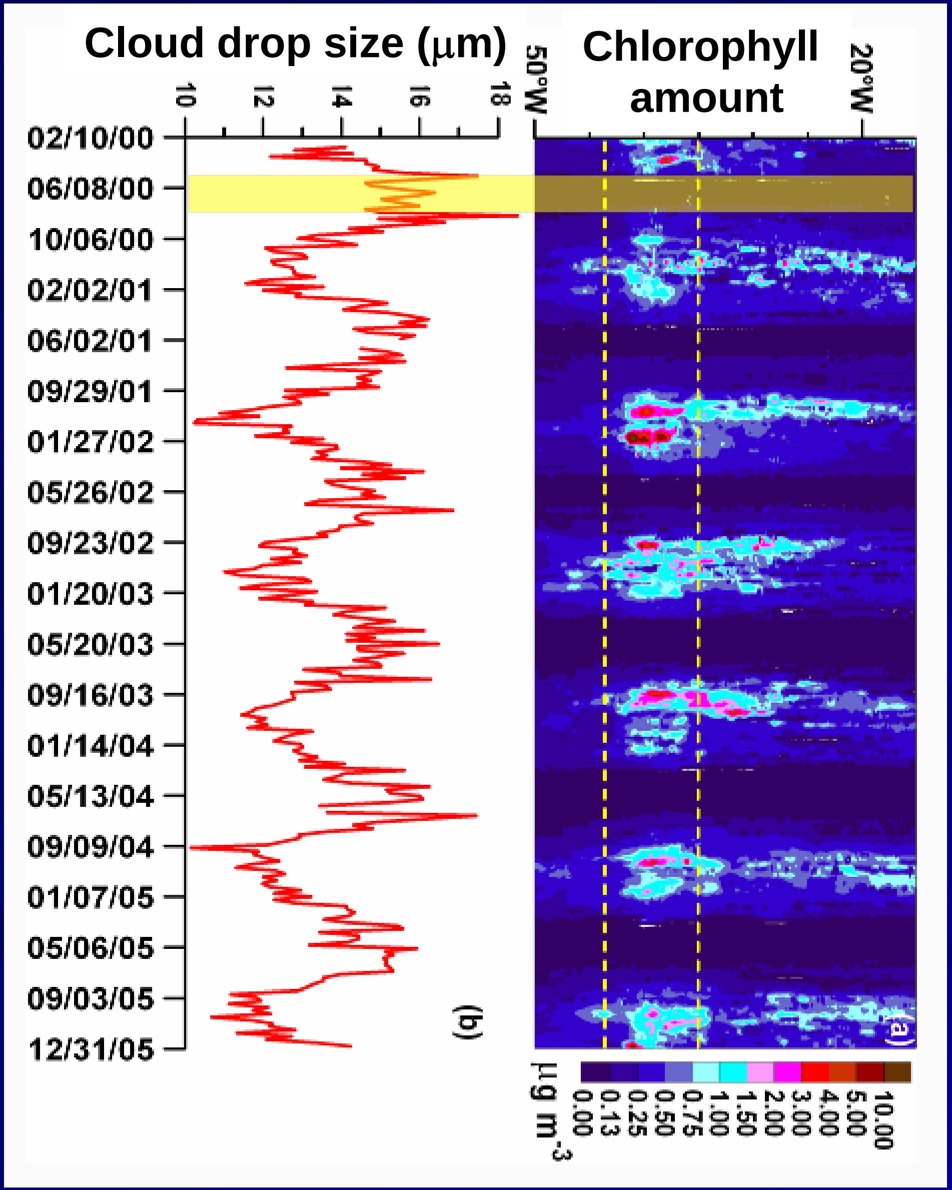

Low chlorophyll period, clouds have large drops (not very reflective)

Meskhidze and Nenes, Science, 2006

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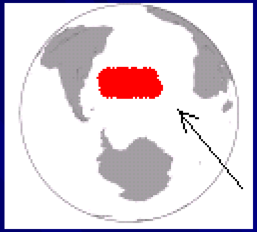

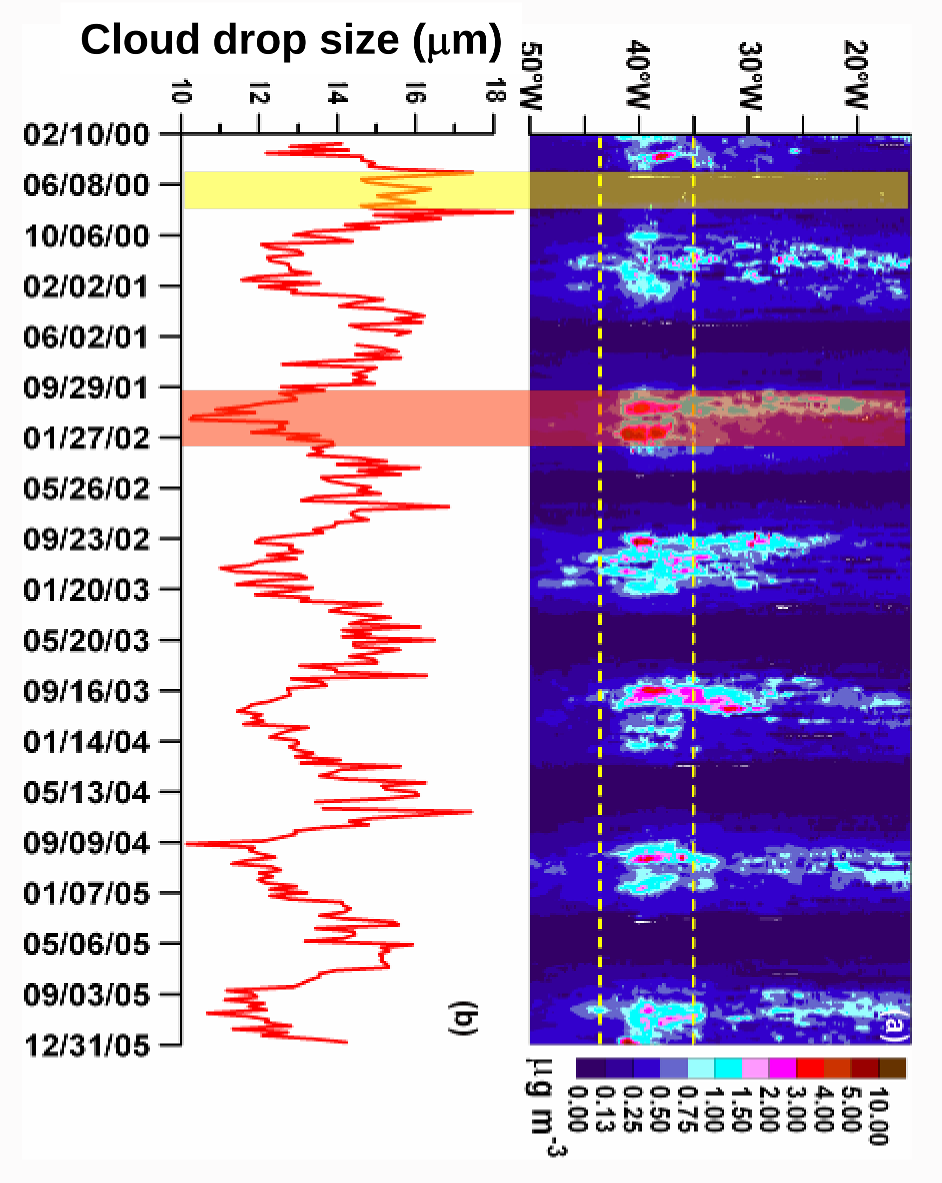

Low chlorophyll period, clouds have large drops (not very reflective)

High Chlorophyll period, Clouds have small drops (very reflective)

Meskhidze and Nenes, Science, 2006

Phytoplankton affect clouds too…

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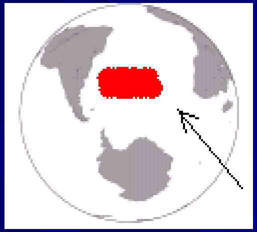
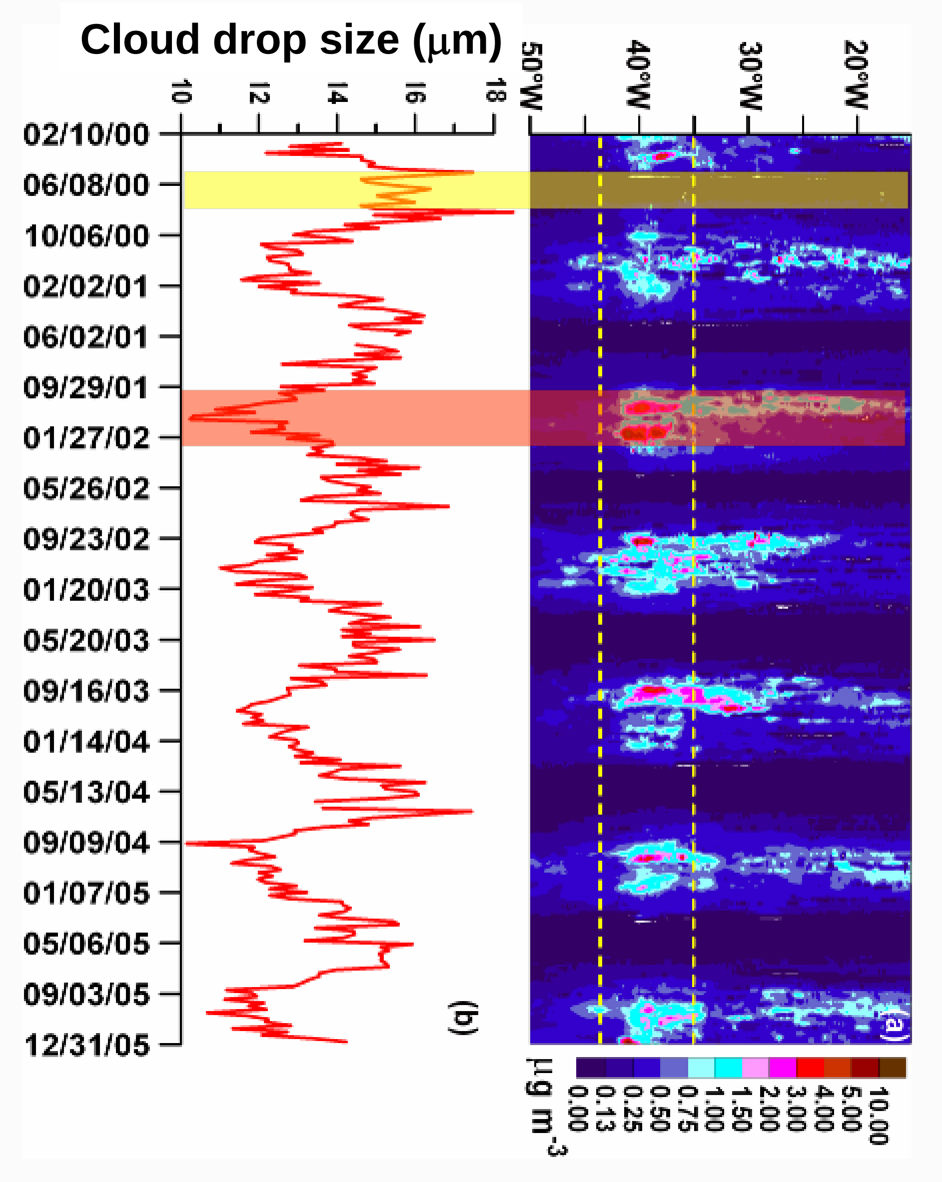




Low chlorophyll period, clouds have large drops (not very reflective)

High Chlorophyll period, Clouds have small drops (very reflective)

Phytoplankton emissions increase particle loads, and strongly impact clouds.

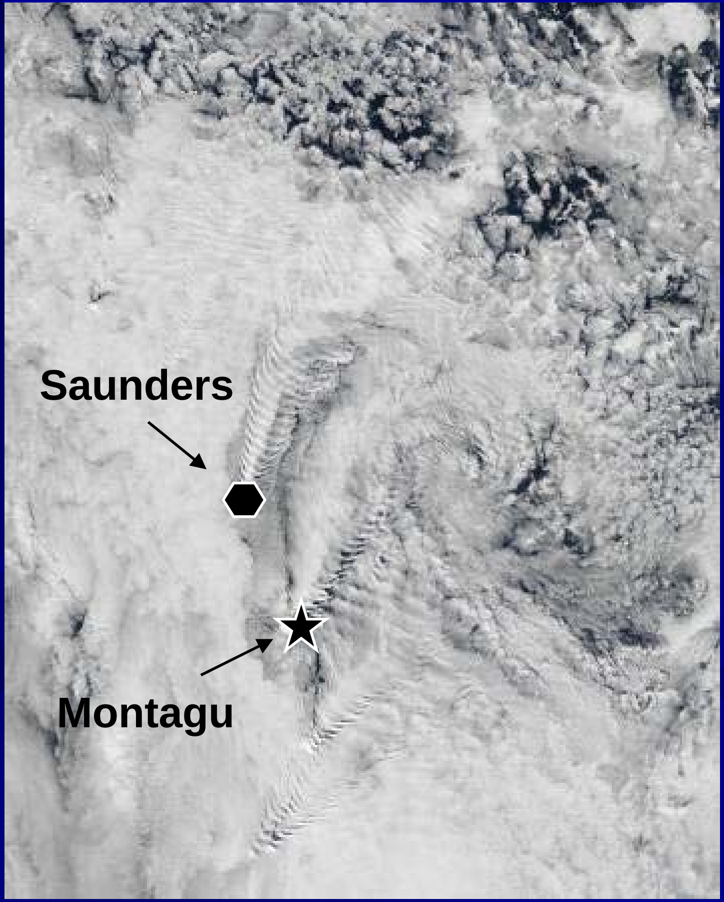
Changes are comparable to contrasts between polluted and clean environments (forcing ~ -15 W m^{-2}).

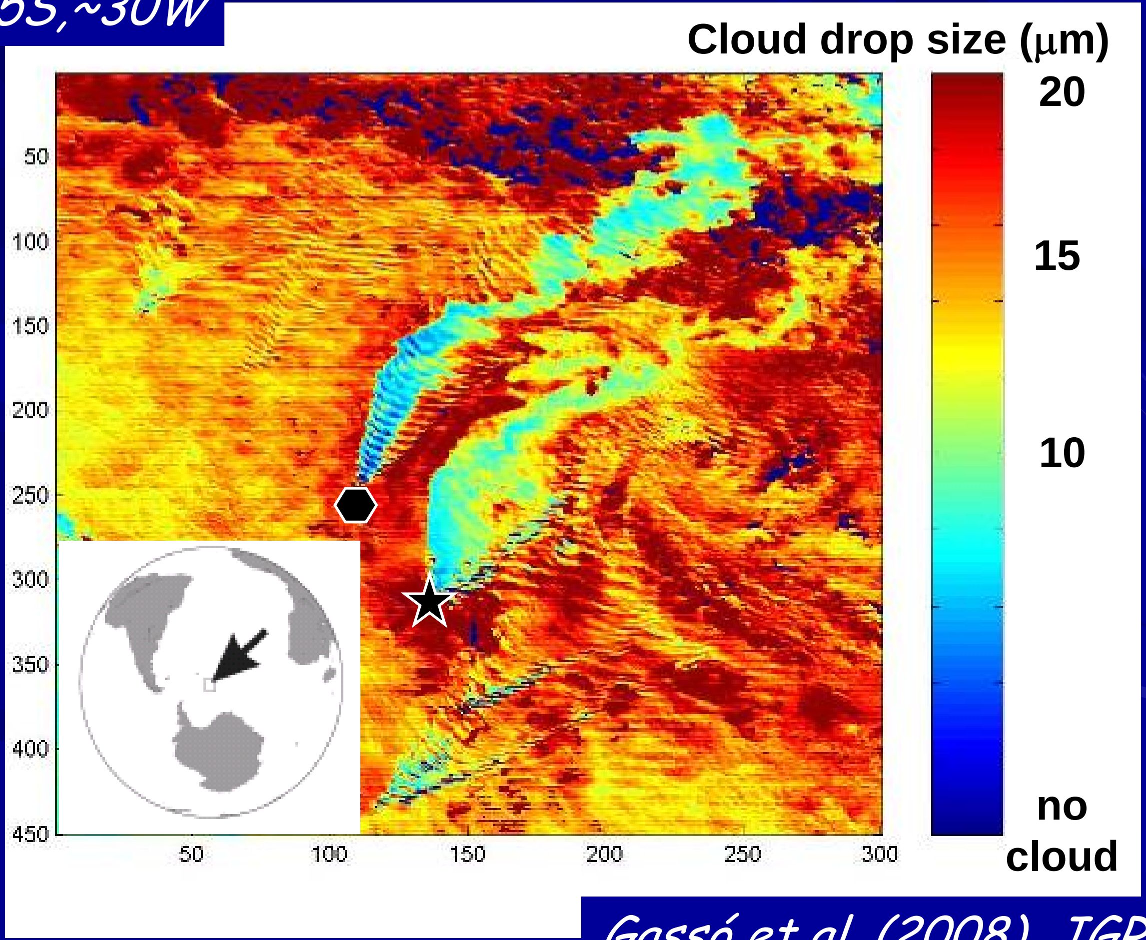
Meskhidze and Nenes, Science, 2006

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.

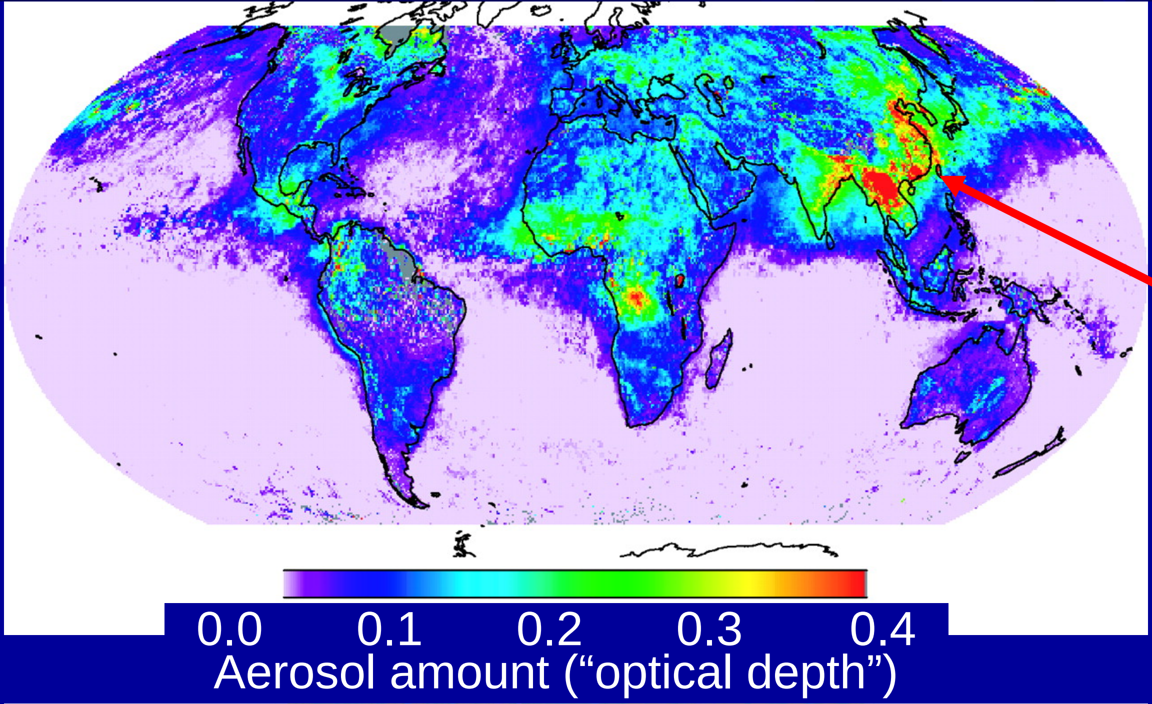
Location: Sandwich Islands , ~55S,~30W



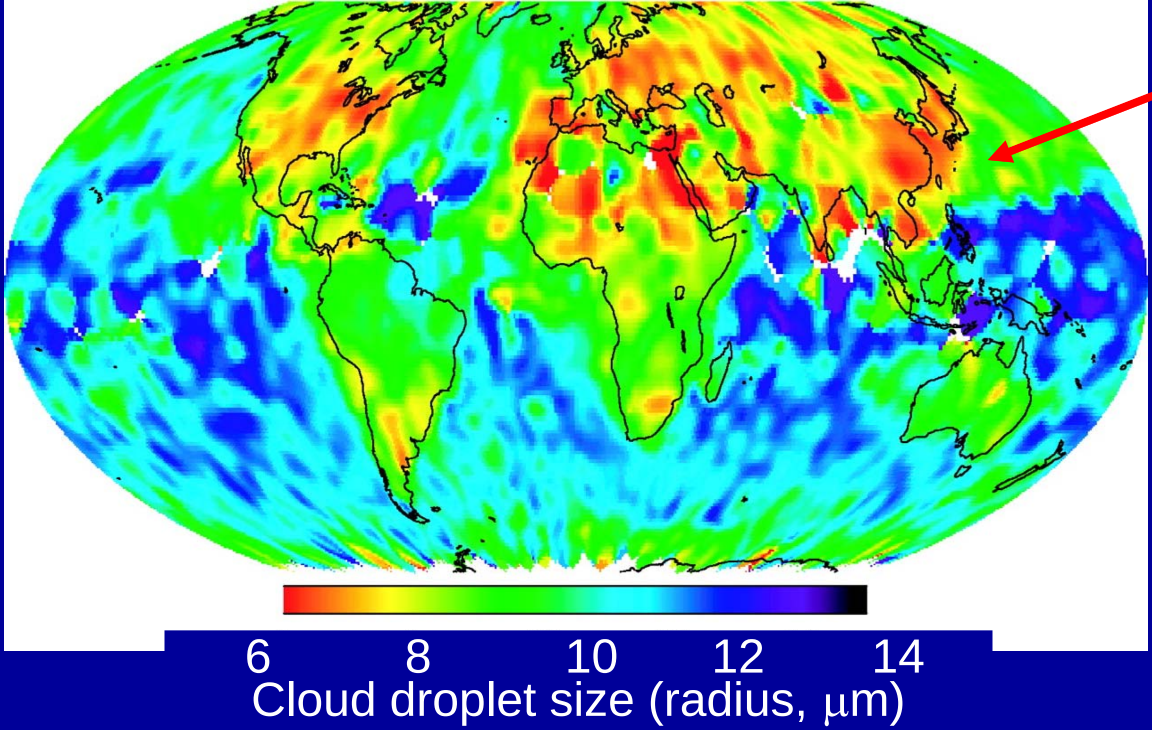


Gassó et al. (2008), JGR

A remote sensing global picture…



A lot of aerosol…



…gives smallest cloud droplets

We see the same on all satellite platforms…

Breón et al. (2002)

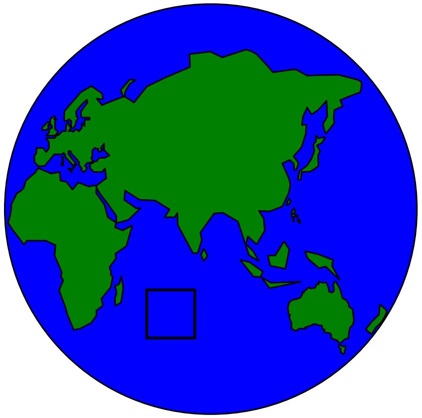
Global Climate Models: Tools of understanding

- Divide the Earth into small parts (“grid cells”).

- Write differential equations that describe conservation of
    - Energy, Water and other Chemical components
    - Incorporate chemical reactions
    - Incorporate phase changes (for example water-ice-vapor)

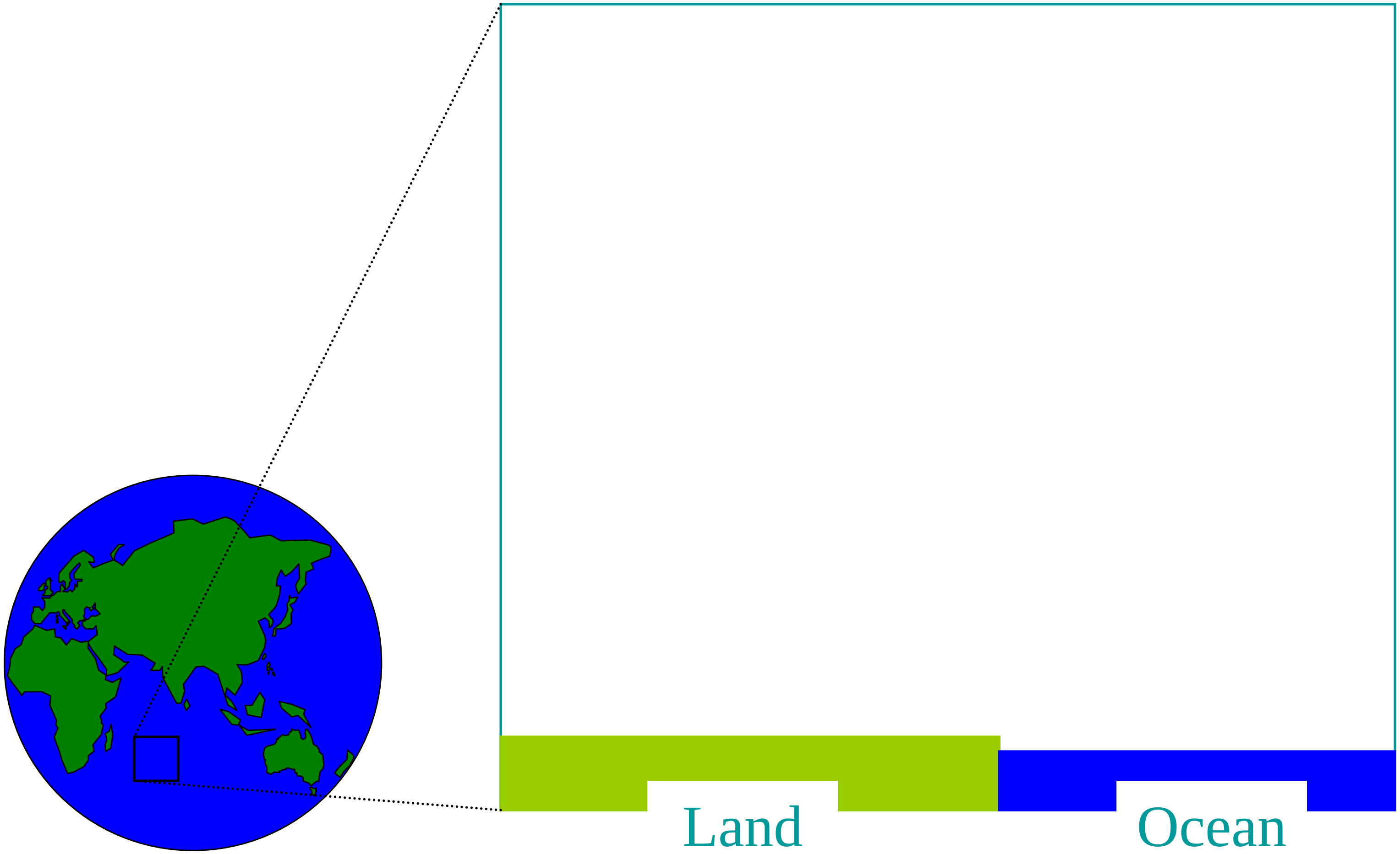
- Prescribe initial conditions (temperature everywhere).

- Integrate the equations (numerically) over time.



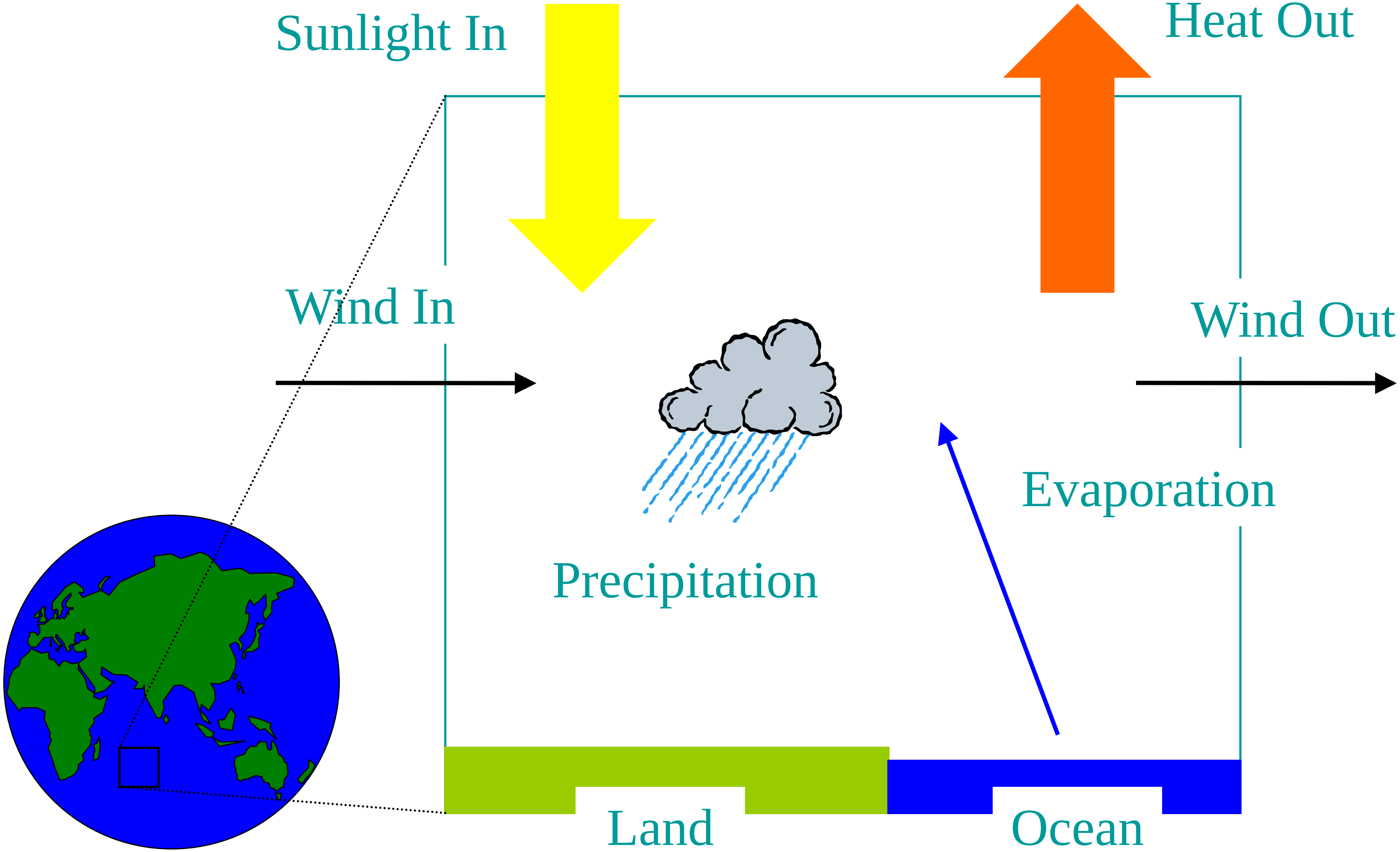
How Computer Climate Models Work

Example: conservation of energy in the atmosphere



How Computer Climate Models Work

Example: conservation of energy in the atmosphere



Sunlight In

Heat Out

Wind In

Wind Out

Evaporation

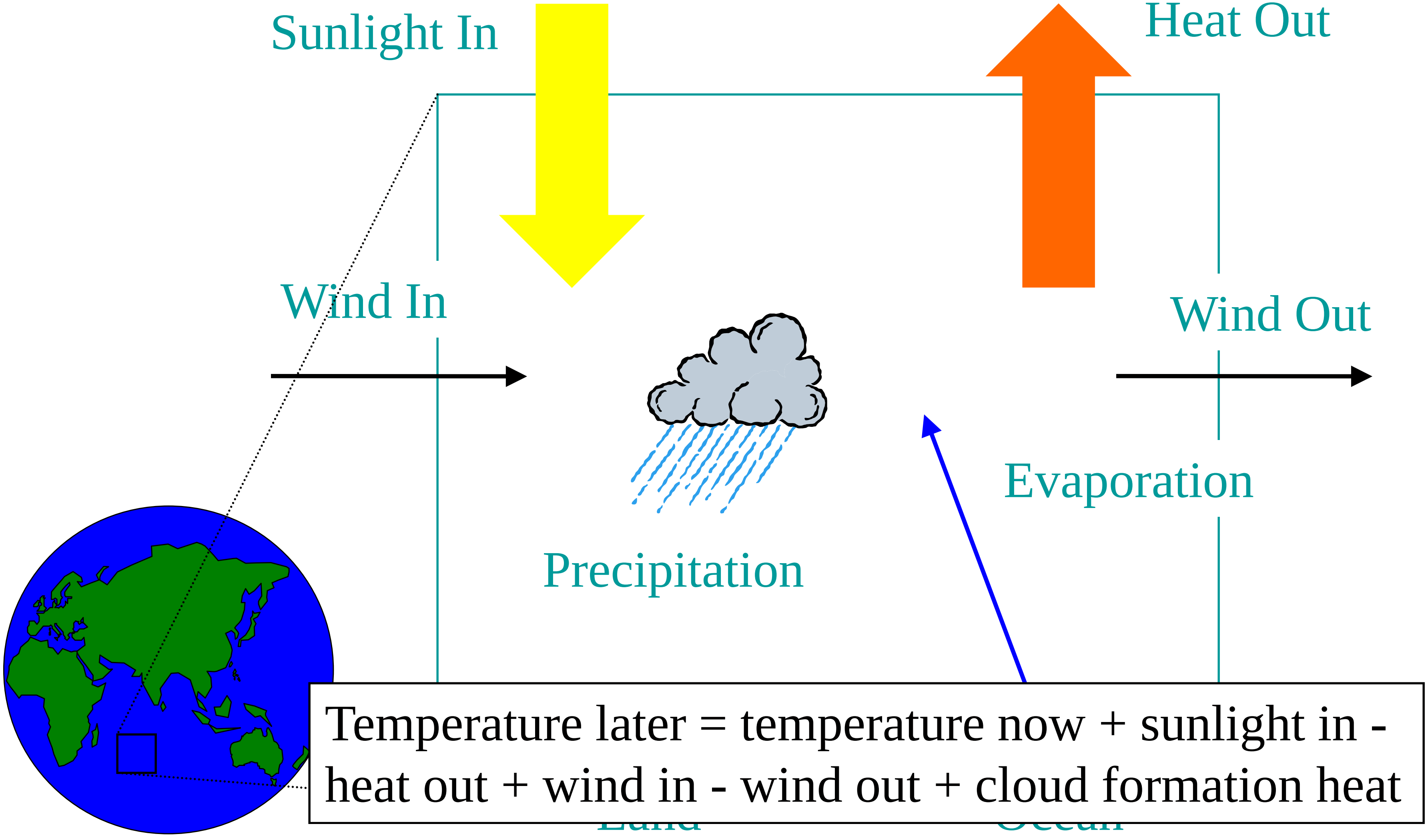
Precipitation

Land

Ocean

How Computer Climate Models Work

Example: conservation of energy in the atmosphere



Temperature later = temperature now + sunlight in - heat out + wind in - wind out + cloud formation heat

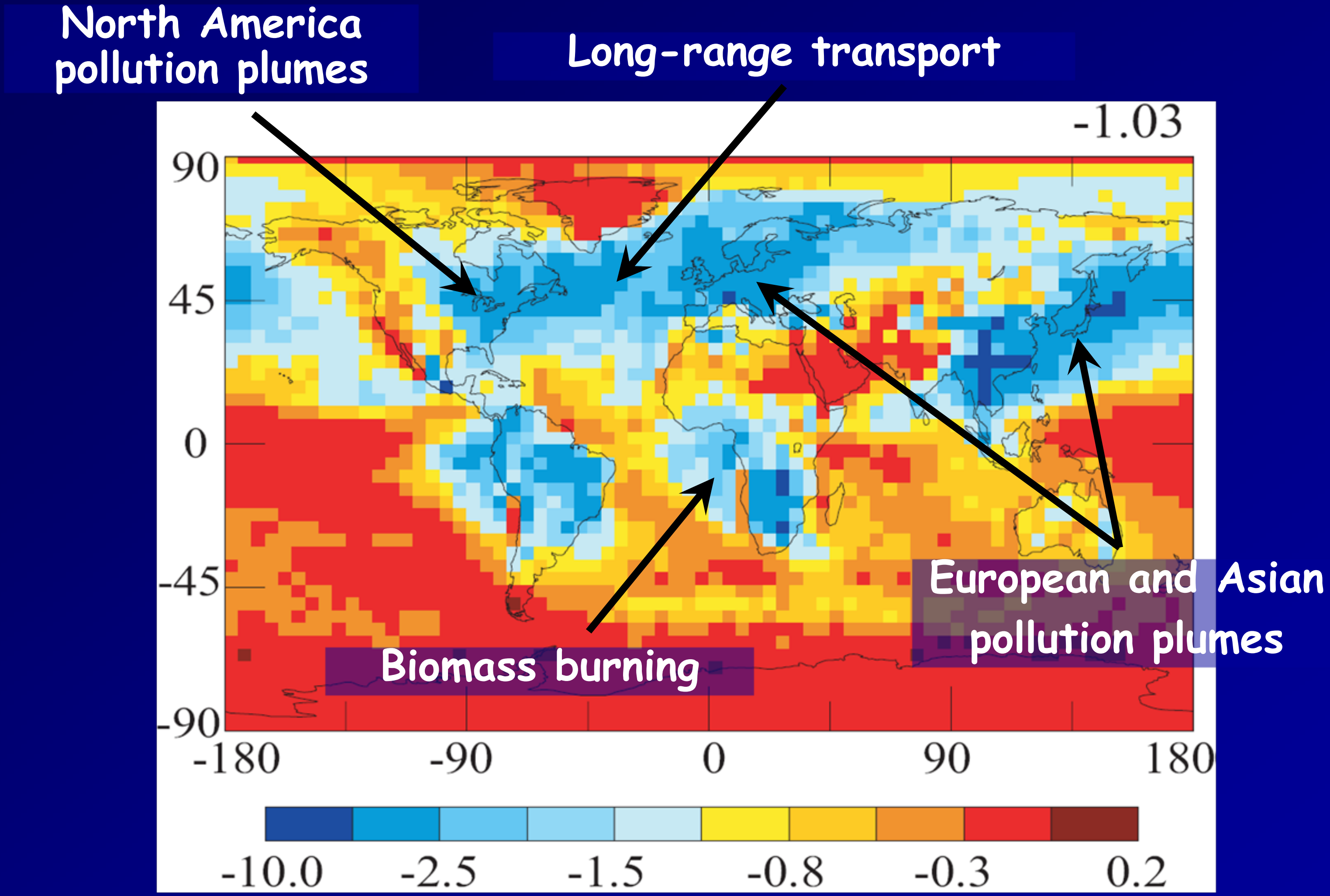
Anthropogenic Indirect Effect:
How do we estimate its global impact?

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})



North America pollution plumes

Long-range transport

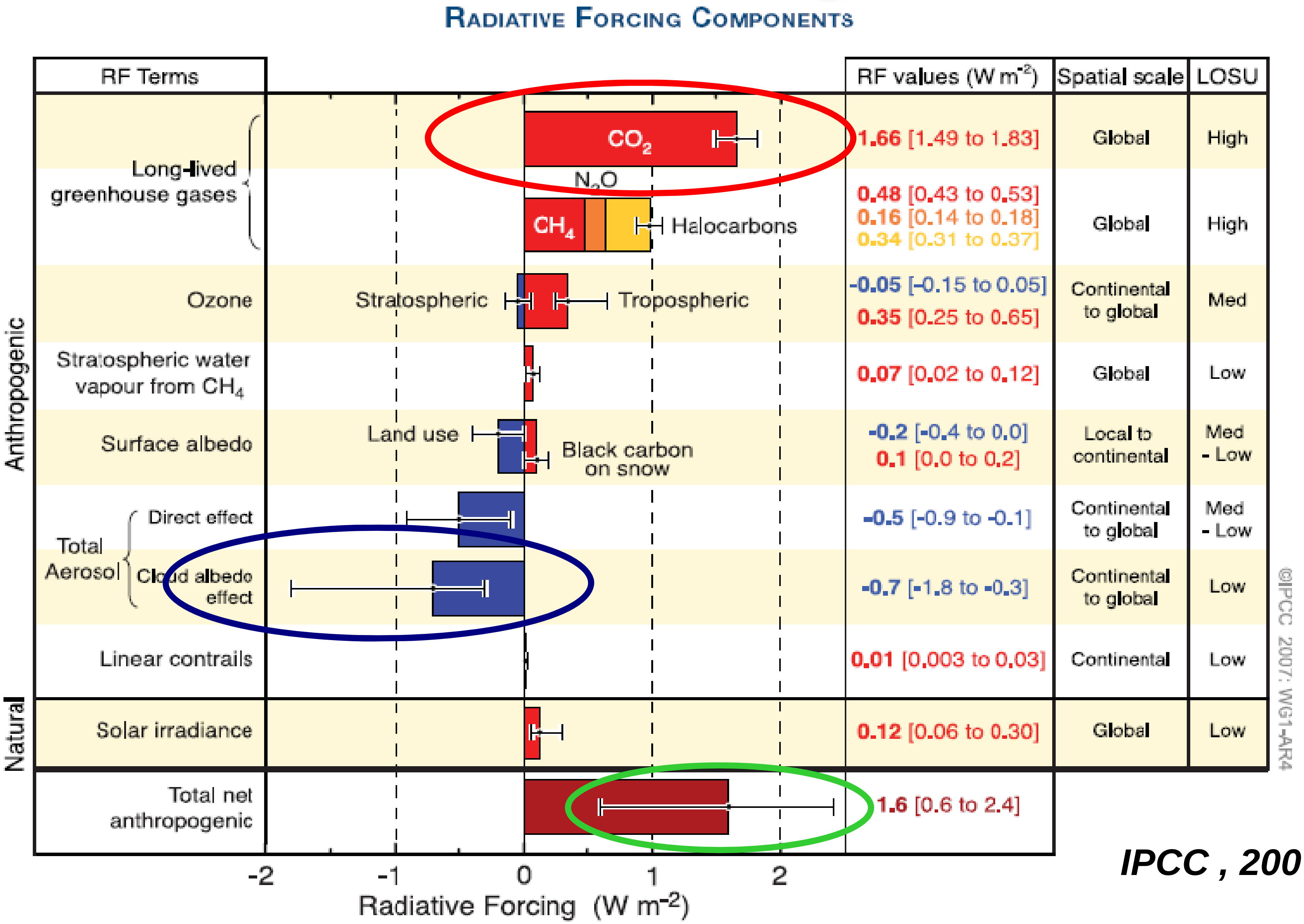
European and Asian pollution plumes

Biomass burning

Sotiropoulou et al., 2007

Spatial pattern of IF follows that of aerosol variations

Components of Human-Induced Climate Change



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
	N_2O CH_4 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
	Total Aerosol: 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

The importance of reducing IE uncertainty

IE diminishes the "punch" of global warming, but we don't know by **how much**.

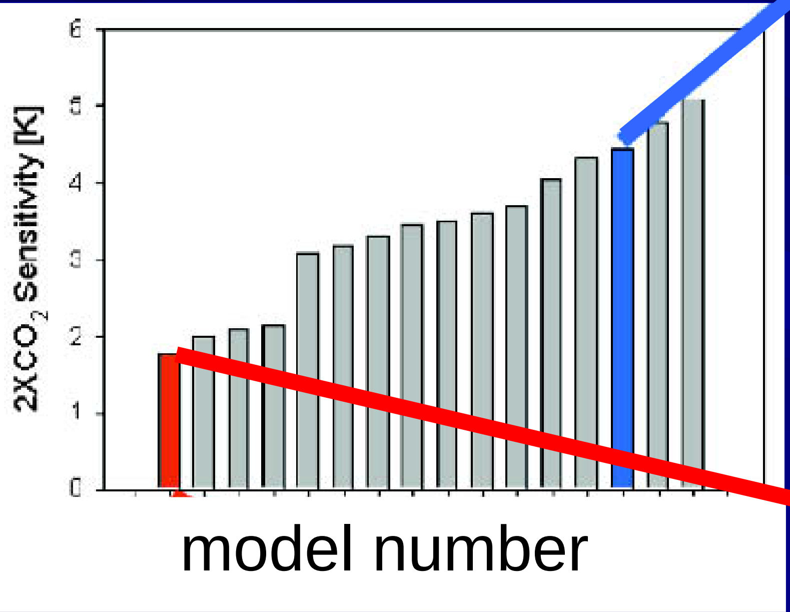
- If IE is low (~ -0.3 Wm^{-2}) then we are "feeling" most of the warming (2.3-0.3=2.0 Wm^{-2}) from greenhouse gases.
- If IE is high (~ -2.0 Wm^{-2}) then we are "feeling" very little of the warming (2.3-2.0=0.3 Wm^{-2}) from greenhouse gases.

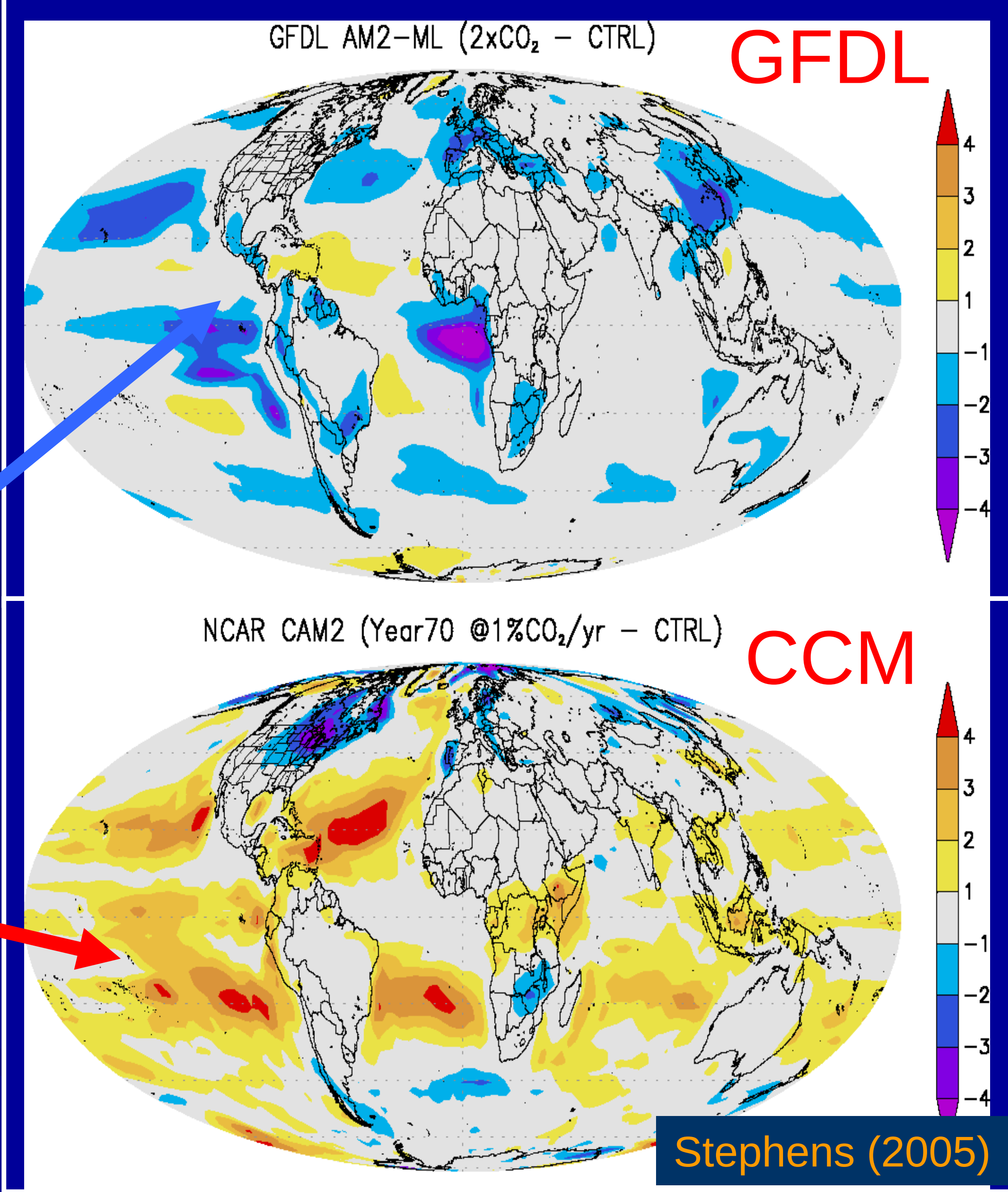
The difference in climate sensitivity to CO_2 is *very large.*

Improving air quality will lead to accelerated global warming.

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

Treatment of clouds largely determine climate sensitivity…

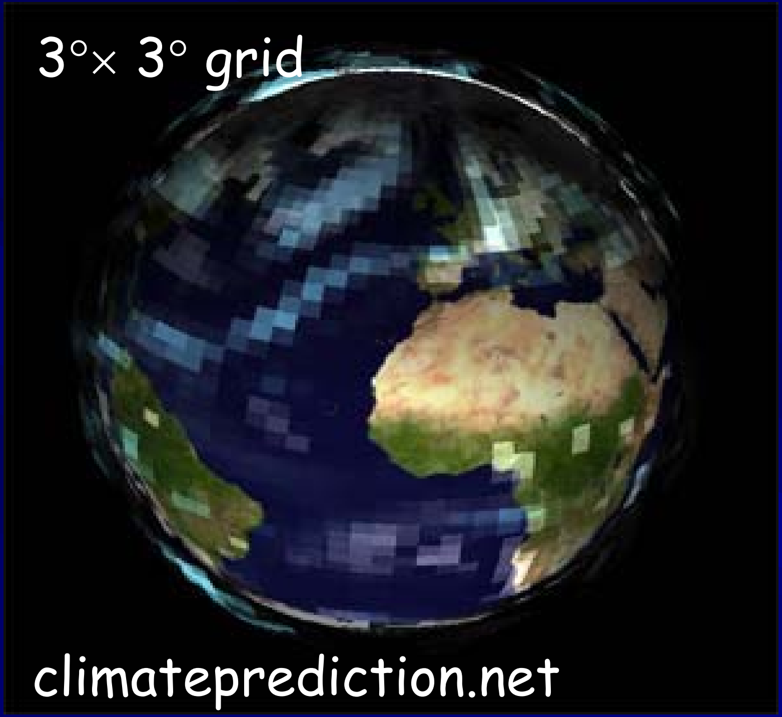




Stephens (2005)

remains the largest source of uncertainty in future climate predictions.

Problems with GCM assessments of aerosol indirect effect

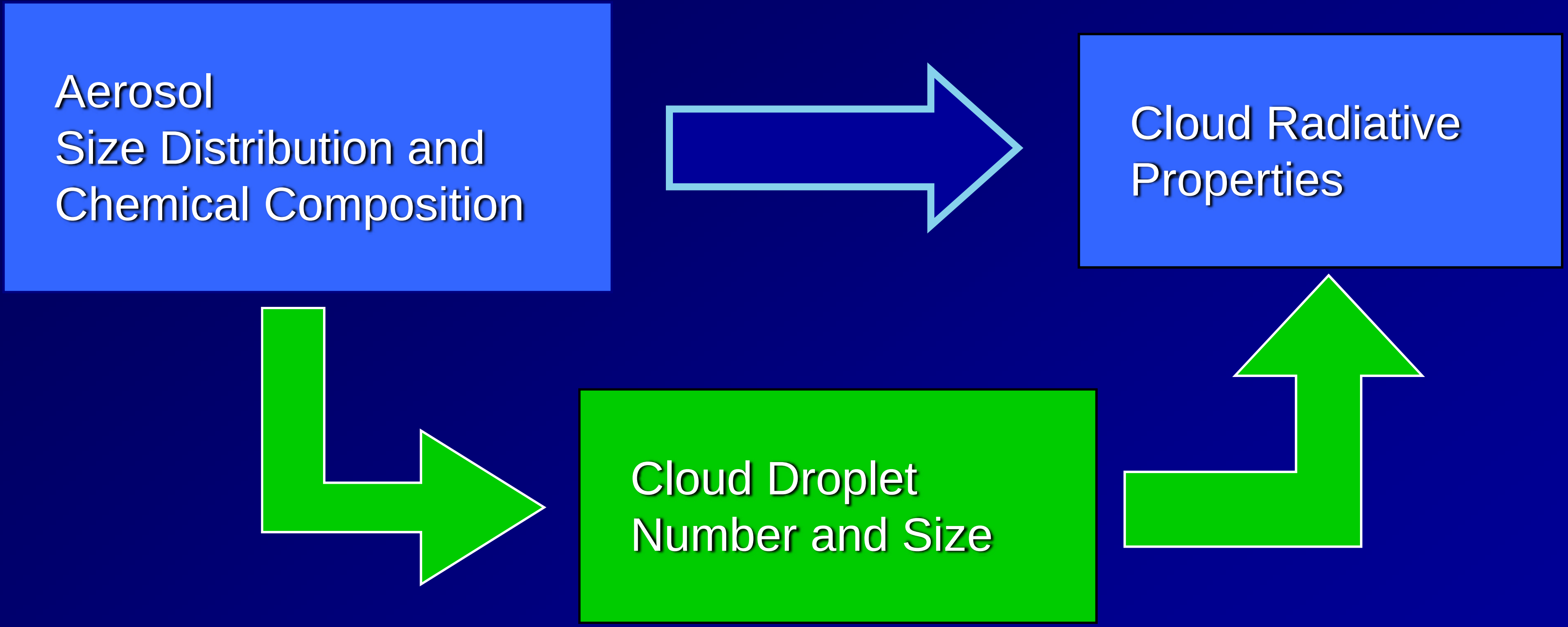


climateprediction.net

- Cloud formation happens at smaller spatial scales than global climate models can resolve.
- Aerosol-cloud interactions are complex.
- Climate models provide limited information about clouds and aerosols.

- Describing cloud formation explicitly in global models is VERY expensive. These calculations need to be simplified ("parameterized").

Quantification of the Indirect Effect in Global Climate Models (GCMs)



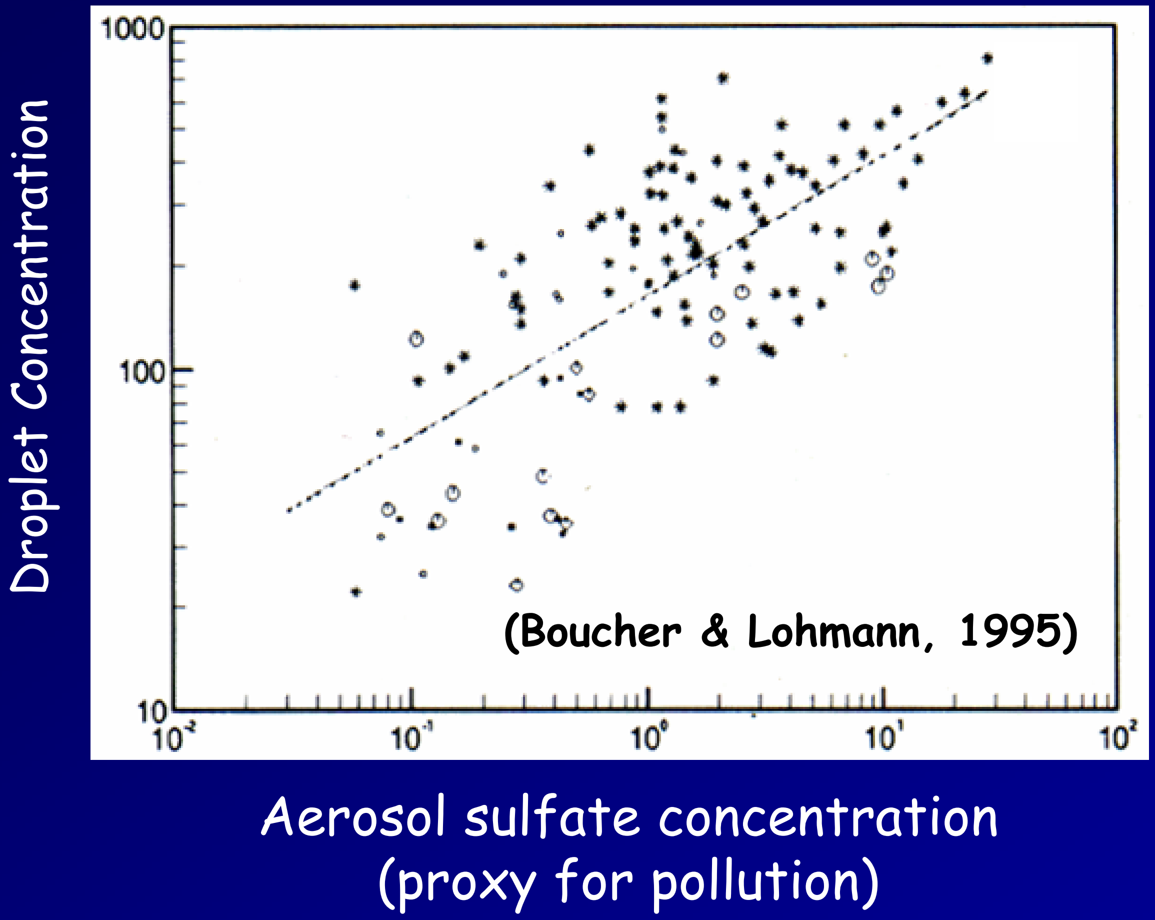
Aerosol
Size Distribution and
Chemical Composition

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



(Boucher & Lohmann, 1995)

Large variability.

Why?

Unaccounted:

- Meteorology
- Cloud microphysics
- Composition
- etc…

Many studies still utilize this type of approach.

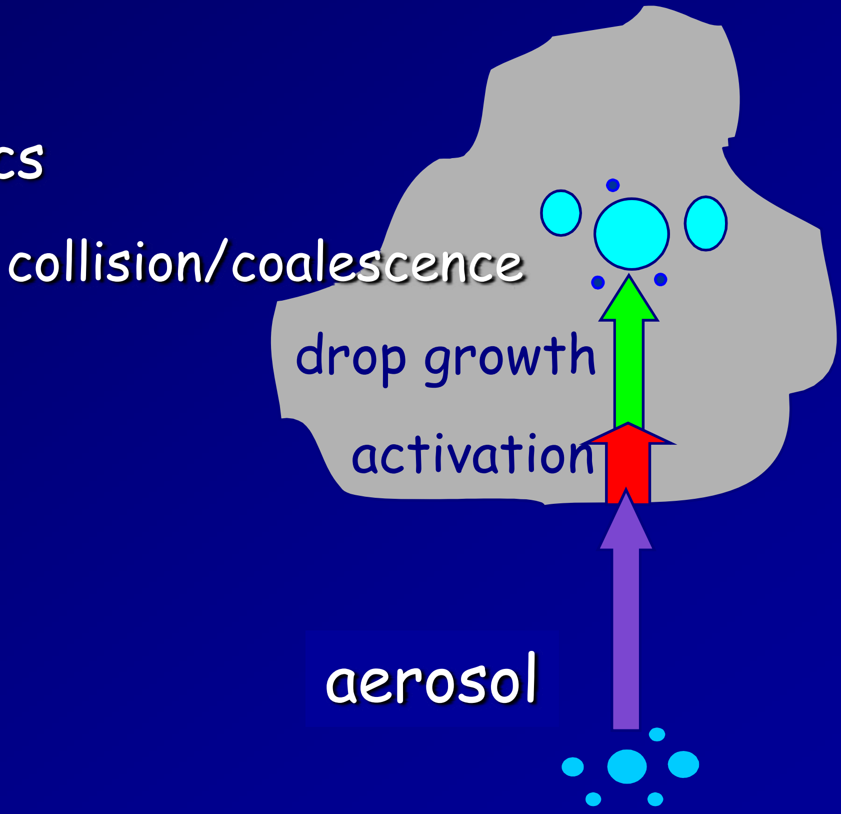
Large predictive uncertainty, without "chances" of improving.

Current Direction: Use simplified but physically based 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

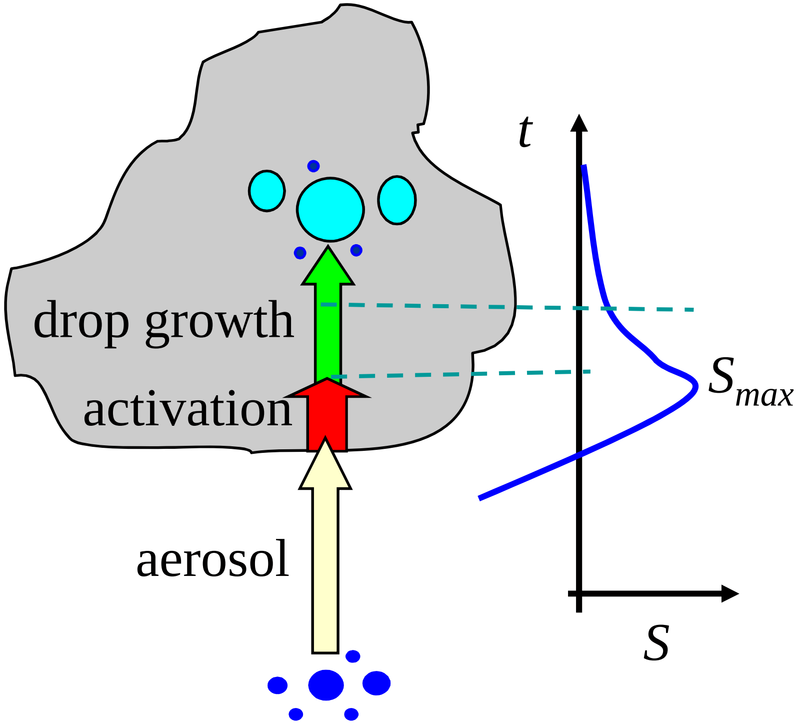


All the links need to be incorporated in global models
The links need to be COMPUTATIONALLY feasible.

Including explicit physics in GCMs is possible…

Approach: **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.

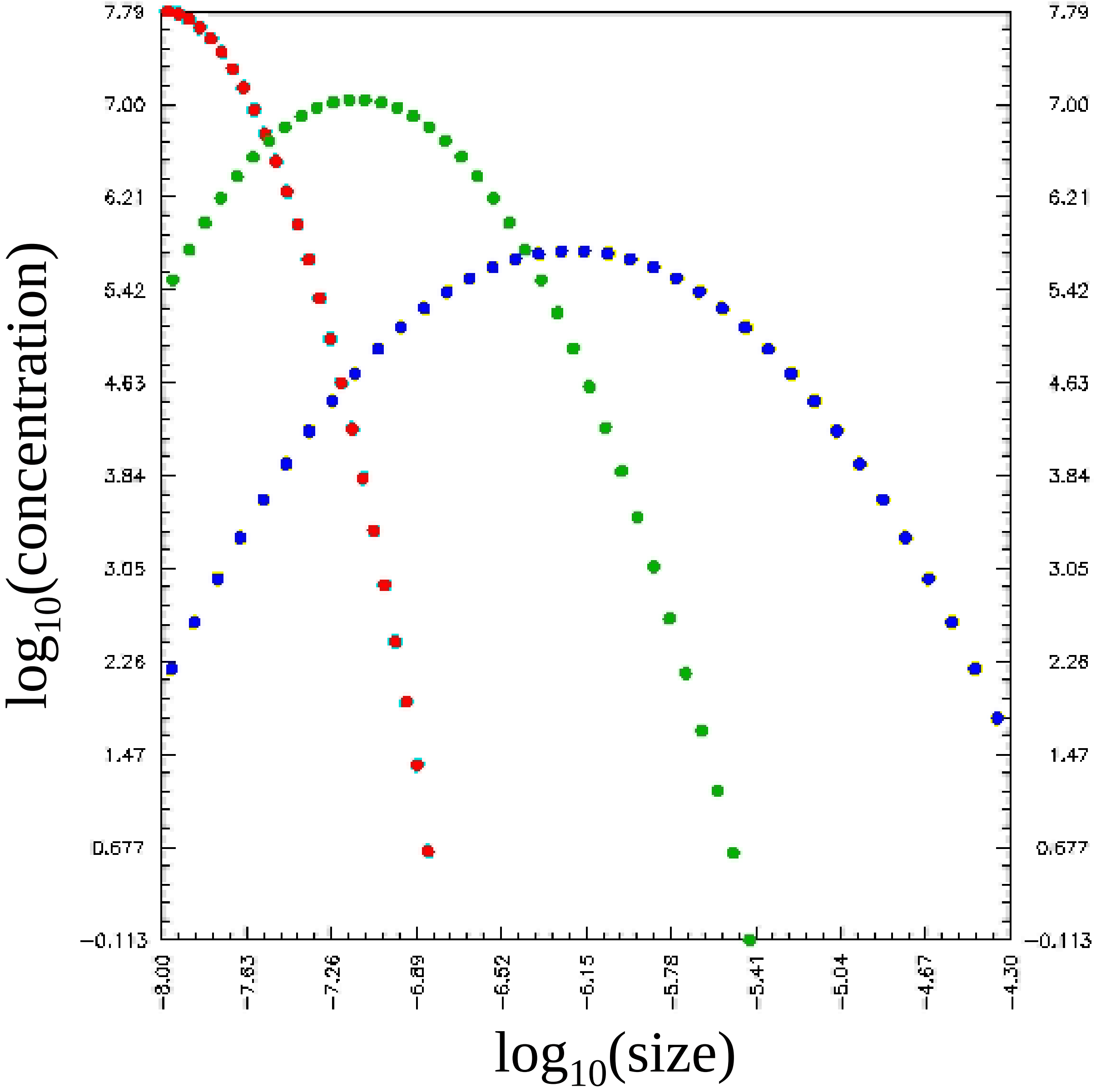


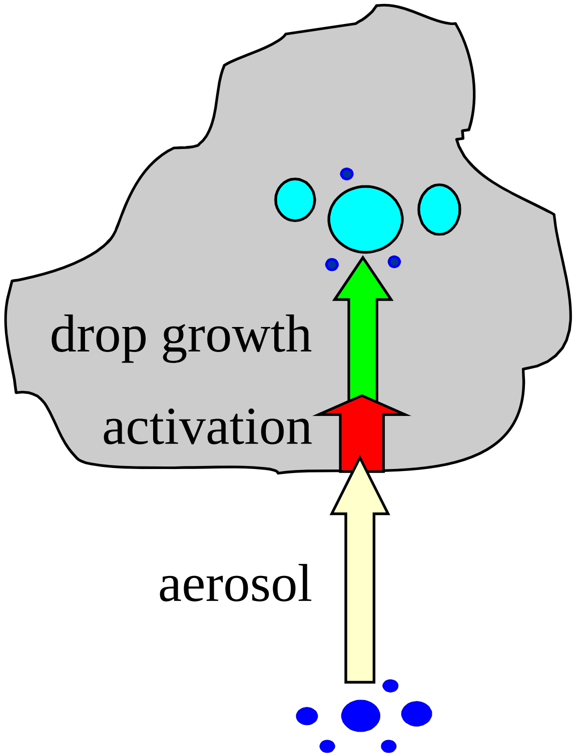
Steps are:

- 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

A "classical" nucleation problem

Cloud droplet formation in updrafts





drop growth

activation

aerosol

Including explicit physics in GCMs is possible…

The "good" news:

This theory is well established

The "bad" news:

It is (very, very) SLOW (you have to solve 100's of stiff ODE's).

Fortunately, there is a solution:

"Physically-based" parameterizations. They don't solve the "full problem" but only what's important for calculating N_d: s_{max} and the CCN(s_{max})

“Mechanistic” Cloud Parameterizations efficiently solve the drop formation problem

Input: P,T, vertical wind, particle size distribution,composition.
Output: Cloud properties (droplet number, size distribution).
How: Solve **one 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:

- 10^3-10^6 times *faster* than numerical parcel models.
- some can treat very complex chemical composition.
- simplified physics, but still you get physically-based links between aerosols and clouds.

Are parameterizations “good enough”?

Let’s apply them for real clouds. In-situ airborne platforms is the ideal laboratory for studying clouds.

CIRPAS Twin Otter

NOAA P3

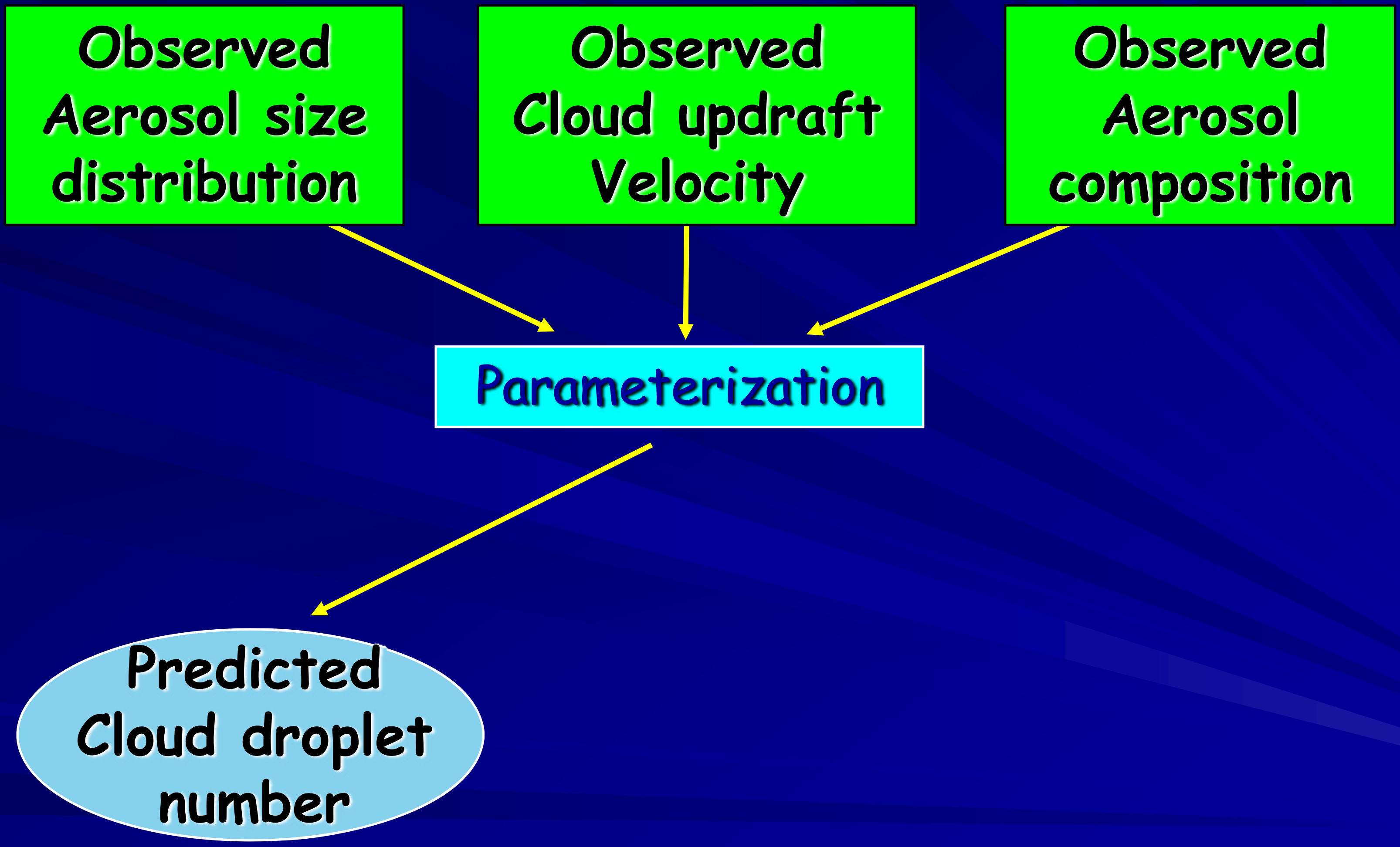
Parameterization Evaluation
CDNC "closure"

Observed
Aerosol size
distribution

Observed
Cloud updraft
Velocity

Observed
Aerosol
composition

Parameterization Evaluation
CDNC "closure"



Observed Aerosol size distribution

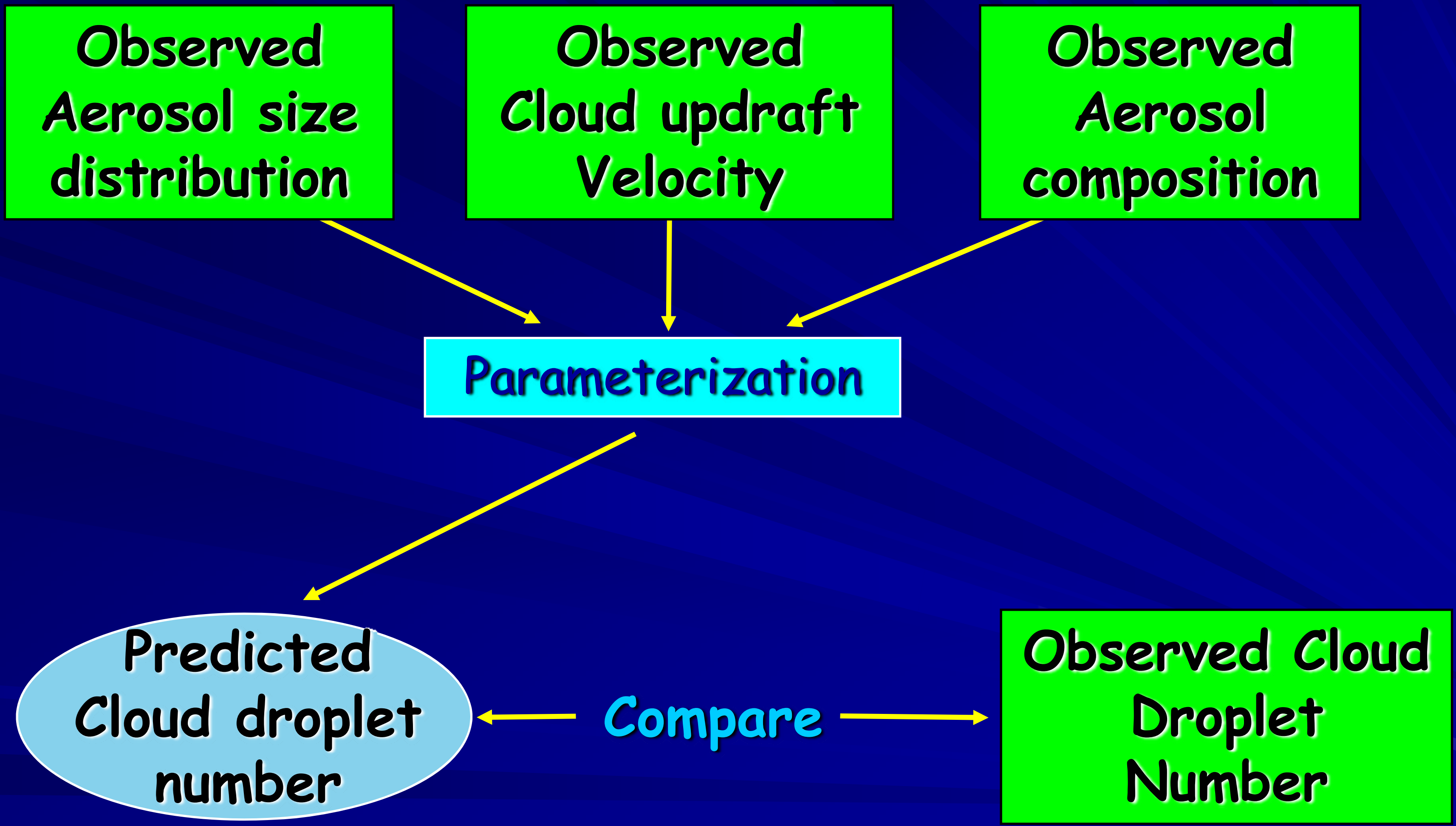
Observed Cloud updraft Velocity

Observed Aerosol composition

Parameterization

Predicted Cloud droplet number

Parameterization Evaluation
CDNC "closure"



Observed Aerosol size distribution

Observed Cloud updraft Velocity

Observed Aerosol composition

Parameterization

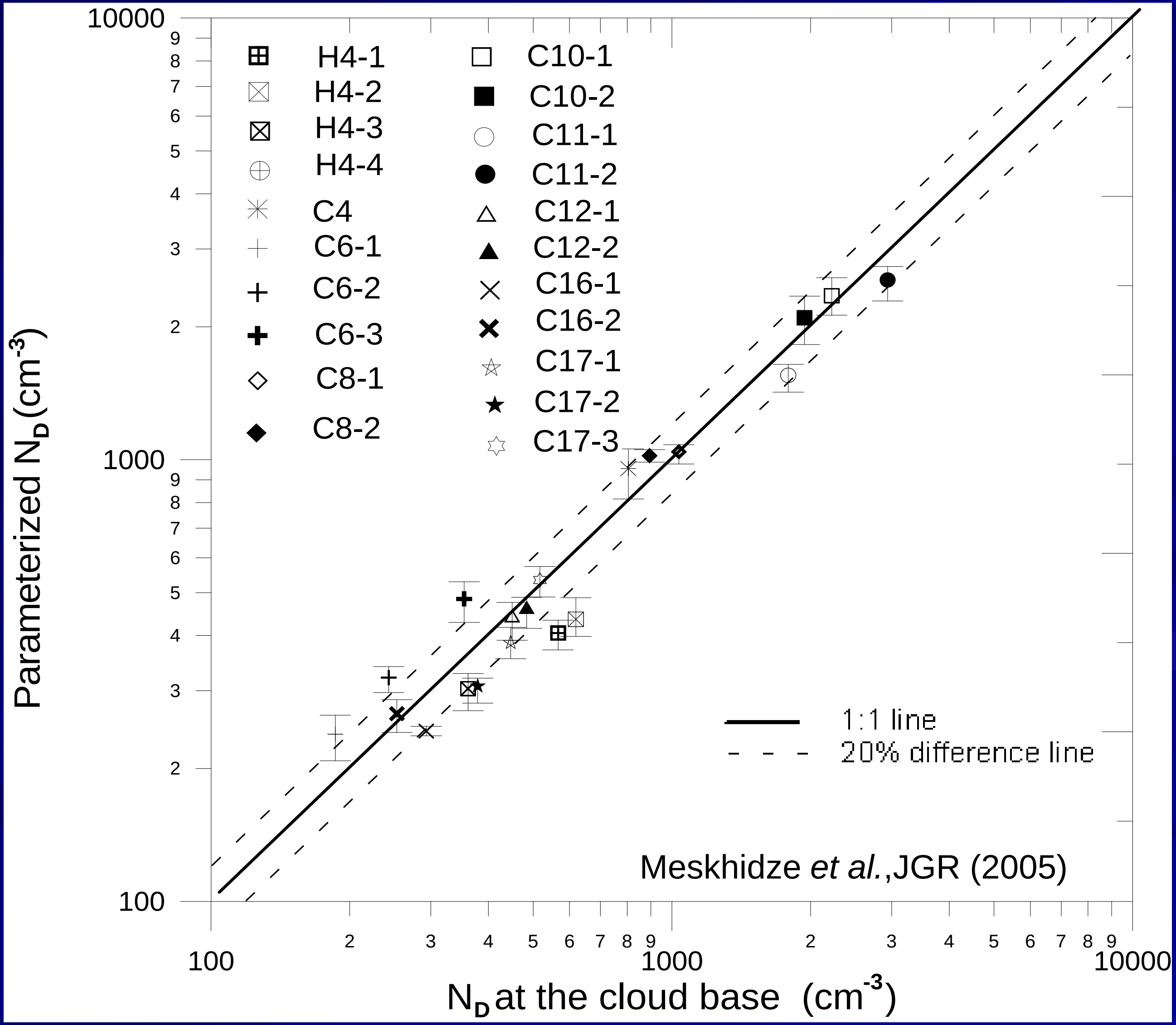
Predicted Cloud droplet number

Compare

Observed Cloud Droplet Number

CRYSTAL-FACE (2002)
Cumulus clouds

CIRPAS Twin Otter



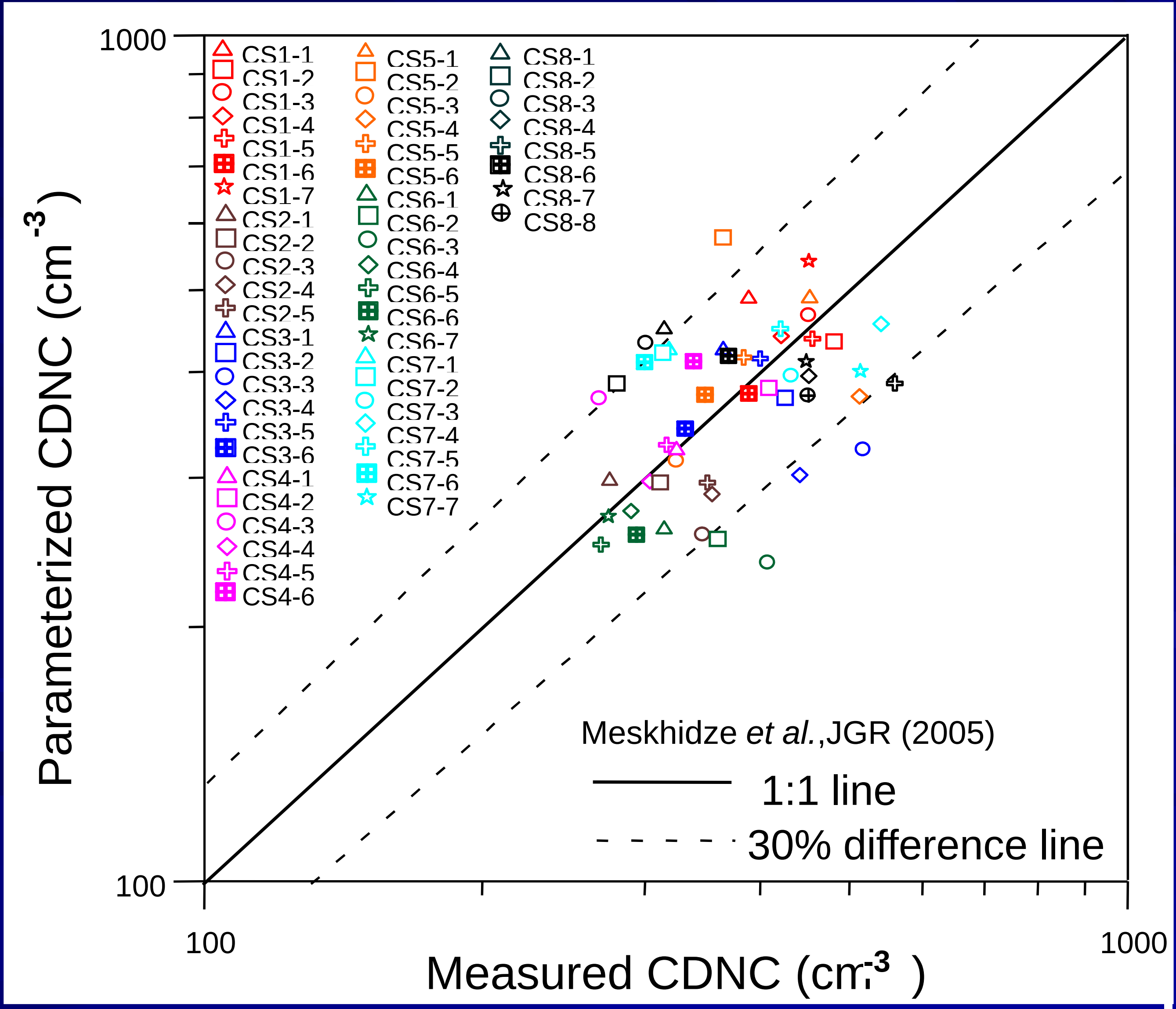
Meskhidze *et al.*,JGR (2005)

Parameterization agrees with observed CDNC

Agreement to within a few % (on average) !

CSTRIPE (2003)
Coastal Stratus Clouds

CIRPAS Twin Otter

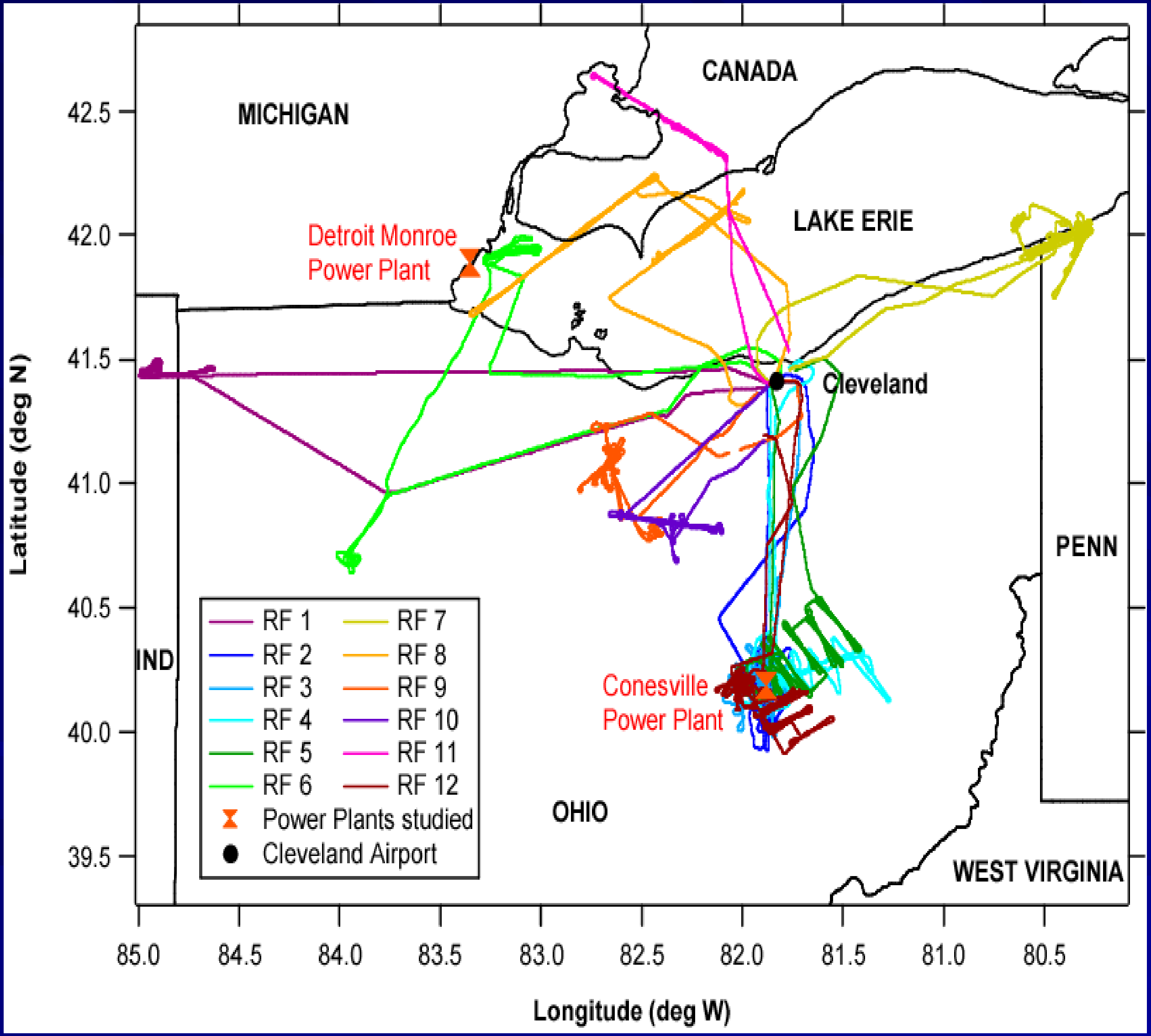


Parameterization agrees with observed CDNC

Agreement to within a few % (on average) !

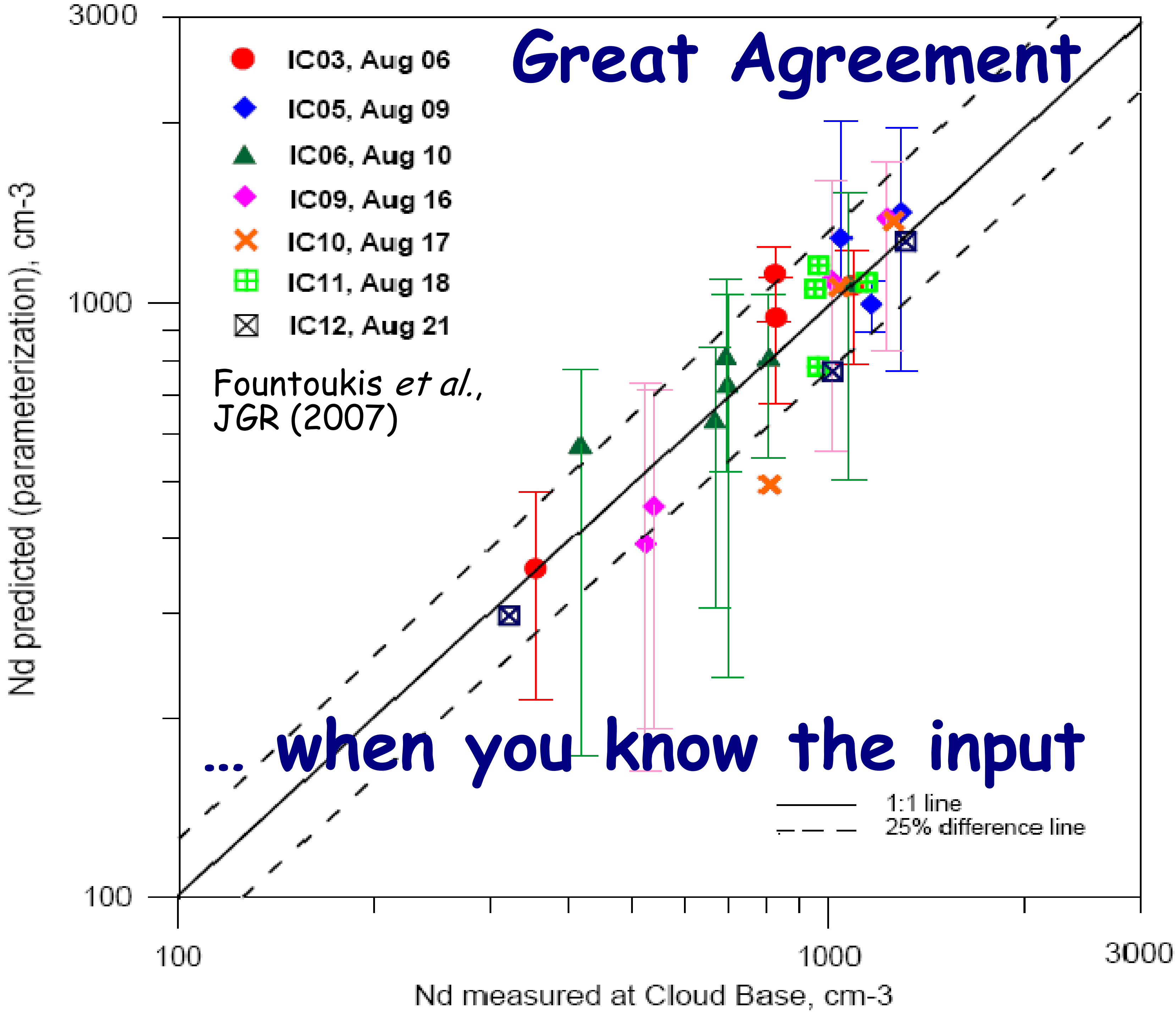
This is a very important class of clouds.

CDNC closure during ICARTT (Aug.2004)



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

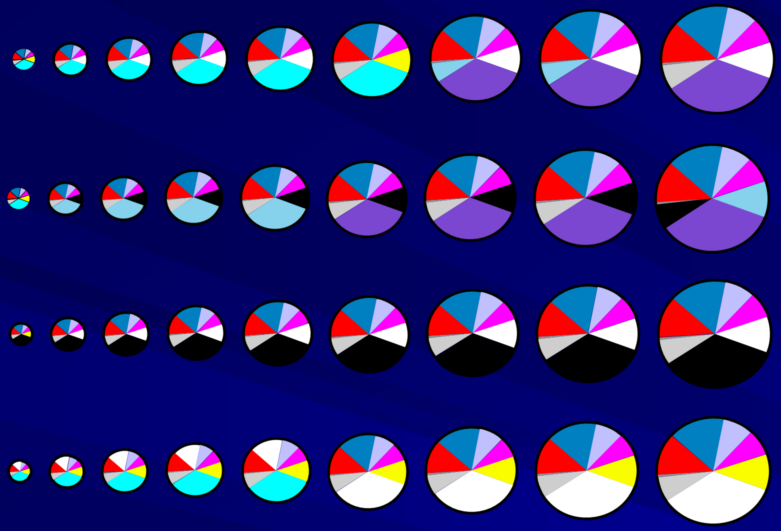
Great Agreement



Fountoukis *et al.*,
JGR (2007)

… when you know the input

Problem: Global Aerosol is Vastly Complex



An integrated "soup" of

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

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$^{-1/2}$).

Predicting CCN concentrations requires knowledge of size distribution and chemical composition.

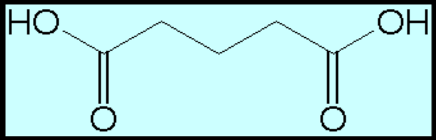
This aerosol is highly complex and variable

Some inorganics:

- $(NH_4)_2SO_4$, NH_4HSO_4
- Sulfuric acid
- Seasalt (NaCl)
- Crustals (Ca,Mg,K salts)
- Nitrates (NH_4NO_3,$NaNO_3$)
- Chlorides (KCl, NH_4Cl)

Some organics:

- Glutaric Acid (biomass)
- Pinonic Acid (terpenes)
- Hydroxy Methyl Benzoic Acid (gasoline)
- Palmitic Acid (plant waxes)
- *and hundreds more…*



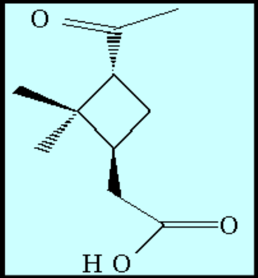


Photo: NASA

The … headache of organic species:

- They can act as surfactants and facilitate cloud formation.
- They can increase the affinity of particles for water (add solute) and facilitate cloud formation.
- "Waxy" films can form and slow down droplet growth

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

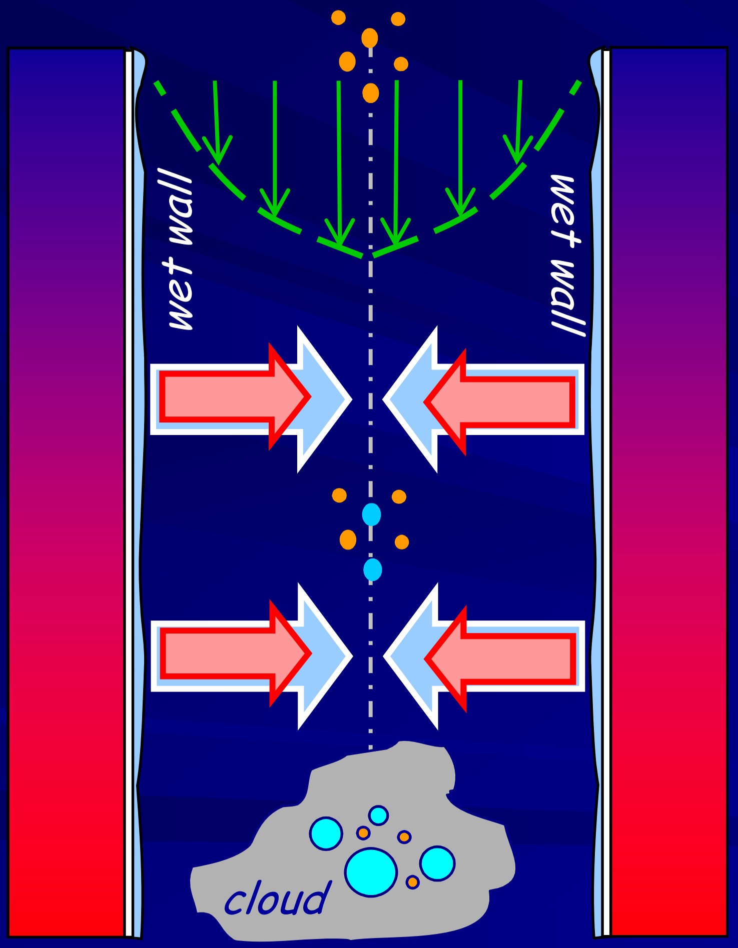
Creative use of CCN activity measurements to "constrain" the importance of organics and mixing state on droplet formation for ambient aerosol.

Desired information:

-Average molar properties (molar volume, solubility)
-Droplet growth kinetics
-Chemical heterogeneity (mixing state) of aerosol
-Surface tension depression

Measuring CCN activity of ambient particles: Cloud Chamber

Inlet: Aerosol



Outlet: [Droplets] = [CCN]

Roberts and Nenes (2005), Patent pending

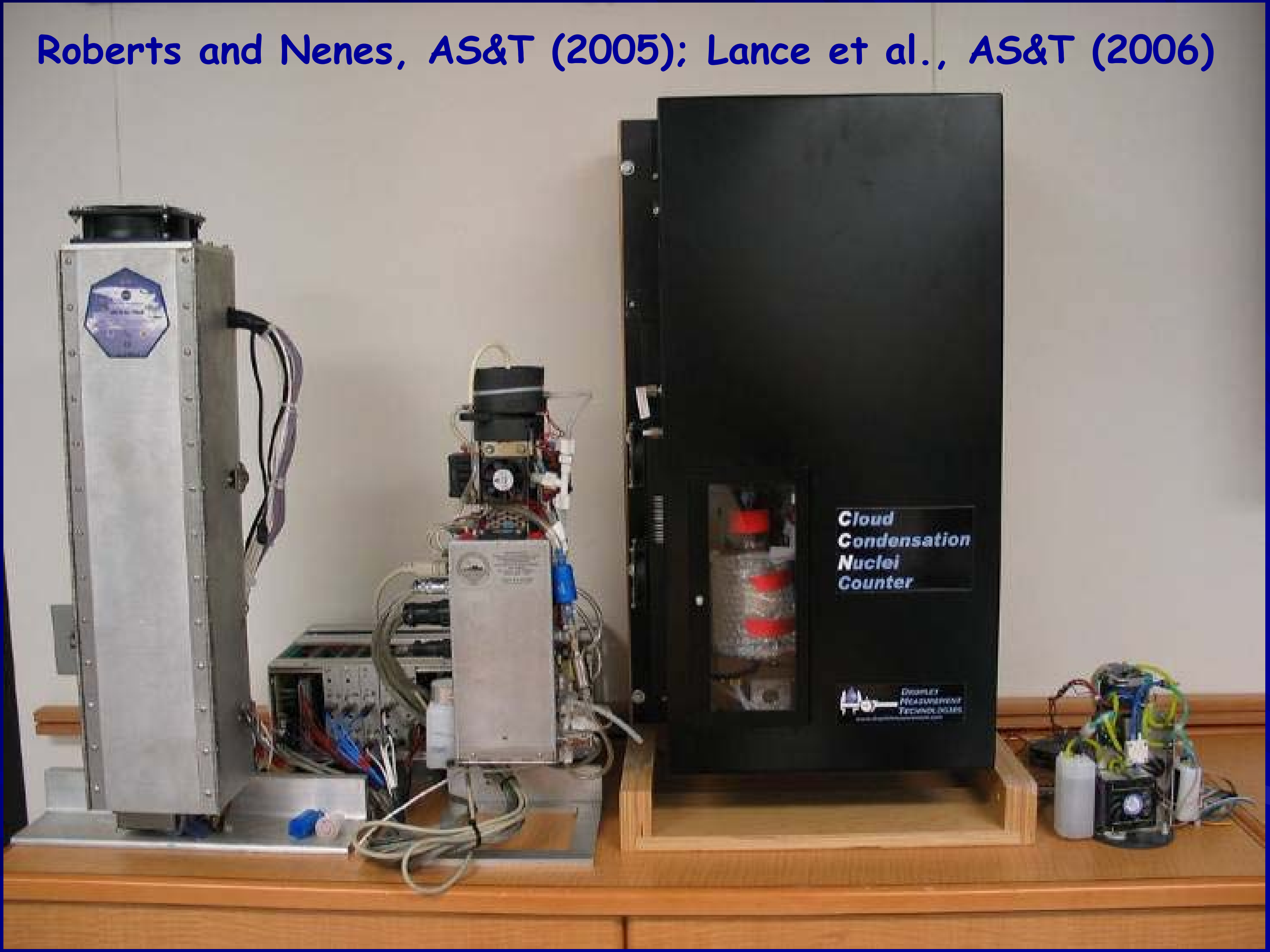
Metallic cylinder with walls wet. Apply T gradient, and flow air.

- 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.

Development phases of cloud chamber

scale = 1 m



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

1st version
April 2002

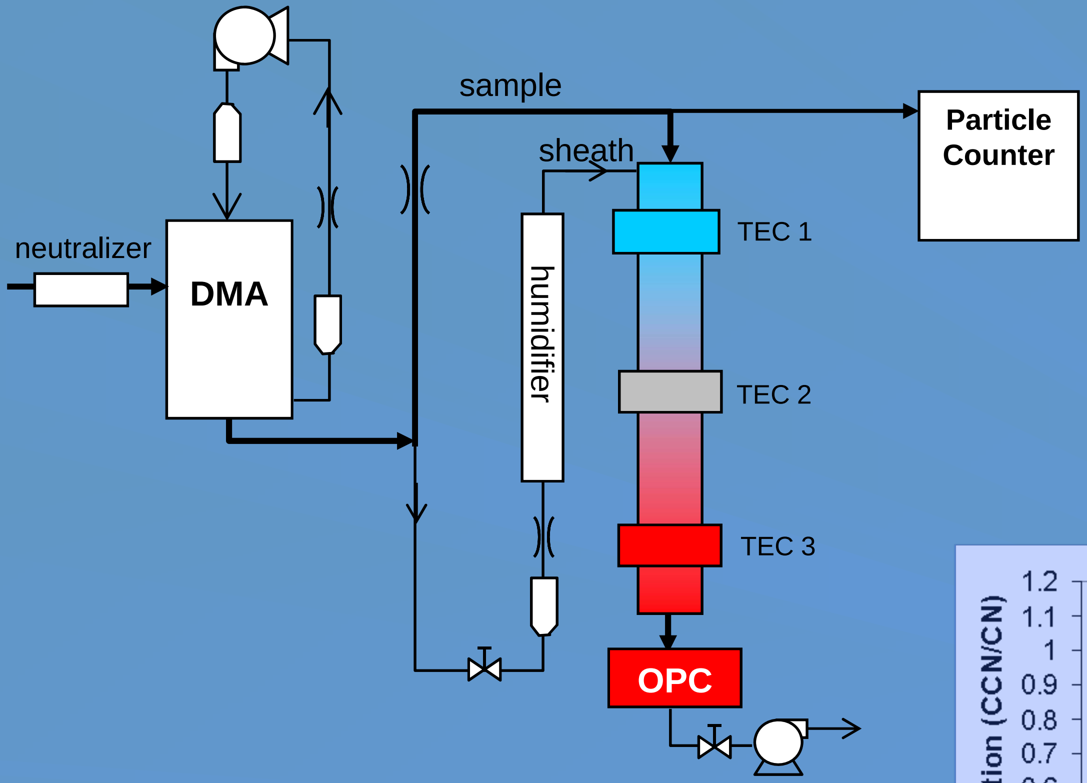
2nd version
January 2003

Commercial ver.
July 2004

mini version
August 2006

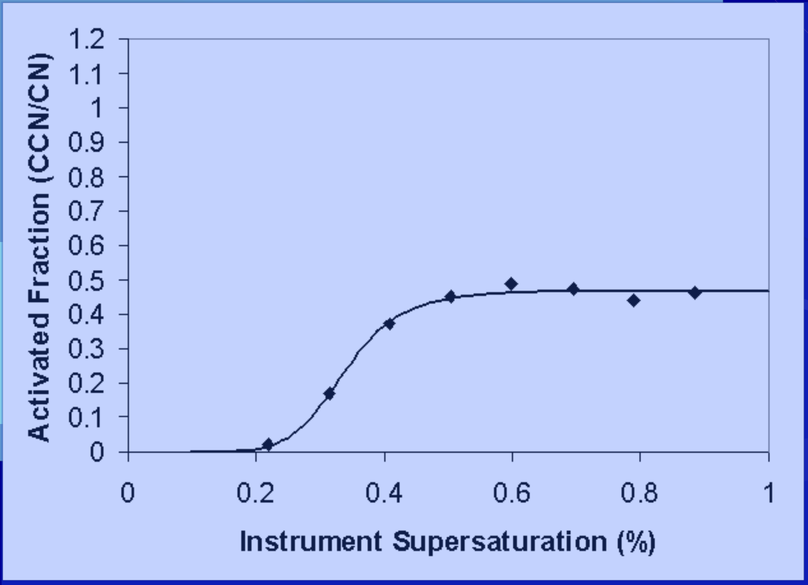
Key source of information:
Size-resolved CCN activity measurements

Measure CCN activity of aerosol with known diameter

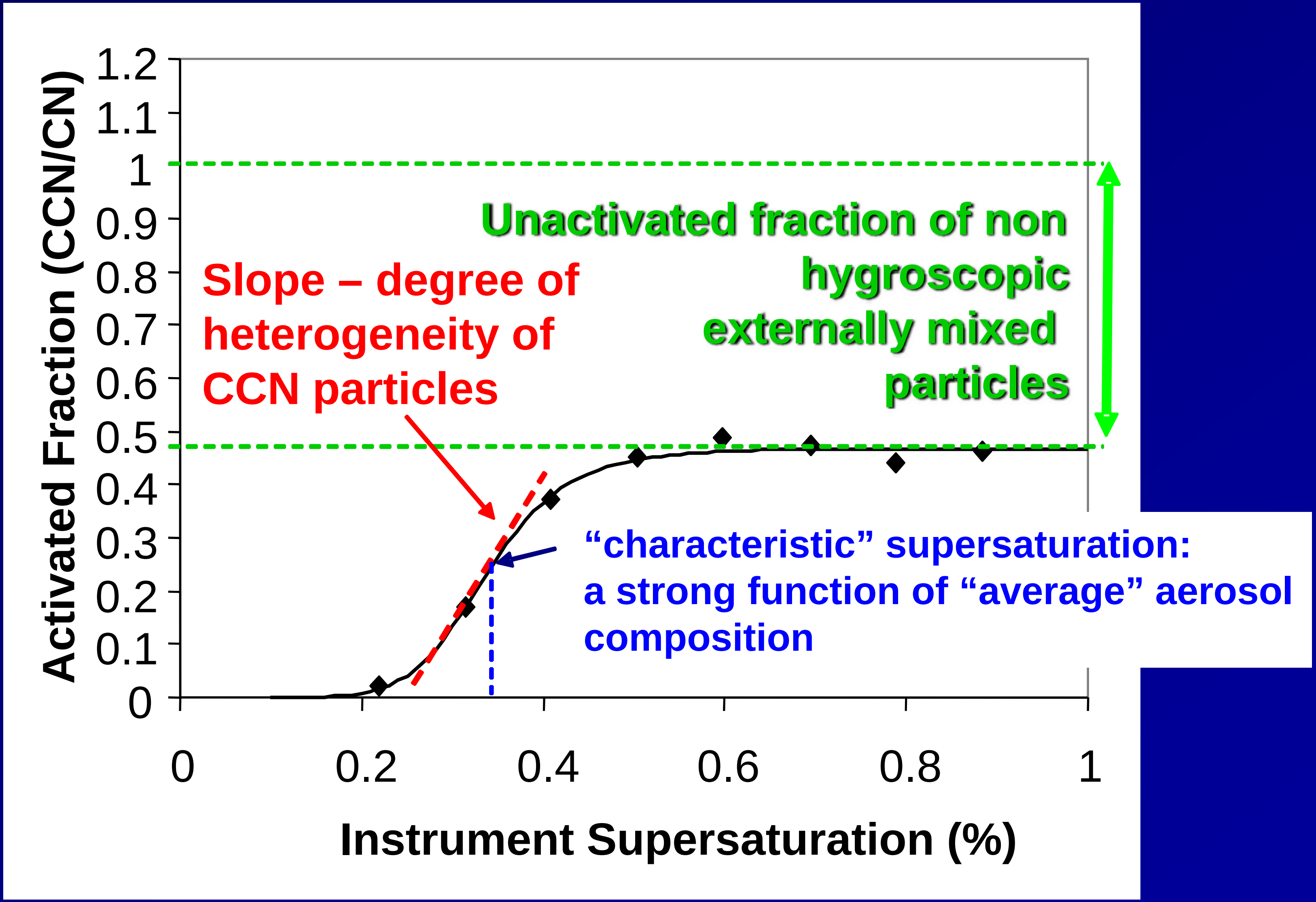


$$\text{Activated Fraction} = \frac{\text{CCN}}{\text{CN}}$$

Results: "activation curves"



What the Activation Curves tell us



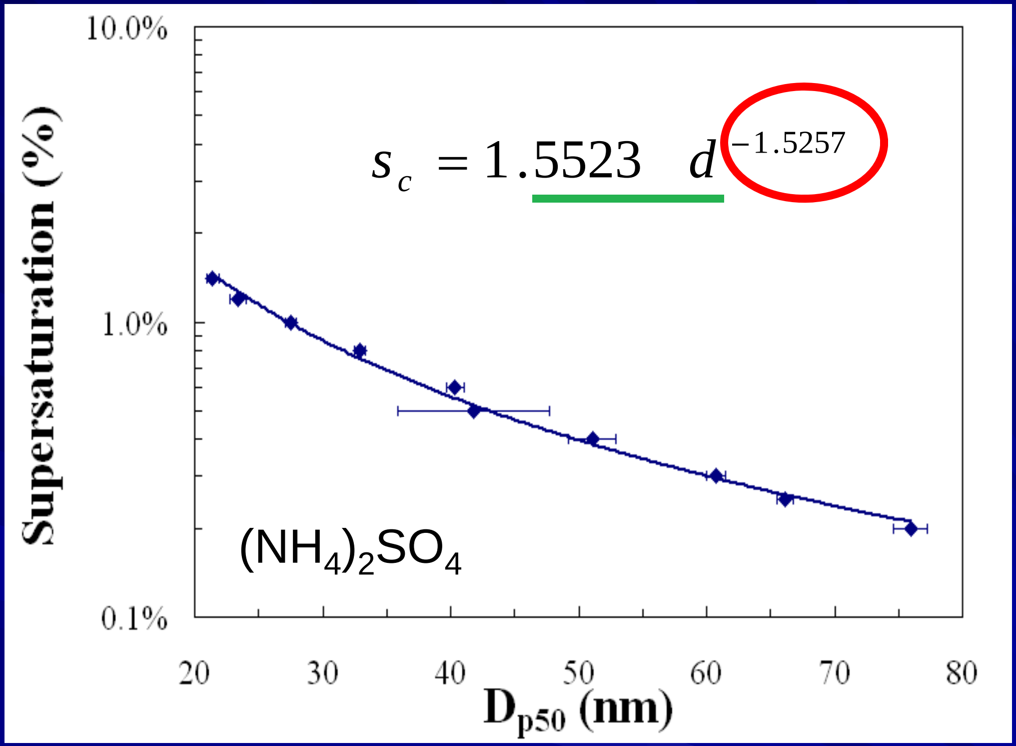
Unactivated fraction of non hygroscopic externally mixed particles

Slope – degree of heterogeneity of CCN particles

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

Inferring Average Molecular Weight from CCN measurements

- 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:



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

Padró *et al.*, ACP; Asa-Awuku *et al.*, ACP; Moore et al., GRL

… and then solve for average molecular weight of organics.

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

Average Molecular weight of organics

Data from CCN measurements

From ion chromatography

A simple and comprehensive way to quantify the contribution of organics to cloud formation.

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.
- Organic Aerosol from biogenic VOC emissions
  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 samples collected in California (2003,2005,2007), New Hampshire (2004), Cleveland (2004), Houston (2006), Greece (2007), Finland (2007), Arctic (2008).

What varies mostly is not the "thermodynamic" properties of the complex organic "soup" but the fraction of soluble material (we can simulate this)…

Complexity sometimes simplifies things for us.

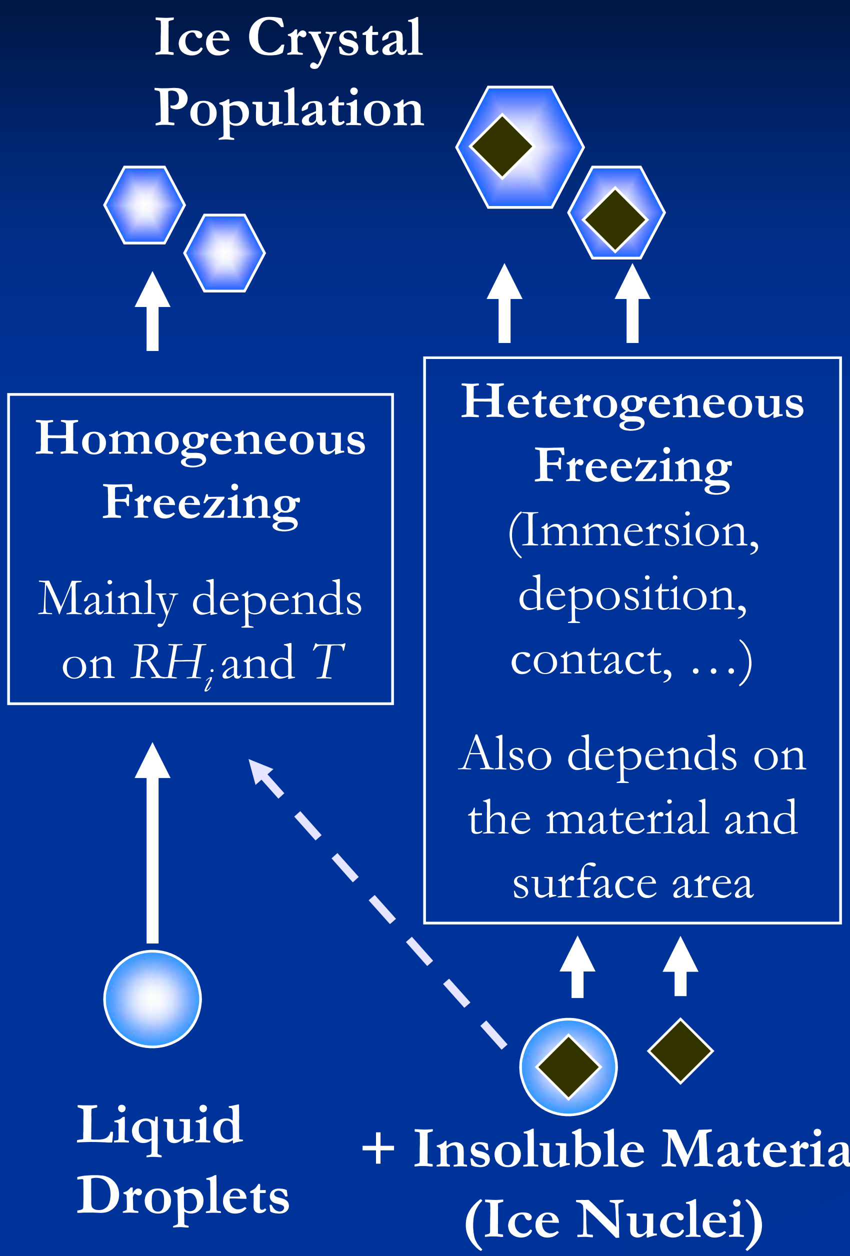
Current efforts: Ice (cirrus) clouds


http://www.alanbauer.com

- Cirrus clouds, composed of ice crystals, are important for:
  - Radiative transfer: they tend to warm climate
  - Stratospheric moisture and circulation
  - Regulation of the ocean temperature
  - Heterogeneous chemistry

Cirrus may be affected by aircraft emissions, transport of dust and pollution. Aerosol effects on cirrus (and climate) are highly unknown (worse than for liquid clouds)!!

Cirrus (Ice) Cloud Formation



Ice Crystal Population

Homogeneous Freezing

Mainly depends on RH_i and T

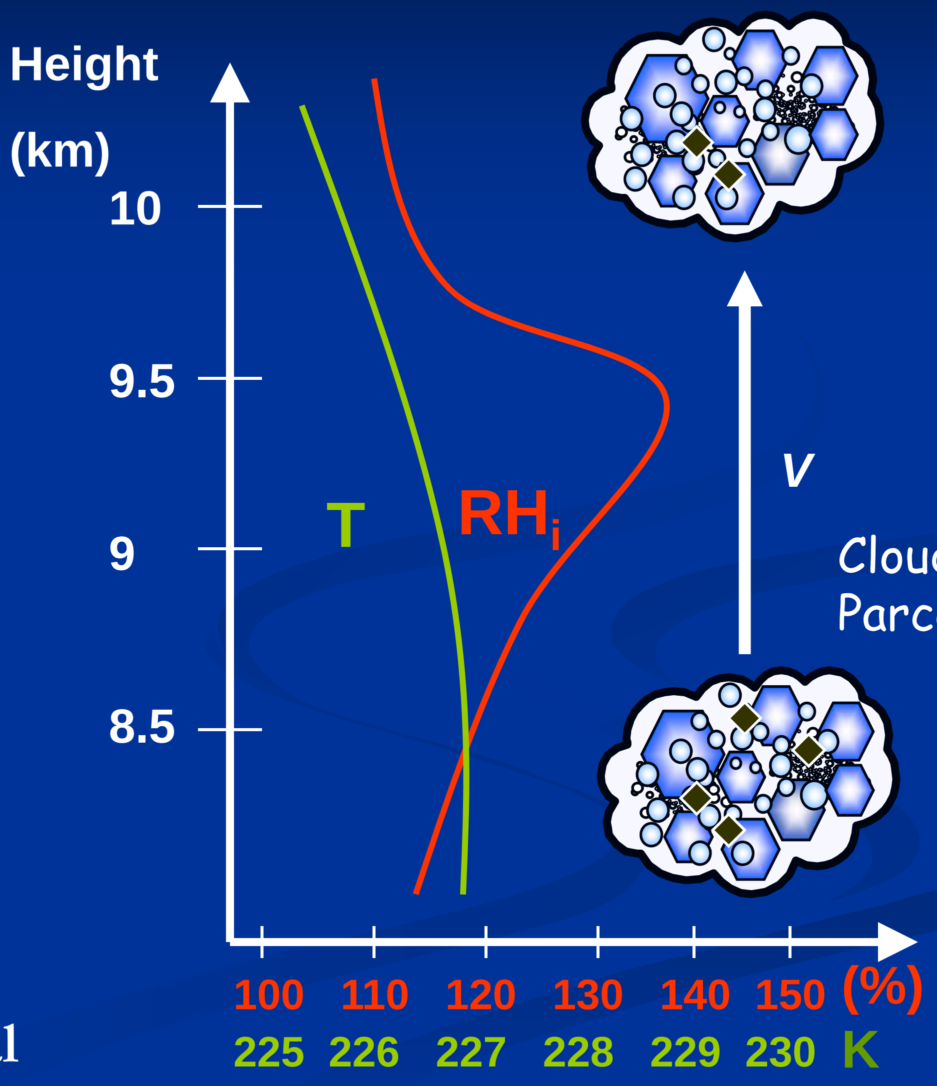
Heterogeneous Freezing

(Immersion, deposition, contact, …)

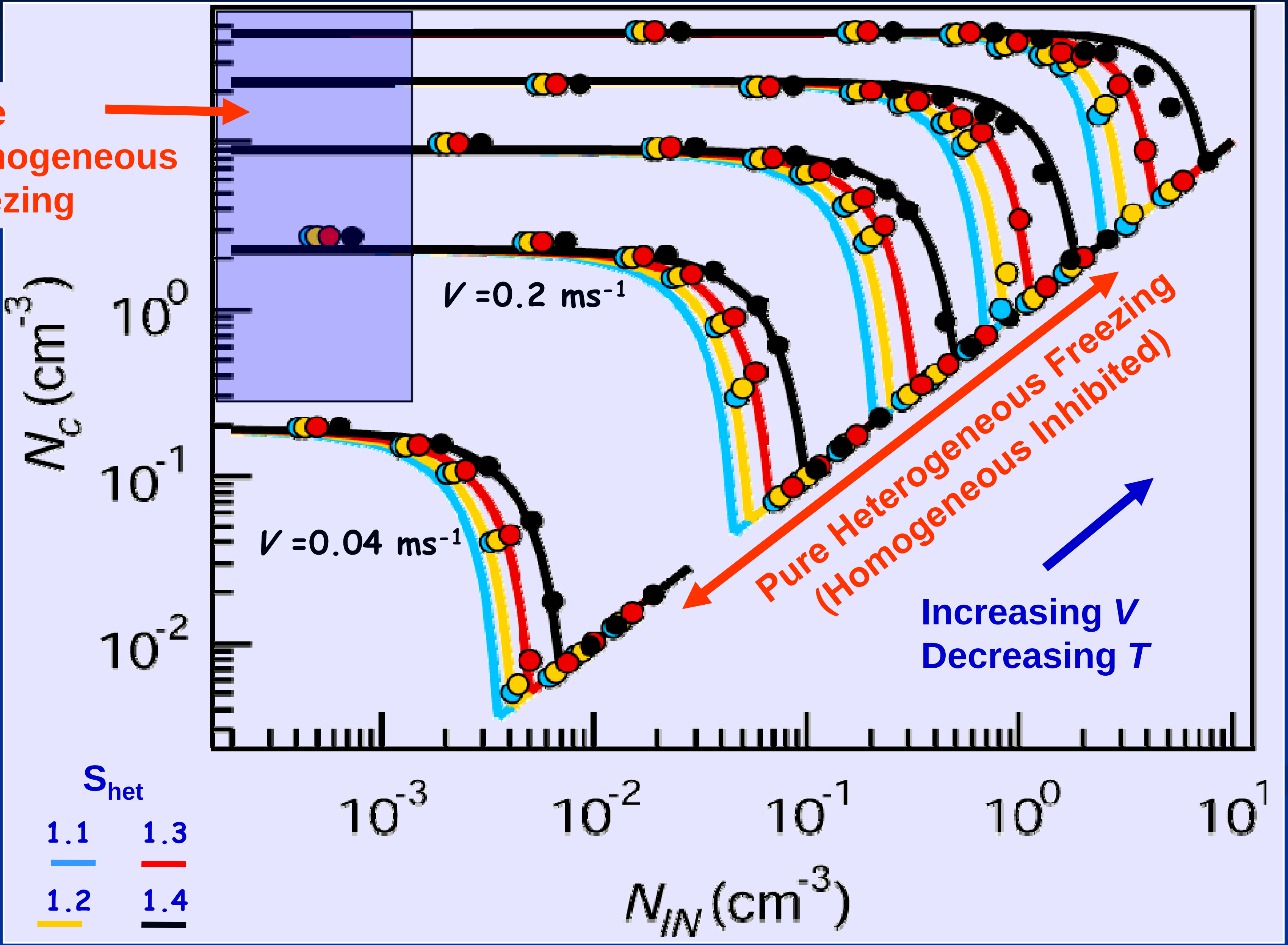
Also depends on the material and surface area

Liquid Droplets

+ Insoluble Material (Ice Nuclei)



Parameterizing Ice Formation Possible



Barahona and Nenes, *ACP*, 2009.

Summary

- 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.
- A large amount of uncertainty arises from the treatment of clouds and aerosol-cloud interactions in climate models.
- The indirect effect can regionally be **dominant (like in Georgia)**. 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 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.