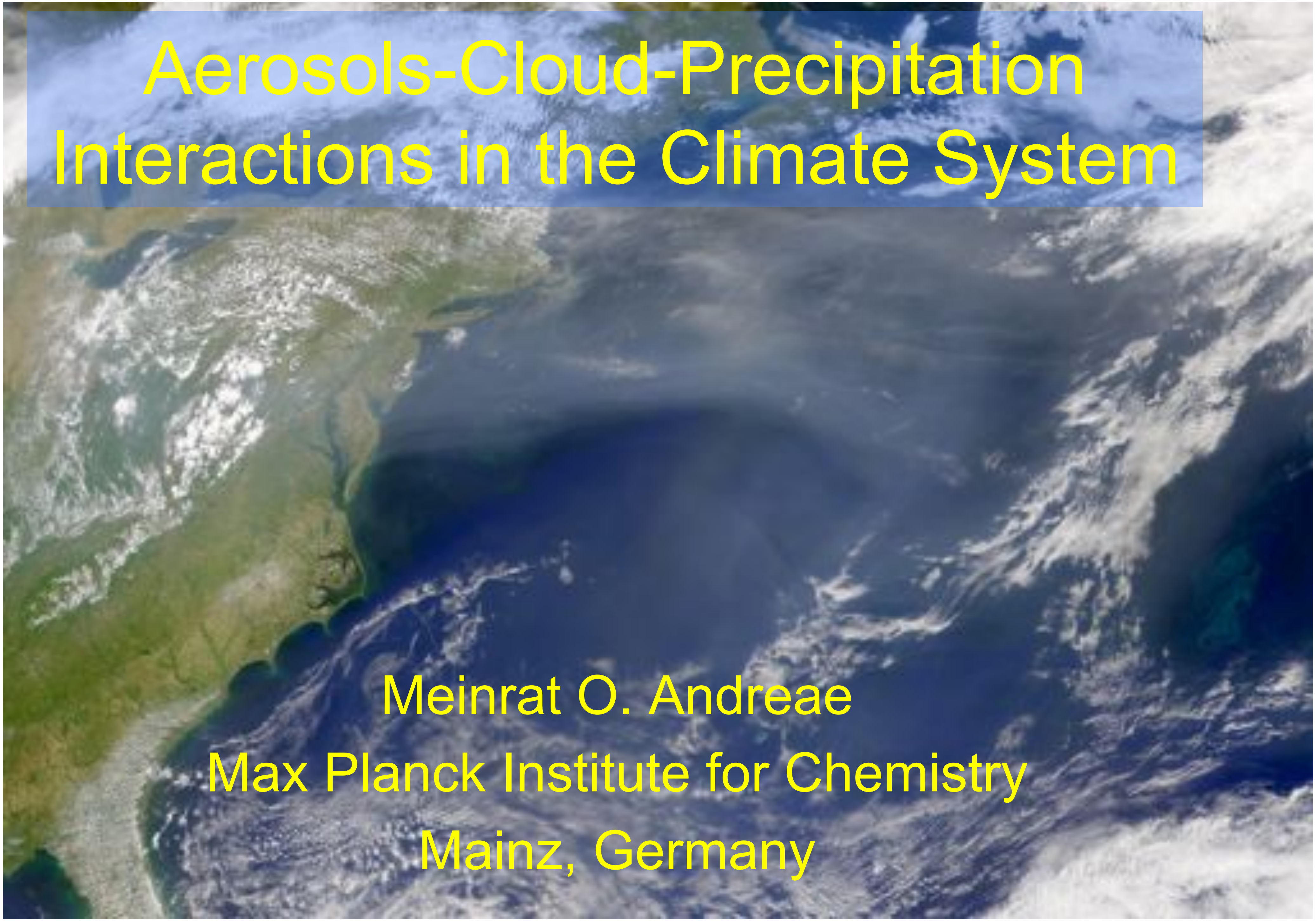
Aerosols-Cloud-Precipitation Interactions in the Climate System



Meinrat O. Andreae
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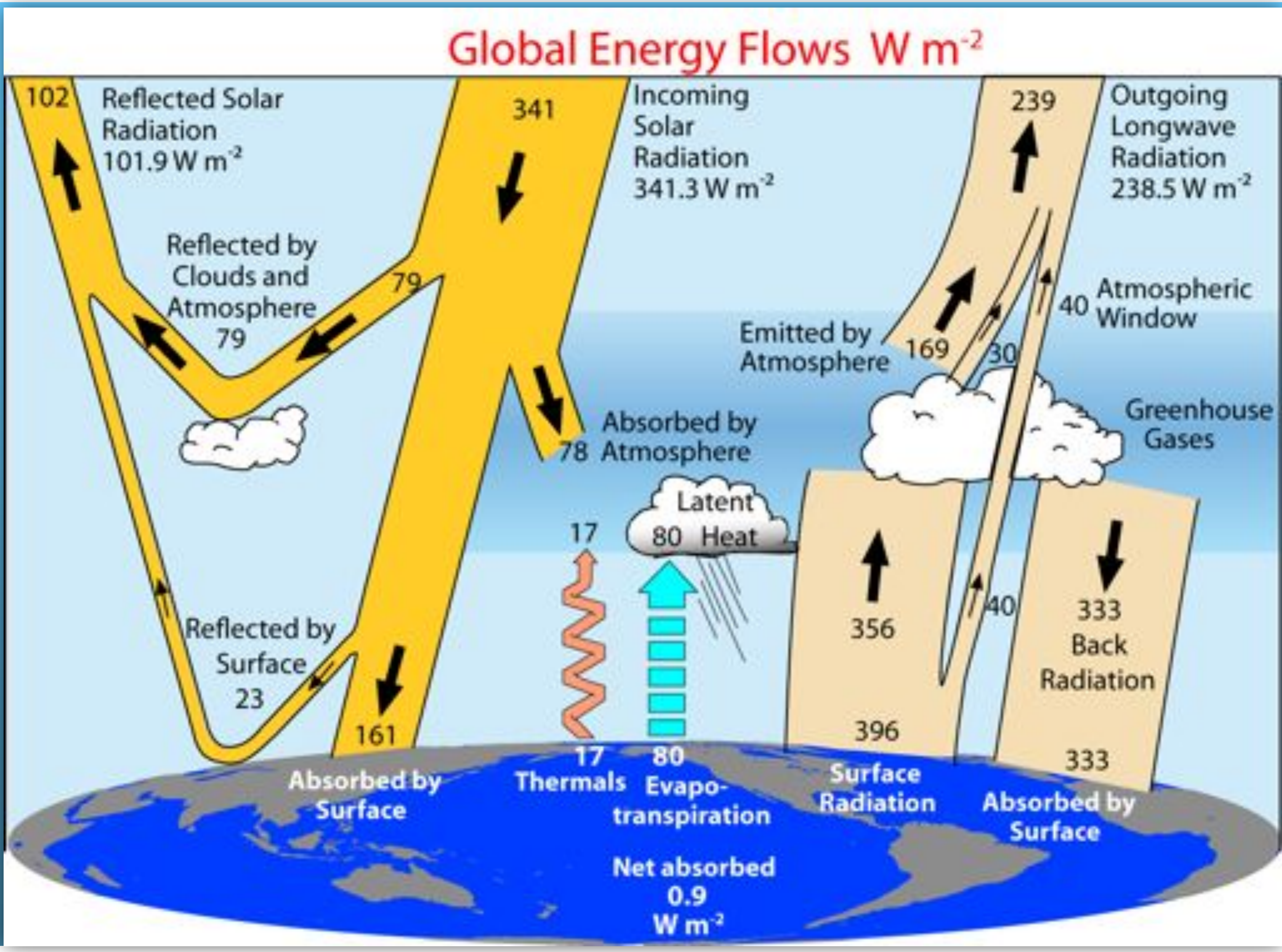
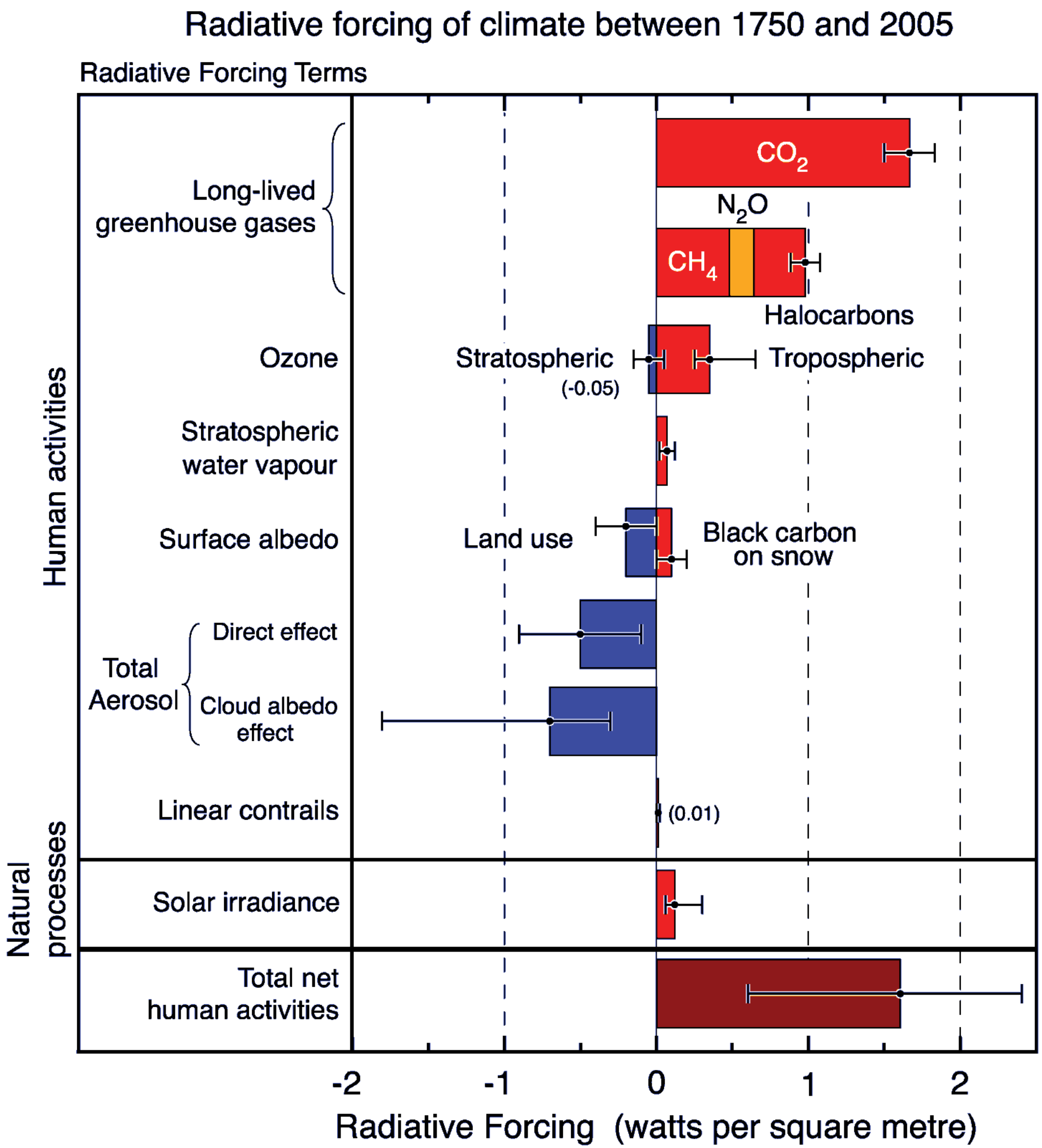


The global annual mean Earth's energy budget for 2000-2004 (W m^{-2}). The arrows indicate the flow of energy in proportion to their importance (Trenberth et al., 2009).



Climate Forcing

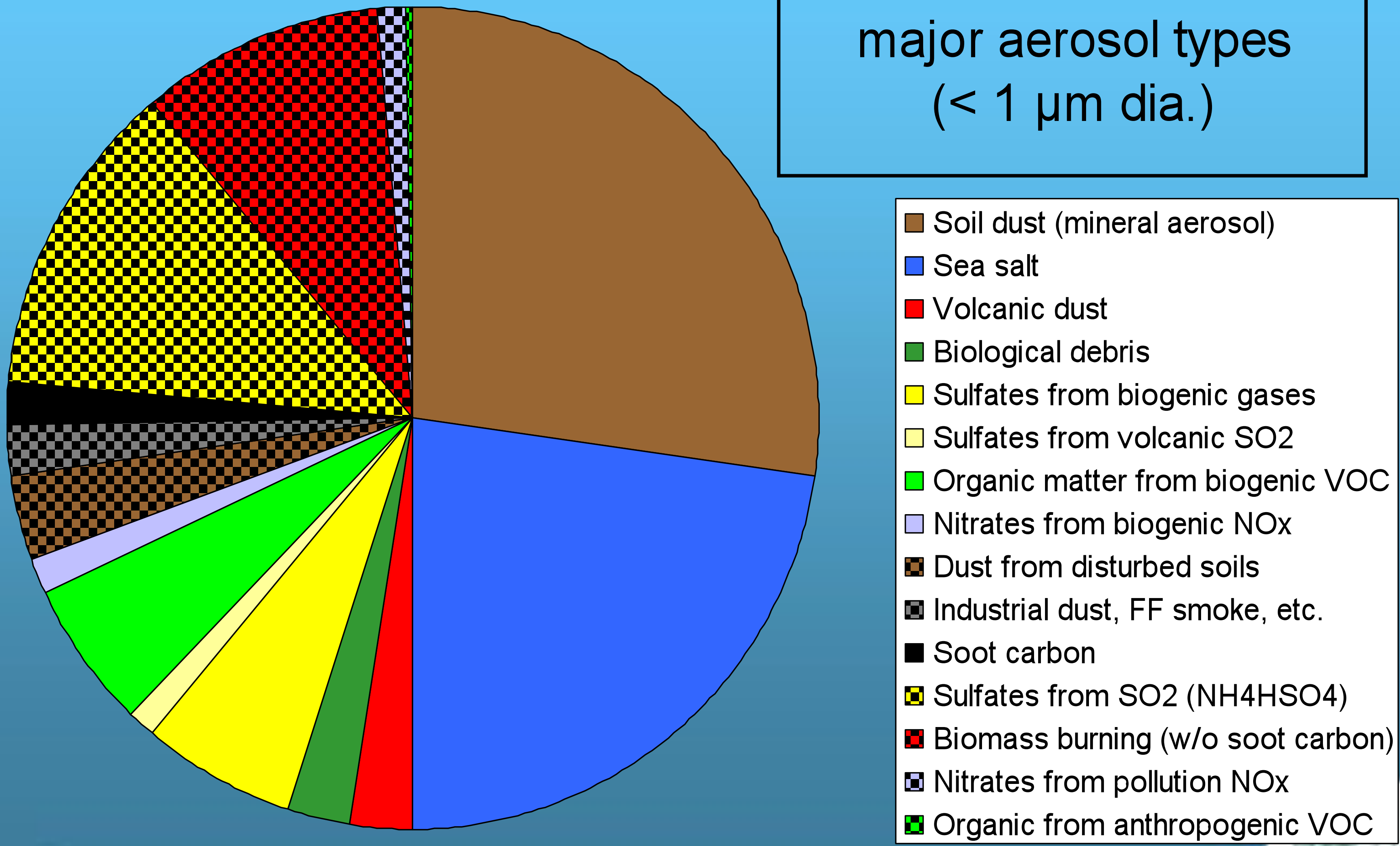
Aerosols are the major uncertainty

Outline

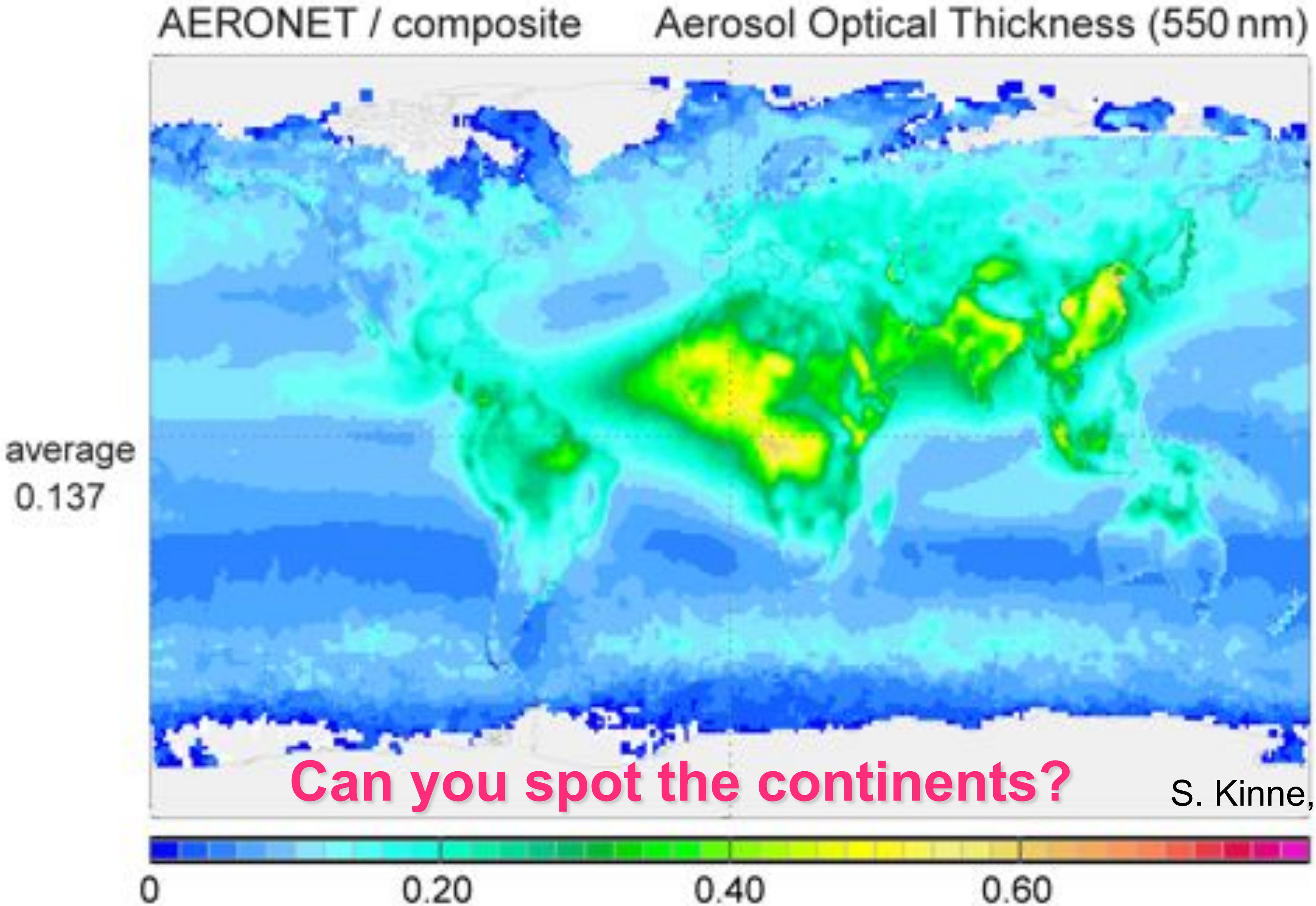
- **Aerosol sources and distributions**
- CCN properties of aerosols
- Aerosol climate effects overview
- Aerosol precipitation interactions

Aerosol Burden from Diverse Sources

Global mass burden for
major aerosol types
(< 1 µm dia.)



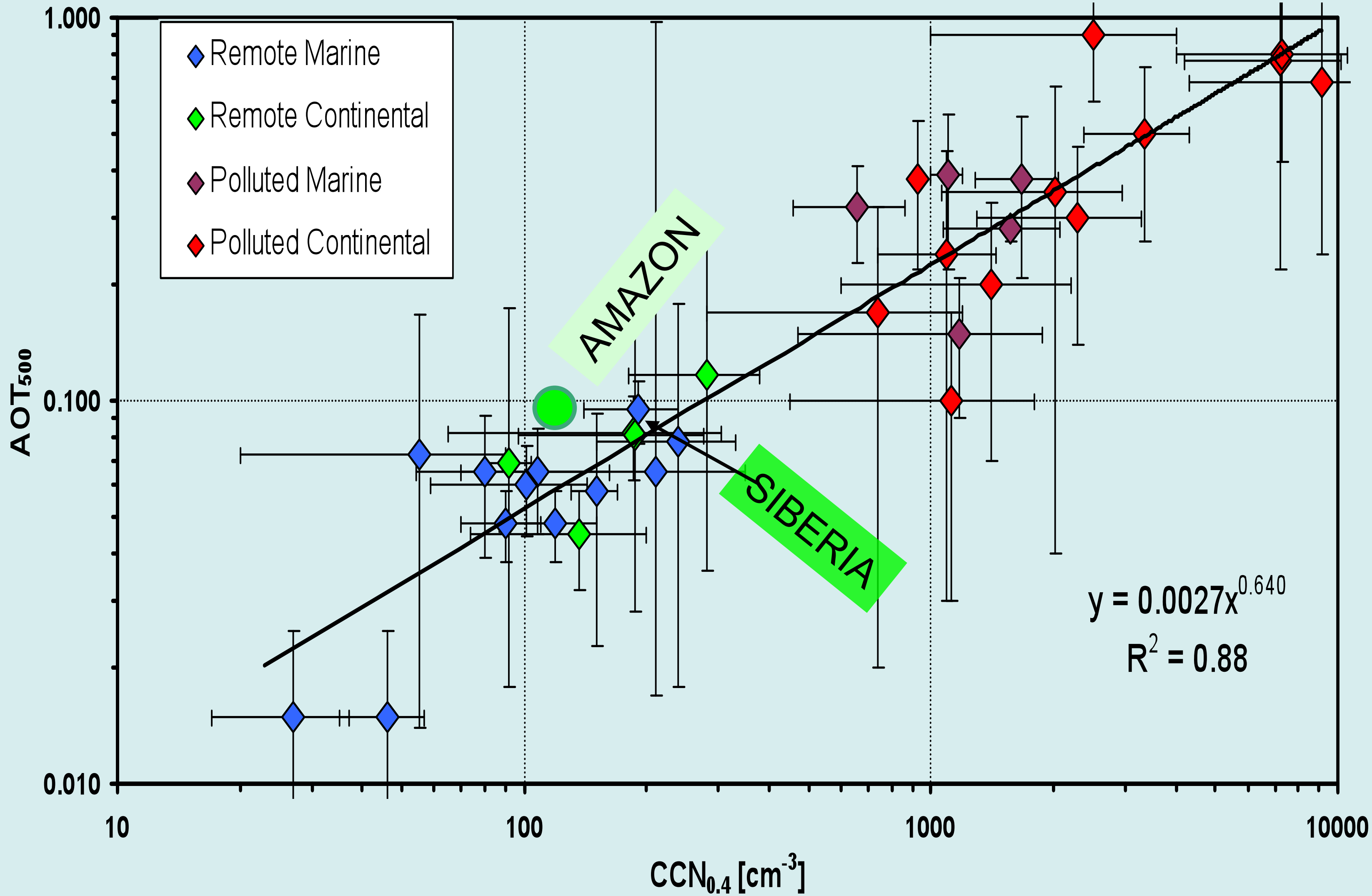
A Global Mean Aerosol Climatology



S. Kinne, 2008

There is no observational support for the old classification into "continental" and "maritime" aerosol (and cloud physics "regimes"!)

Global Distribution of CCN and AOT



CCN concentrations and AOD over the cleanest continental sites are similar to the cleanest marine sites!

Observations:

- We see strong signals from pollution, dust, smoke and salt, but very low AOT over the remaining regions
- There is surprisingly little contrast in AOT and CCN between clean ocean and clean continents
- This is supported by in situ measurements in Amazonia and Siberia

Outline

- Aerosol sources and distributions
- <u>**CCN properties of aerosols**</u>
- Aerosol climate effects overview
- Aerosol precipitation interactions

Cloud Condensation Nuclei (CCN)

- A CCN is a particle that can nucleate a cloud droplet at a particular water vapor supersaturation
- Without CCN, a supersaturation of about 800% would be required (because of the Kelvin effect). Cloud base would be at about 5 km.
- CCN help droplet nucleation by supplying solutes that help keep the water molecules in the liquid phase (the Raoult effect)

Kelvin's Law

Equilibrium water partial pressure over a flat surface (from Clausius-Clapeyron):

$$e_s(T) = 6.112 \exp\left(\frac{17.67T}{T + 243.5}\right)$$

Saturation ratio: $$\frac{e}{e_s(T)} = S_w$$

Kelvins law describes the increase in vapor pressure with decreasing diameter (or radius of curvature)

$$\frac{e}{e_s(T)} = S_w = \exp\left[\frac{4M_w\sigma_w}{RT\rho_w D}\right]$$

(for pure water)

Cloud formation in “pure air”

- To form stable water clusters that can grow into cloud droplets, a saturation ratio of ca. 8-9 would be required
- For typical conditions (dew point ca. 13°C), this would require cooling air to ca. -15 to -20°C
- This would result in cloud base at about 4-5 km altitude
- In real atmosphere, clouds form lower, so something must come to help

Raoult's Law

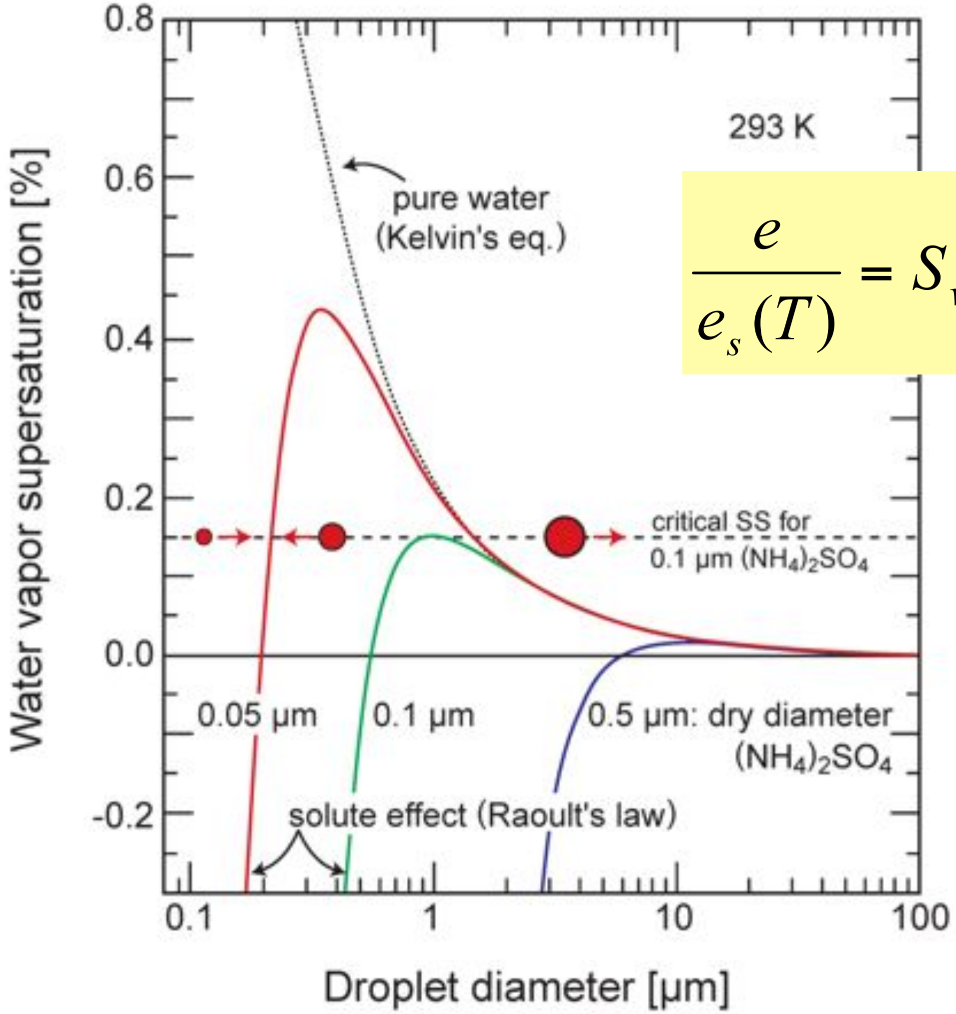
Water equilibrium vapor pressure is lower over salt solutions:

$$\frac{e}{e_s(T)} = A \cdot \exp\left[-\frac{vX_s M_w}{M_s \rho_w}\right] = A \cdot \exp\left[-\frac{6vm_s M_w}{M_s \rho_w \pi D^3}\right]$$

Kelvins law describes the decrease in vapor pressure with increasing solution concentration

Combining Kelvin's and Raoult's law yields the Köhler equation:

Köhler Theory of Droplet Activation



Kelvin Term

Raoult Term

$$\frac{e}{e_s(T)} = S_w = \exp\left[\frac{4M_w\sigma_w}{RT\rho_w D} - \frac{6vm_s M_w}{M_s\rho_w \pi D^3}\right]$$

$$e_s(T) = 6.112\exp\left(\frac{17.67T}{T+243.5}\right)$$

...tute for Chemistry, Mainz, Germany

Critical supersaturation, S_c

The critical supersaturation, S_c, at which an aerosol particle is activated to become a cloud droplet depends on:

- particle size (d_s – dry particle diameter)
- chemical composition (ε – soluble fraction; v – dissociation factor; ρ_s – density; M_s – molecular weight)

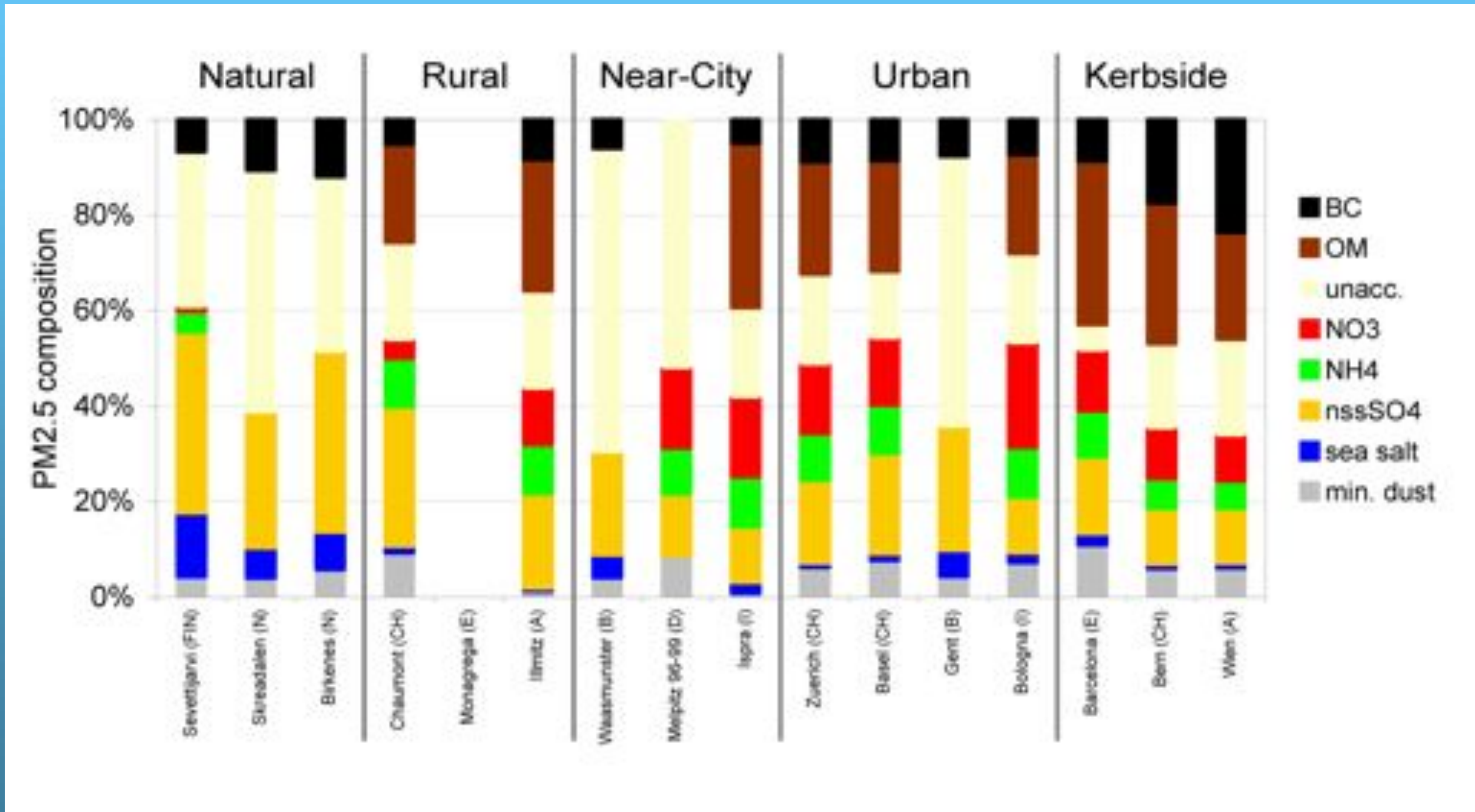
$$s_c \approx \left(\frac{4A^3}{27C \cdot n_i} \right)^{1/2}$$

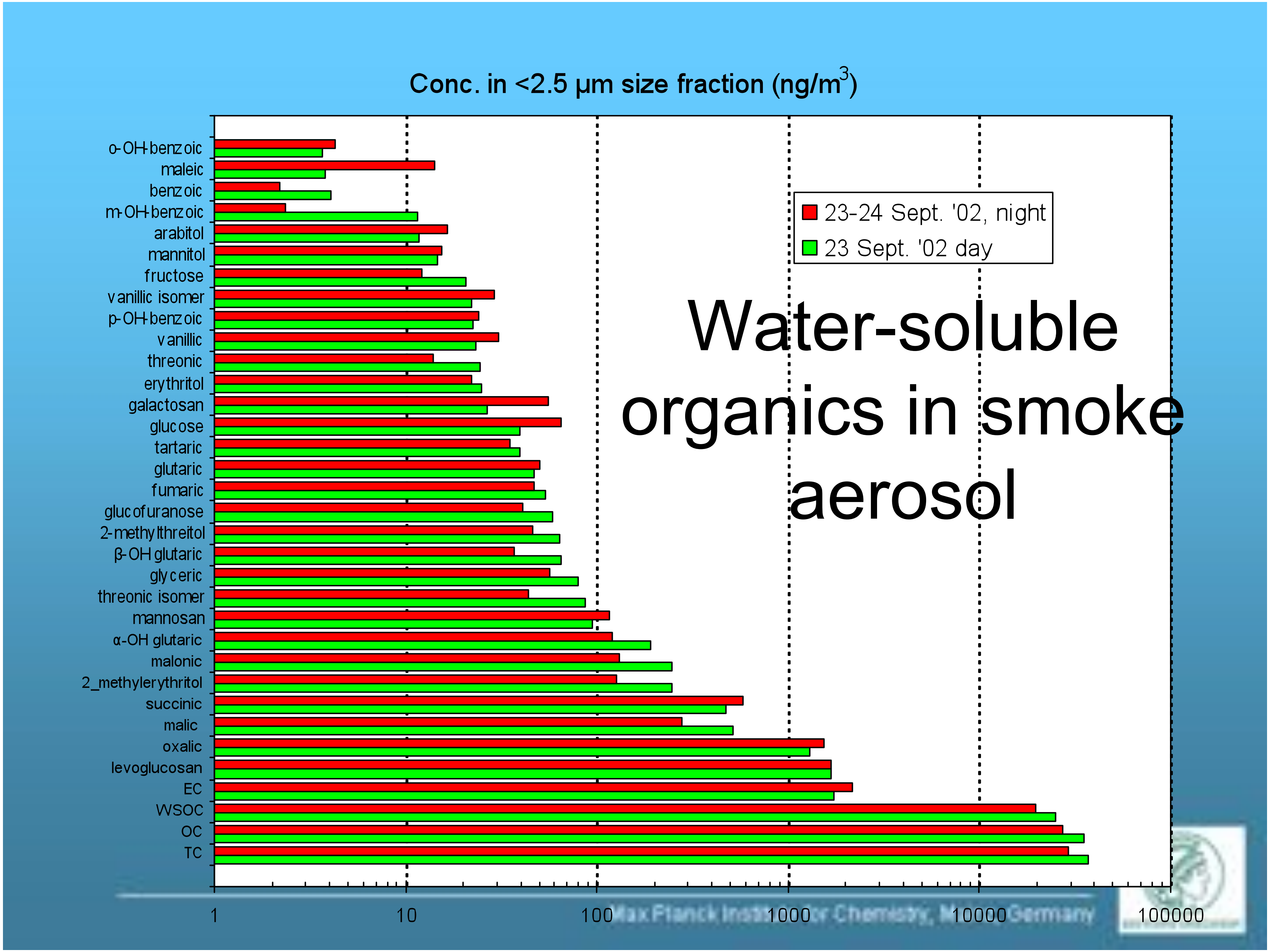
$$n_i = \frac{\pi \cdot d_s^3}{6} \cdot \varepsilon \cdot \upsilon \cdot \frac{\rho_s}{M_s}$$

Note that "chemistry" variables enter at first power, size at THIRD power!

We could derive the critical supersaturation if we know the size and composition of the particle (as long as there is some valid mixing rule)

Chemical composition of aerosols




Conc. in <2.5 µm size fraction (ng/m3)
o-OH-benzoic
maleic
benzoic
m-OH-benzoic
arabitol
mannitol
fructose
vanillic isomer
p-OH-benzoic
vanillic
threonic
erythritol
galactosan
glucose
tartaric
glutaric
fumaric
glucofuranose
2-methylthreitol
β-OH glutaric
glyceric
threonic isomer
mannosan
α-OH glutaric
malonic
2_methylerythritol
succinic
malic
oxalic
levoglucosan
EC
WSOC
OC
TC
1
10
100
1000
10000
100000
23-24 Sept. '02, night
23 Sept. '02 day
Water-soluble organics in smoke aerosol
Max Planck Institute for Chemistry, Mainz, Germany

Additional Complications

- Surface tension reduction (surfactants)
  - Reduces Kelvin effect ("better CCN")
- Limited solubility (organics)
  - Reduces Raoult effect ("worse CCN")
- Insoluble coatings (oily substances)
  - Slow down water uptake
- Soluble gaseous species
  - HNO_3, NH_3; facilitate droplet nucleation

Is it all just too complicated … ?

Critical supersaturation, S_c

The critical supersaturation, S_c, at which an aerosol particle is activated to become a cloud droplet depends on:

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$$s_c \approx \left(\frac{4A^3}{27C \cdot n_i} \right)^{1/2}$$

$$n_i = \frac{\pi \cdot d_s^3}{6} \cdot \varepsilon \cdot \upsilon \cdot \frac{\rho_s}{M_s}$$

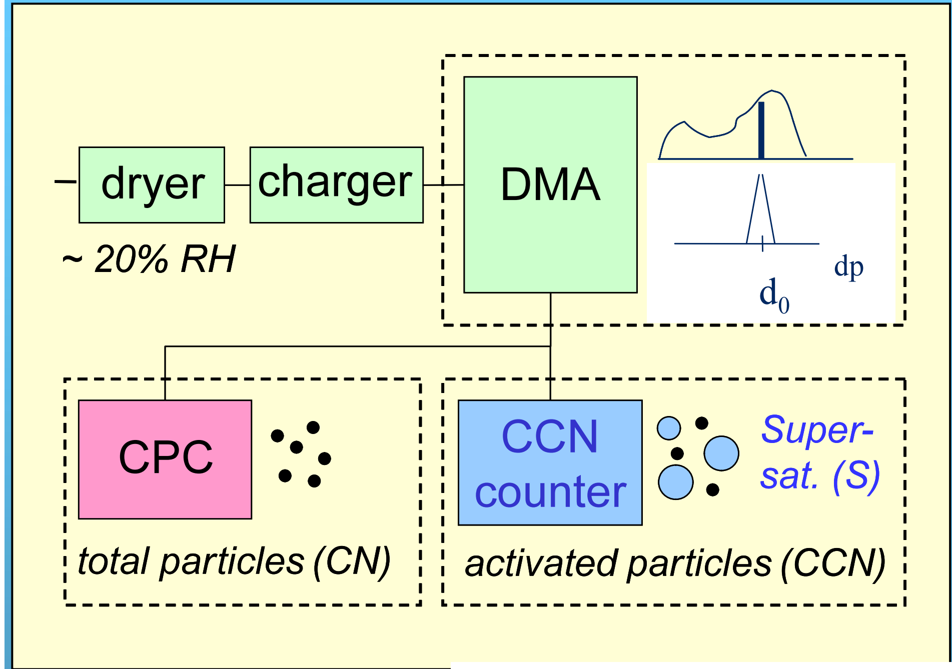
Note that "chemistry" variables enter at first power, size at THIRD power!

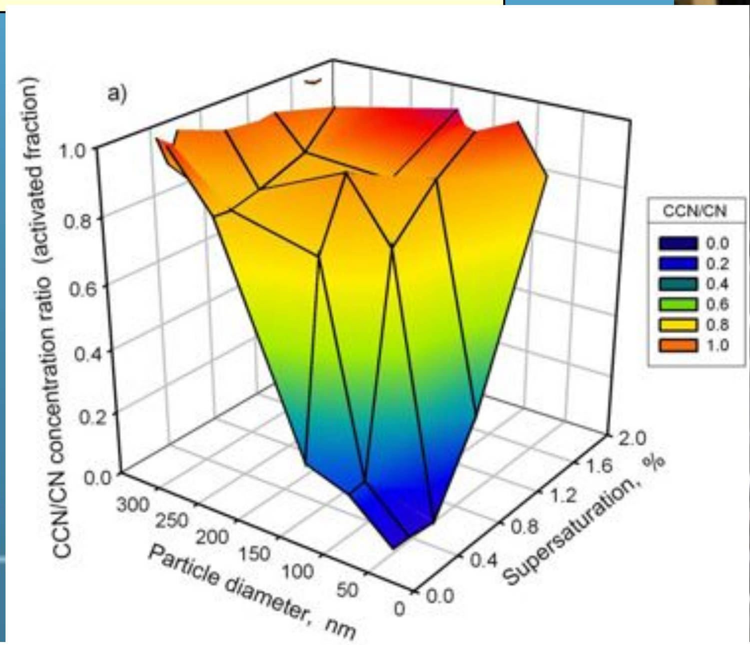
At constant composition, s_c should decrease as a function of d_s with a logarithmic slope of -2/3

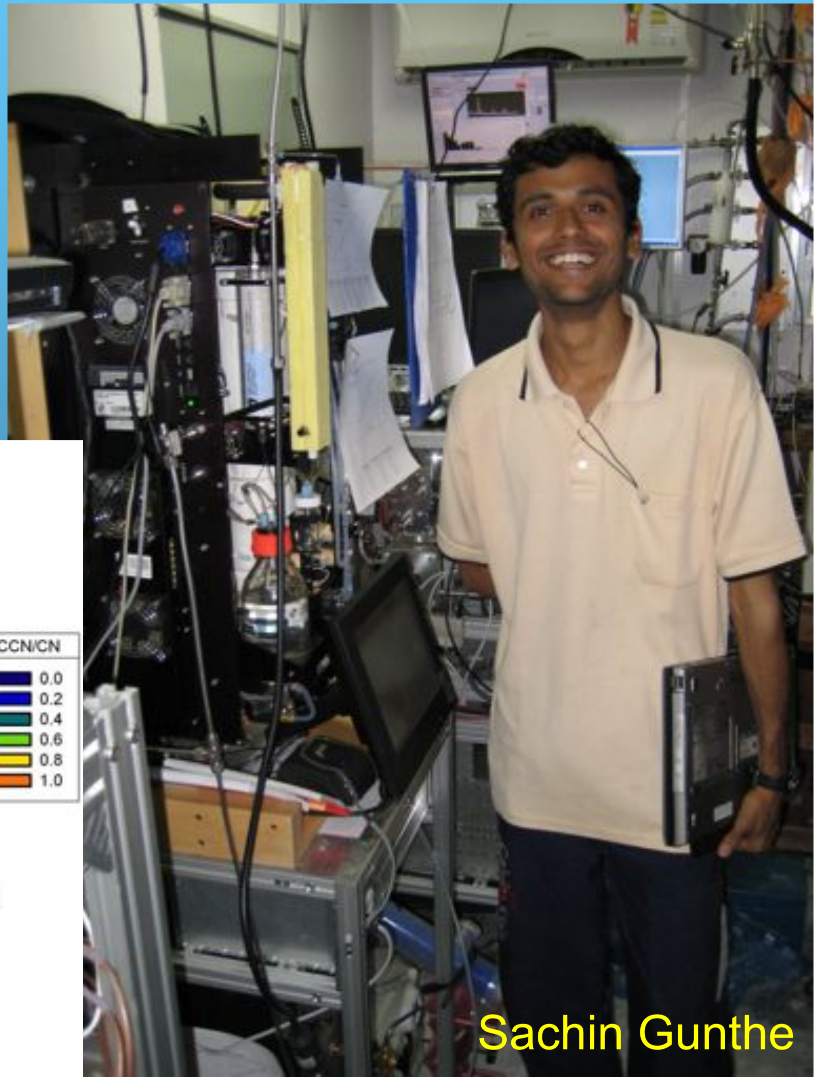
Size resolved measurements eliminate one free variable (size) and are useful to:

1. Directly determine activation diameters of ambient aerosol
2. Study the role of chemical composition for CCN activation

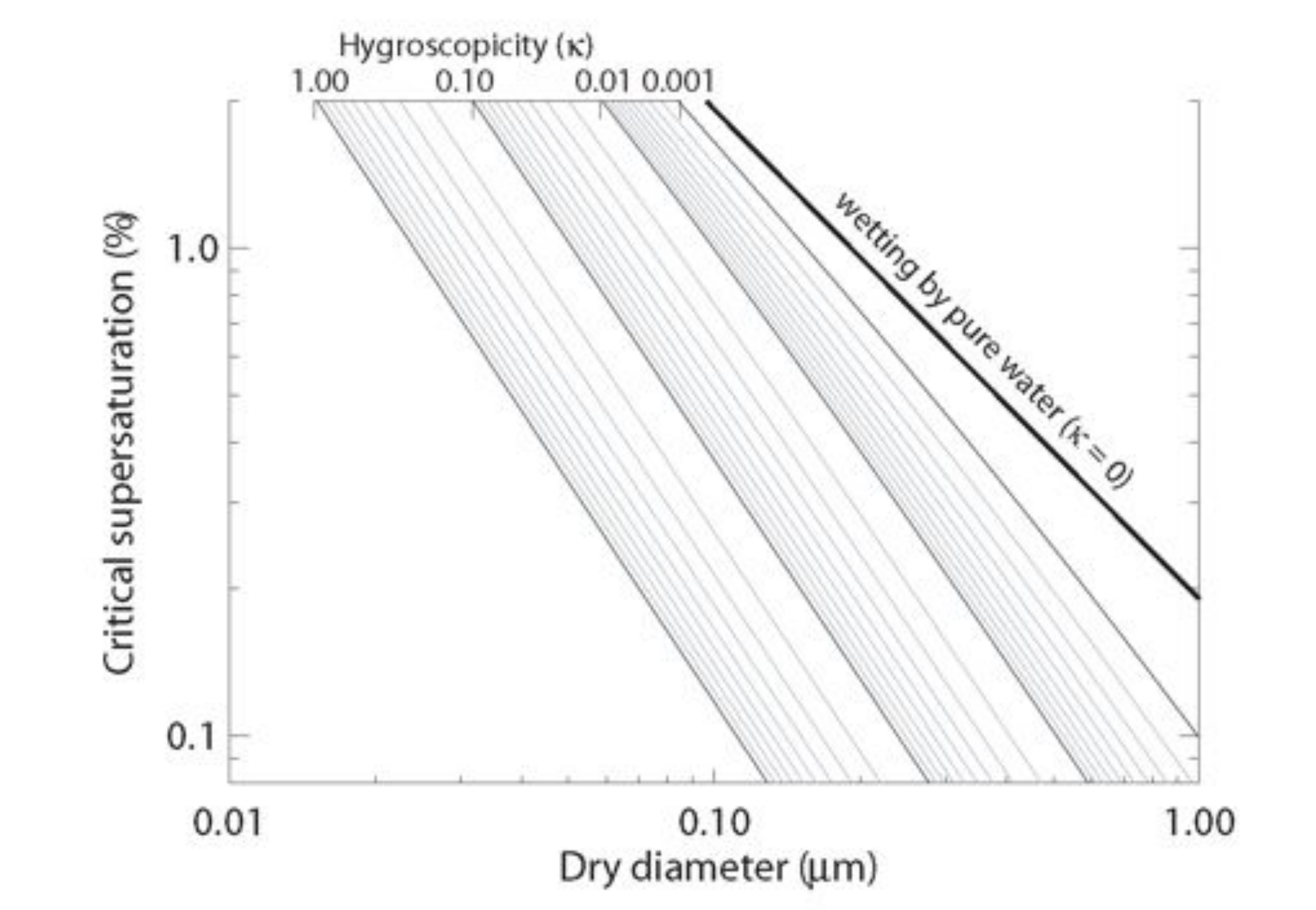
Size-resolved CCN Measurements







S_c-d_p-κ Space:



Definition of Hygroscopicity Parameter κ

The saturation ratio, S_w, over an aqueous solution droplet can be calculated from

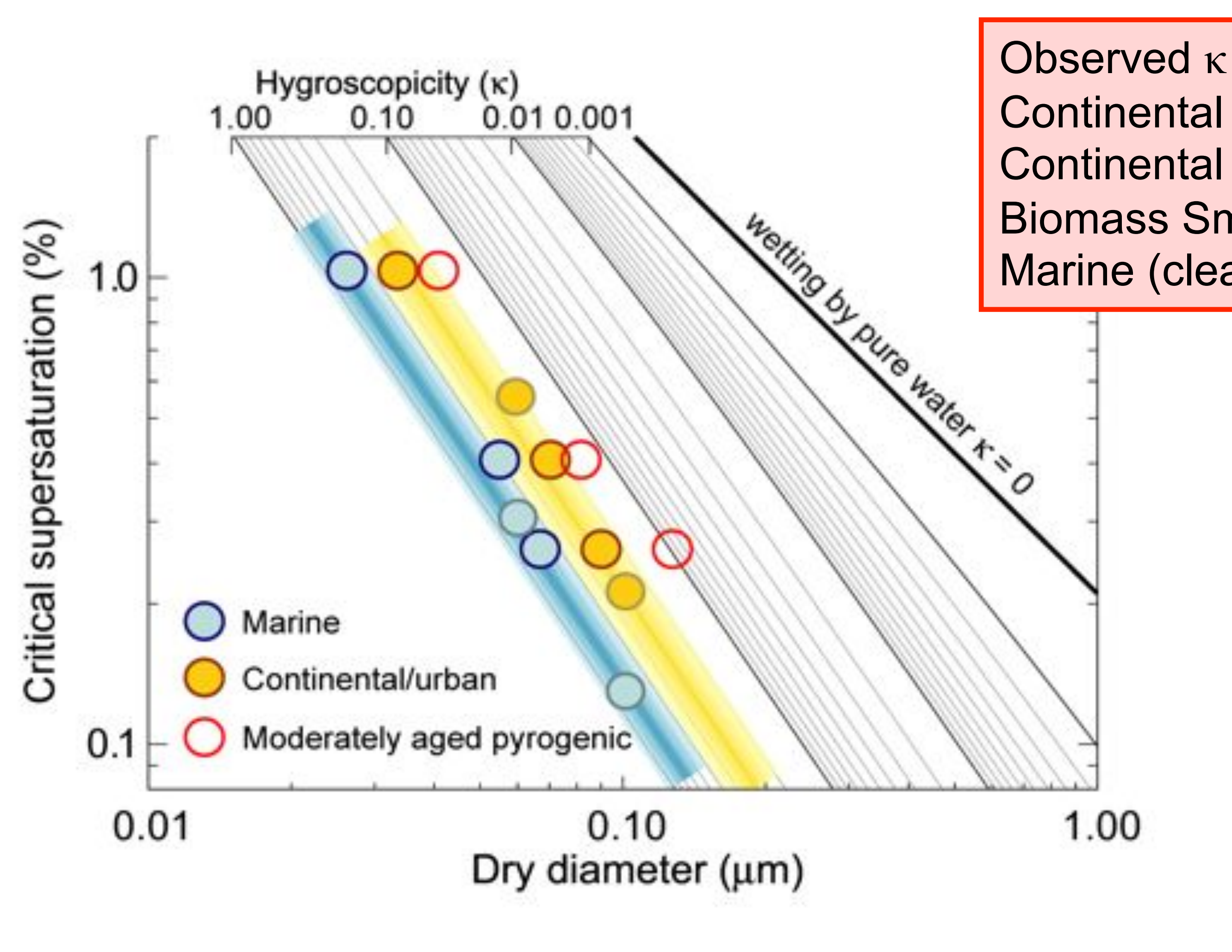
$$S_w = a_w \exp\left(\frac{4\sigma_{sa} M_w}{RT\rho_w D}\right)$$

where a_w is the activity of water in solution, ρ_w is the density of water, M_w is the molecular weight of water, σ_{sa} is the surface tension of the solution/air interface, R is the universal gas constant, T is temperature, and D is the diameter of the droplet. The hygroscopicity parameter is defined through its effect on the water activity of the solution:

$$\frac{1}{a_w} = 1 + \kappa \frac{V_s}{V_w}$$

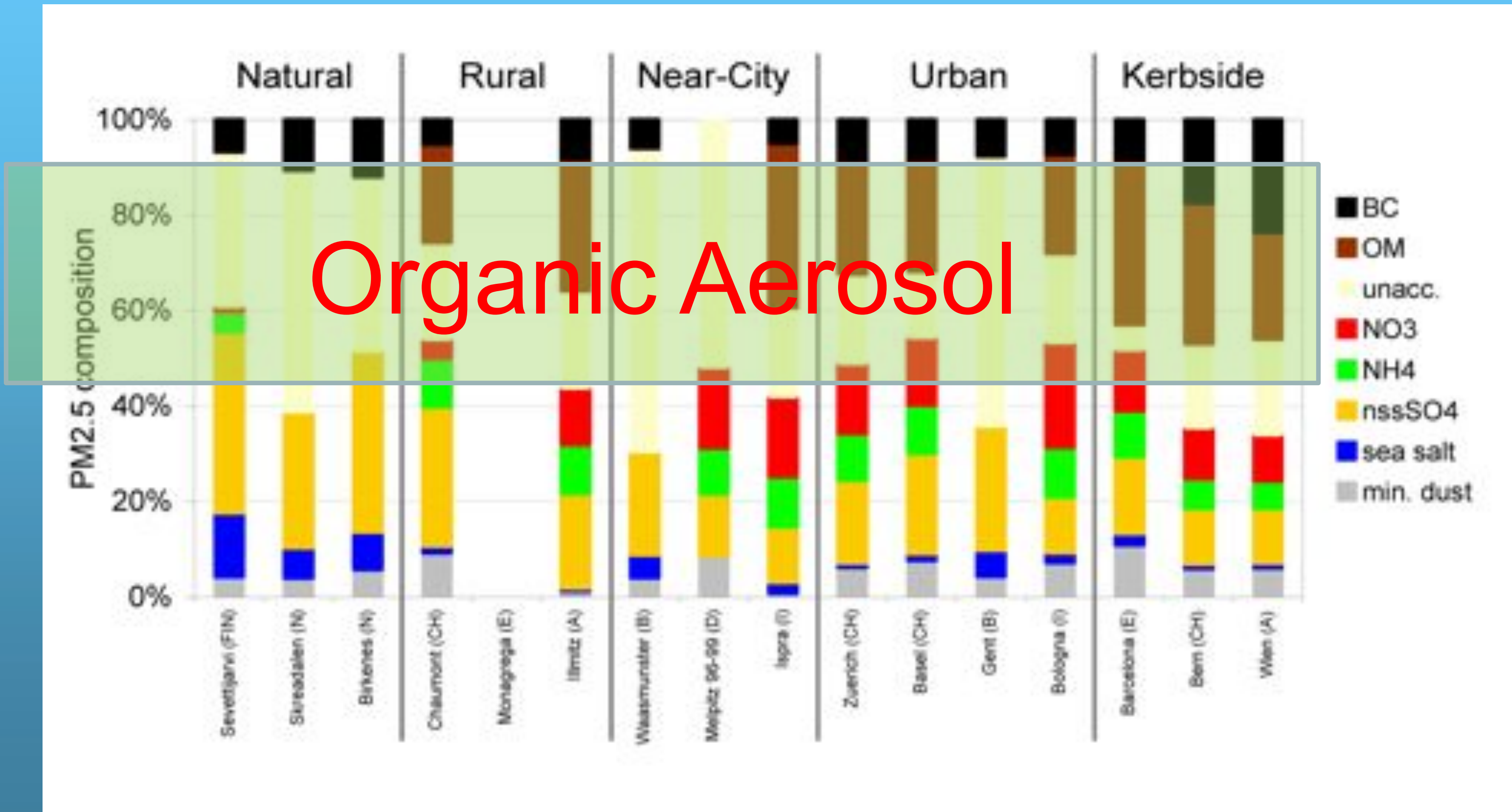
where V_s is the volume of the dry particulate matter and V_w is the volume of the water.

The world in S_c-d_p-κ Space:



Observed κ values:	
Continental (polluted):	0.3±0.1
Continental (clean):	0.2±0.1
Biomass Smoke:	0.2±0.1
Marine (clean):	0.7±0.2

Chemical composition of aerosols



Organic Aerosol Evolution

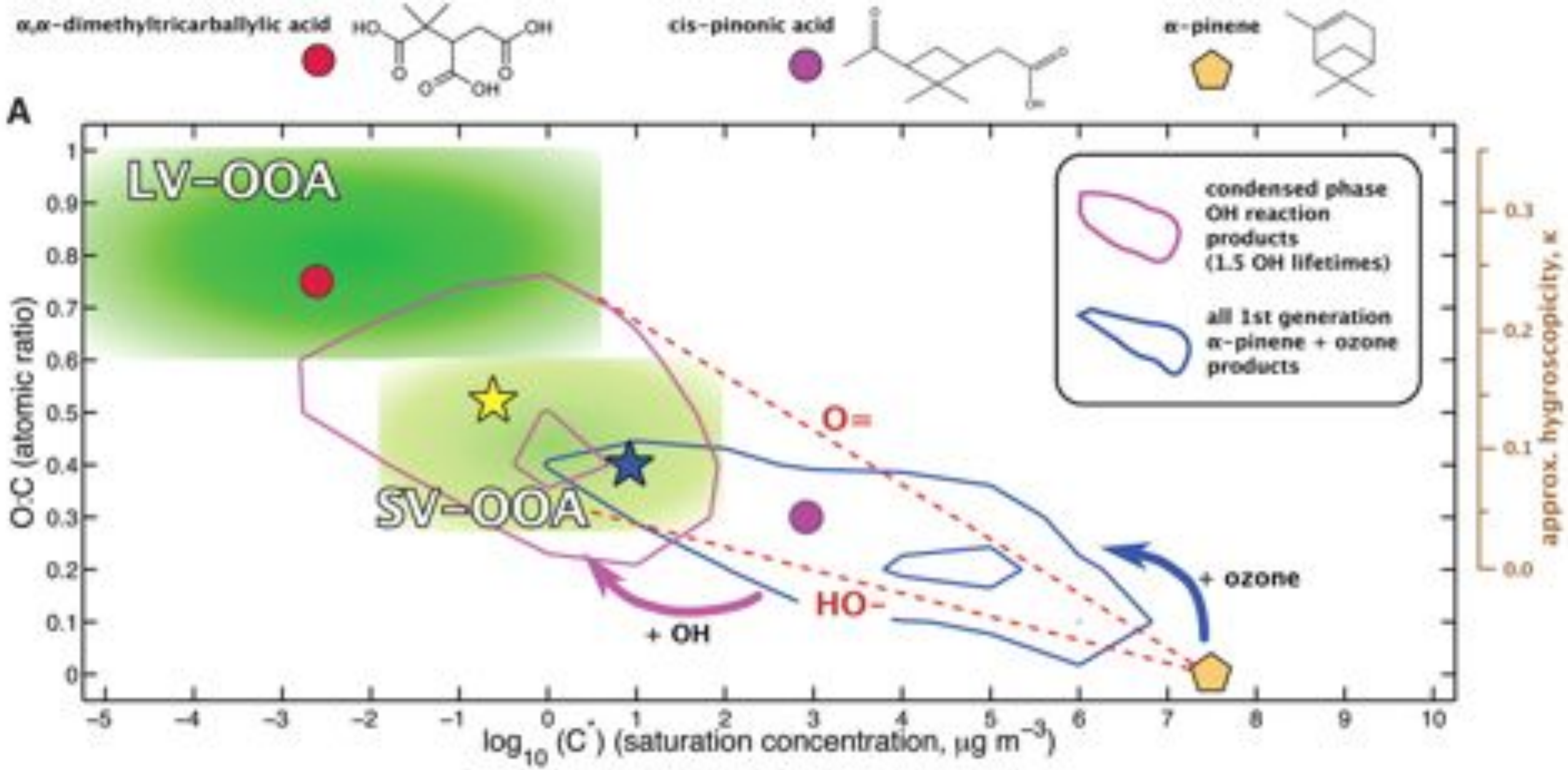


Fig. 4 (A) 2D framework for OA aging

J. L. Jimenez et al., Science 326, 1525-1529 (2009)

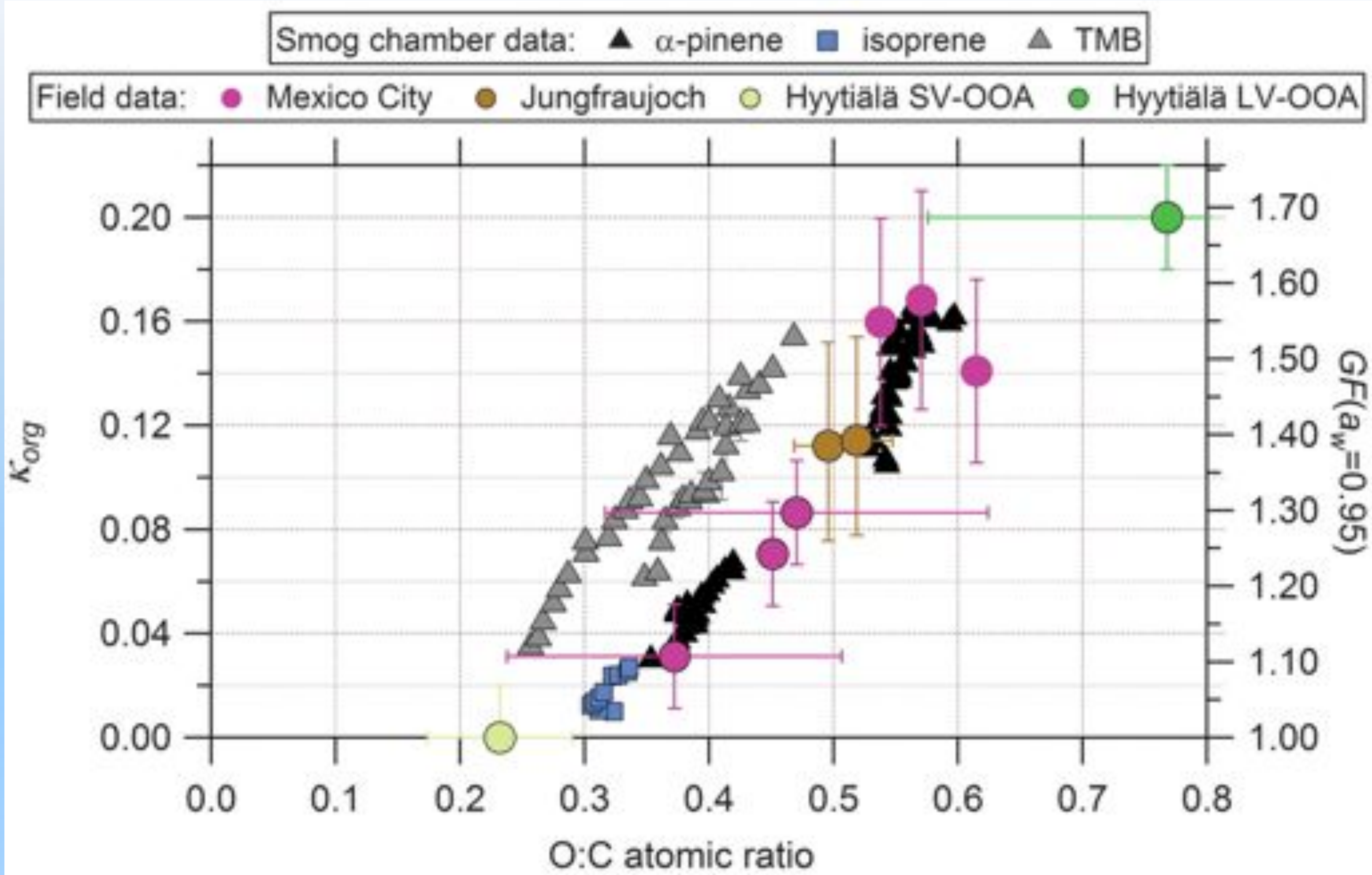
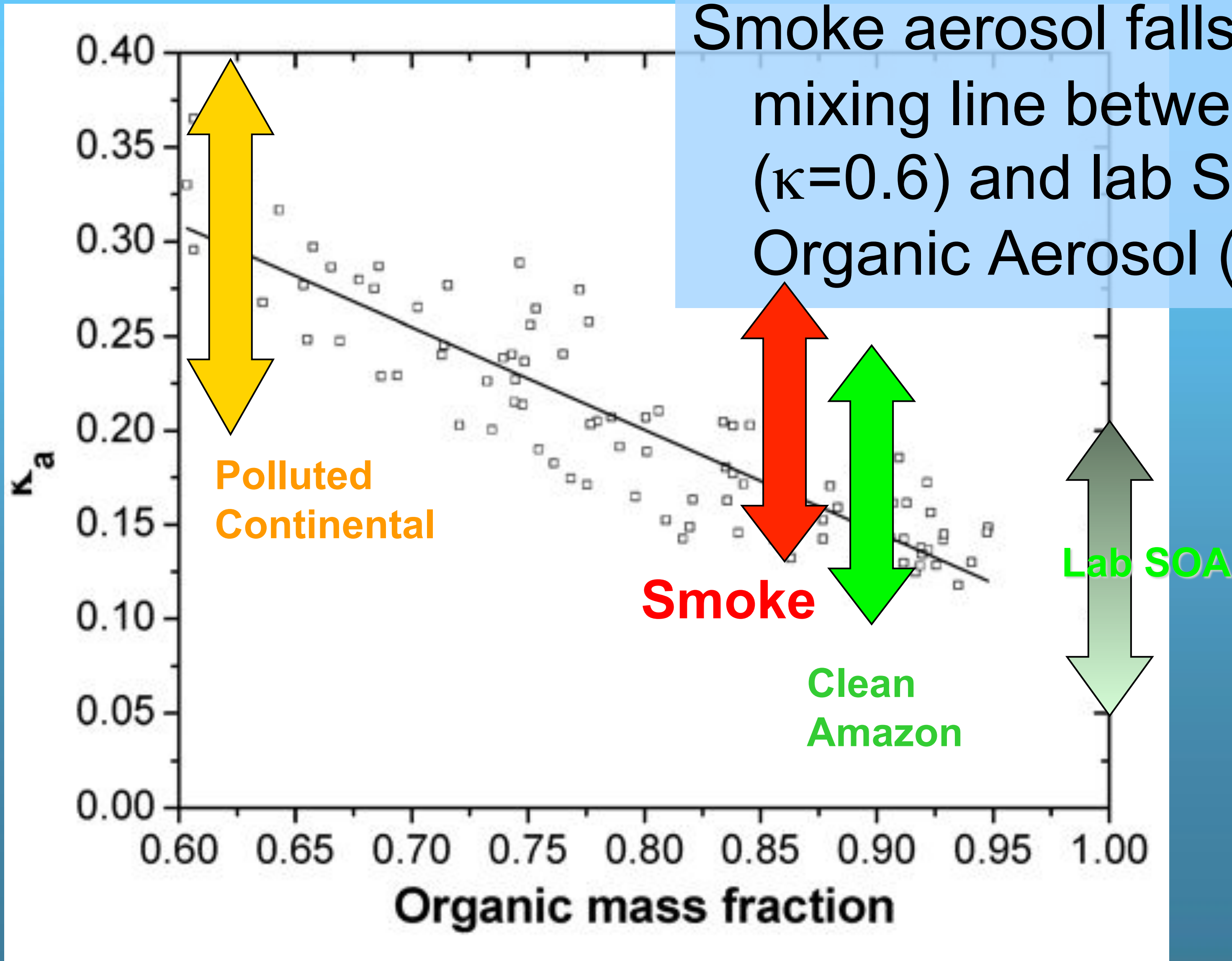


Fig. 3 Relationship between O:C and hygroscopicity (κ, or equivalently the particle growth factor at 95% relative humidity) of OA for several field data sets (a high-altitude site at Jungfraujoch, Switzerland; above Mexico City, a polluted megacity; and at the forested site of Hyytiälä, Finland) and for laboratory smog chamber SOA

J. L. Jimenez et al., Science 326, 1525-1529 (2009)

Influence of composition on hygroscopicity

Smoke aerosol falls on mixing line between sulfate (κ=0.6) and lab Secondary Organic Aerosol ($\kappa \approx 0.1$)



Global distribution of κ

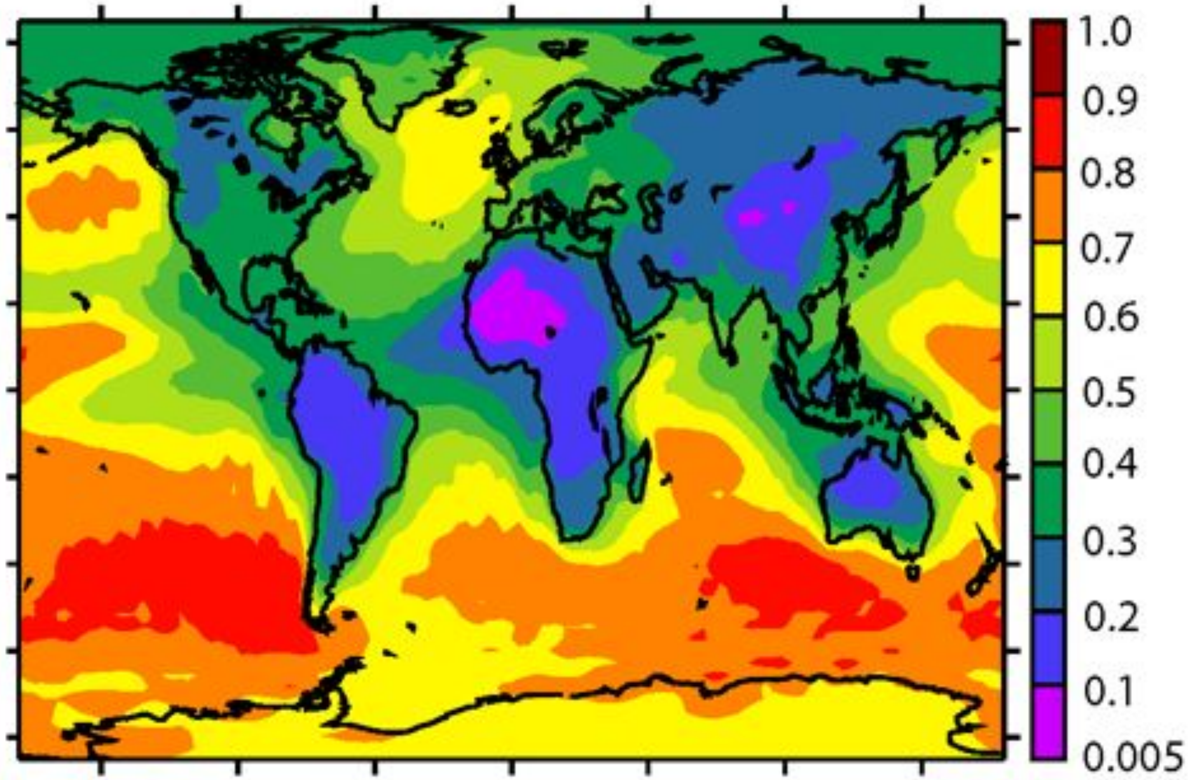
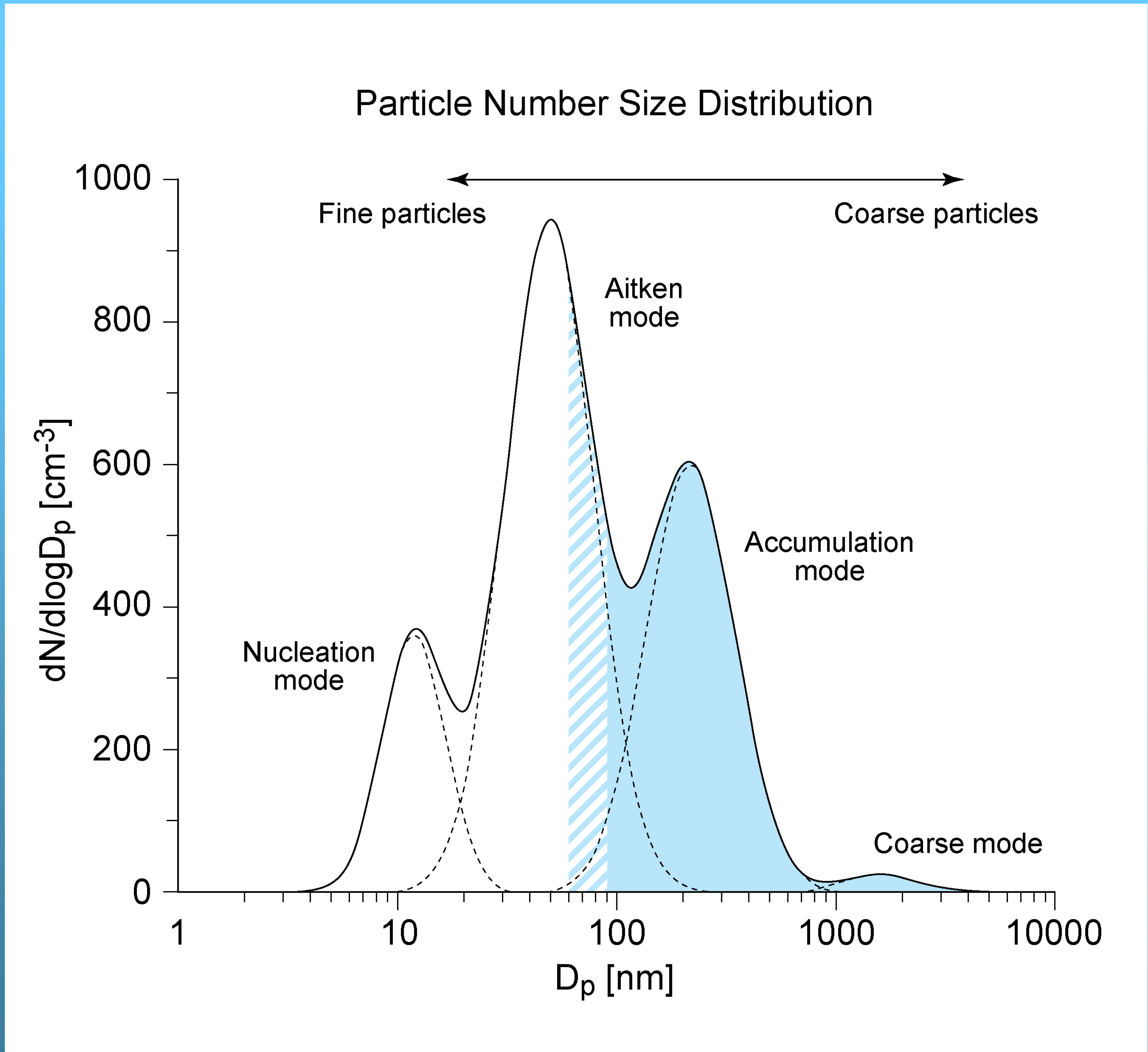


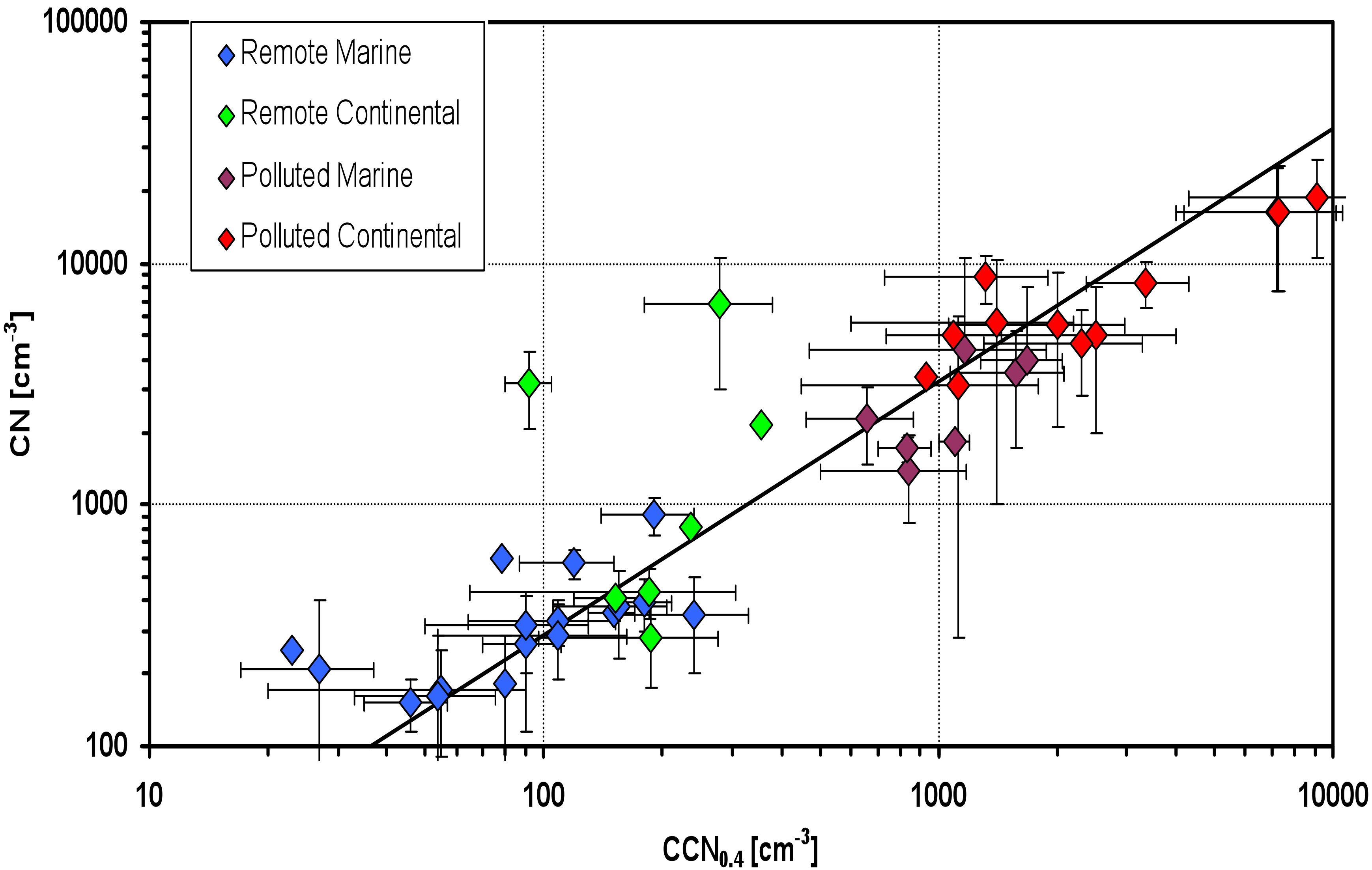
Fig. 2. Annual mean distribution of κ at the altitude of the planetary boundary layer.

Pringle et al., ACP 2010



Particle Number Size Distribution
Fine particles
Coarse particles
Aitken mode
Accumulation mode
Nucleation mode
Coarse mode
1000
800
600
400
200
0
1
10
100
1000
10000
$dN/d\log D_p$ [cm^{-3}]
D_p [nm]

Global CCN/CN Relationship



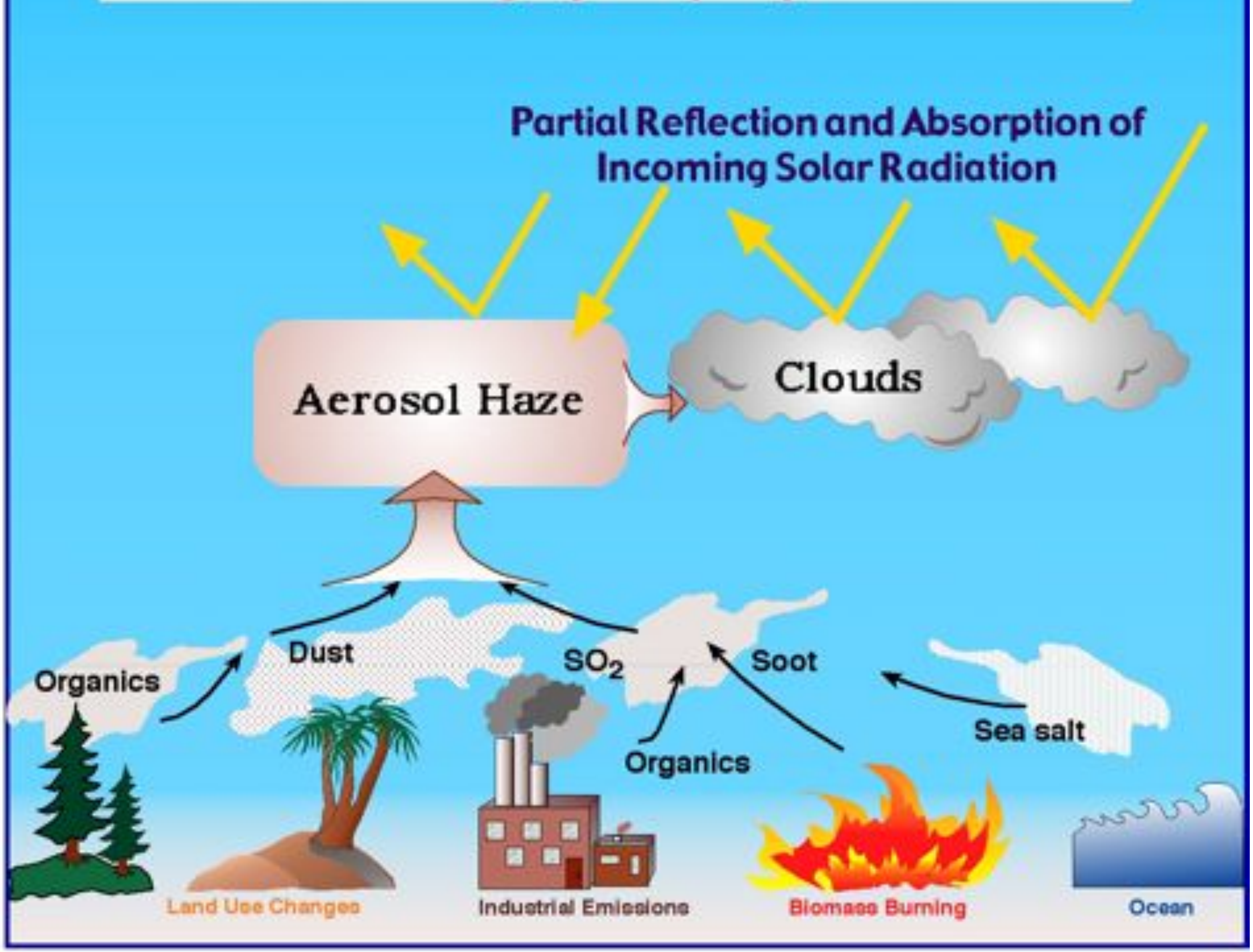
Summary on CCN

- CCN properties are in principle influenced by complex combination of chemical properties and size
- In real-world aerosols, the range of composition converges to a small range of hygroscopicities, dominated by inorganic component
- Size distributions also are variable, but again converge, thus:
- **There is a surprisingly constant ratio of CCN to total particle number concentrations**
- CCN measurements over biologically active ocean and land are similar (low hundreds per cc) – away from biol. activity they are much lower (tens per cc)

Outline, Lecture 2

- Aerosol sources and distributions
- CCN properties of aerosols
- **<u>Aerosol climate effects overview</u>**
- Aerosol precipitation interactions

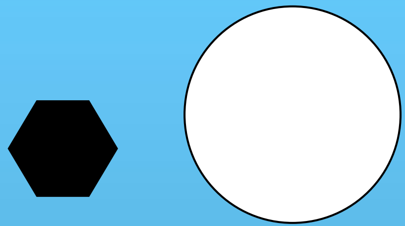
Radiative Forcing by Tropospheric Aerosol



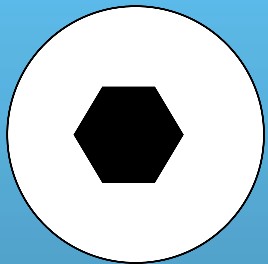

"Direct" Effects
Note: Haze is lighter than surface almost everywhere, especially over water, but darker over the low cloud patch in the Po Valley

Aerosol Mixing State of Black Carbon

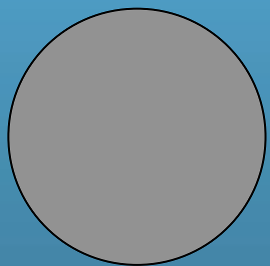
Forcing
(W m^{-2}, Jacobson, 2000)



External Mixing +0.27



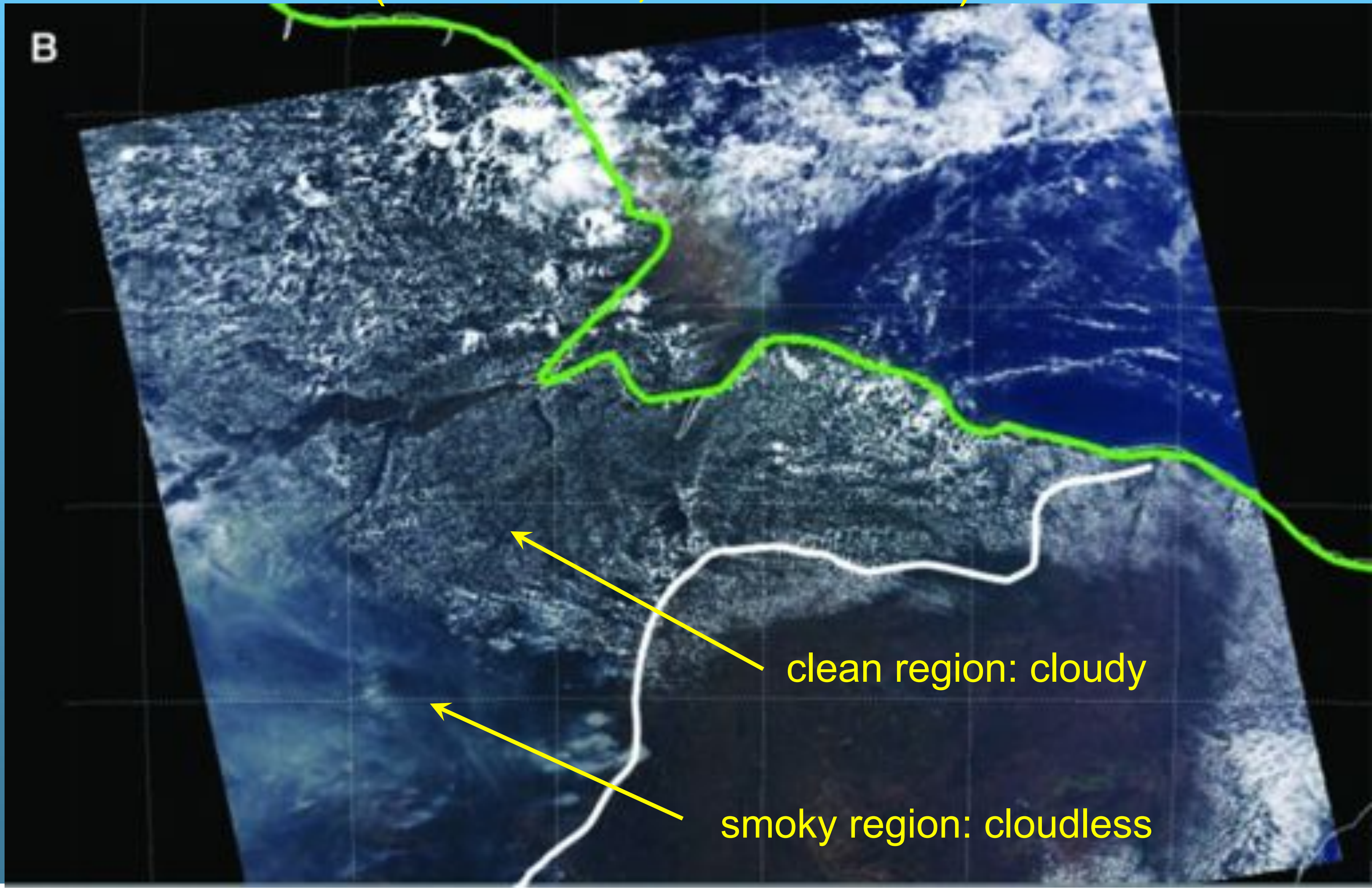
Black Carbon Core +0.54



Internal Mixing +0.78

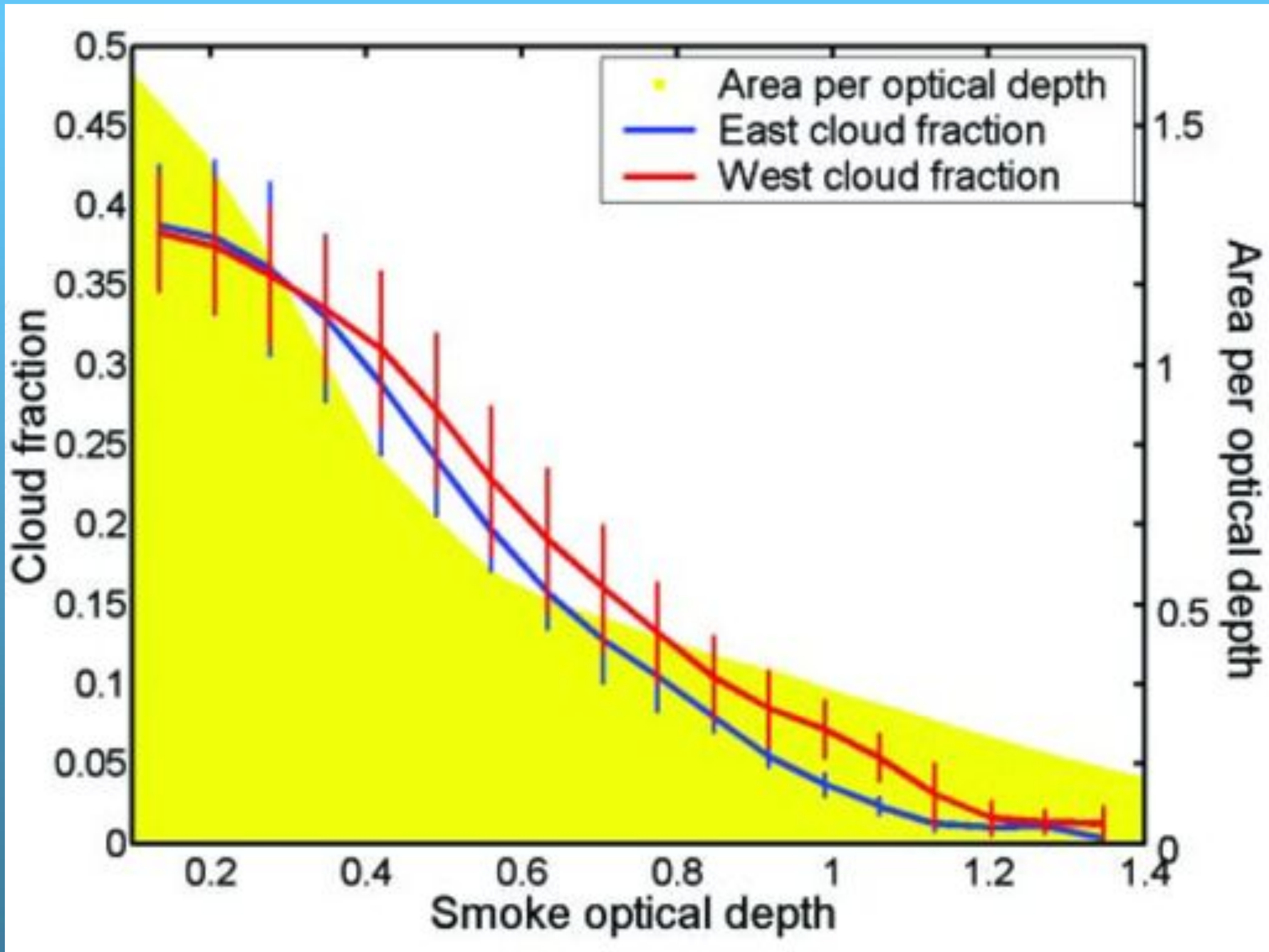
Because of large uncertainties on both the amount of absorbing carbon in the atmosphere, and its absorption efficiency, this forcing is still highly uncertain!!

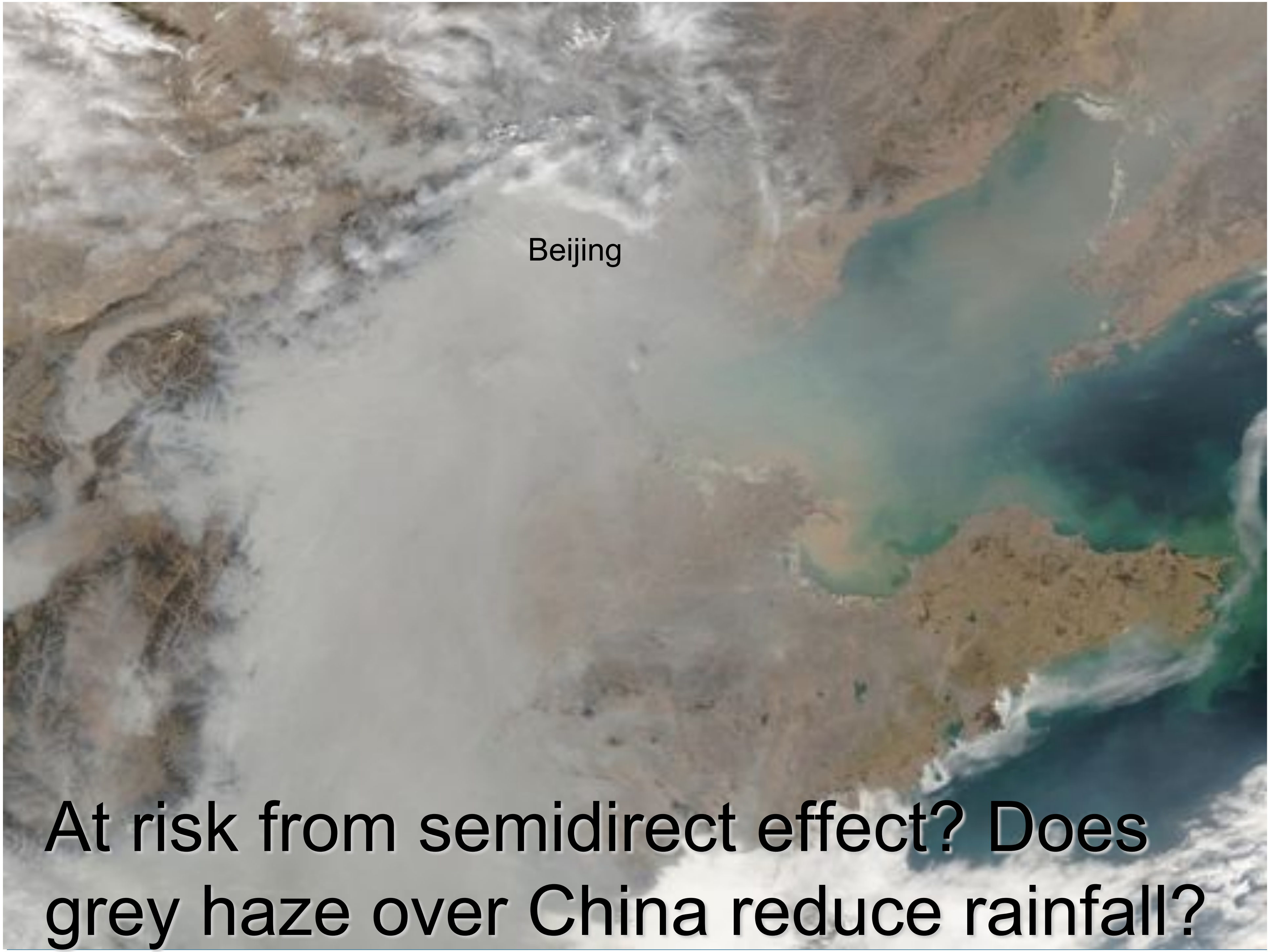
Semidirect Effect:
Cloud suppression by Smoke in the Amazon
(Koren et al., Science 2004)



Cloud suppression by Smoke in the Amazon

(Koren et al., Science 2004)




Beijing
At risk from semidirect effect? Does grey haze over China reduce rainfall?

Indirect Aerosol Effects

- **Each cloud droplet needs a "seed" or nucleus to be able to form: "Cloud Condensation Nucleus" (CCN)**
- **For a given cloud, the more CCN in the air, the more droplets**
- **Since the water supply in a cloud is limited: more droplets means smaller droplets**
- **This increases cloud albedo and cloud amount, and changes cloud microphysics and precipitation formation**

To prove that aerosols are needed to form clouds, a movie:

Aerosol Climate Forcing Mechanisms Overview

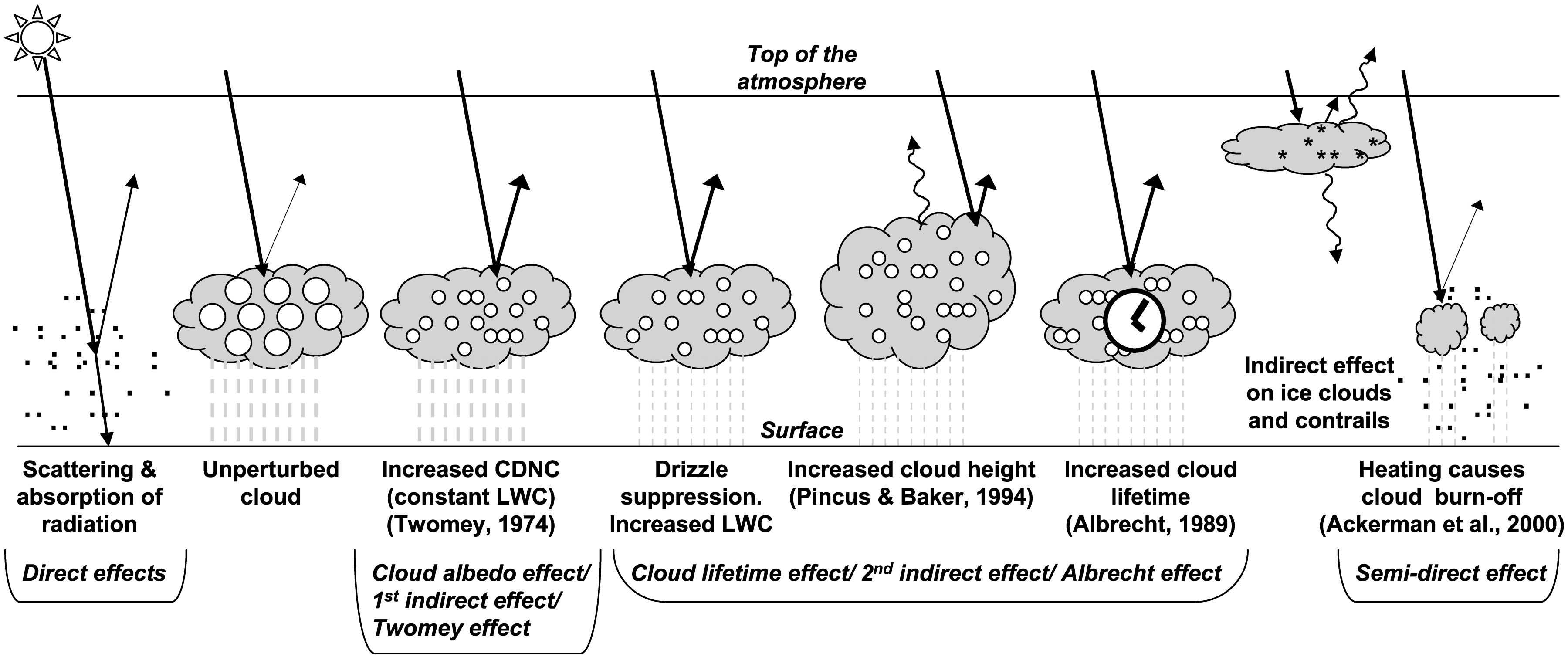


Figure 2.10. *Schematic diagram showing the various radiative mechanisms associated with cloud effects that have been identified as significant in relation to aerosols (modified from Haywood and Boucher, 2000). The small black dots represent aerosol particles; the larger open circles cloud droplets. Straight lines represent the incident and reflected solar radiation, and wavy lines represent terrestrial radiation. The filled white circles indicate cloud droplet number concentration (CDNC). The unperturbed cloud contains larger cloud drops as only natural aerosols are available as cloud condensation nuclei, while the perturbed cloud contains a greater number of smaller cloud drops as both natural and anthropogenic aerosols are available as cloud condensation nuclei (CCN). The vertical grey dashes represent rainfall, and LWC refers to the liquid water content.*

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- Aerosol sources and distributions
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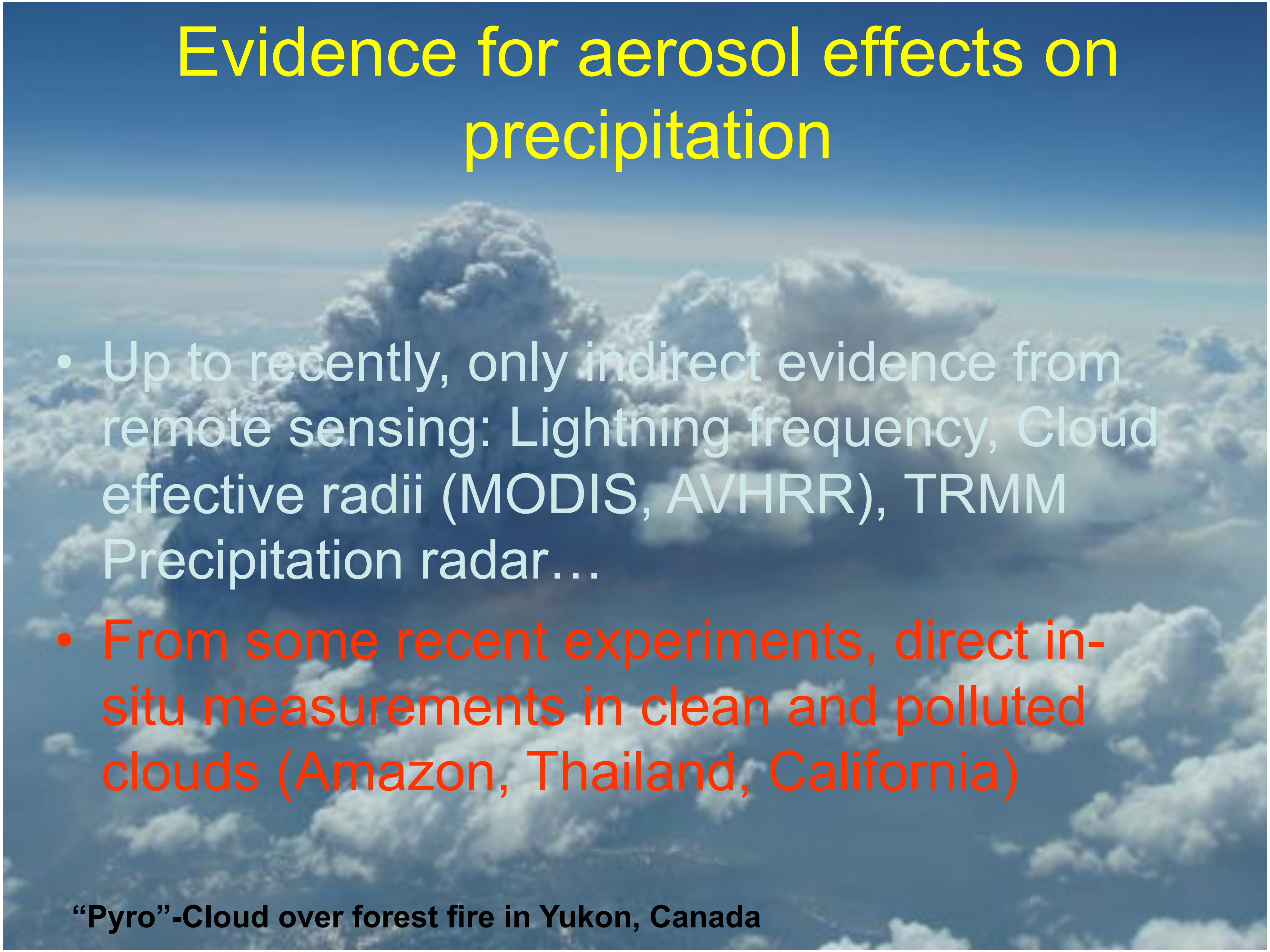
- **Aerosol precipitation interactions**

What is the role of aerosols in determining if clouds are like this:

Evidence for aerosol effects on precipitation

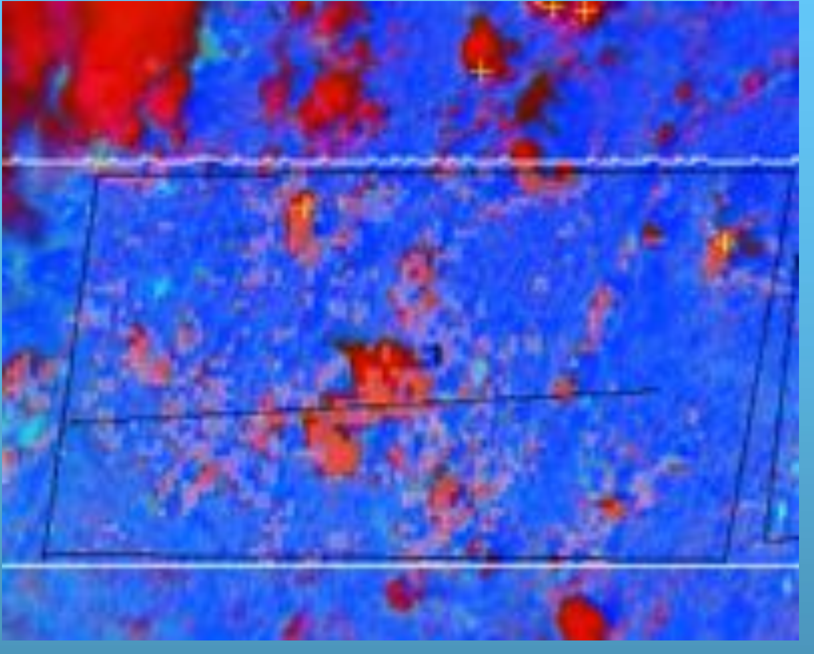
- Up to recently, only indirect evidence from remote sensing: Lightning frequency, Cloud effective radii (MODIS, AVHRR), TRMM Precipitation radar…
- From some recent experiments, direct in-situ measurements in clean and polluted clouds (Amazon, Thailand, California)

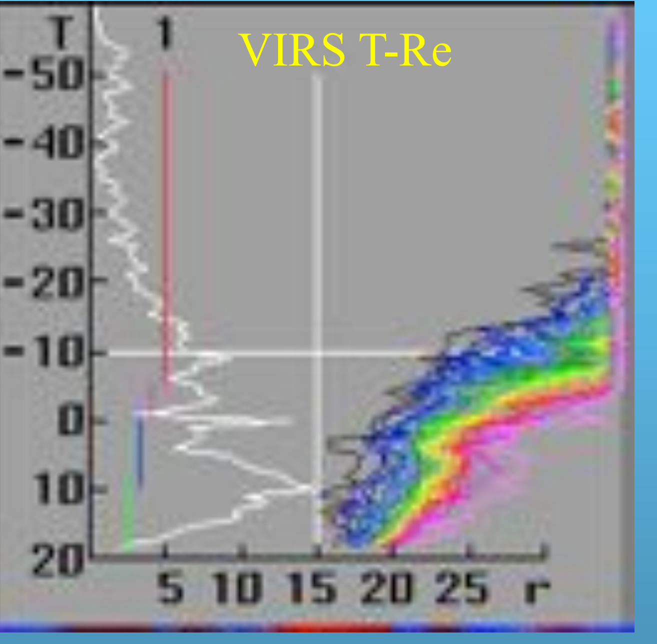


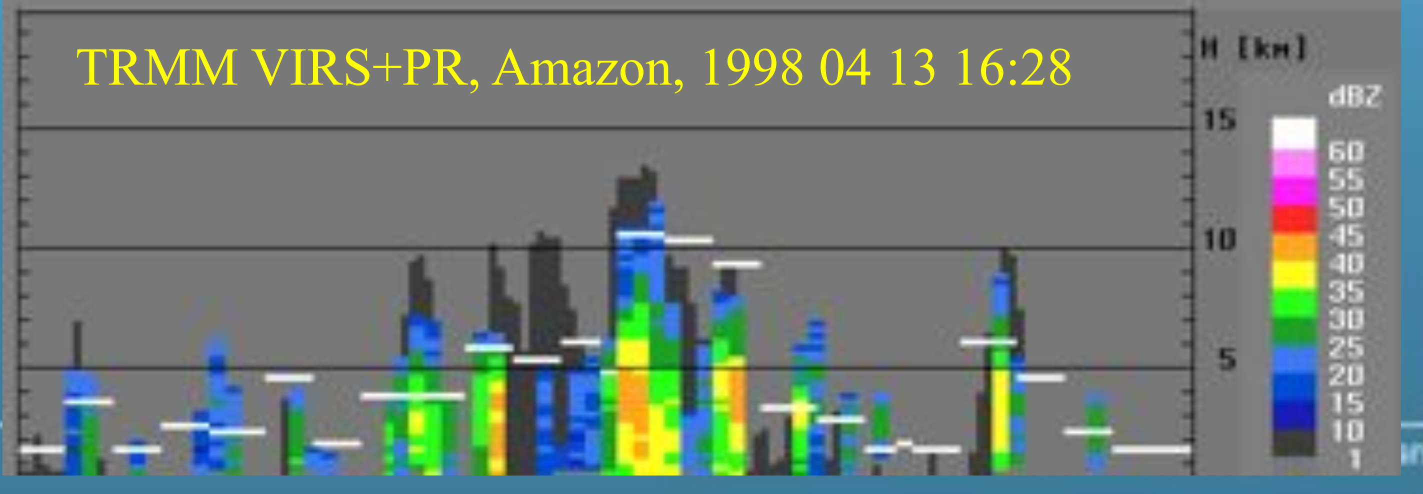
“Pyro”-Cloud over forest fire in Yukon, Canada

The “Green Ocean”: Maritime clouds over the Amazon

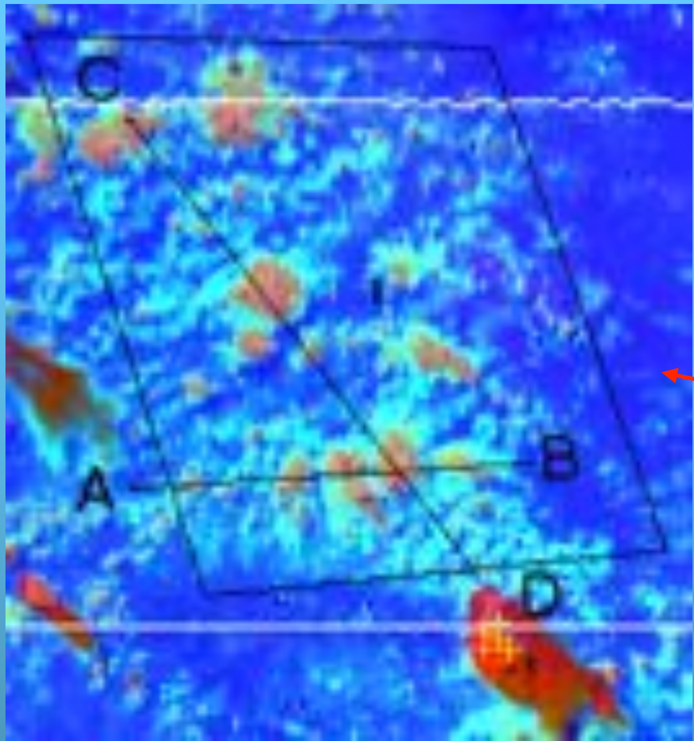
April - the wet and clean time of year: Note the shallow precipitating clouds, extensive warm rainout, glaciation at T>-10°C, and few lightning events

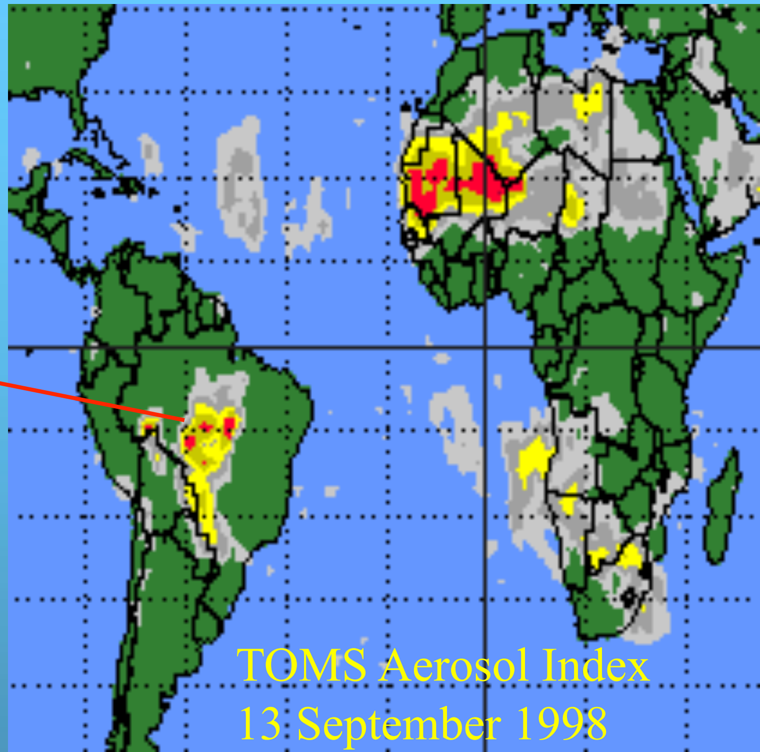






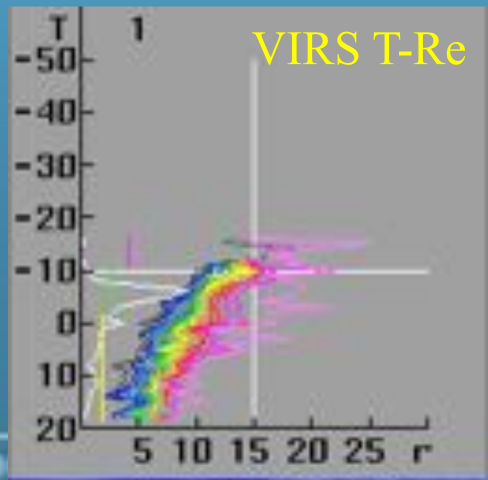
The "Green Ocean" turns dry: Smoky clouds over the Amazon



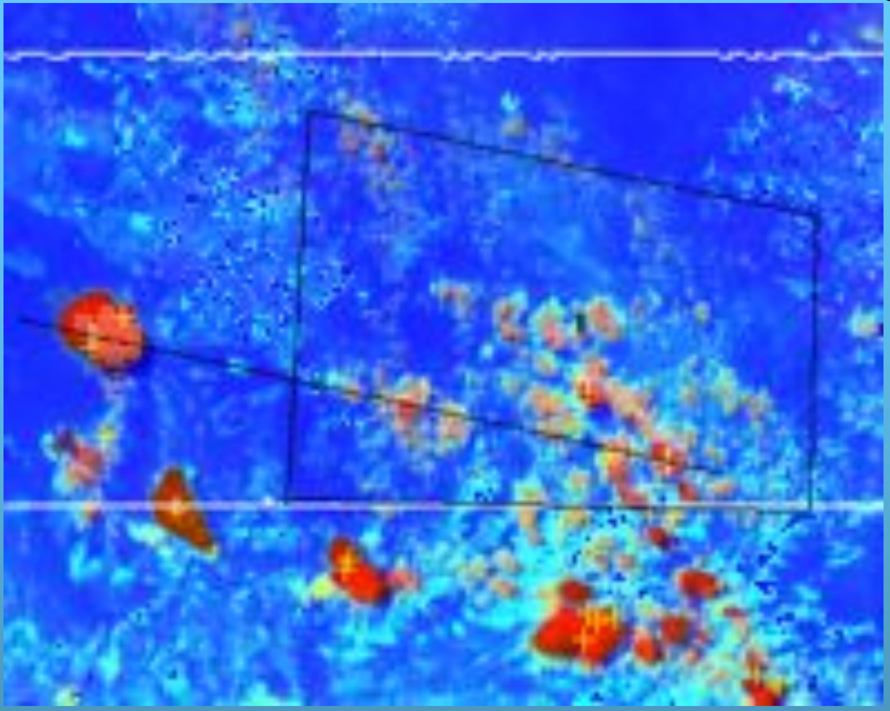


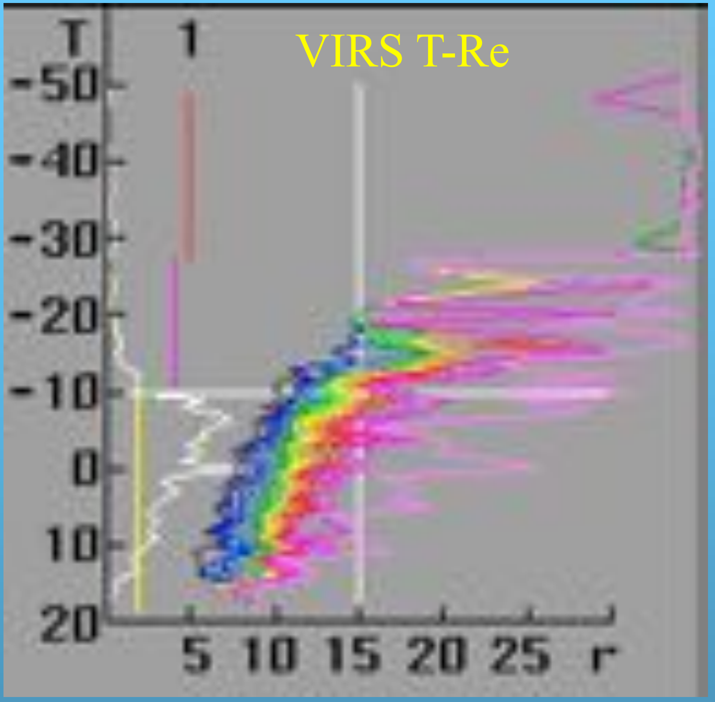
September: The Fire Season

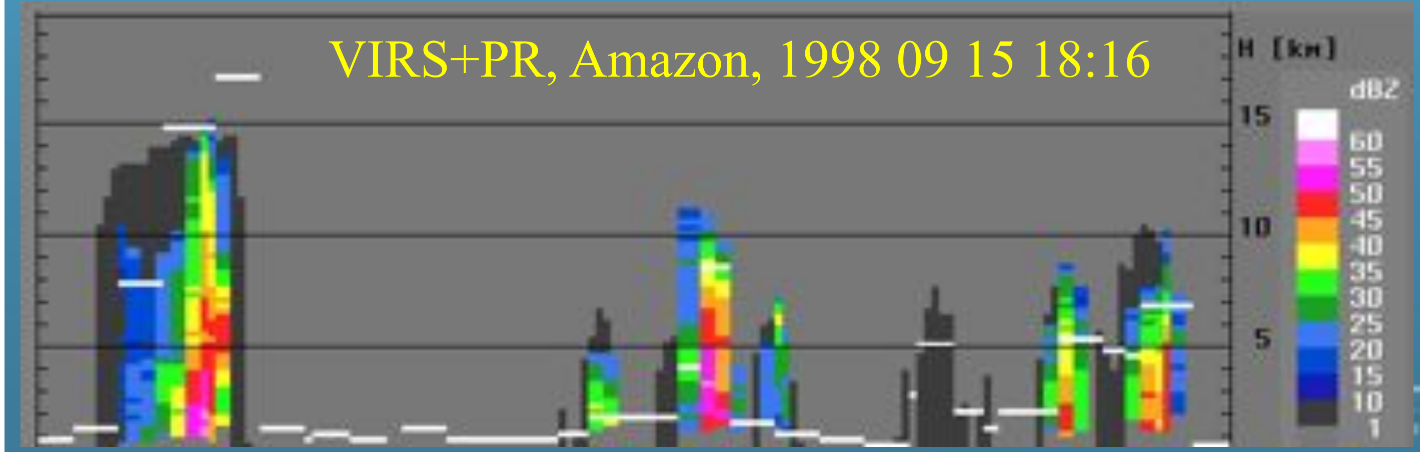
Note that clouds do not precipitate before reaching height of 6.5 km or –12°C isotherm, while containing ample cloud water.

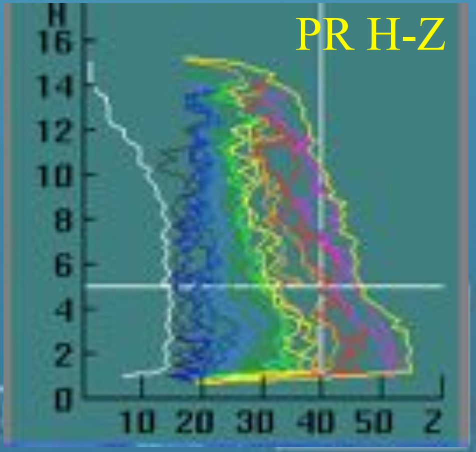


When the “smoky clouds” become Cb, they spark lightning and high Z

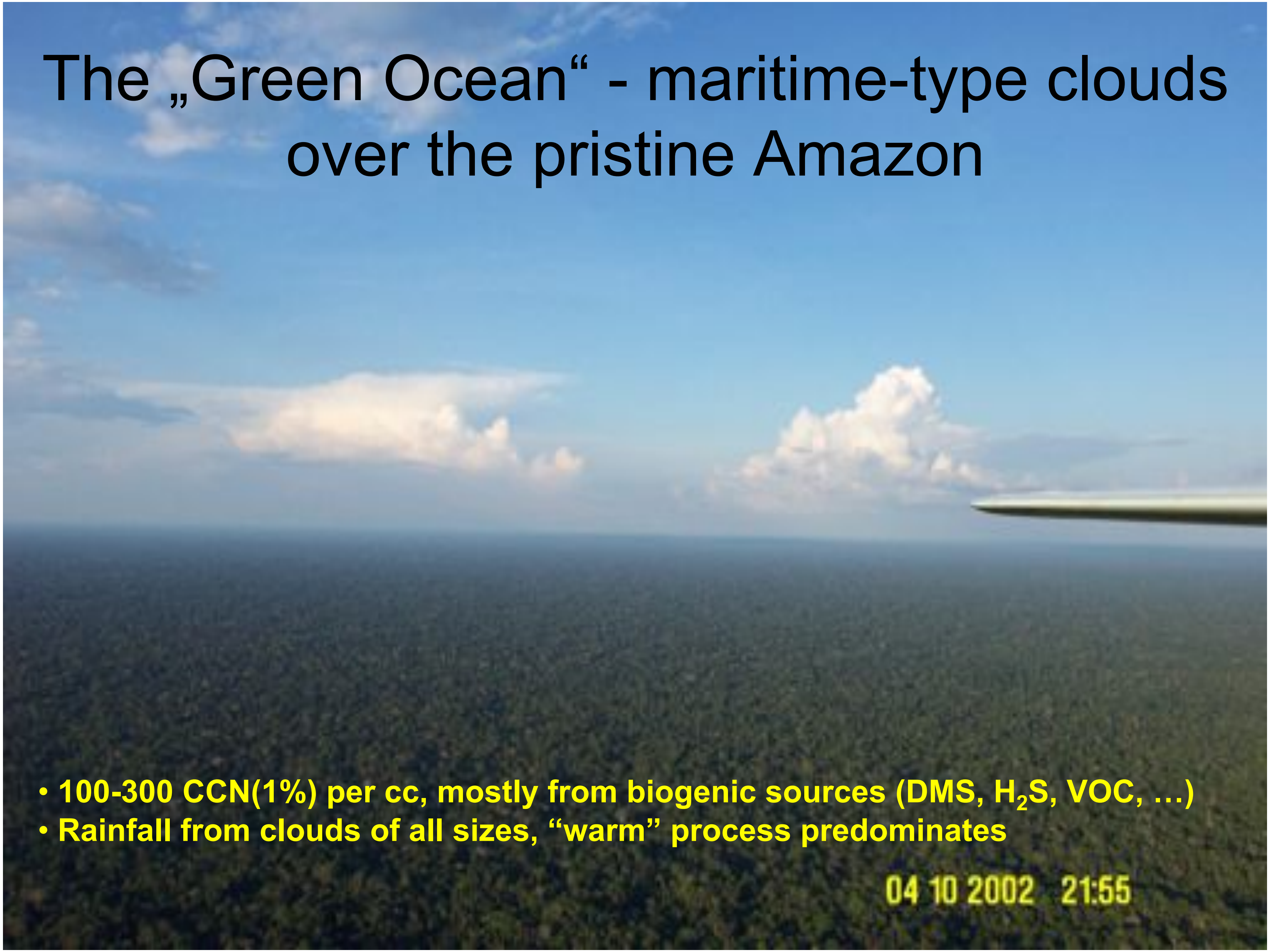




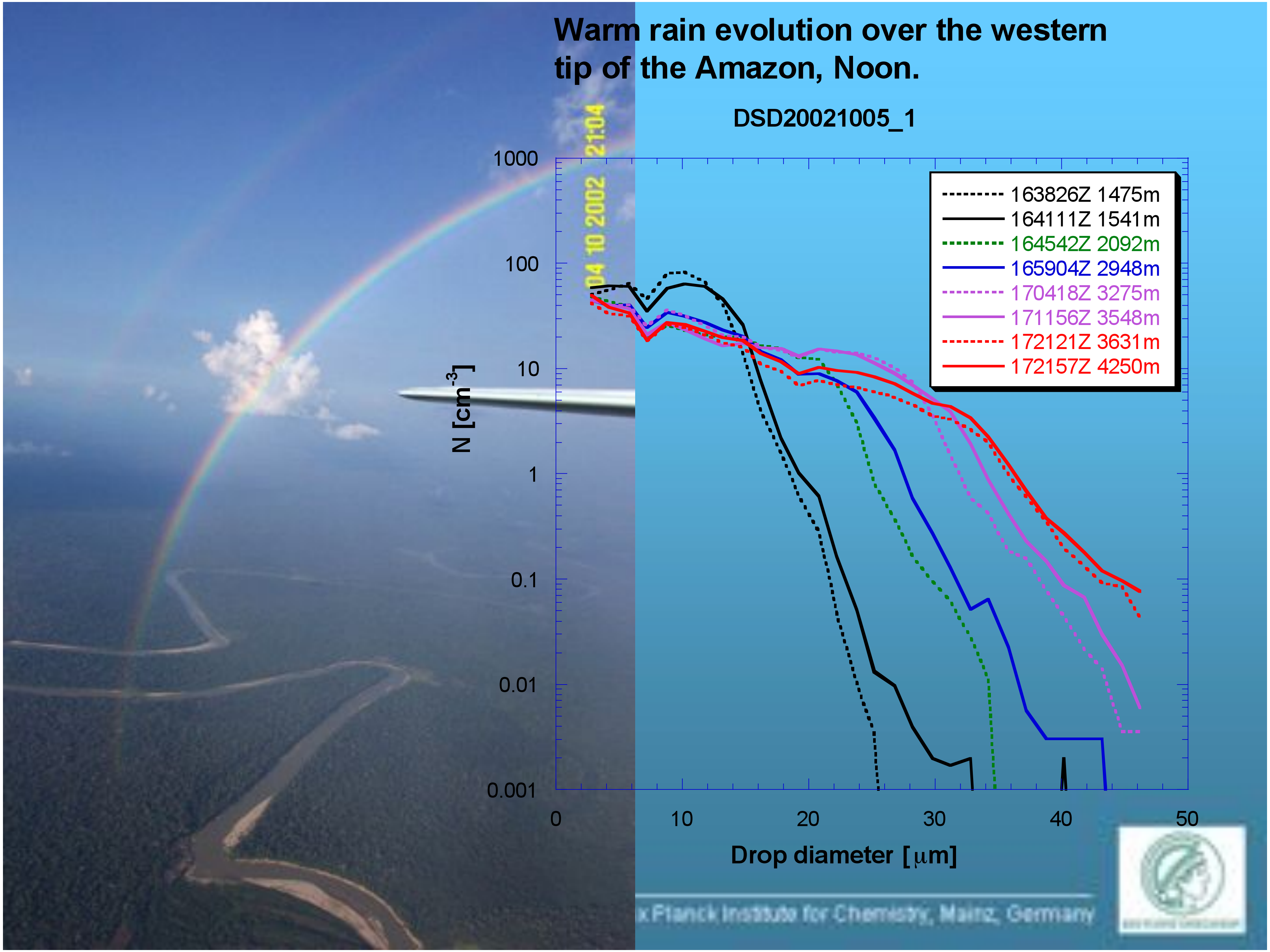




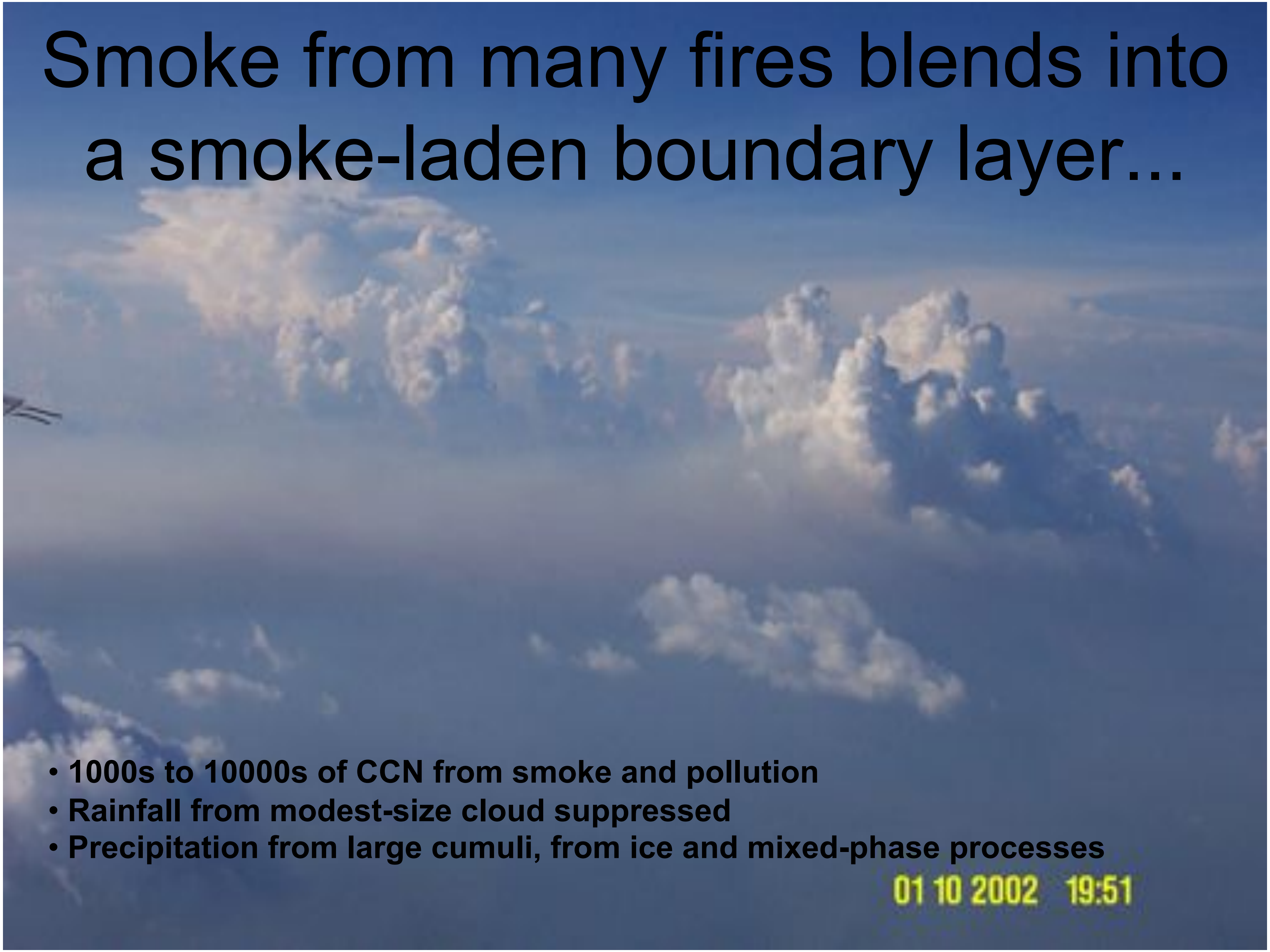
The „Green Ocean“ - maritime-type clouds over the pristine Amazon



- 100-300 CCN(1%) per cc, mostly from biogenic sources (DMS, H_2S, VOC, …)
- Rainfall from clouds of all sizes, “warm” process predominates

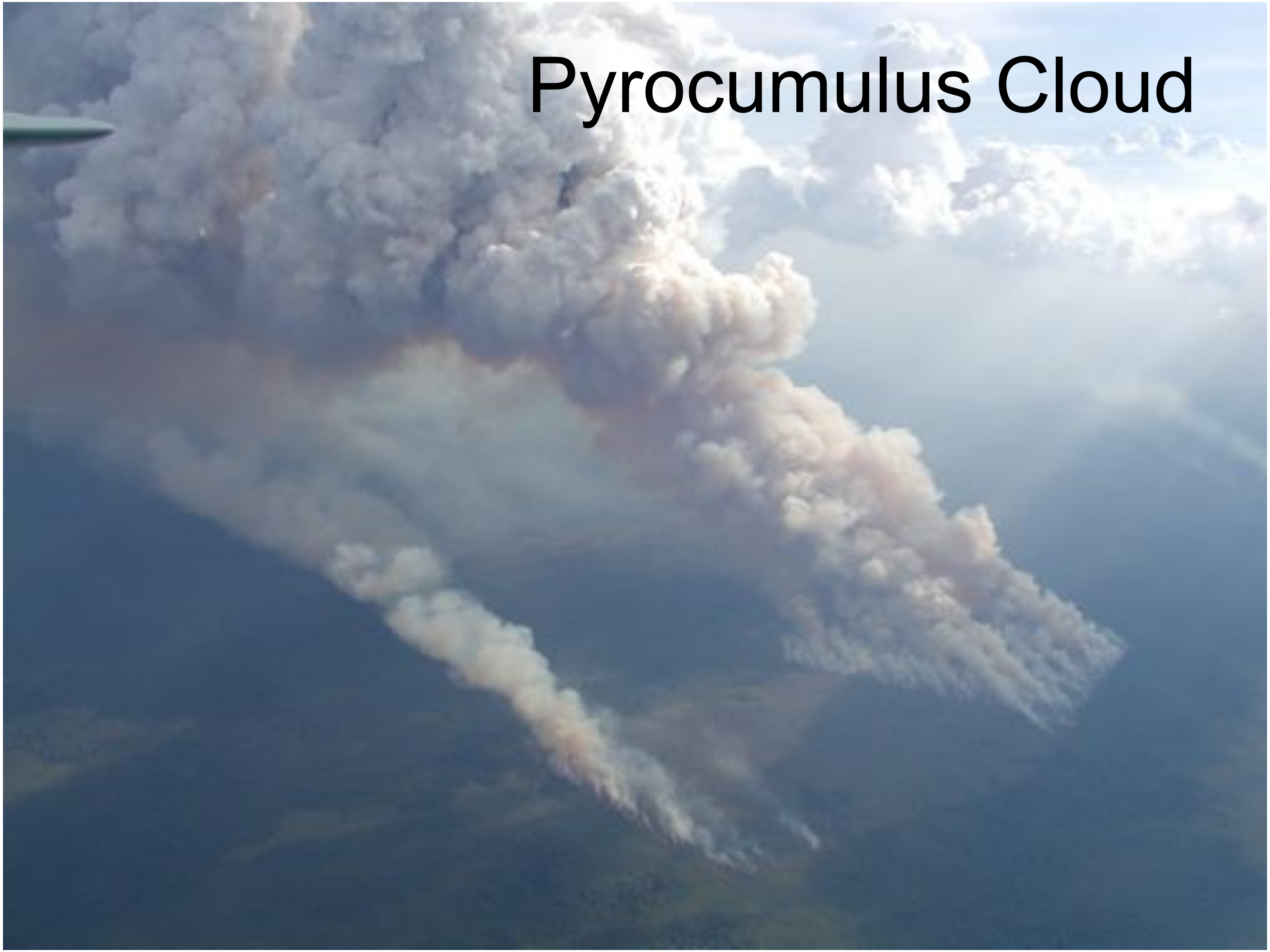

Warm rain evolution over the western tip of the Amazon, Noon.
04 10 2002 21:04
DSD20021005_1
N [cm^-3]
1000
100
10
1
0.1
0.01
0.001
0
10
20
30
40
50
Drop diameter [μm]
163826Z 1475m
164111Z 1541m
164542Z 2092m
165904Z 2948m
170418Z 3275m
171156Z 3548m
172121Z 3631m
172157Z 4250m
x Planck Institute for Chemistry, Mainz, Germany

Smoke from many fires blends into a smoke-laden boundary layer...

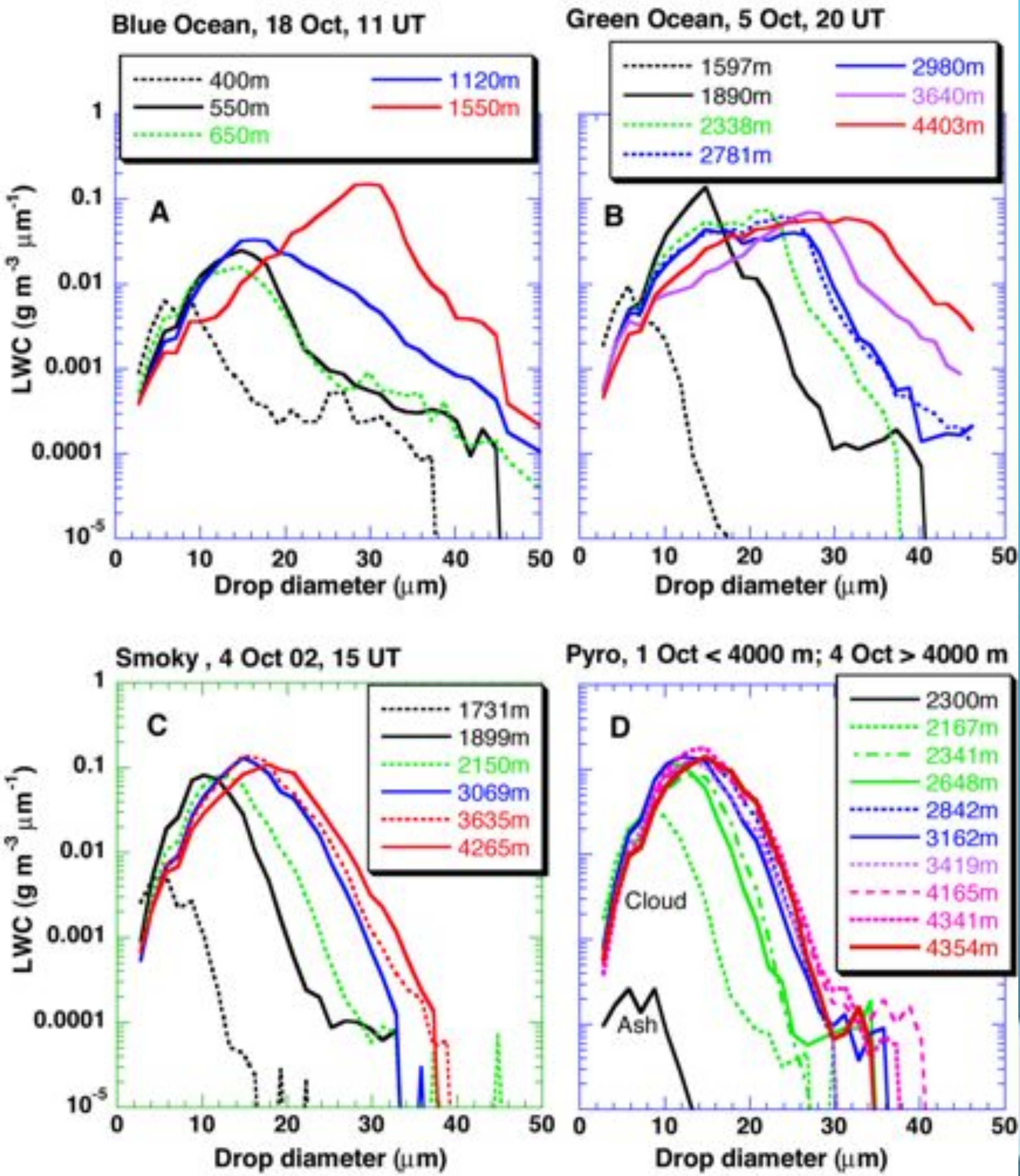


- **1000s to 10000s of CCN from smoke and pollution**
- **Rainfall from modest-size cloud suppressed**
- **Precipitation from large cumuli, from ice and mixed-phase processes**

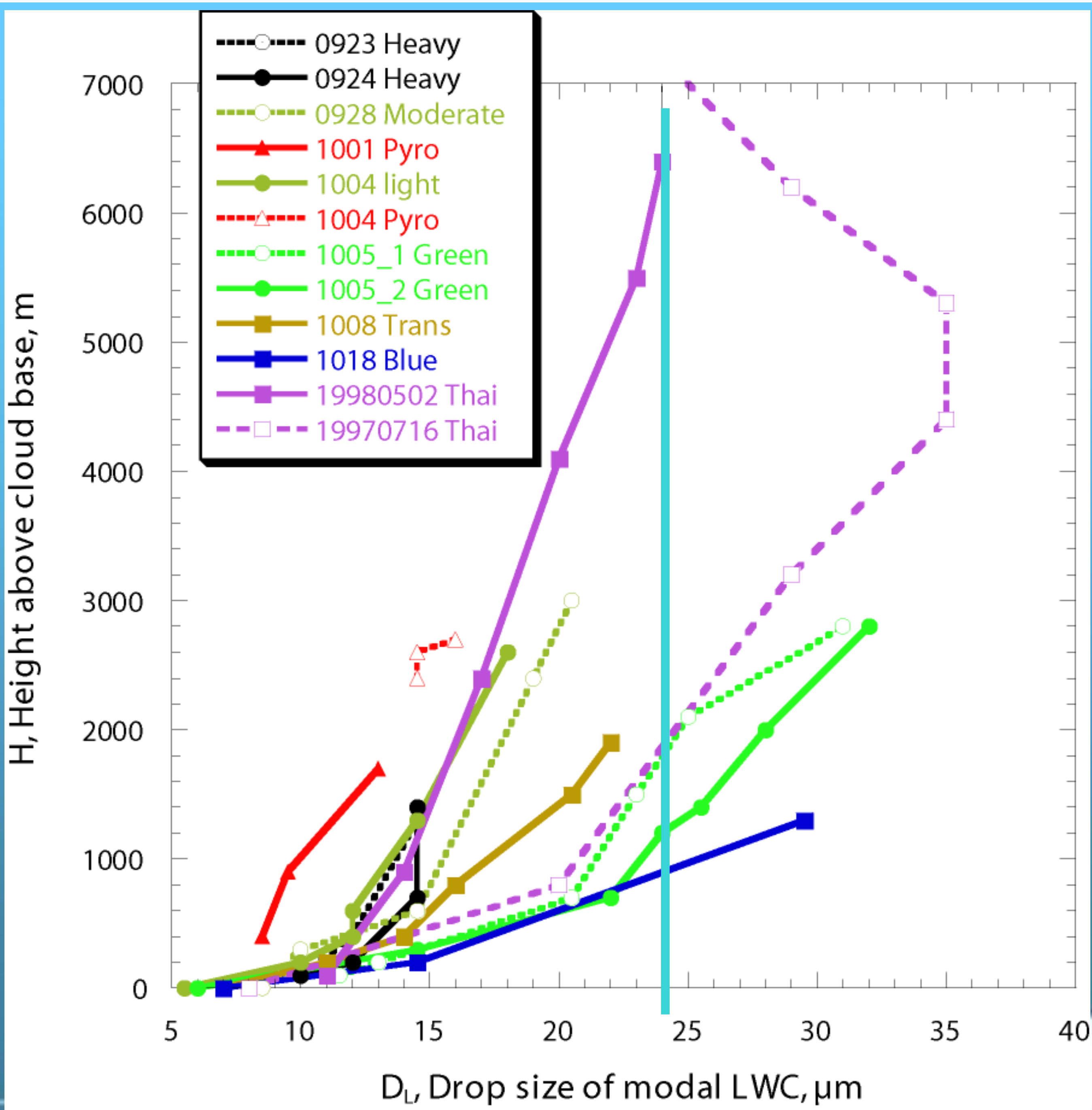
Pyrocumulus Cloud



Addition of pyrogenic CCN has pronounced impact on cloud droplet size spectra!



Aerosol pollution increases the height where cloud drops grow big enough to grow by coagulation and form "warm" rain



Andreae et al., SCIENCE, 303(2004)1337-1342

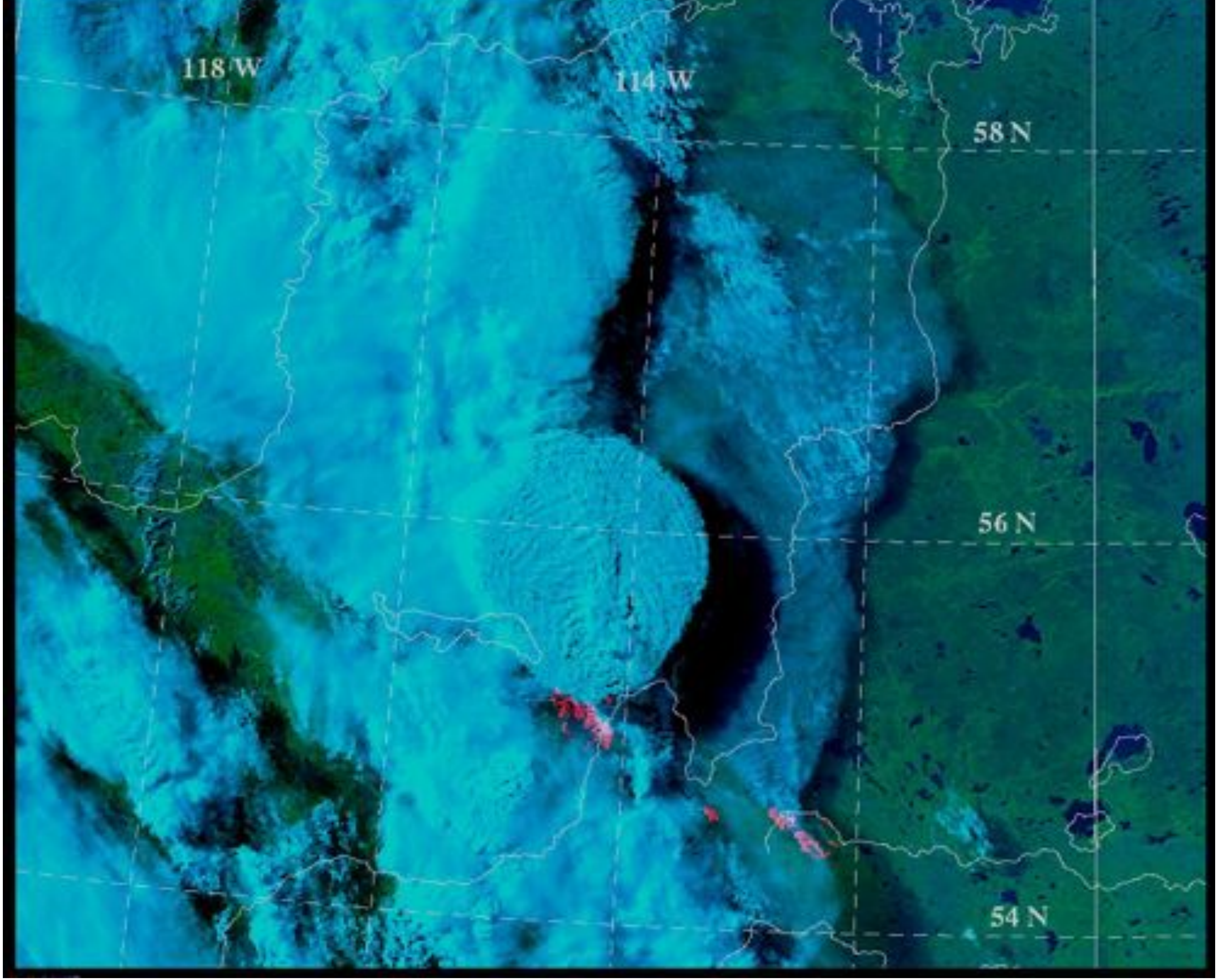
Deforestation fire, Brazil: a “Smoking Cloud”

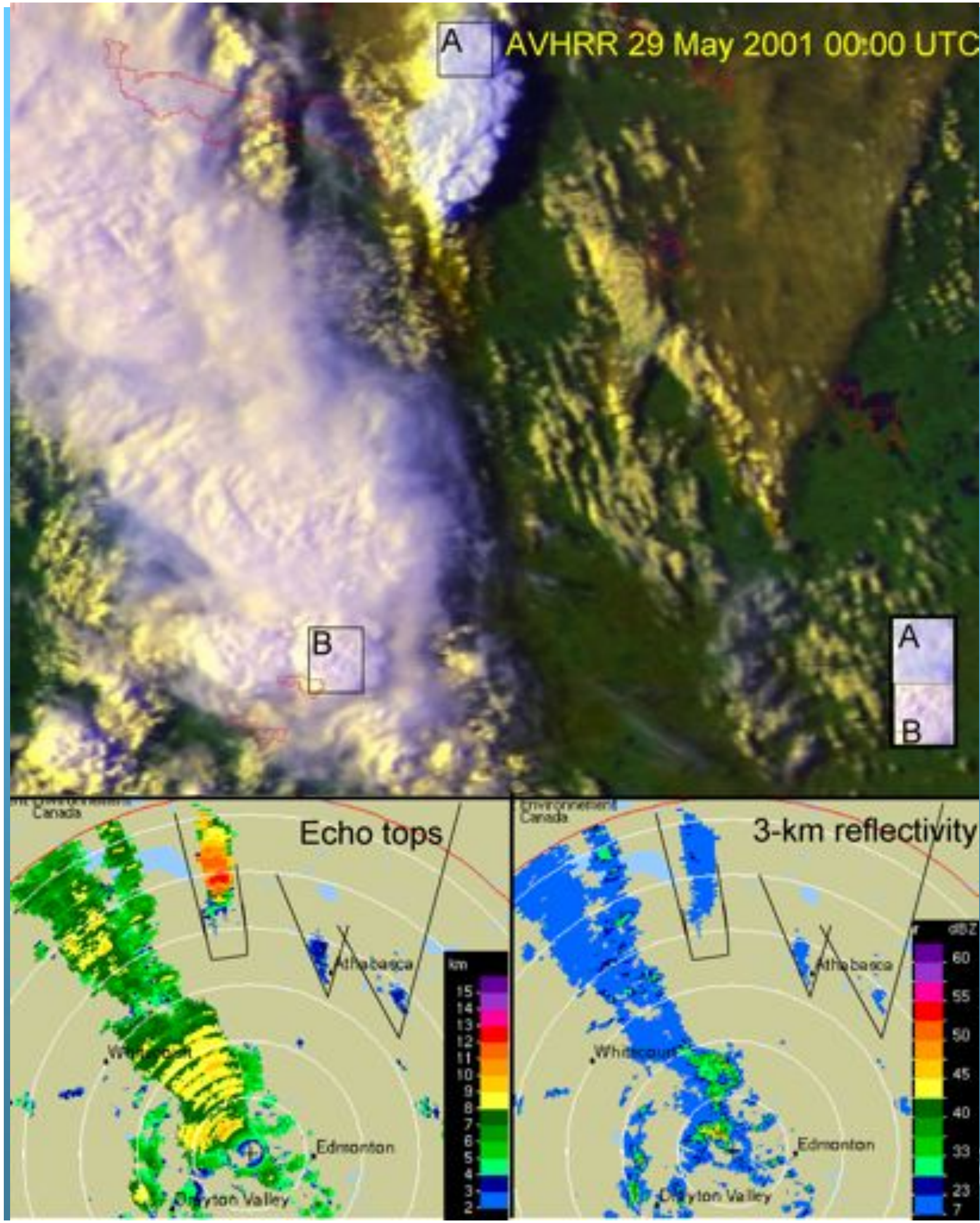

Smoke detraining from cloud top

Chisholm – The Perfect Storm

Total heat release equivalent to ~1200 Hiroshima bombs

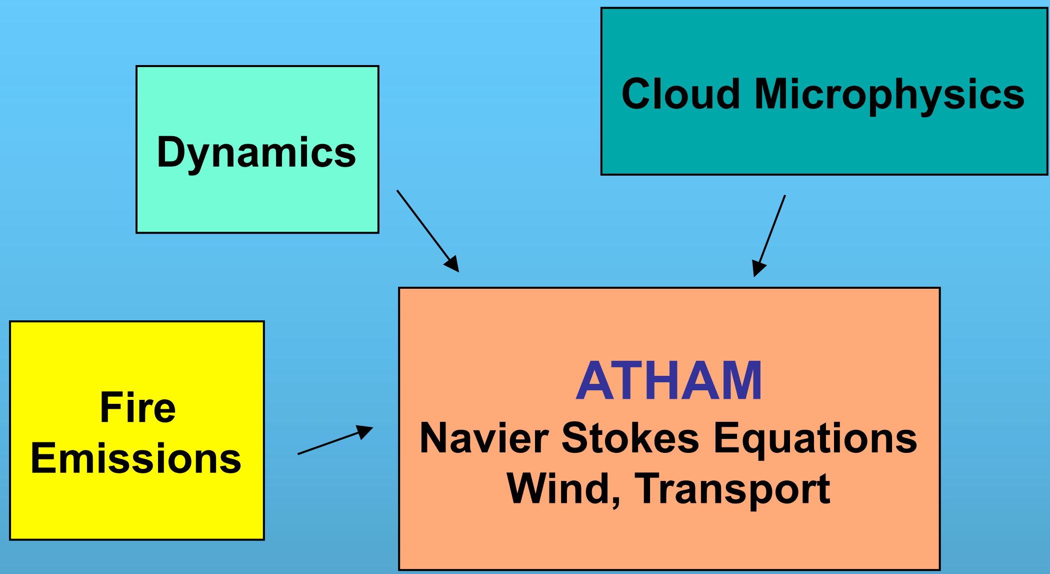
Radar-acp-2006-0235-mv1.avi


118 W
114 W
58 N
56 N
54 N



In spite of the very high echo tops, the Chisholm pyro-cumulus shows weak reflectivity: little precipitation

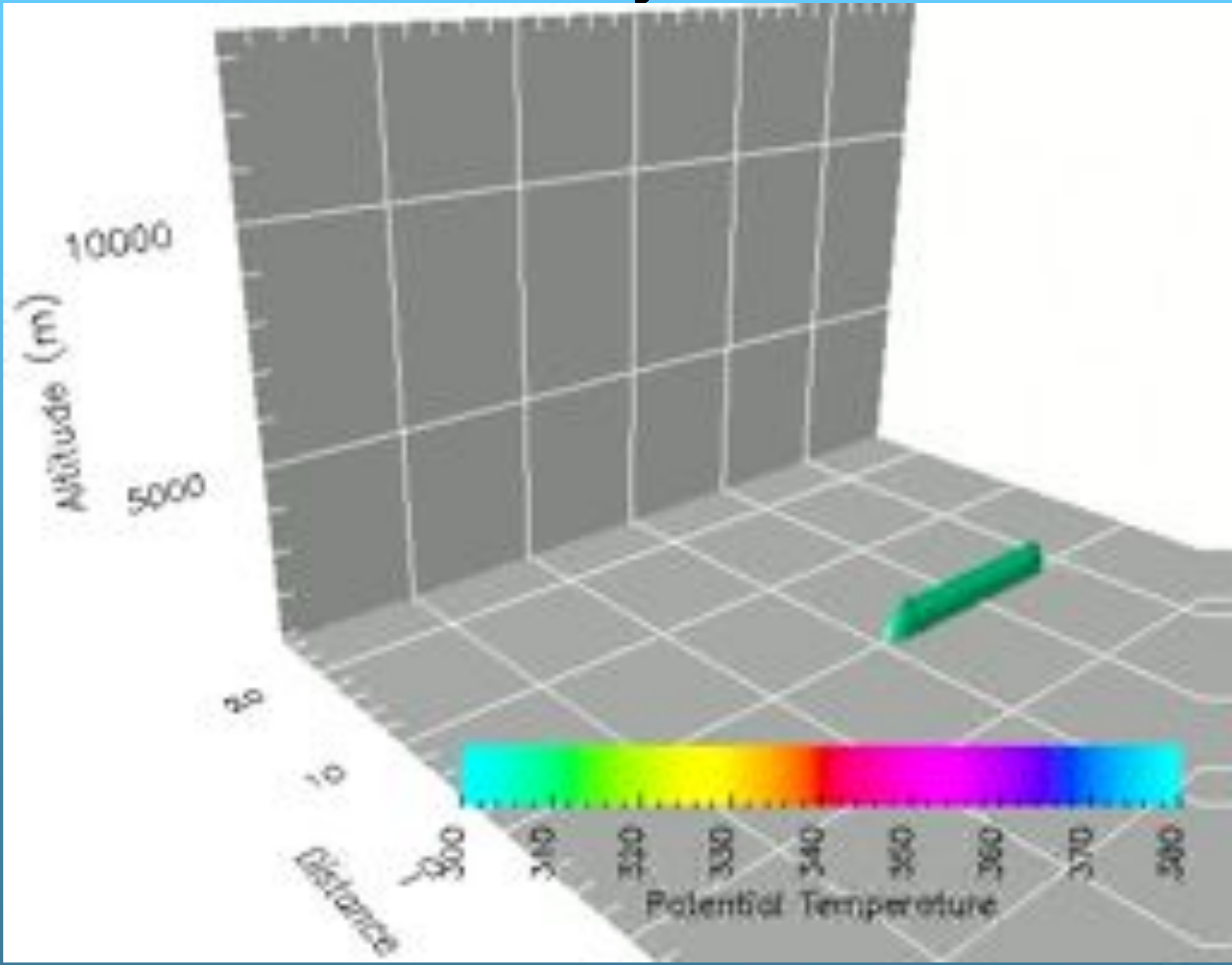
Active tracer high-resolution atmospheric model (ATHAM)



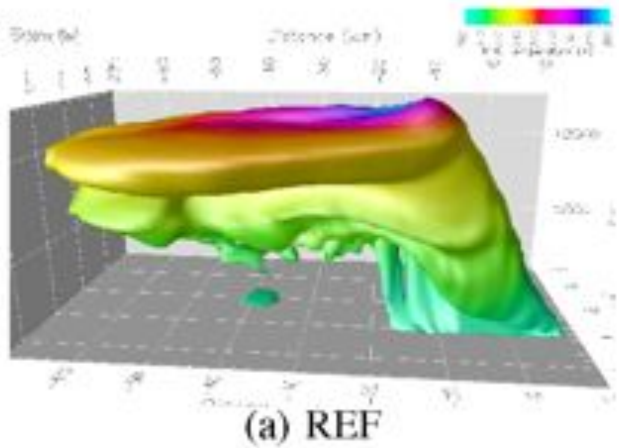
Biomass burned: 10 kg/m^2
→ **total energy flux: 530 kJ/m^2/s** (50% contributes to convection)
→ **total sensible heat flux: 265 kJ/m^2/s** (= 2000 GW)
Water Vapor Emission: 26 g H_2O/m^2/s (55% from combustion)
→ **Potential Latent Energy Flux: 74 kJ/m^2/s**
Aerosol Emission: 400 mg/m^2/s

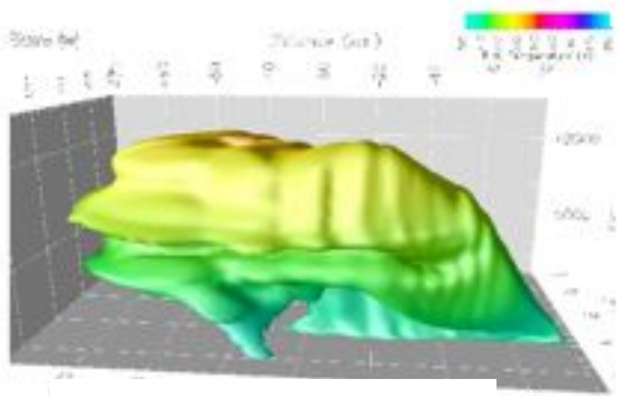
Model domain: x = y = 60 km; z = 28 km;
Grid resolution: dx = 500 m; dy = 100 m; dz = 50 m (at fire)
Size of fire front: x = 15 km; y = 500 m, area: 7.5 km^2
Rate of spread: 1.5 m/s
Time Step: 1 s

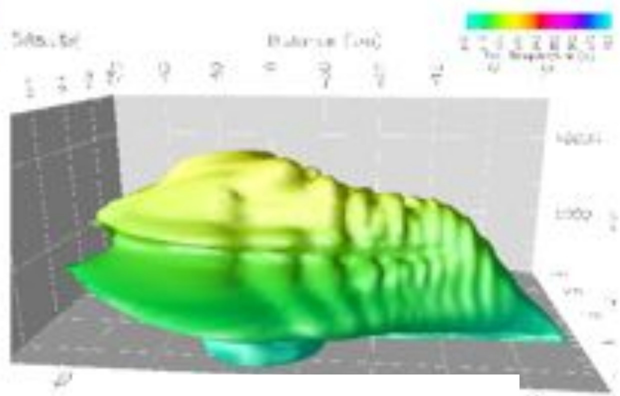
The Chisholm Pyro-Cb modeled

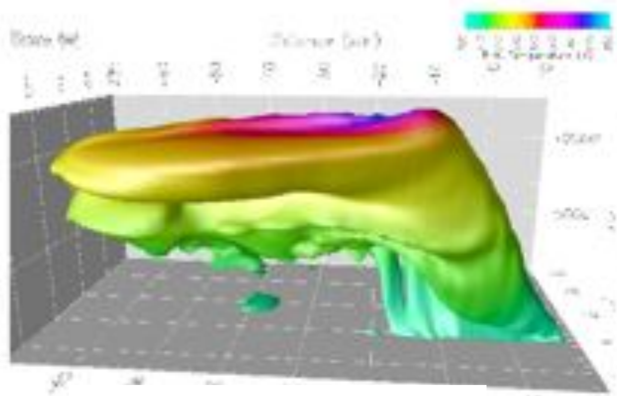


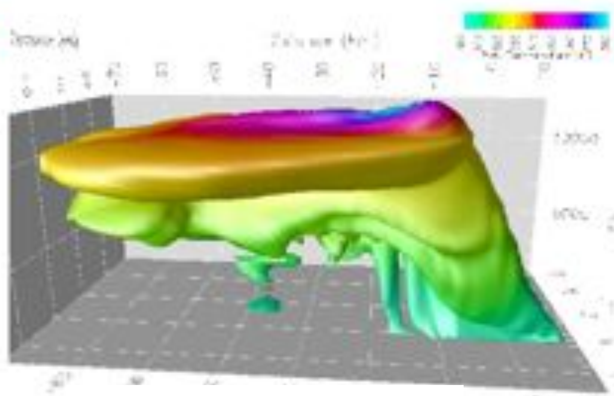
Sensitivity of pyroCb development to changing input of heat, water vapor and CCN from the fire




Sensible Heat 50%


Sensible Heat 25%


No H2O from Fire


CCN = 200 cm-3

Luderer et al., ACP (2006)

Germany

Stratospheric injection requires heat input from fire:

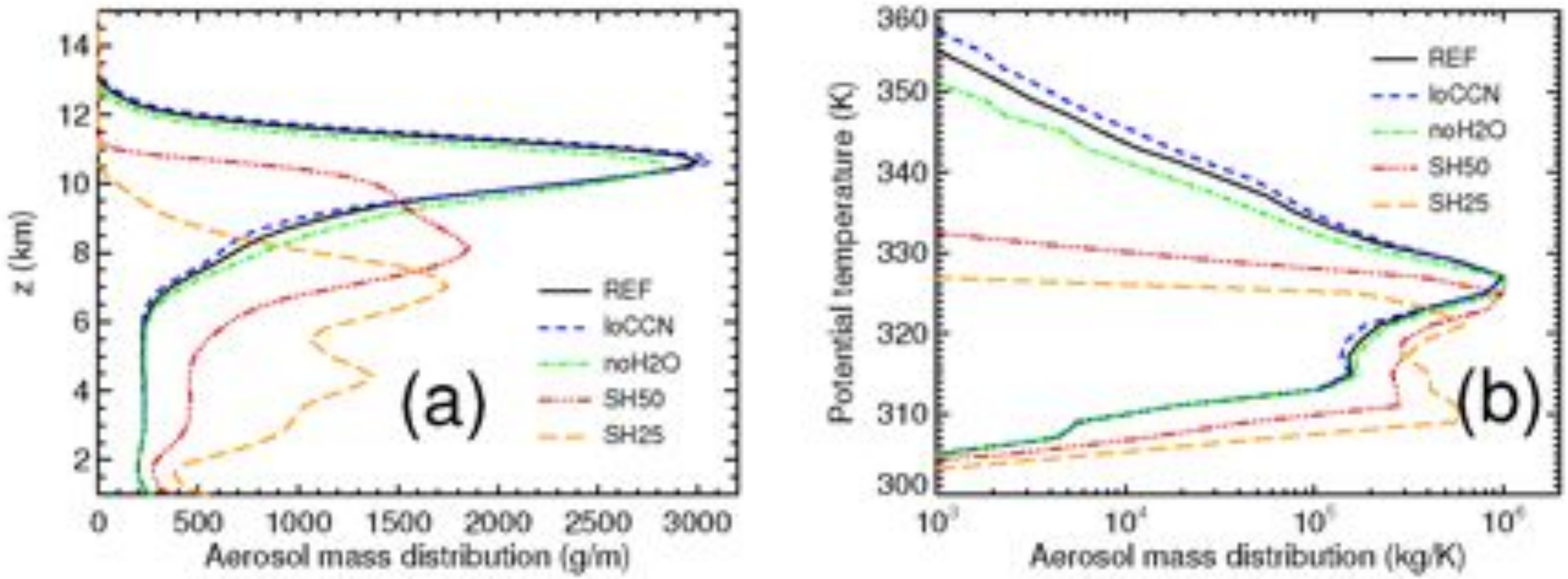


Fig. 4. Vertical distribution of aerosol mass as a function of altitude (a) and as a function of potential temperature (b) after 40 min integration time. The 2 PVU tropopause is located at 332 K/11.0 km.

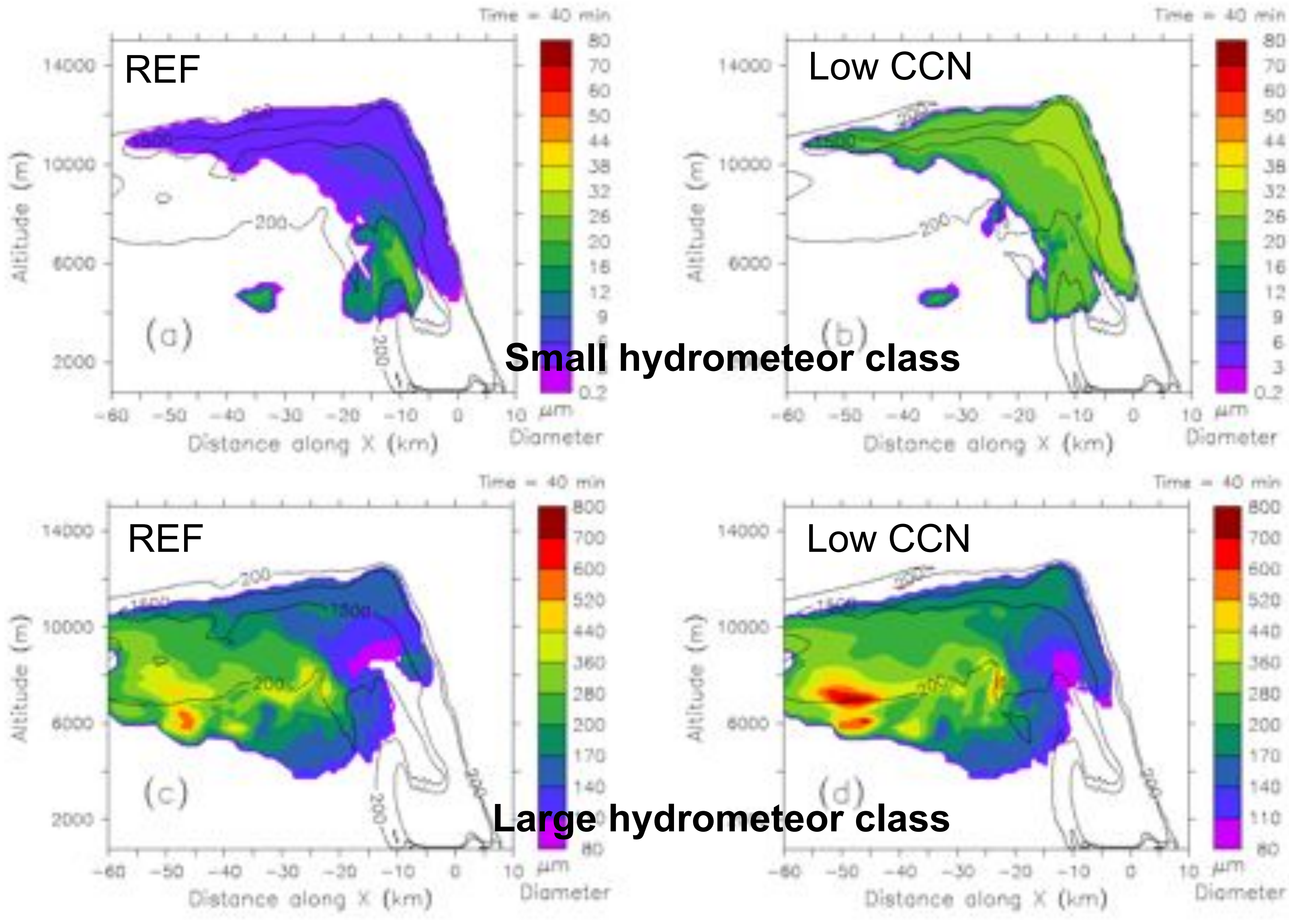
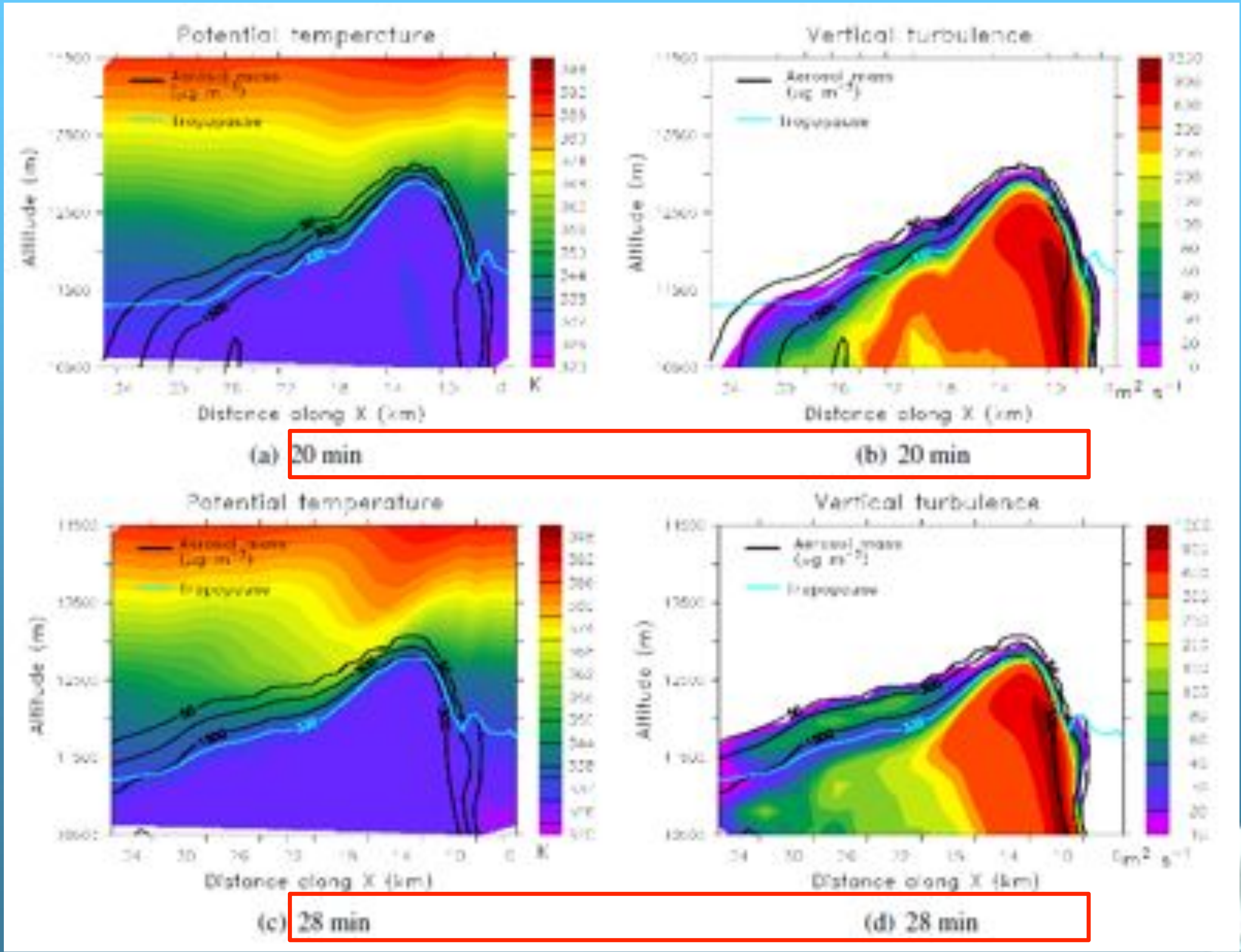
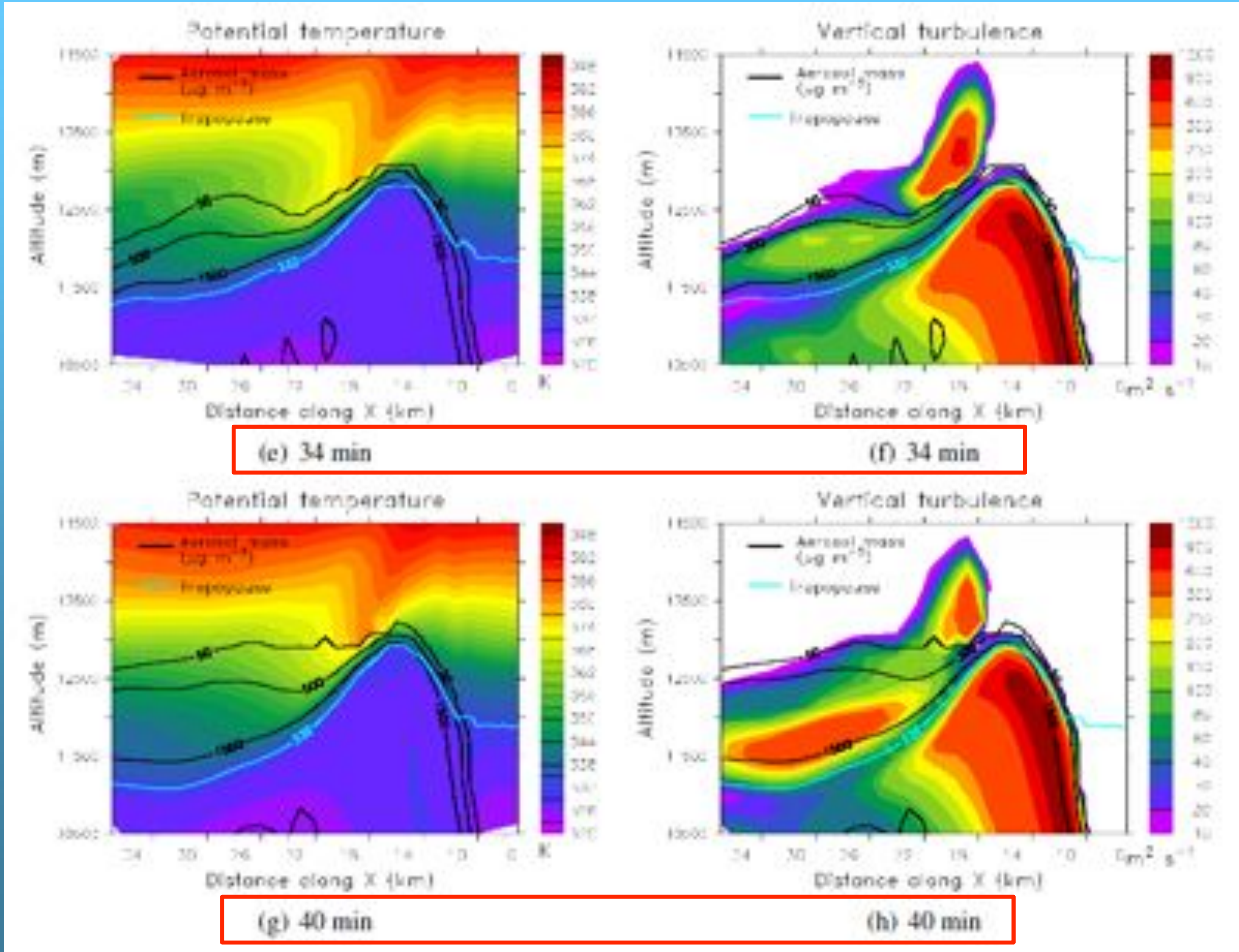


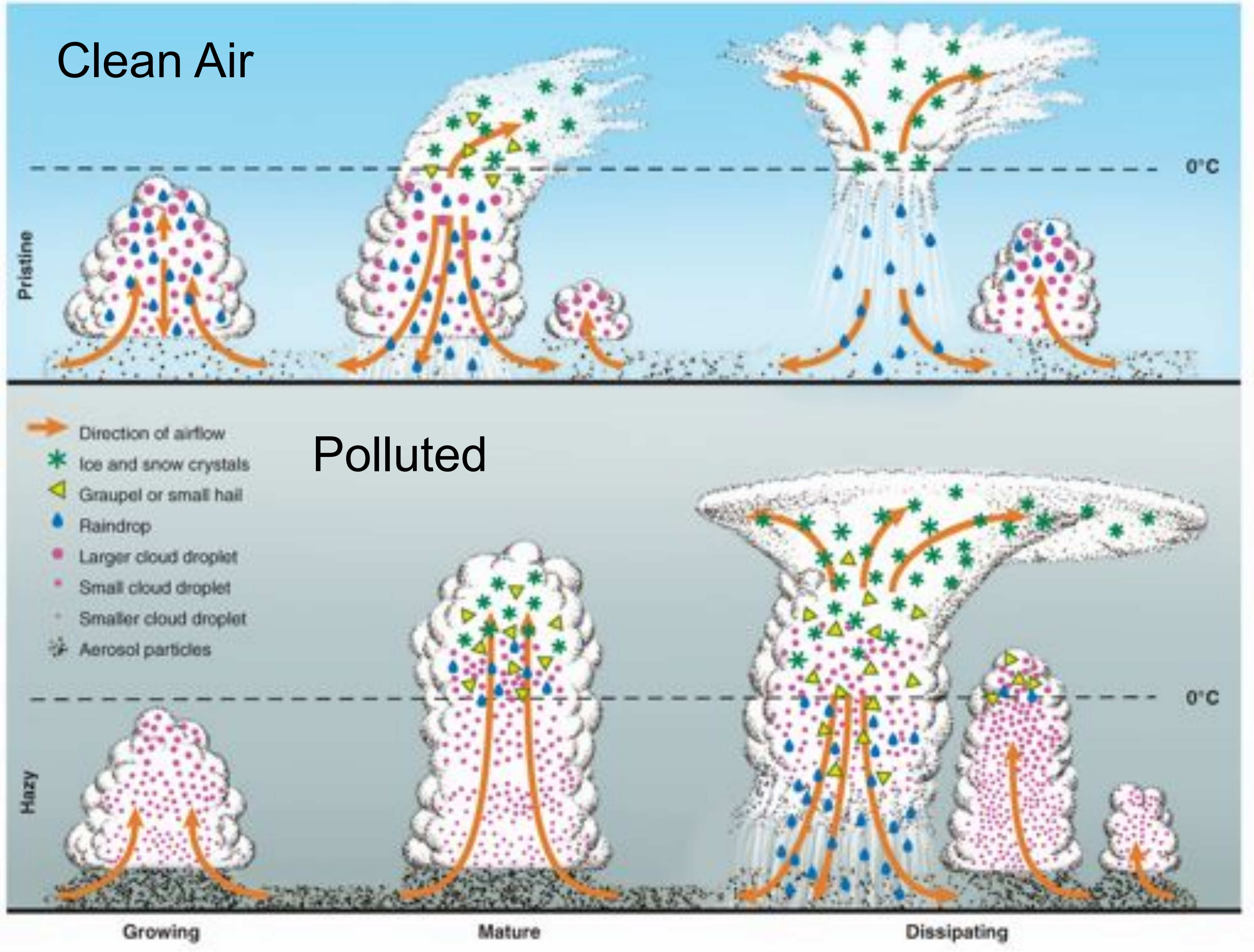
Fig. 6. Sensitivity of hydrometeor volume mean diameter (VMD) to CCN abundance. Diameters are depicted for (a) small size classes (cloud droplets and ice crystals) in REF, (b) small size classes in loCCN, (c) large size classes (rain drops and graupel) in REF, (d) large size class in loCCN. Contour lines give aerosol concentration in $\mu g\ m^{-3}$.

Wave-breaking at Tropopause…

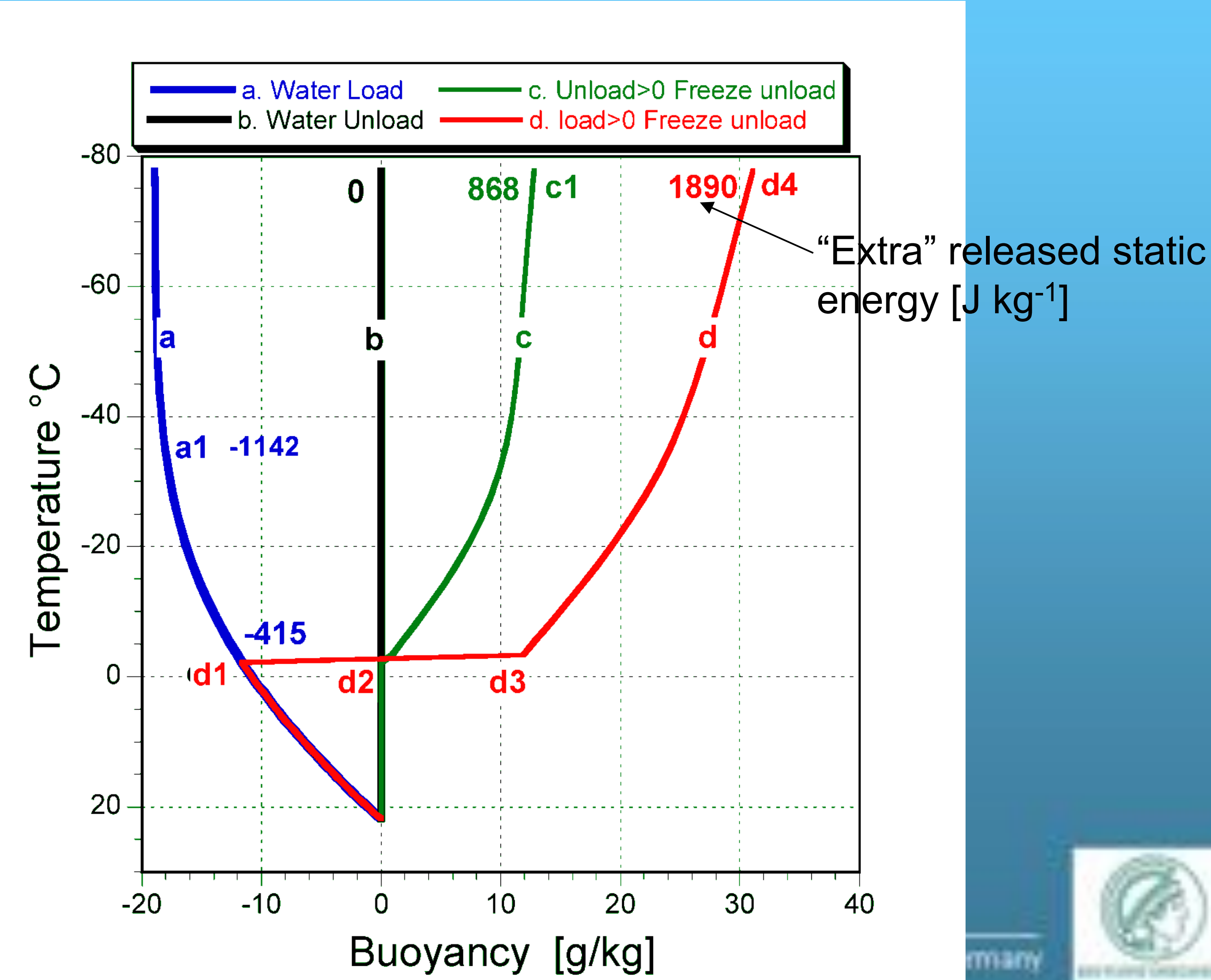


…injects smoke into Stratosphere

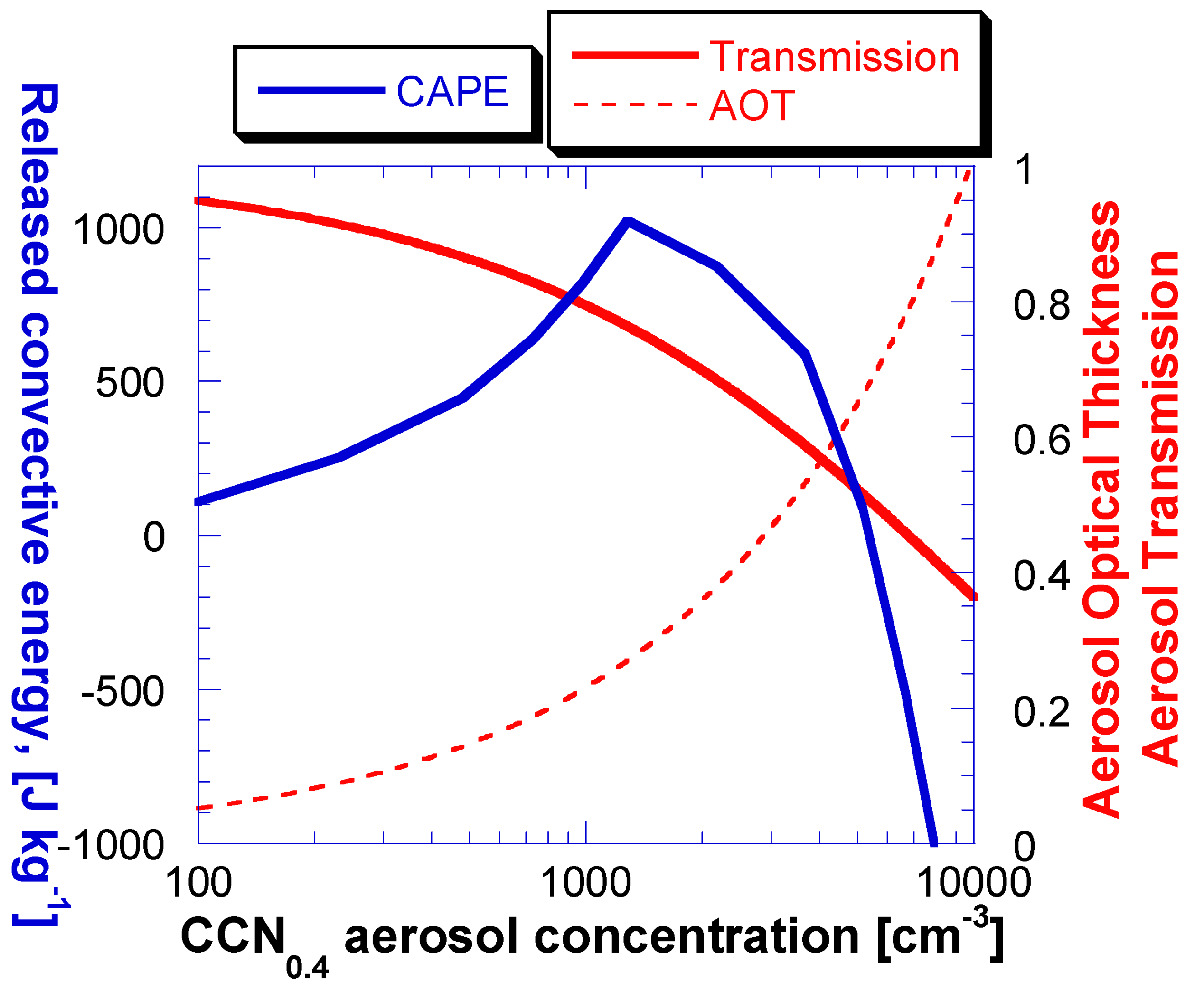



Clean Air
0°C
Pristine
Polluted
Direction of airflow
Ice and snow crystals
Graupel or small hail
Raindrop
Larger cloud droplet
Small cloud droplet
Smaller cloud droplet
Aerosol particles
0°C
Hazy
Growing
Mature
Dissipating

Thermodynamic consequences of warm precipitation suppression by aerosols



Radiative and microphysical effects on cloud convection intensity



Observational evidence for non-monotonic response of clouds to aerosols

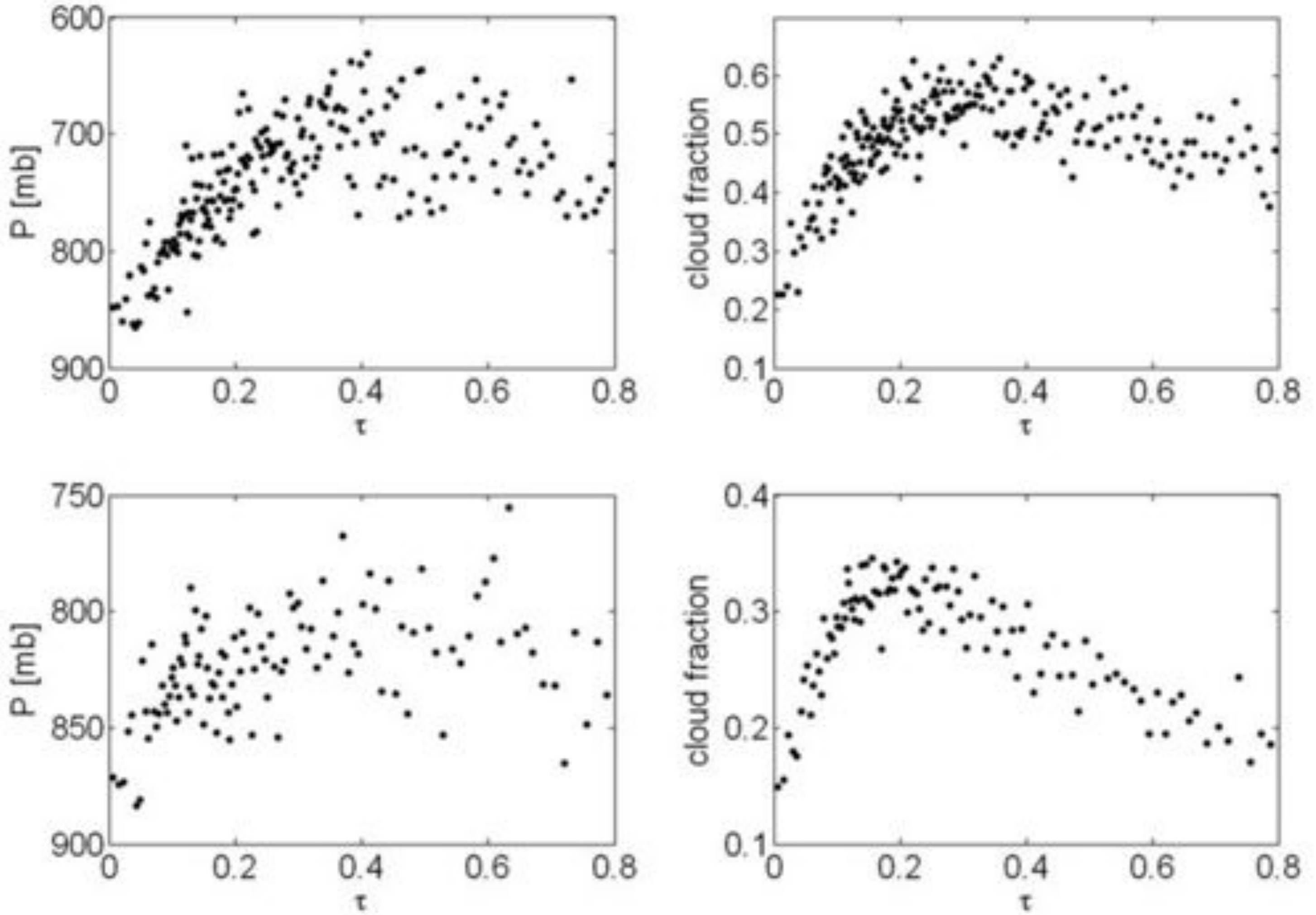


Fig. 2. Relationships between cloud properties and aerosol loading (estimated by τ). (**Left panels**) P versus τ. Lower P may indicate taller convective clouds that reach to higher levels of the atmosphere. (**Right panels**) Cloud fraction versus τ. The upper row shows all data and the lower row shows data restricted to a cloud fraction <0.5.

Koren et al., Science 2008

Summary on aerosol-precipitation interactions

- Aerosols act on convection by radiative and microphysical effects
- Radiative effect is monotonically increasing suppression
- Microphysical effect leads first to invigoration by shift to ice processes, then to weakening by suppressing cold precipitation as well
- This has consequences at all scales, from local (rain intensity, hail, lightning) to global (general circulation)

But, there are many other processes that connect aerosol and precipitation, e.g., …

- In warm clouds, suppression of early precipitation forces increased evaporation, which destabilizes the convective layer, and thus invigorates convection and precipitation.
- Over the ocean, higher windspeeds enhance both aerosol production and evaporation, thus increasing both AOD **and** rainfall
- …

→ We are still a long way from a comprehensive and quantitative understanding of the processes that control the interactions between aerosols, clouds and precipitation.

- The joint iLEAPS/IGAC/GEWEX project Aerosol-Clouds-Precipitation-Climate (ACPC) was created with the goal to improve this understanding