It has been shown that small atmospheric particles can impact climate in two ways (IPCC, 2001). First, aerosols absorb and scatter solar and terrestrial radiation in what is known as the direct effect. For example, a decrease in surface temperature can be correlated with an increase in atmospheric aerosol loading. This is commonly referred to as ‘global dimming’.

Particulate matter also initiates the formation of clouds. Thus, a second impact on climate is attributed to changes in aerosols (e.g., number density, atmospheric residence time, chemical composition, etc.) as they indirectly affect cloud properties. Within the indirect effect are a number of distinct aerosol-cloud interactions but, for simplicity, they are normally separated according to liquid, ice, and mixed-phase clouds.

Hygroscopicity and size are the determining factors in whether an aerosol will take up gas-phase water, grow, and form a droplet within a warm cloud. Freezing is a more complex process. Particles are known to initiate the formation of ice clouds via two distinct mechanisms. Heterogeneous freezing can occur at temperatures as high as 0 degrees C and saturations as low as that of ice but requires the presence of rare and chemically distinct particles known as ice nuclei. Homogeneous freezing, conversely, is initiated by common aerosols which are aqueous mixtures of sulfates, organics, ammonia, nitrates, and other species but can only occur at temperatures below about -40 degrees C and saturations approaching that of liquid water.

Combustion particles play a major role in both cloud formation and climate. Black carbon behaves in a significantly different manner than sulfate aerosols with respect to absorption of radiation. Biomass burning aerosols have been associated with pyrocumulus cloud formation. The role of these aerosols in ice nucleation remains uncertain with sometimes contradictory results from laboratory studies. For these reasons a comprehensive examination of the effect of combustion generated particles is required for a better understanding of global climate.

Intergovernmental Panel on Climate Change, Climate Change 2001, The Scientific Basis (2001). J. T. Houghton et al., eds. Cambridge University, Cambridge, 291-335.

Combustion Aerosols, Clouds, and Climate

Daniel Cziczo and Ulrike Lohmann

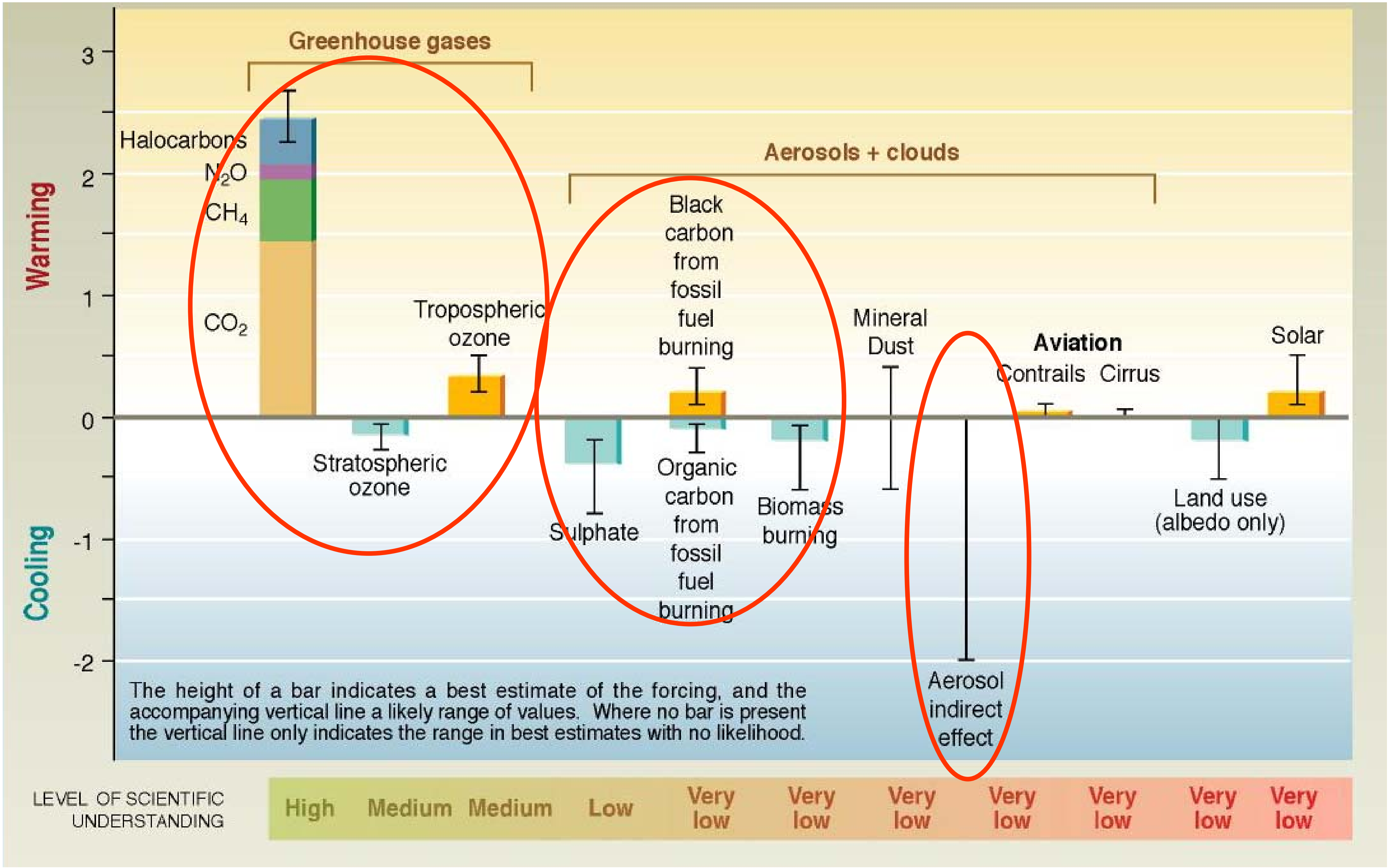
Institute for Atmospheric and Climate Science

ETH Zurich

10th ETH Conference on Combustion Generated Nanoparticles

21.8 - 23.8.2006

Why Study Aerosol / Cloud Interactions ?



The height of a bar indicates a best estimate of the forcing, and the accompanying vertical line a likely range of values. Where no bar is present the vertical line only indicates the range in best estimates with no likelihood.

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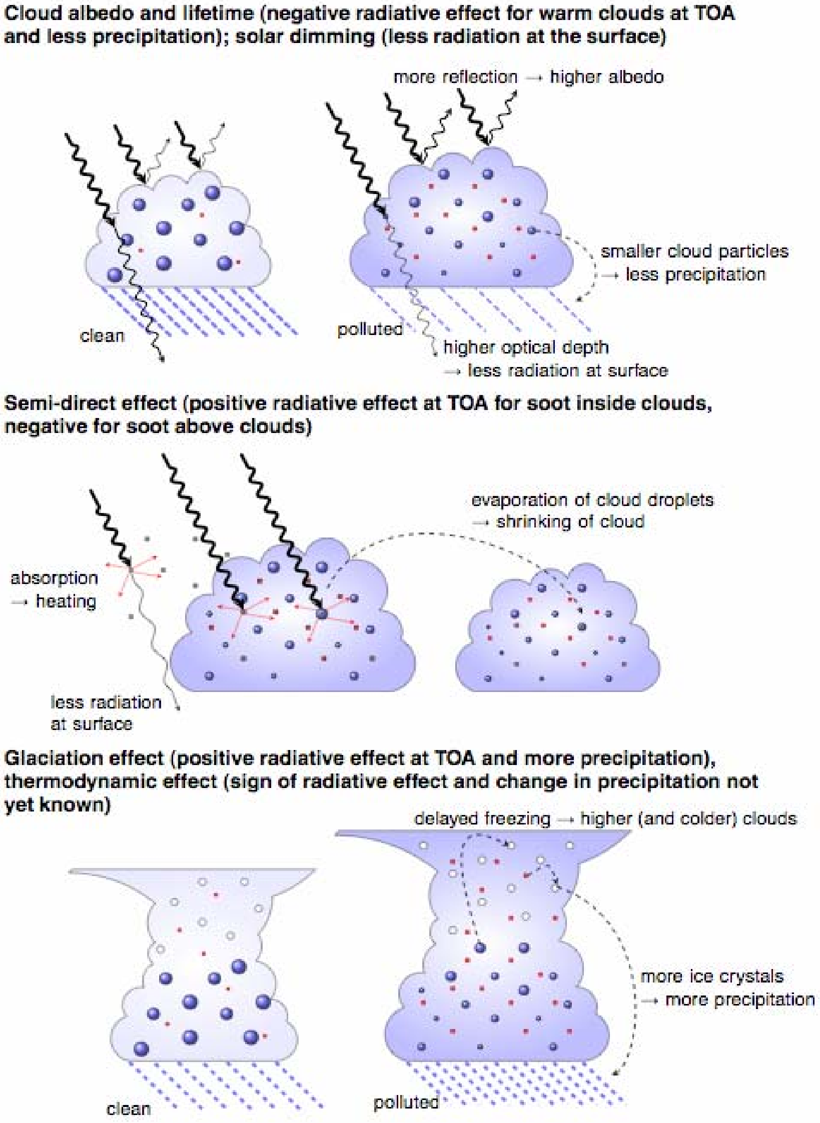
IPCC, 2001

The Indirect Effects of Aerosols



Cloud albedo and lifetime (negative radiative effect for warm clouds at TOA and less precipitation); solar dimming (less radiation at the surface)

Semi-direct effect (positive radiative effect at TOA for soot inside clouds, negative for soot above clouds)

Glaciation effect (positive radiative effect at TOA and more precipitation), thermodynamic effect (sign of radiative effect and change in precipitation not yet known)

Warm Clouds:
1st and 2nd Effect

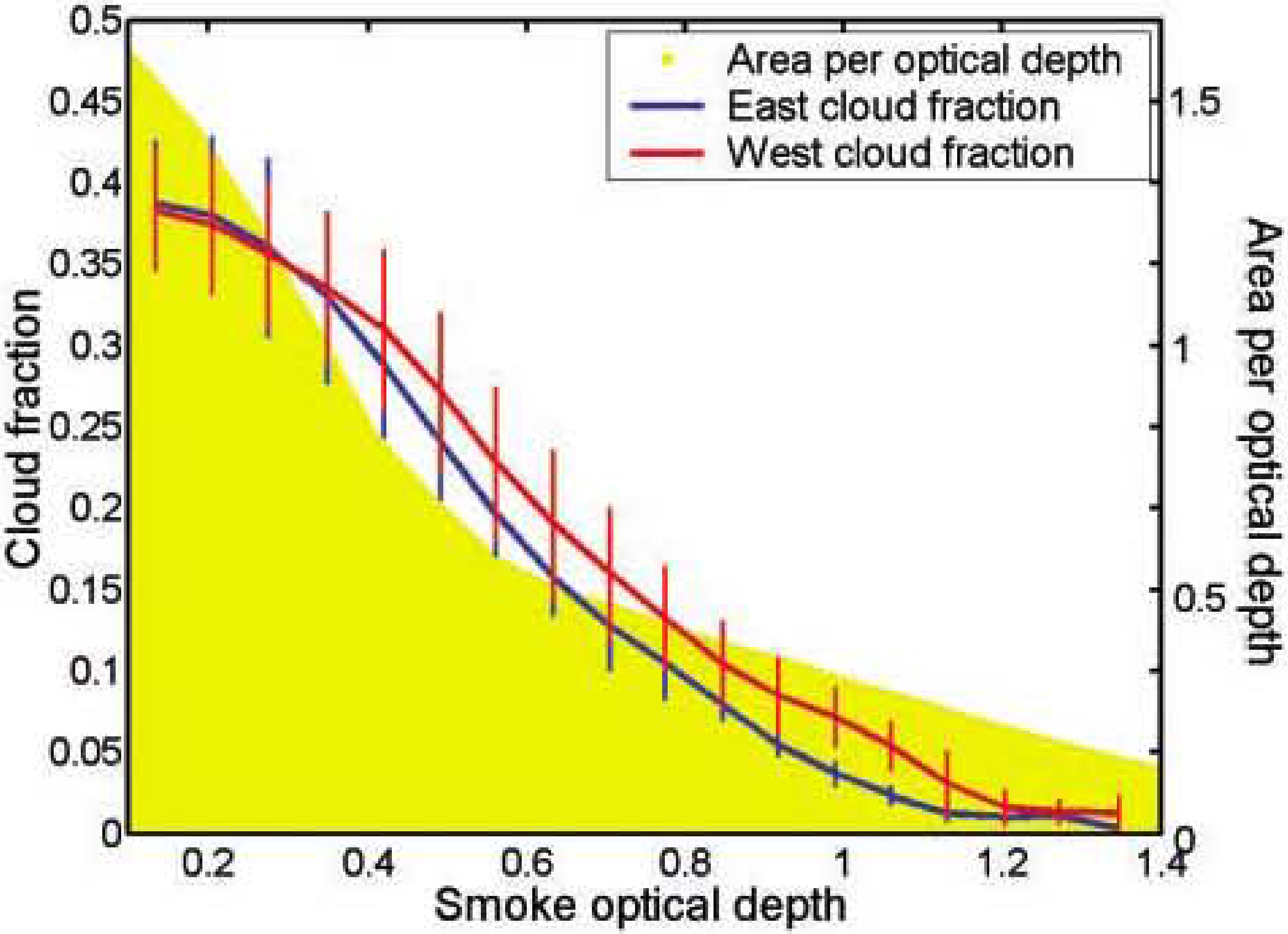
Semi-Direct Effects

Cold Clouds :
Glaciation/Thermo Effects

Courtesy of RP

Semi-direct Effect

(a.k.a. Hansen Effect or'Cloud Burning')



Koren et al., Science, 2004

Indirect Effects : Warm Clouds

- 1st Indirect Effect (‘Twomey Effect’): more aerosols = more cloud condensation nuclei (CCN) ? = smaller size = more reflection = *cooling*

- 2nd Indirect Effect (‘Albrecht Effect’): more aerosols = more cloud drops ? = smaller size = slower fall/coagulation/ coalescence = no precipitation = longer life = *cooling*

Indirect Effects : Warm Clouds

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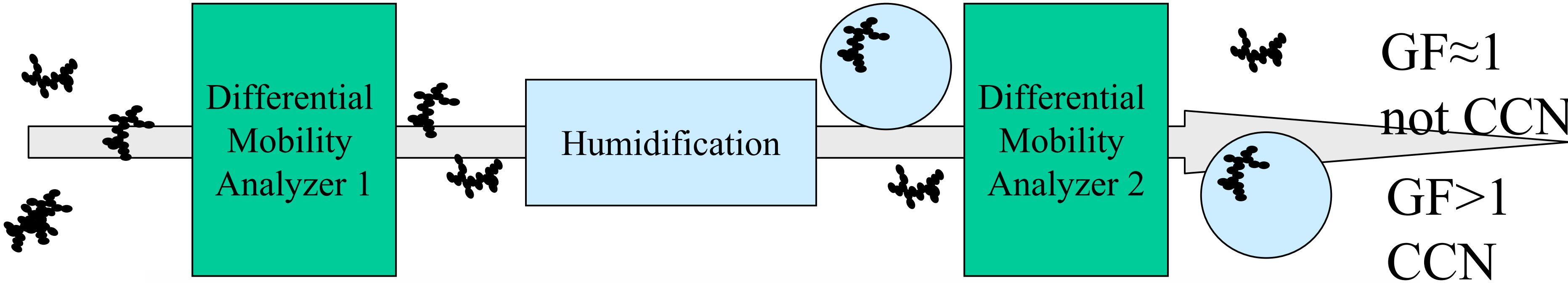
How Have We Studied Warm Cloud Effects (HTDMA)?

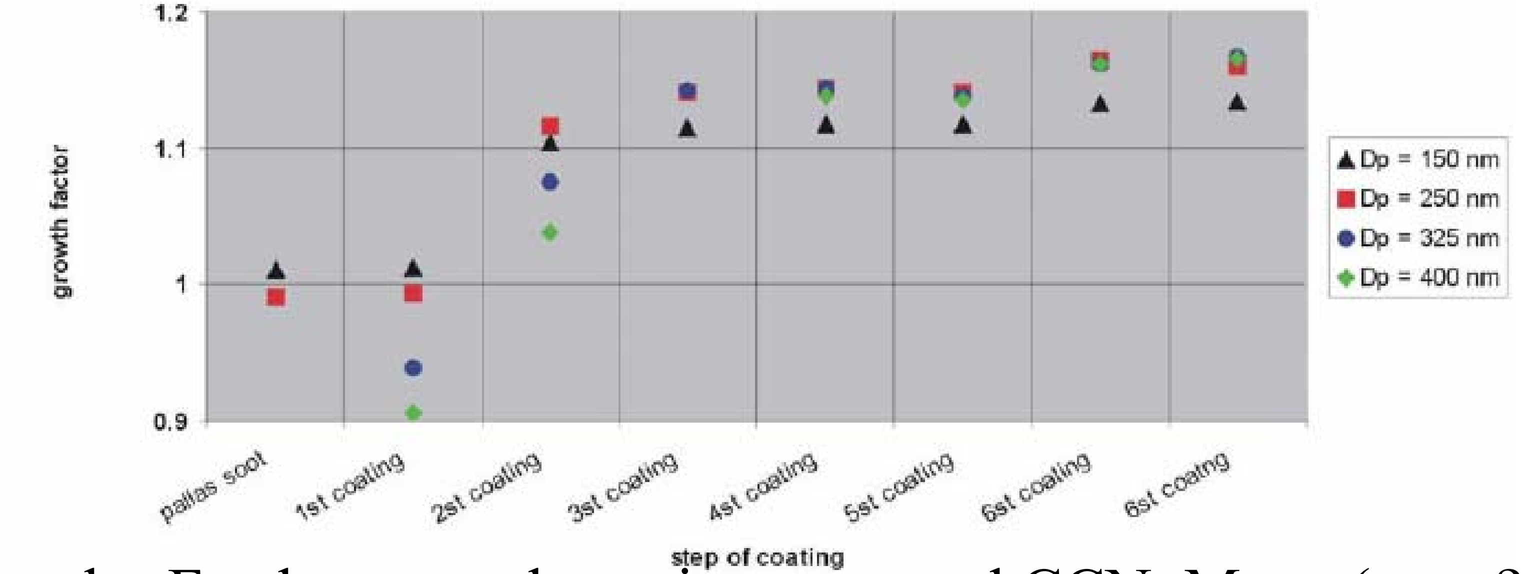

Differential Mobility Analyzer 1

Humidification

Differential Mobility Analyzer 2

GF≈1 not CCN

GF>1 CCN



Result : Fresh soot, at least, is not a good CCN. Many (most?) combustion-generated organics are also poor CCN.

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Wiedensohler et al., EAC, 2003.

Indirect Effects : Cold Clouds

- Glaciation Effect : more aerosols = more ice nuclei (IN; freezing begins above ~-40°) ?
  = more precipitation = ?

- Thermodynamic Effect : more aerosols = more homogeneous freezing nuclei (HFN; freezing begins below ~-40°) ?
  = more/smaller cloud elements = ?

Indirect Effects : Cold Clouds

- Glaciation Effect : more aerosols = more ice nuclei (IN; freezing begins above ~-40°) ?
  = more precipitation = ?

- Thermodynamic Effect : more aerosols = more homogeneous freezing nuclei (HFN; freezing begins below ~-40°) ?
  = more/smaller cloud elements = ?

How Have We Studied Cold Cloud Effects (Optically)?

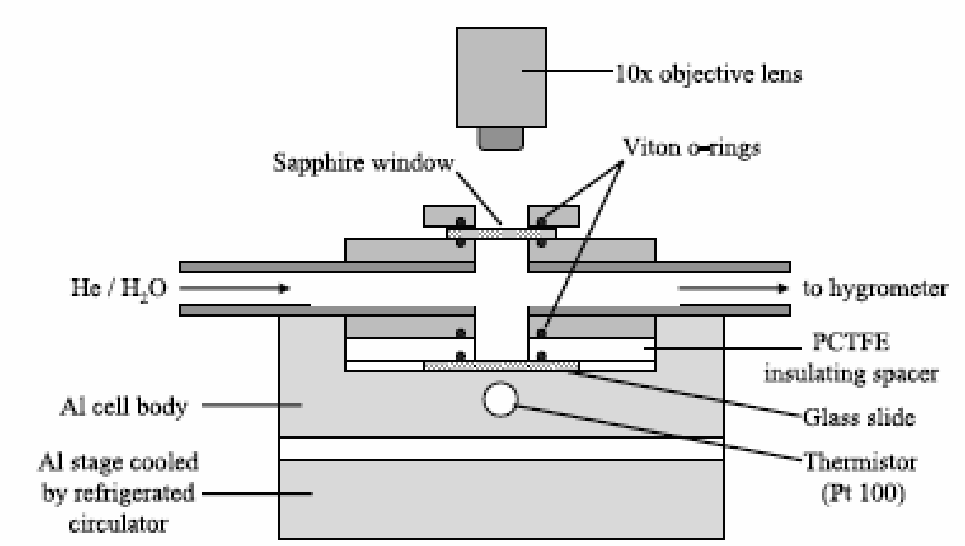

Figure 1. Flow cell and location of microscope objective (Al = aluminum and PCTFE = polychlorotrifluoroethylene).

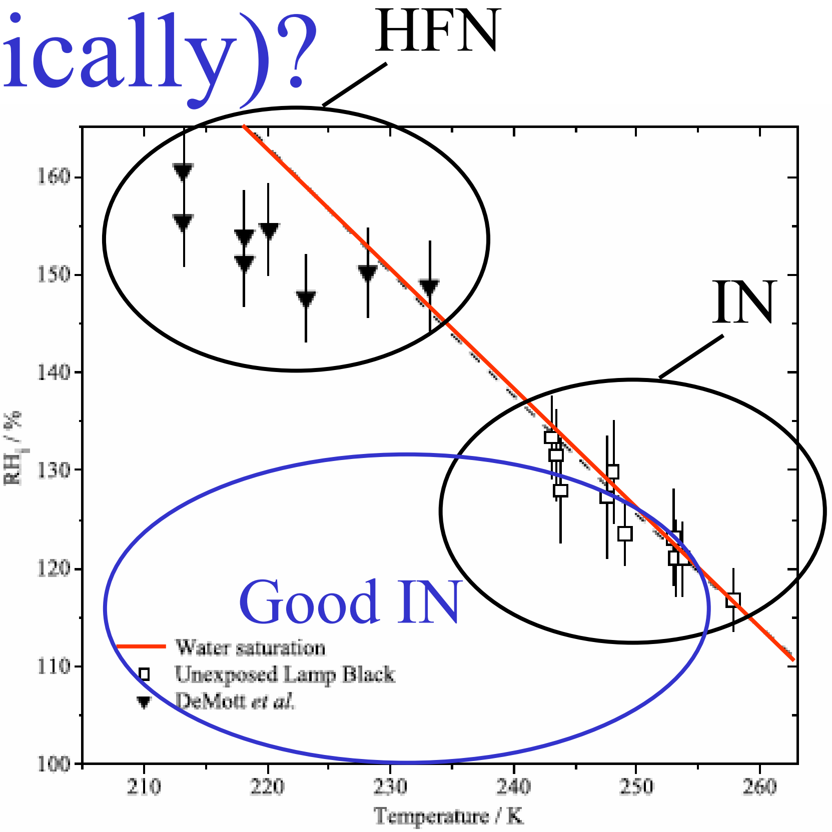

Figure 6. A comparison of our results for Lamp Black 101 (open squares) with those of *DeMott et al.* [1999] (solid triangles). Our data points correspond to the conditions at which water droplets were observed using soot particles ranging in size from 1 to 40 μm in diameter. In these experiments, water droplets were always observed first. If ice did form it was only after the appearance of water droplets. The results from DeMott et al. correspond to the onset for which 1% of Lamp Black soot particles (a number mean diameter of 240 nm) nucleated ice.

Dymarska et al., JGR, doi:10.1029/2005JD006627, 2006.

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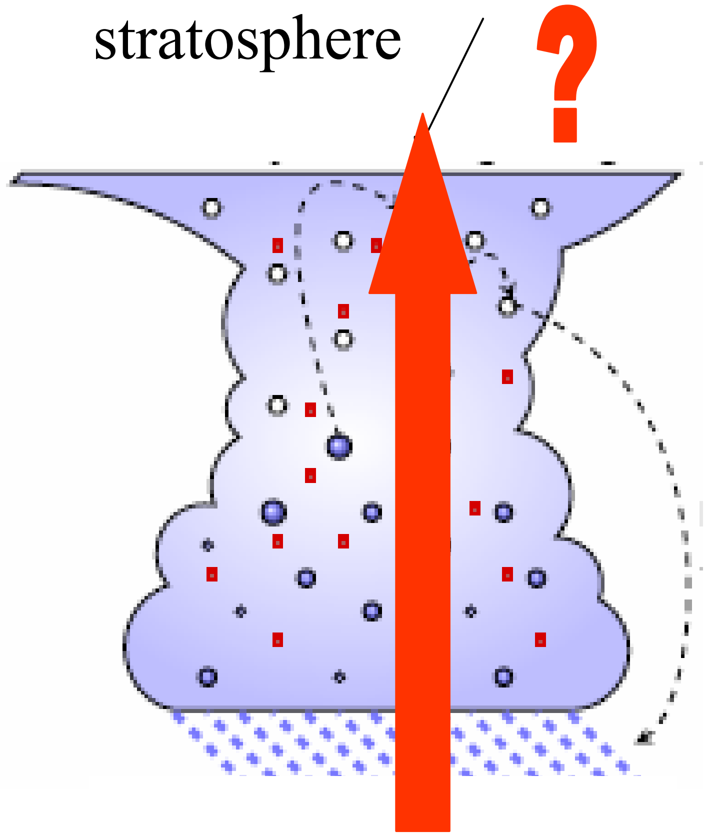
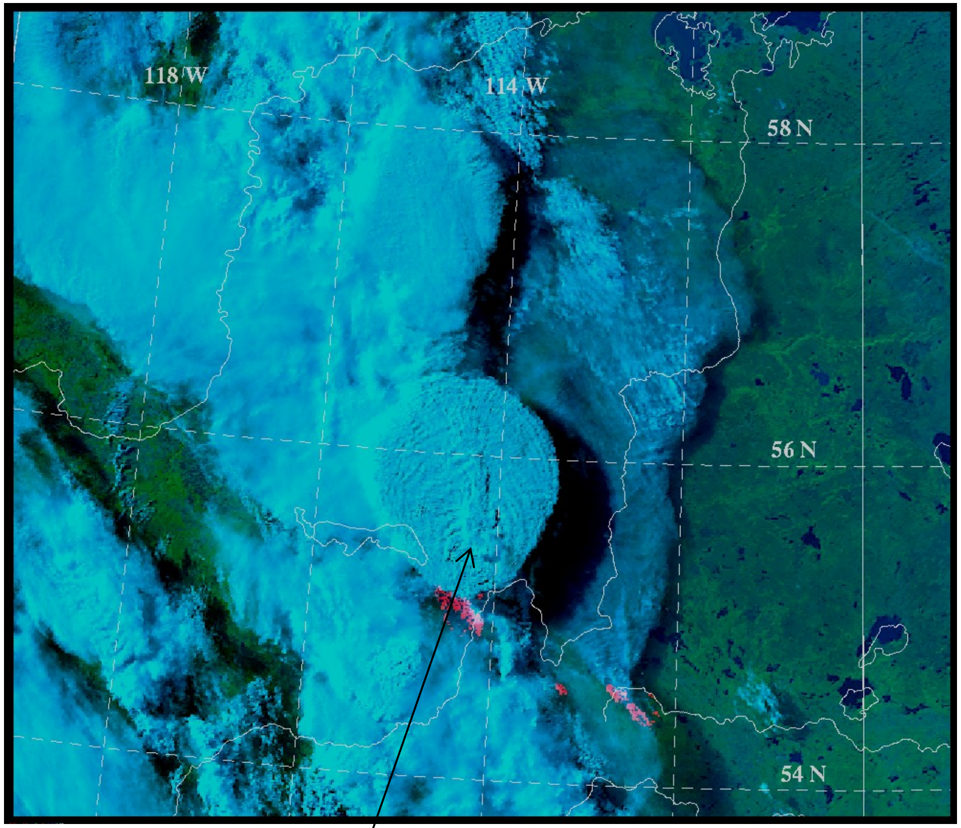
An Atmospheric Example

Flat top ('anvil') is the delineation of turbulent troposphere and stable stratosphere



Side view of a cumulonimbus cloud



Fromm and Servranckx, GRL, 30, 1542, 2003.

'Overshooting' sometimes occurs…

Biomass Burning in the Stratosphere

Alaskan biomass plume (soot, organic etc.), courtesy of Japan Airlines. Up to half of atmospheric BC is from biomass burning (Ramanathan et al., Science, 2001).

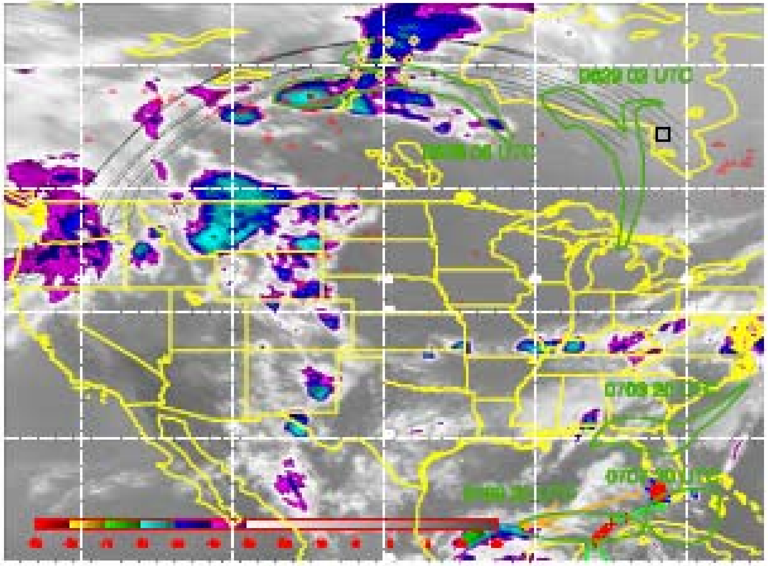


Figure 4. Cloud infrared brightness temperature from GOES weather satellite from June 28, 2002 05:45 UTC, showing at the top center the convective system likely responsible for the pumping up of the smoke plume. We estimate a minimal extent of the plume by advecting (indicated by dark yellow arrow) the highest CO points (red +) measured on the flight of July 7 (blue trace off SW Florida) to July 9 (dark yellow +), combining them with the measured points (red +) from July 9 (green trace) and assuming the plume to be contiguous within the green ellipse. This ellipse was then advected backward isentropically (θ = 382 K) in time to June 28 (shapes are labeled in MMDD HH format). The black lines are back trajectories initialized at and around the POAM footprint (white square), and the yellow diamonds mark the position at the time of the GOES image. Red triangles corresponds to MODIS fire locations in the period June 25 through June 29 above 40°N.

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Jost et al., GRL, 31, L11101, 2004.

Biomass Burning in the Stratosphere

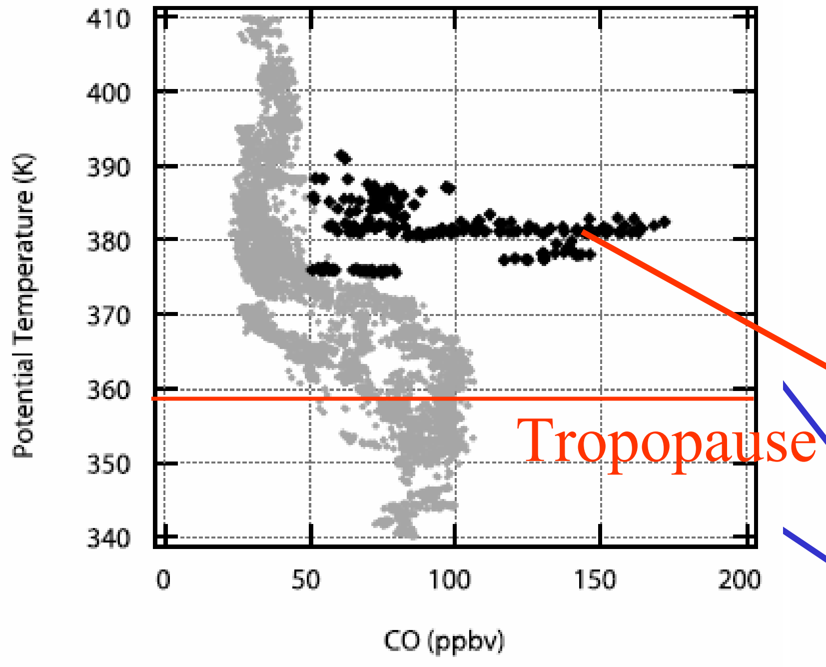

Figure 3. Profile of CO versus potential temperature for the 9 July 2002 WB-57F flight. Light shaded points indicate data from the entire flight and dark shaded points are in the "plume" portion of the flight as discussed in the text.

Ray et al., JGR, 109, 18304, 2004.

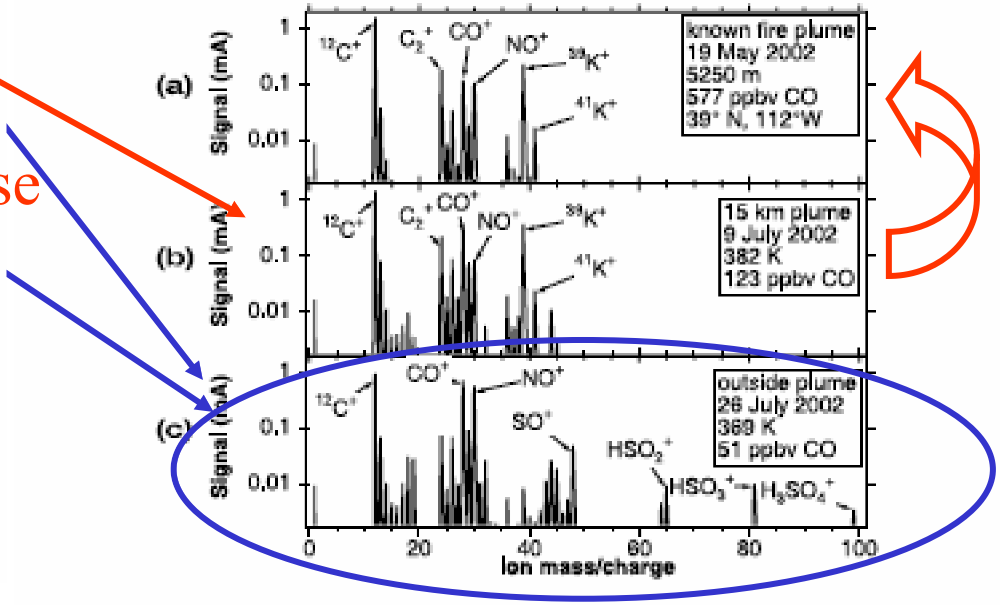

Figure 2. Single particle positive ion mass spectrum recorded by the PALMS instrument. (a) Mass spectrum of a particle of known, 2 hours old forest fire plume. (b) Mass spectrum of a particle in the high CO layer. (c) Mass spectrum of a representative, stratospheric sulfate particle.

Jost et al., GRL, 31, L11101, 2004.

Results

- Soot aerosols are VERY good absorbers of solar radiation. Addition of soot will warm the atmosphere (direct effect).

- Soot present near cloud top will heat the local environment and burn off the cloud. Addition of soot will warm the atmosphere (semi-direct effect).

- Fresh soot aerosols are *not* good CCN, IN, or HFN. Soot is not effectively removed via precipitation mechanisms and, unless scavenged, will exist in the atmosphere for longer than other species* (i.e., until processed or coagulated).

- The effect of soot entering the stratosphere has not been extensively studied.

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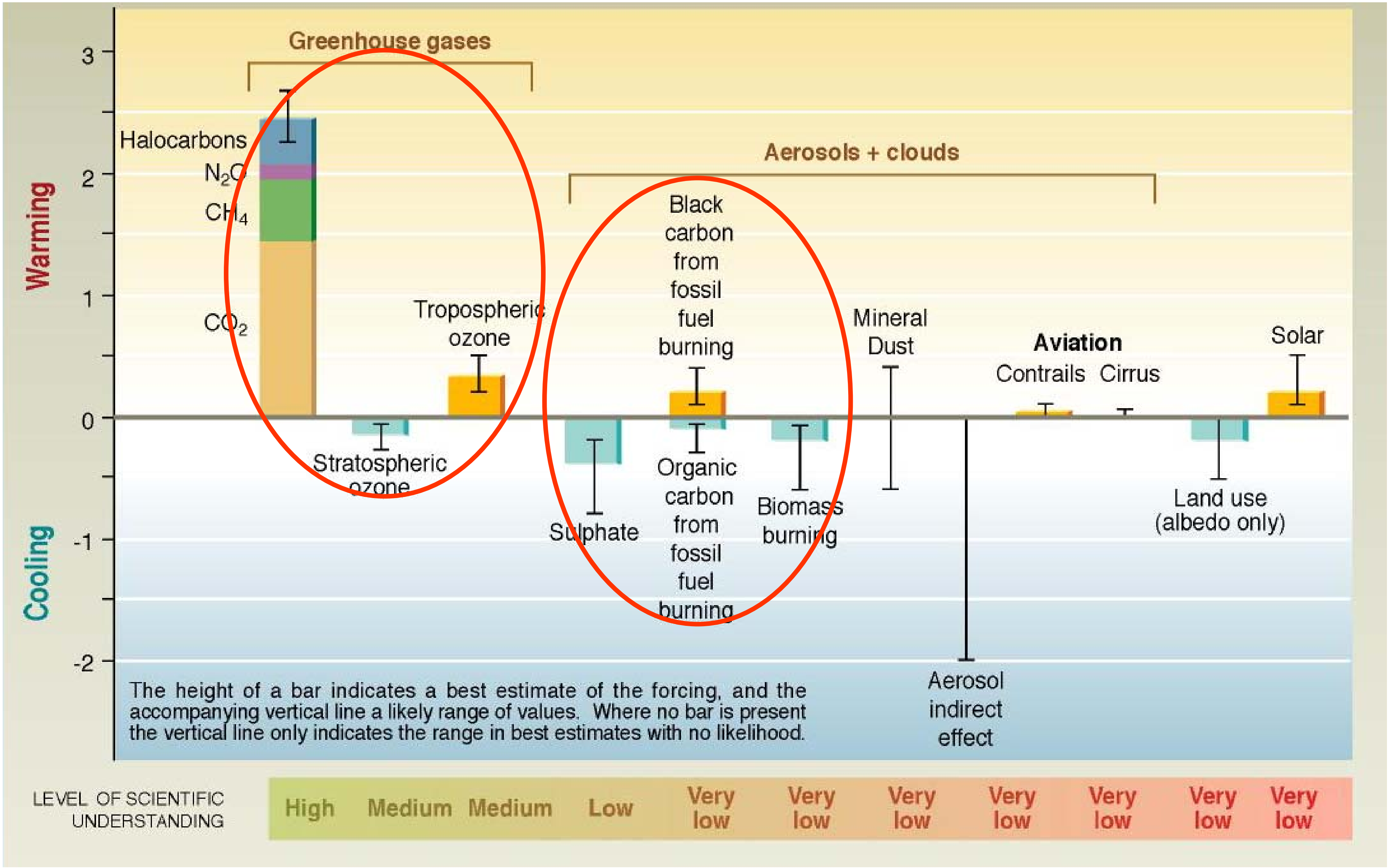
*Up to twice the lifetime : Lesins et al., JGR, 10.1029/2001JD000973, 2002.

Questions?

Why Study Aerosol / Cloud Interactions ?



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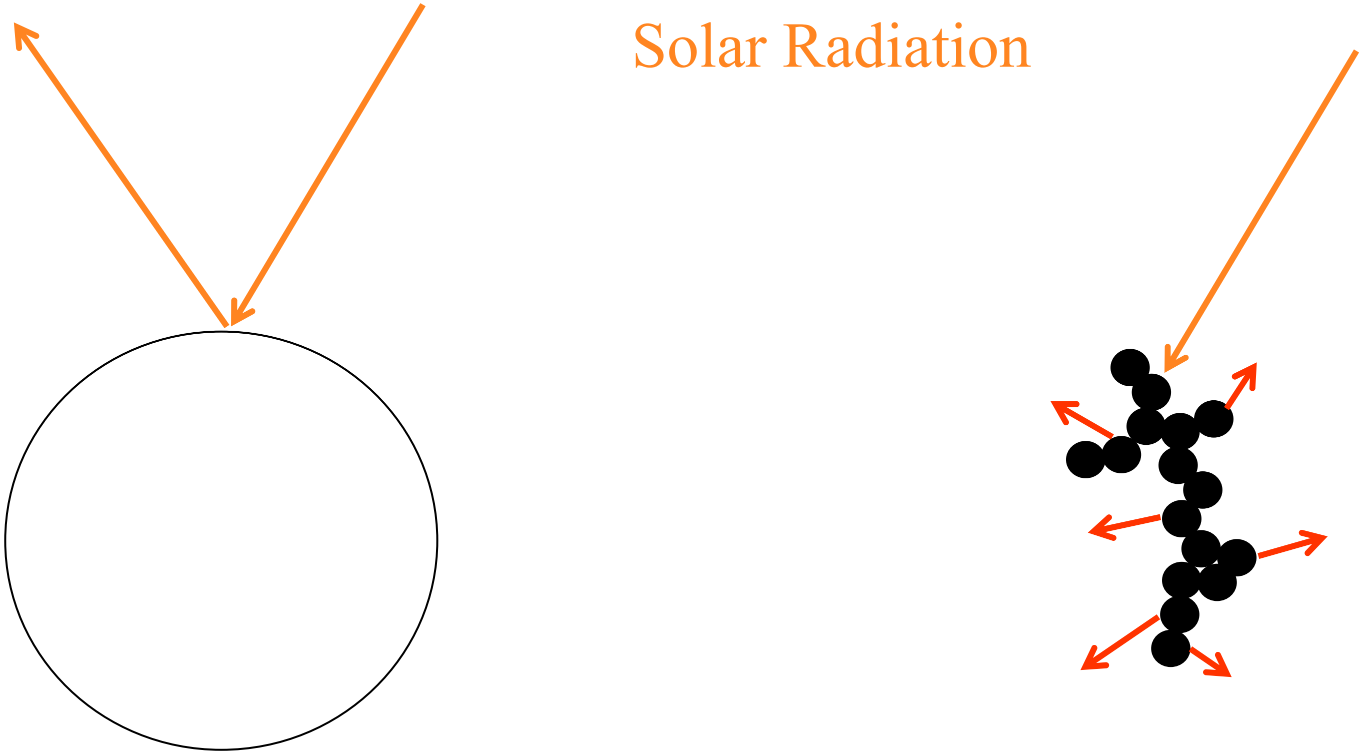
IPCC, 2001

Direct Effect of Aerosols



Solar Radiation

sulfate / organic aerosol =
scattering = *cooling**

soot aerosol =
absorbing = *warming*

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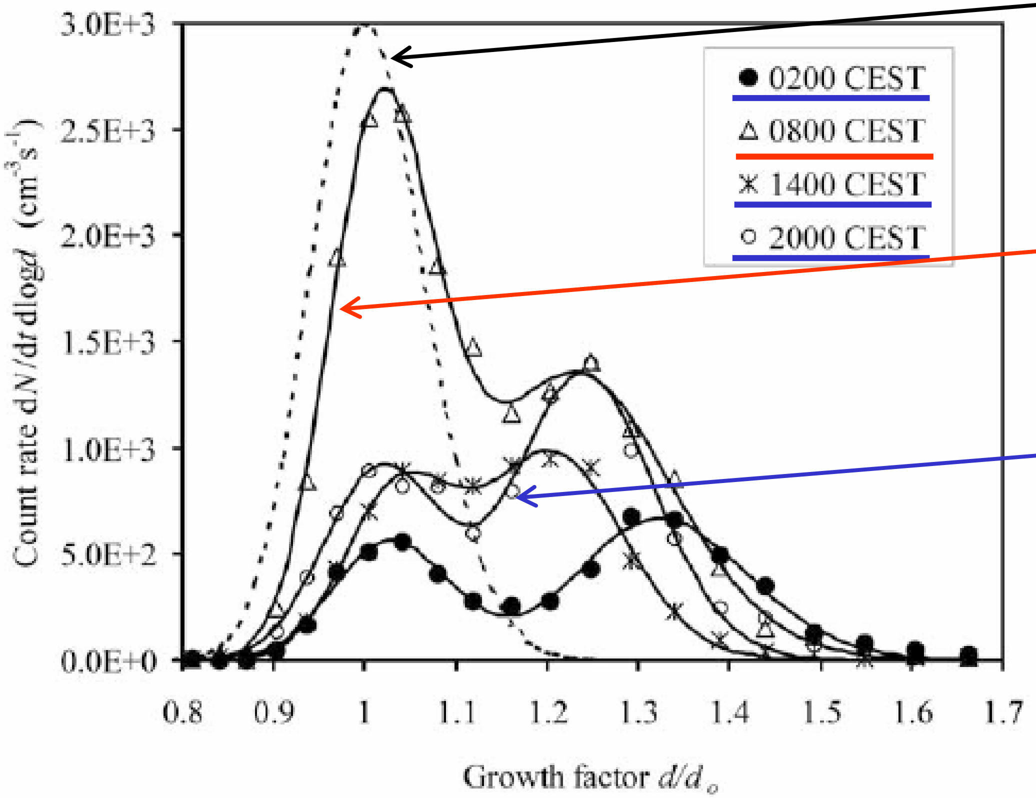
* Global Dimming : e.g., Stanhill and Cohen, Journal of Climatology, 10, 2078-2086, 2001.

Atmospheric Effects : Milan



Initial Aerosol Size

‘Rush Hour’ = mainly soot

Other times = mixed

Figure 6. Temporal evolution of the growth factor (d/d_0) distribution on 2 June 1998 during IOP 2. Dry monodisperse particles (d_0 = 100 nm, dashed line) were exposed to RH = 90% at 0200, 0800, 1400, and 2000 CEST (points). The solid lines are bimodal fits of the less and more hygroscopic modes. *Y* axis units correspond to CPC counts normalized to both the diameter interval and the scan time during such an interval. Lines denote a monomodal/bimodal lognormal fit.

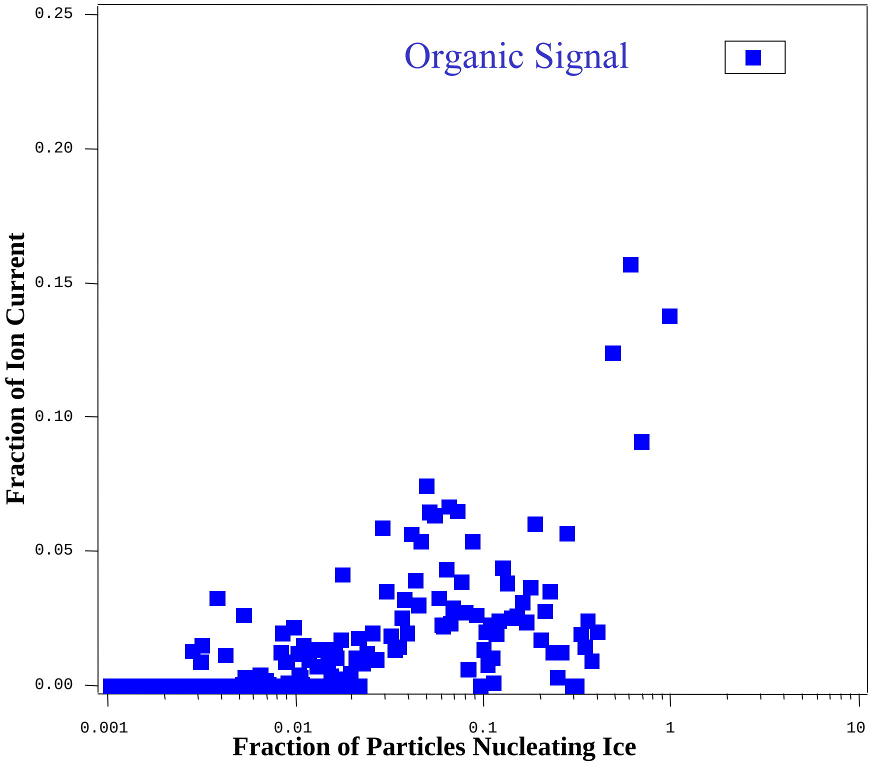
Baltensperger, JGR, 2002.

Atmospheric Effects

IN

HFN

Collection of Ice and Snow,
Analysis of Central Particles:
- mineral dust
- metal oxides



Result : Soot and organics are neither good IN or HFN

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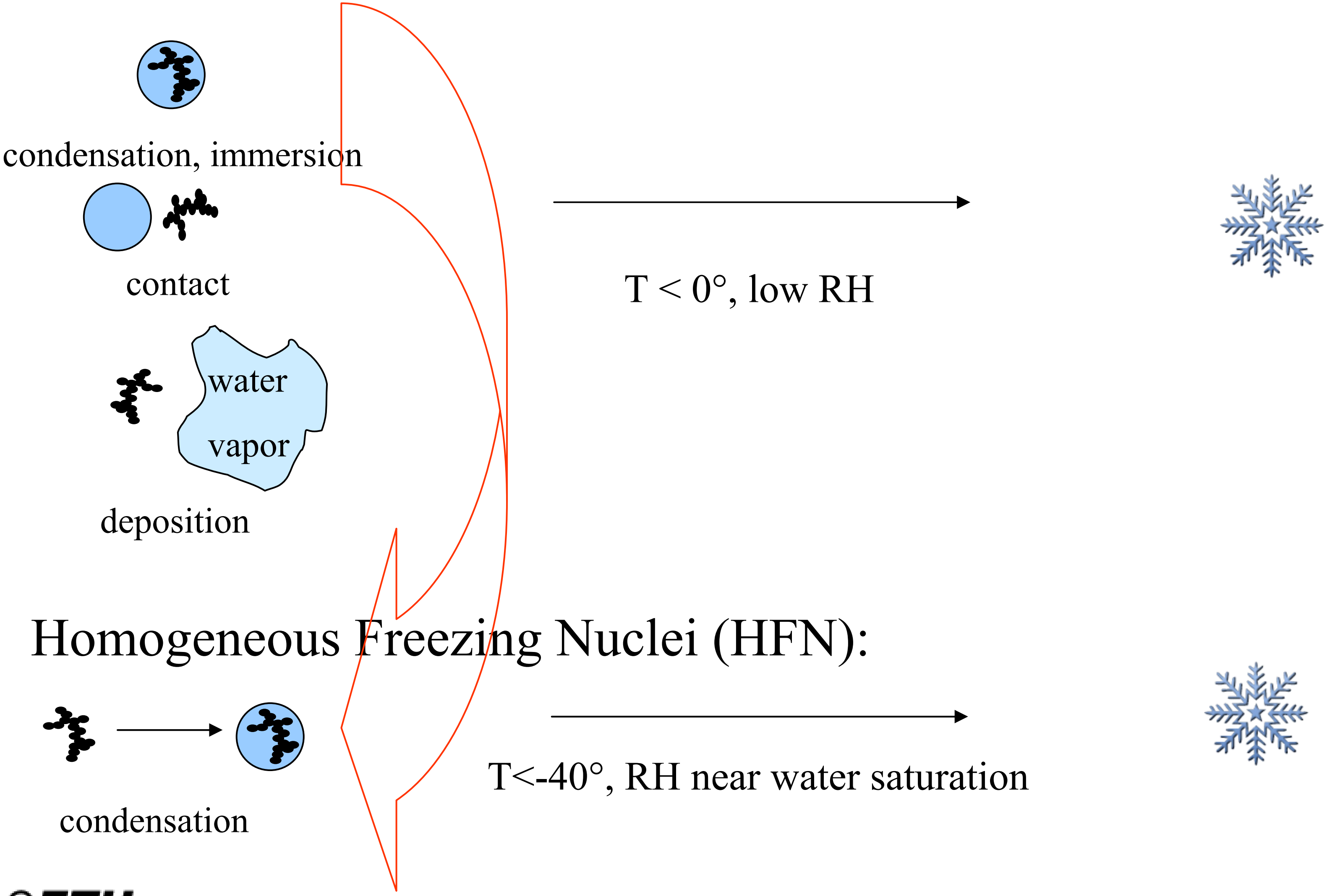
Cziczo et al., GRL, 2004

Digression : Freezing Mechanisms



Ice Nuclei (IN):

condensation, immersion

contact

water

vapor

deposition

T < 0°, low RH

Homogeneous Freezing Nuclei (HFN):

condensation

T<-40°, RH near water saturation