Aerosol Effects on Water and Ice Clouds

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Contributions from
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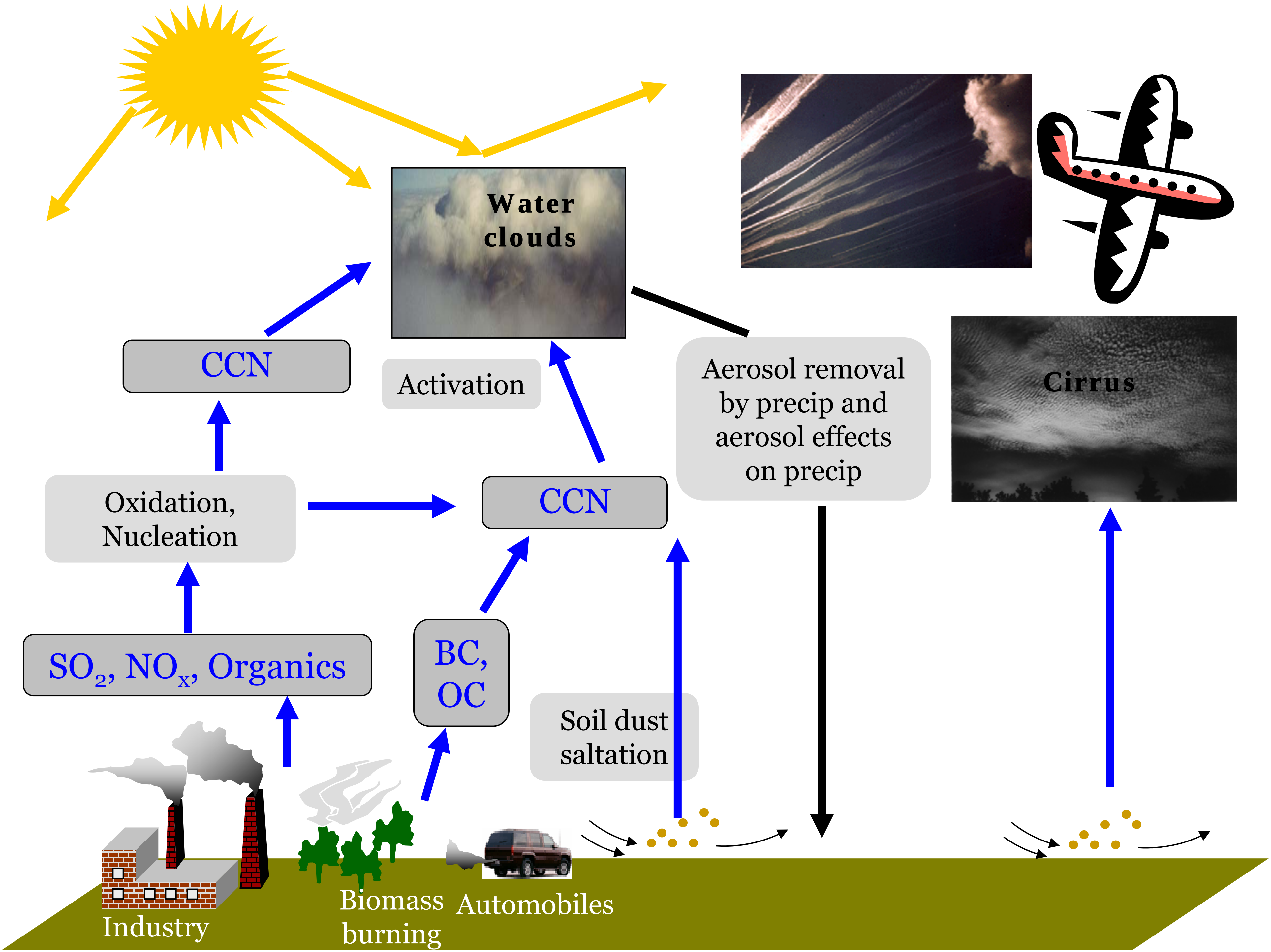
Outline of this Lecture

- Different observations of aerosol effects on clouds
- Aerosol effects on *water clouds*
- Aerosol effects on *cirrus clouds and contrails*
- Aerosol effects on the *hydrological cycle*
- Conclusions



Water clouds

CCN

Activation

Aerosol removal by precip and aerosol effects on precip

Cirrus

Oxidation, Nucleation

CCN

SO_2, NO_x, Organics

BC, OC

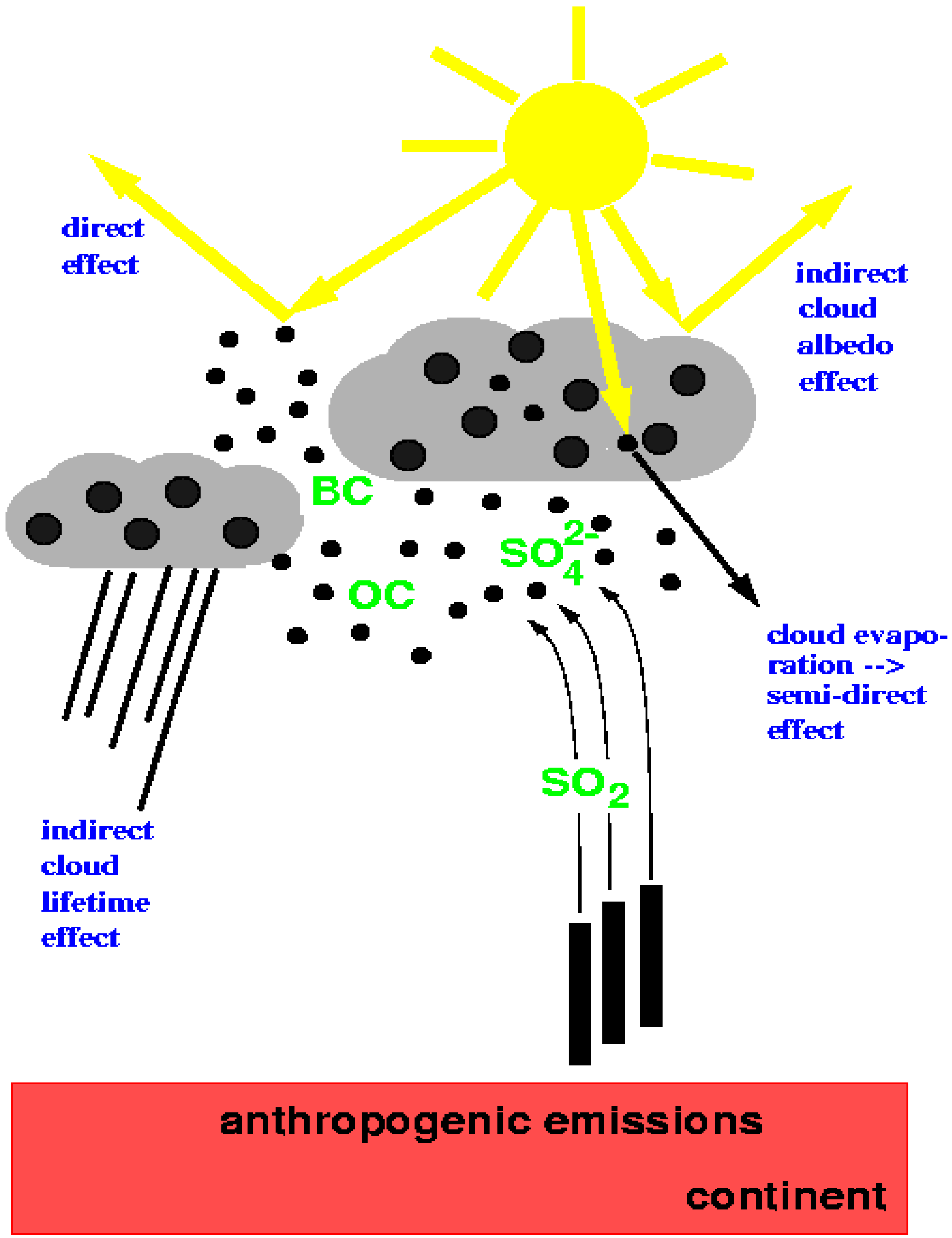
Soil dust saltation

Industry

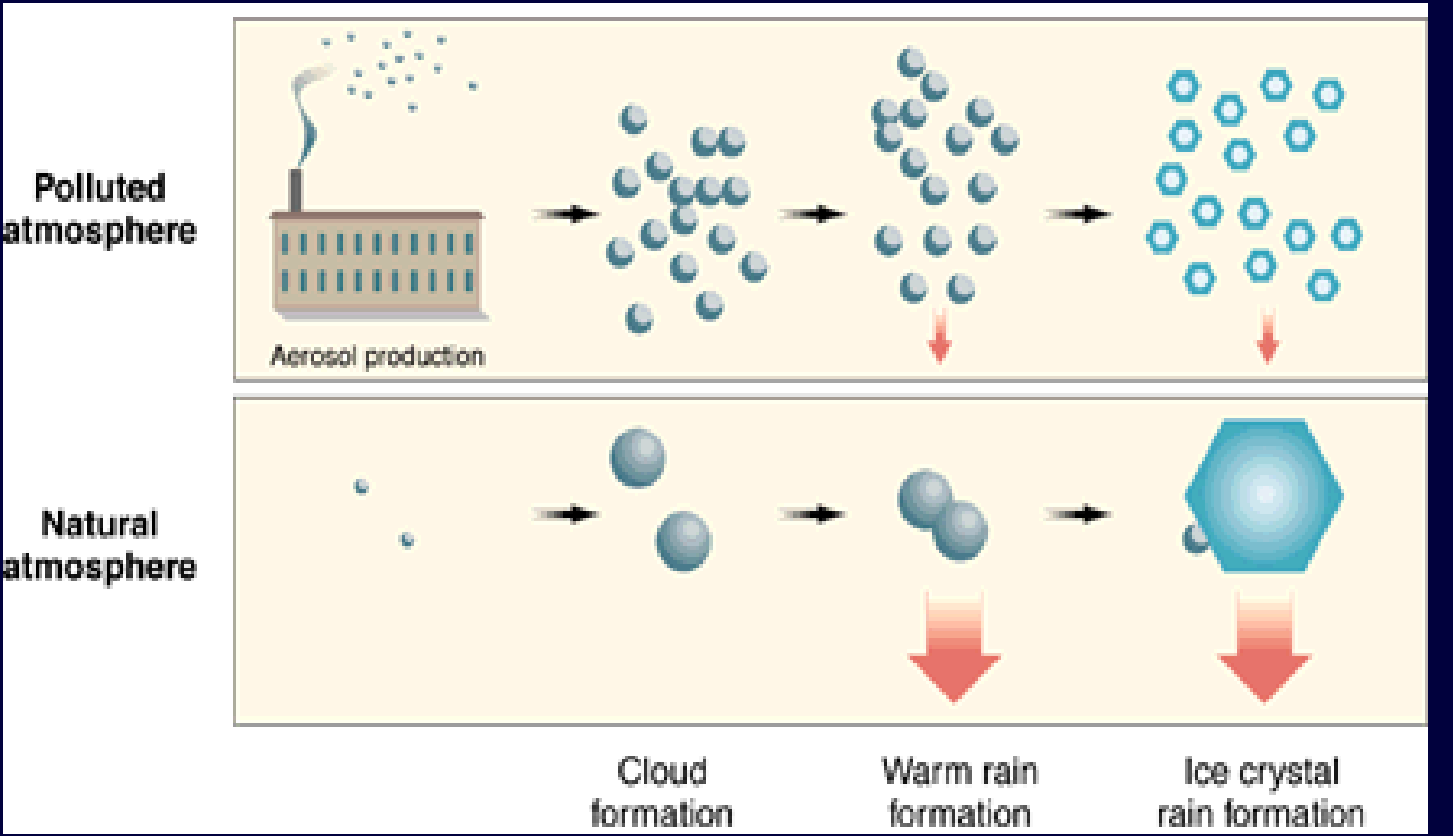
Biomass burning

Automobiles

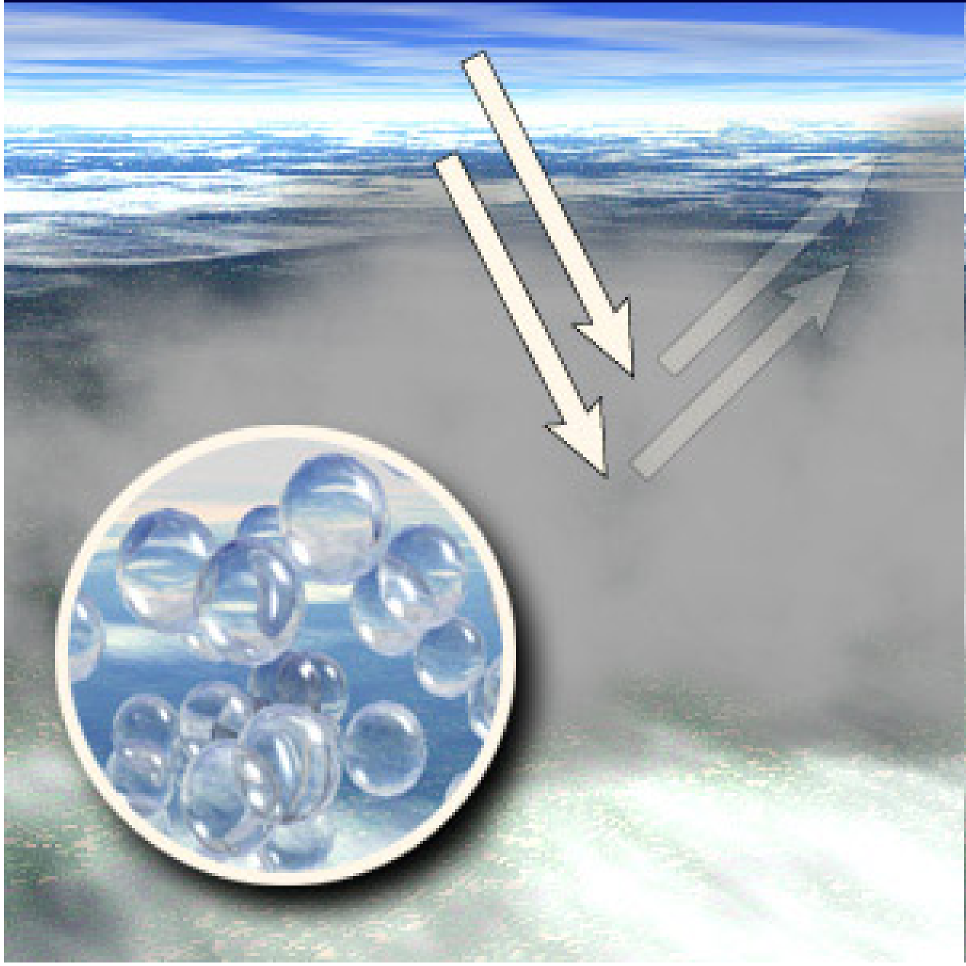
Different Aerosol Effects on Water Clouds



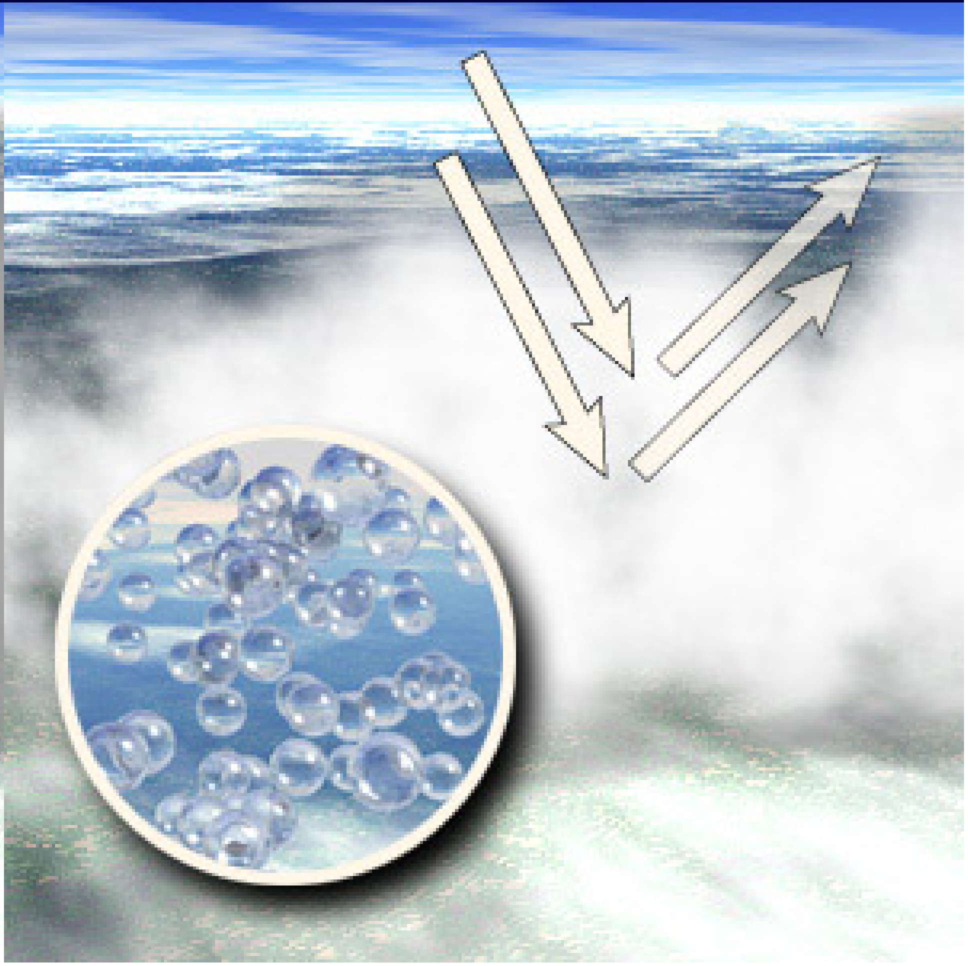
Cloud Formation in a Clean and Polluted Atmosphere



This cloud has only few cloud droplets, hence, reflects less sunlight (darker cloud).



This cloud has more cloud droplets, hence, reflects more sunlight (lighter cloud).



Aerosol Modification of Marine Stratus Clouds

[Durkee et al., 2000]

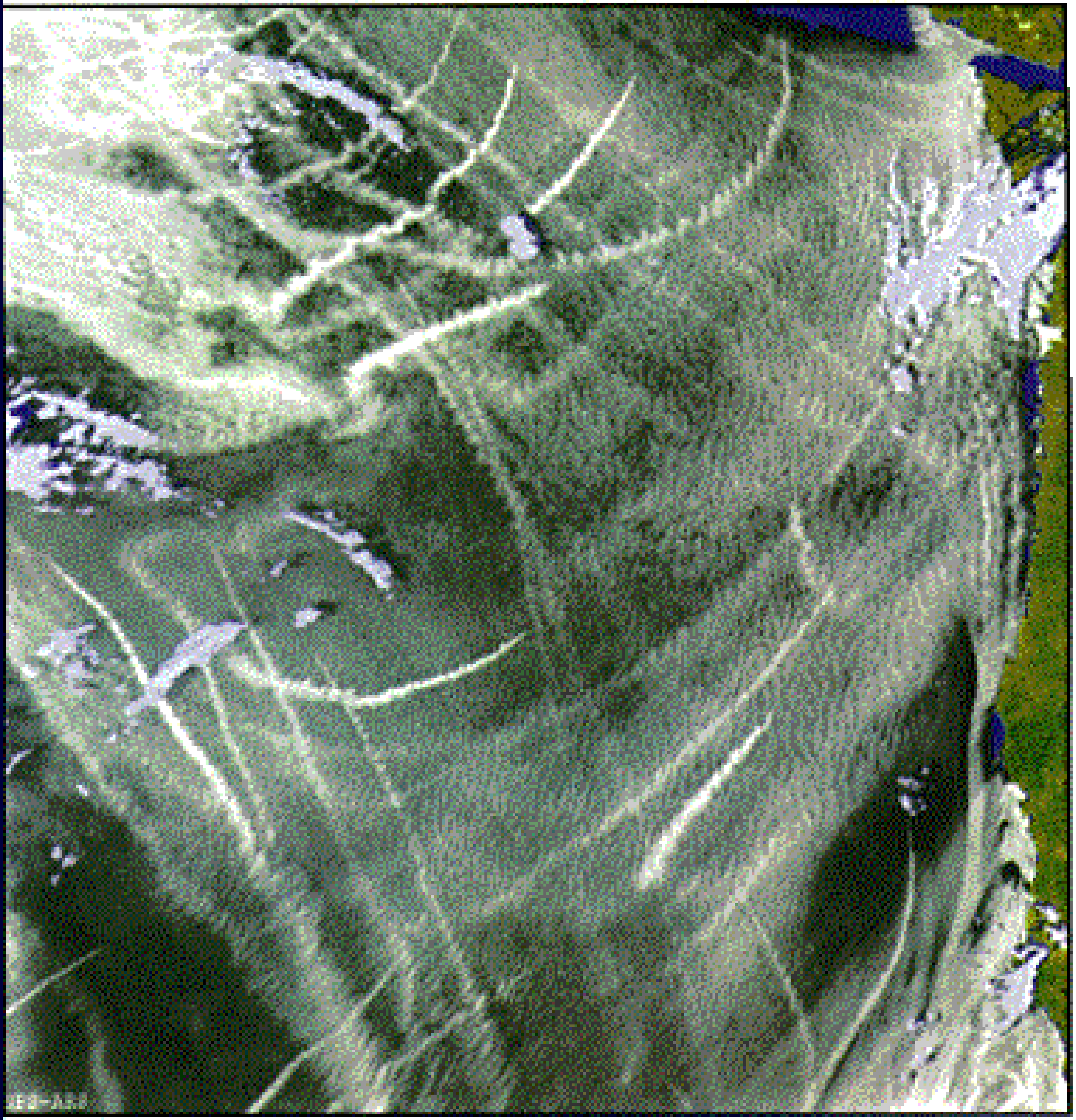
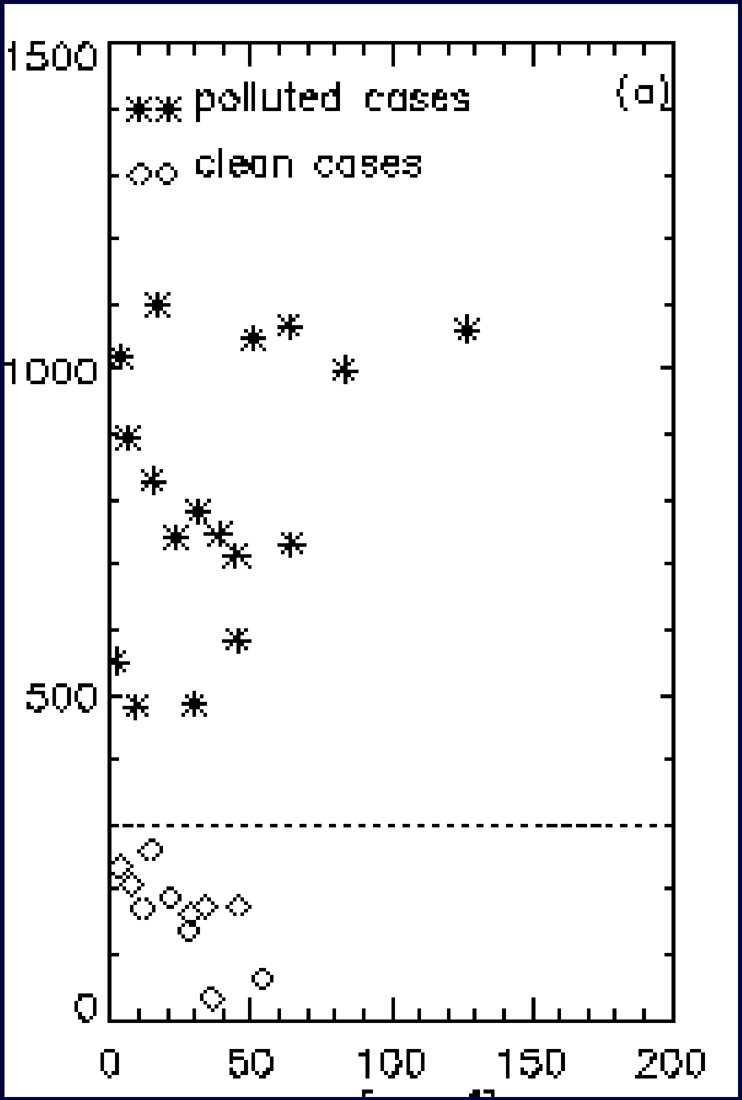


Figure 1: Ship tracks off the coast of Washington

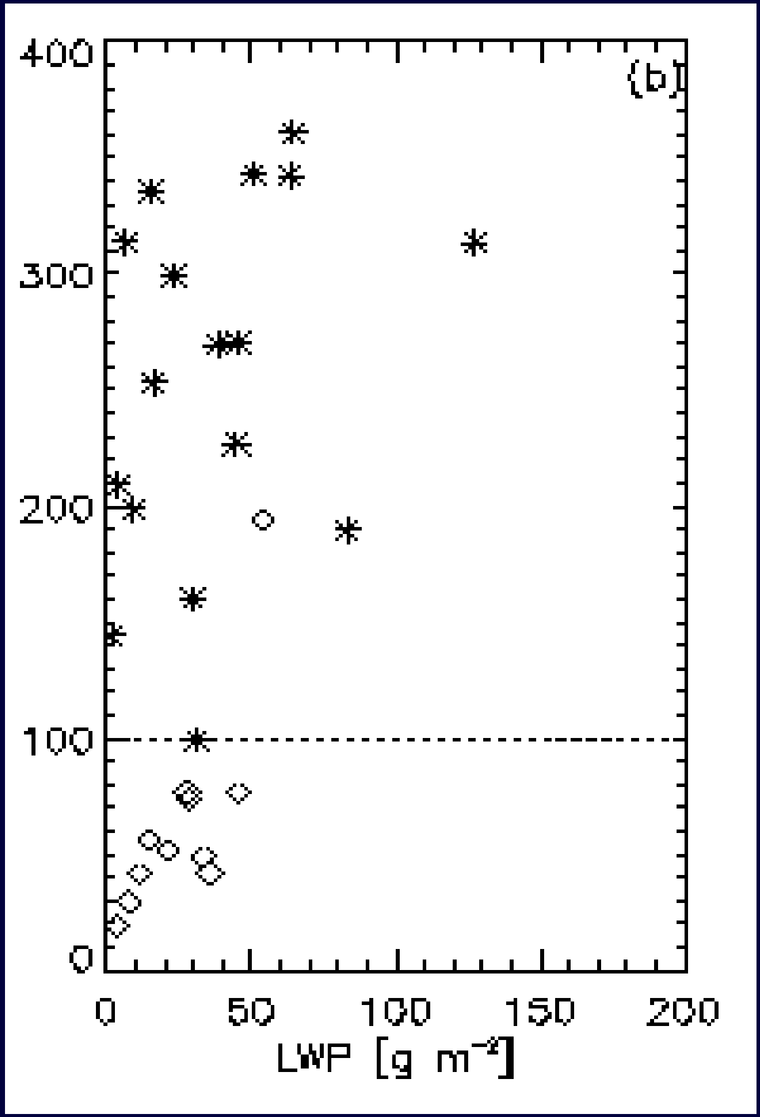
Data from Canadian Field Experiments

[Peng et al., 2002]



Number of aerosols (cm^{-3})

Cloud water (g m^{-2})



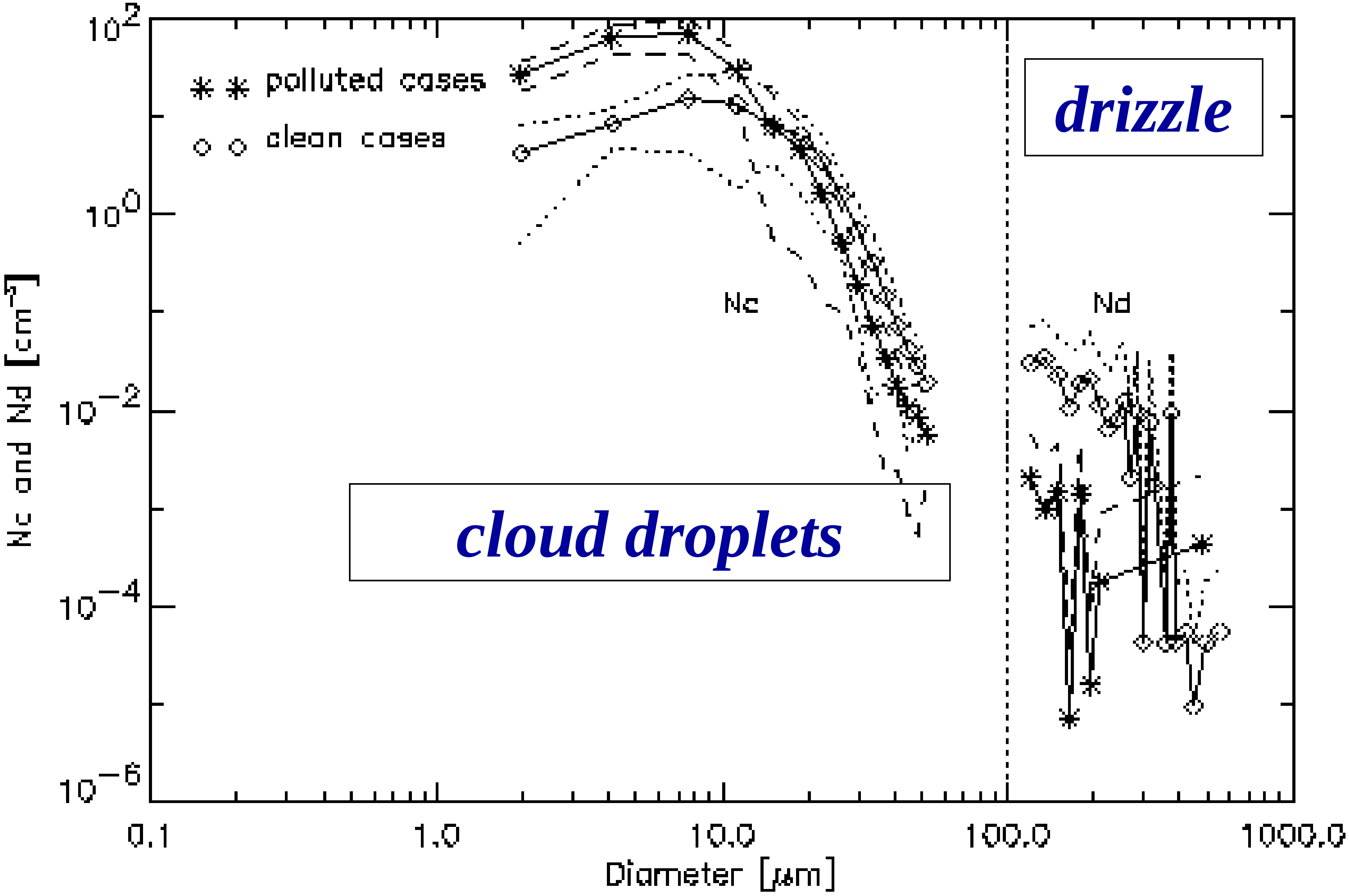
Number of cloud droplets (cm^{-3})

Cloud water (g m^{-2})

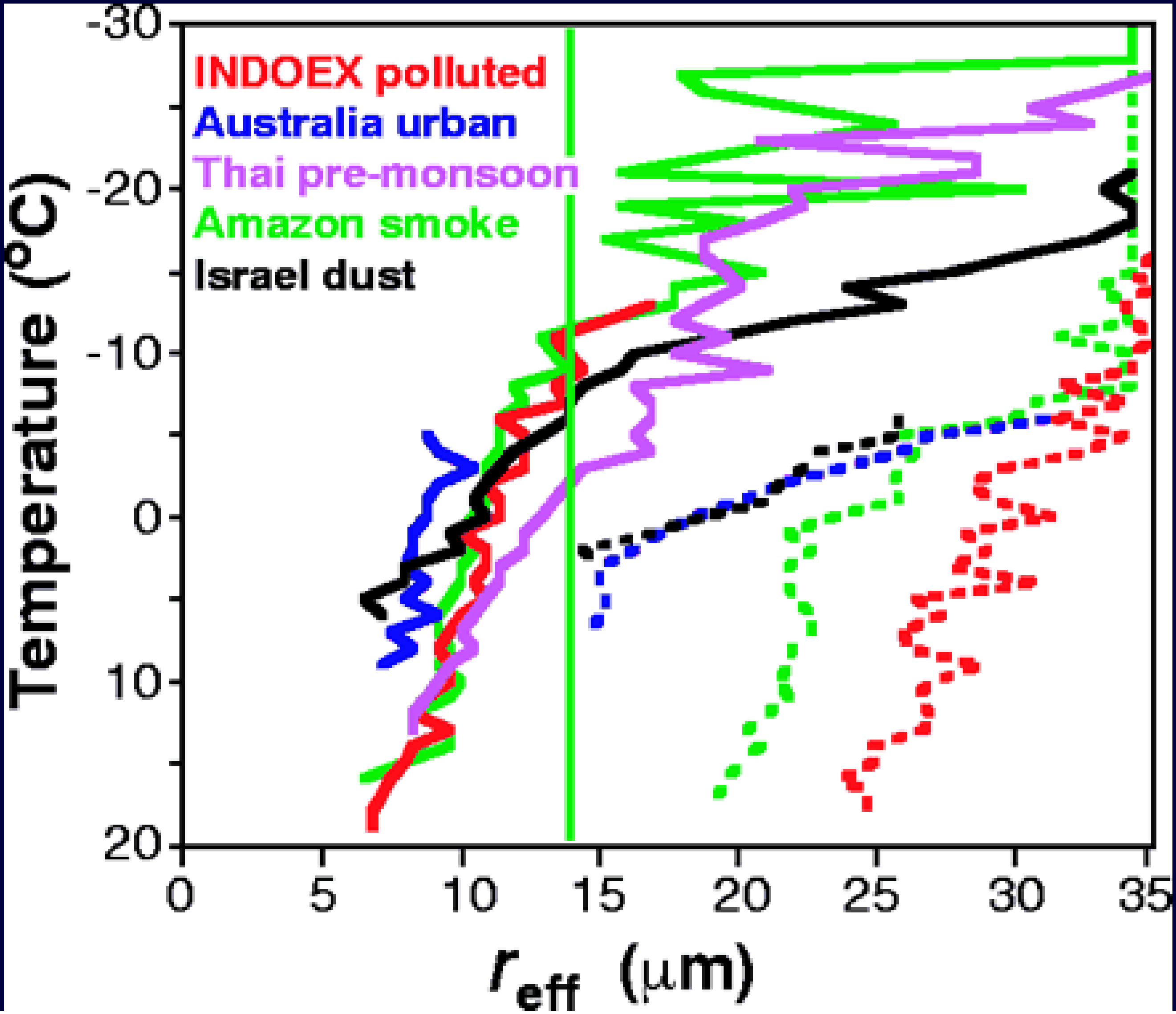
Data from Canadian Field Experiments

[Peng et al., 2002]

Number of Cloud Droplets and Drizzle



Satellite-retrieved median effective radius of particles near the top of deep convective clouds at various stages of their vertical development



polluted clouds:
solid lines

clean clouds:
dotted lines

Ramanathan et al. [2001]

Effect of Aerosols on Deep Convective Clouds

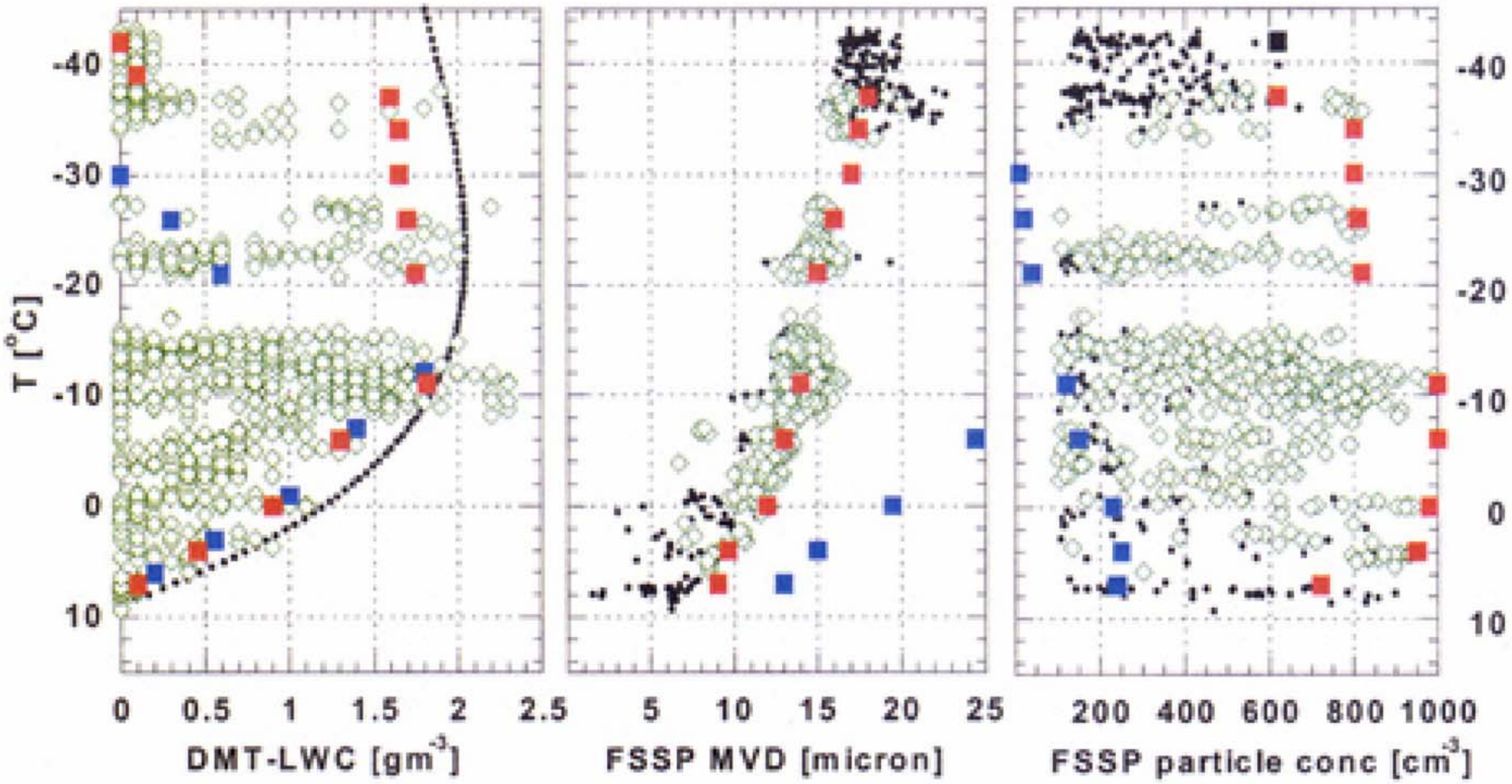
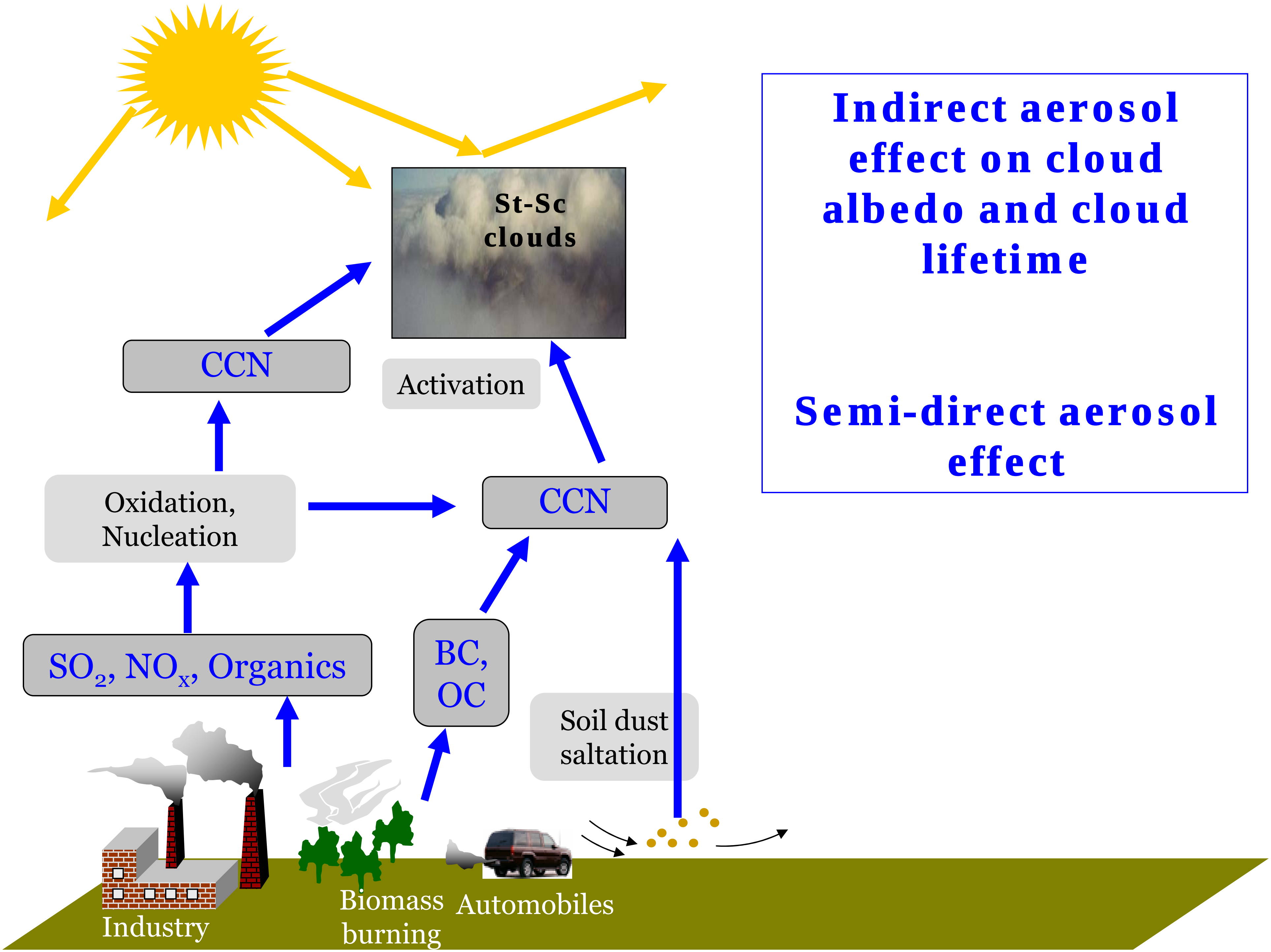


Figure 3. Vertical profiles of maximum values of (a) cloud water content (CWC), (b) the mean volume diameter and (c) droplet concentration observed in the control run at 250 m below the growing cloud top, presented on the background of the aircraft observations (Rosenfeld and Woodley, 2000), shown in green (CWC>0.2 gm^{-3} and black (CWC≤0.2 gm^{-3}). The blue and red squares denote model calculated values for the low and high CCN concentrations. The black square in the concentration panel (c) denotes the model ice concentrations.



St-Sc clouds

CCN

Activation

Oxidation, Nucleation

CCN

SO_2, NO_x, Organics

BC, OC

Soil dust saltation

Industry

Biomass burning

Automobiles

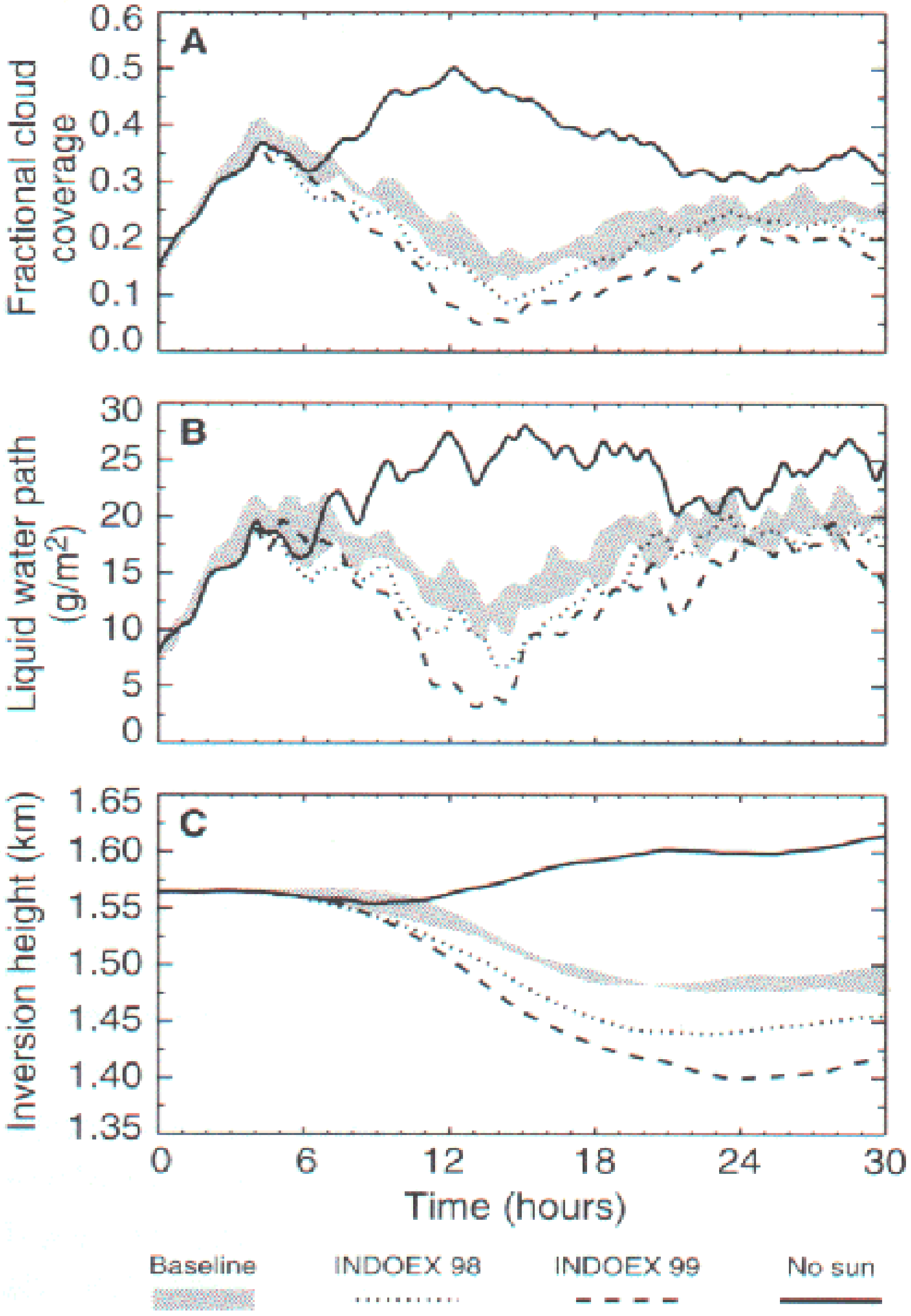
Indirect aerosol effect on cloud albedo and cloud lifetime

Semi-direct aerosol effect

Reduction of Cloudiness by Soot?

Large eddy simulation studies guided by data taken during the Indian Ocean Experiment (INDOEX)

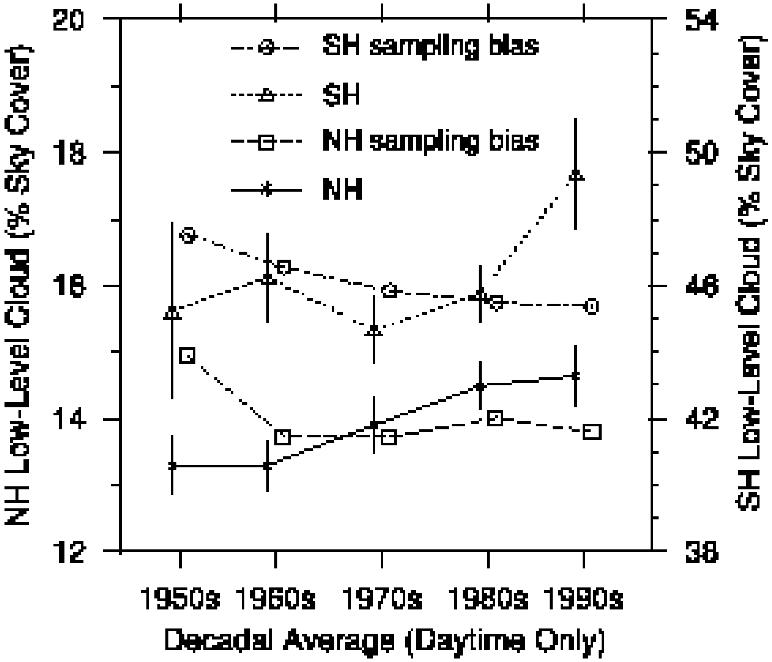
[Ackerman et al., 2000]

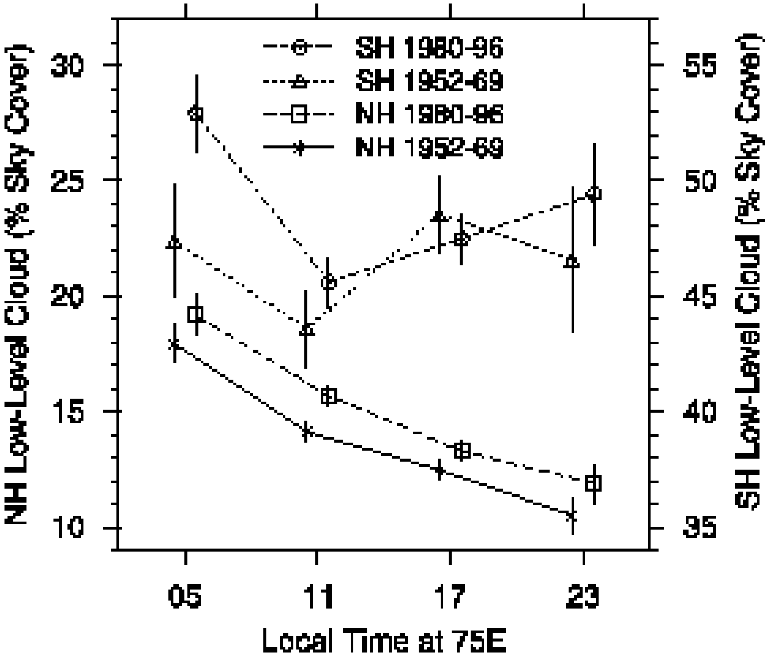


Reduction of Cloudiness by Soot?

Analysis of surface station cloud observations over the last 50 years

[Norris et al., 2001]

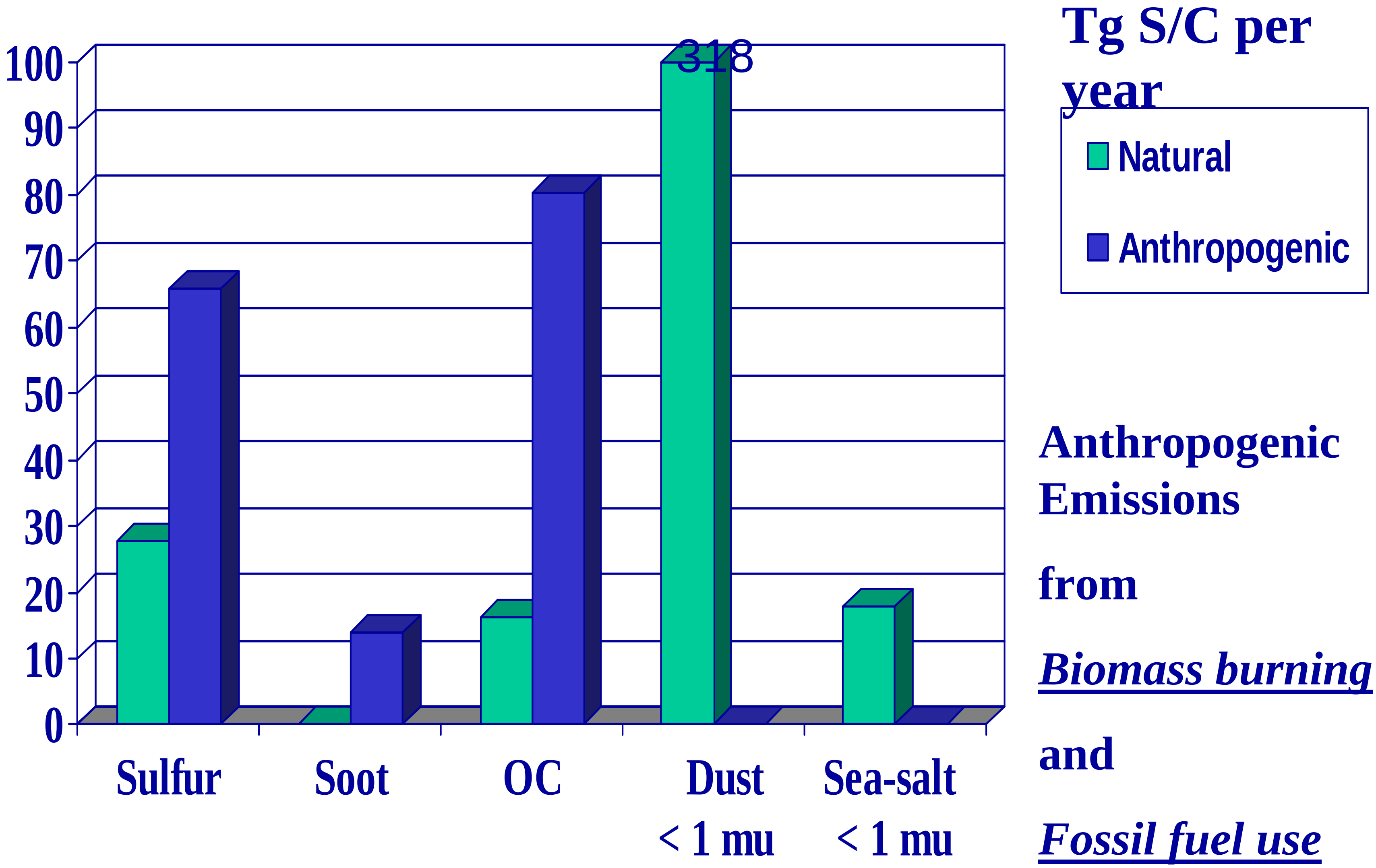




How Important is the Semi-Direct Aerosol Effect Globally? *[Lohmann and Feichter, 2001]*

- Pre-industrial (PI) simulations: No fossil fuel use and no biomass burning
- Indirect aerosol effect: Difference in shortwave radiation at the top of the atmosphere between present-day (PD) and PI simulations
- Conduct 3 pairs of 5 year T30 simulations:
- *INDIRECT*: Cloud albedo and cloud lifetime effect
- *DIRECT*: Direct and semi-direct aerosol effect
- *COMBINED*: All aerosol effects on water clouds

Global Mean Aerosol Emissions



Anthropogenic Emissions

from

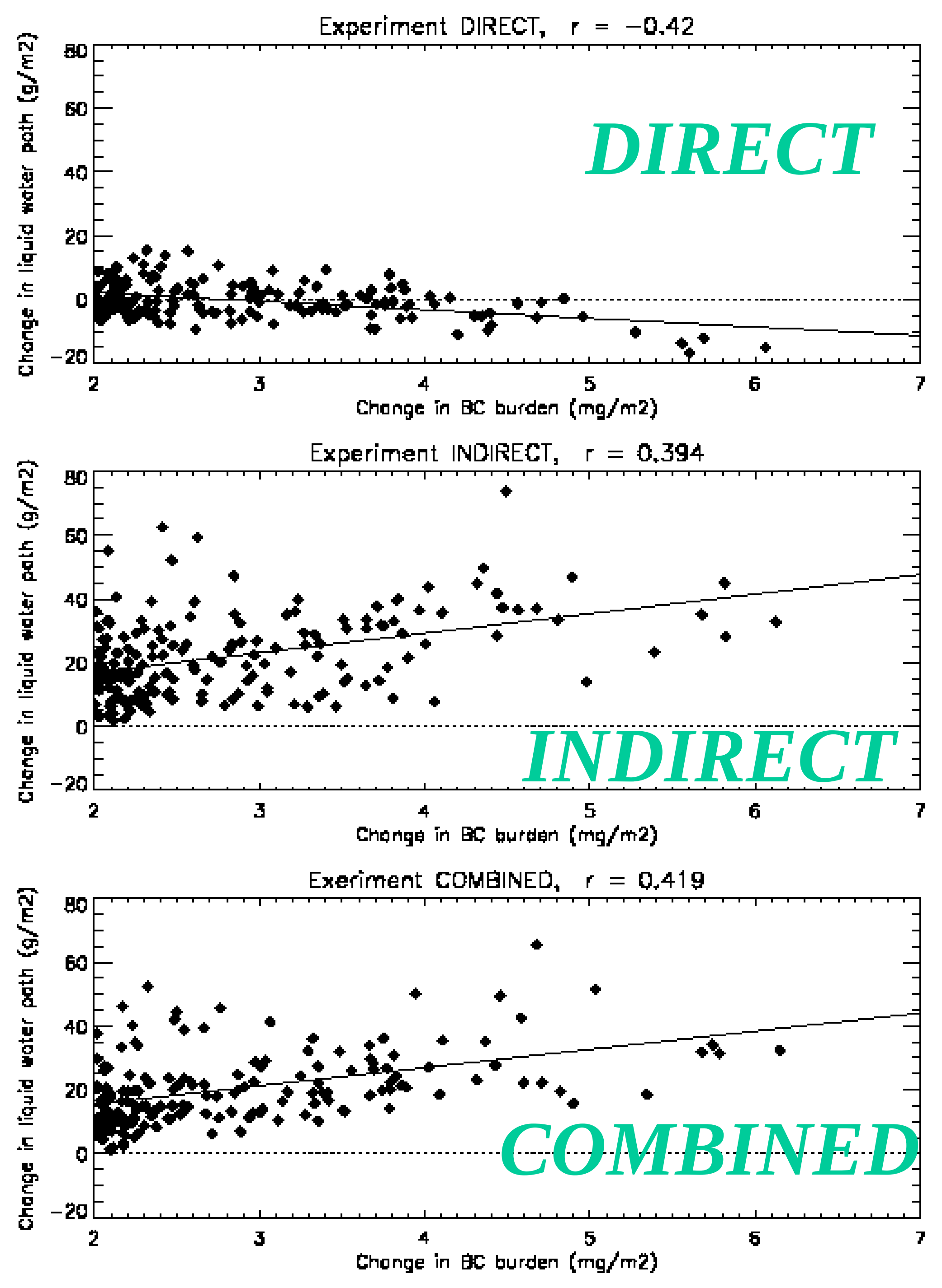
<u>Biomass burning</u>

and

<u>Fossil fuel use</u>

Correlation of the change in black carbon burden with LWP for the experiments *DIRECT*, *INDIRECT* and *COMBINED*

[Lohmann and Feichter, 2001]


Experiment DIRECT, r = −0.42

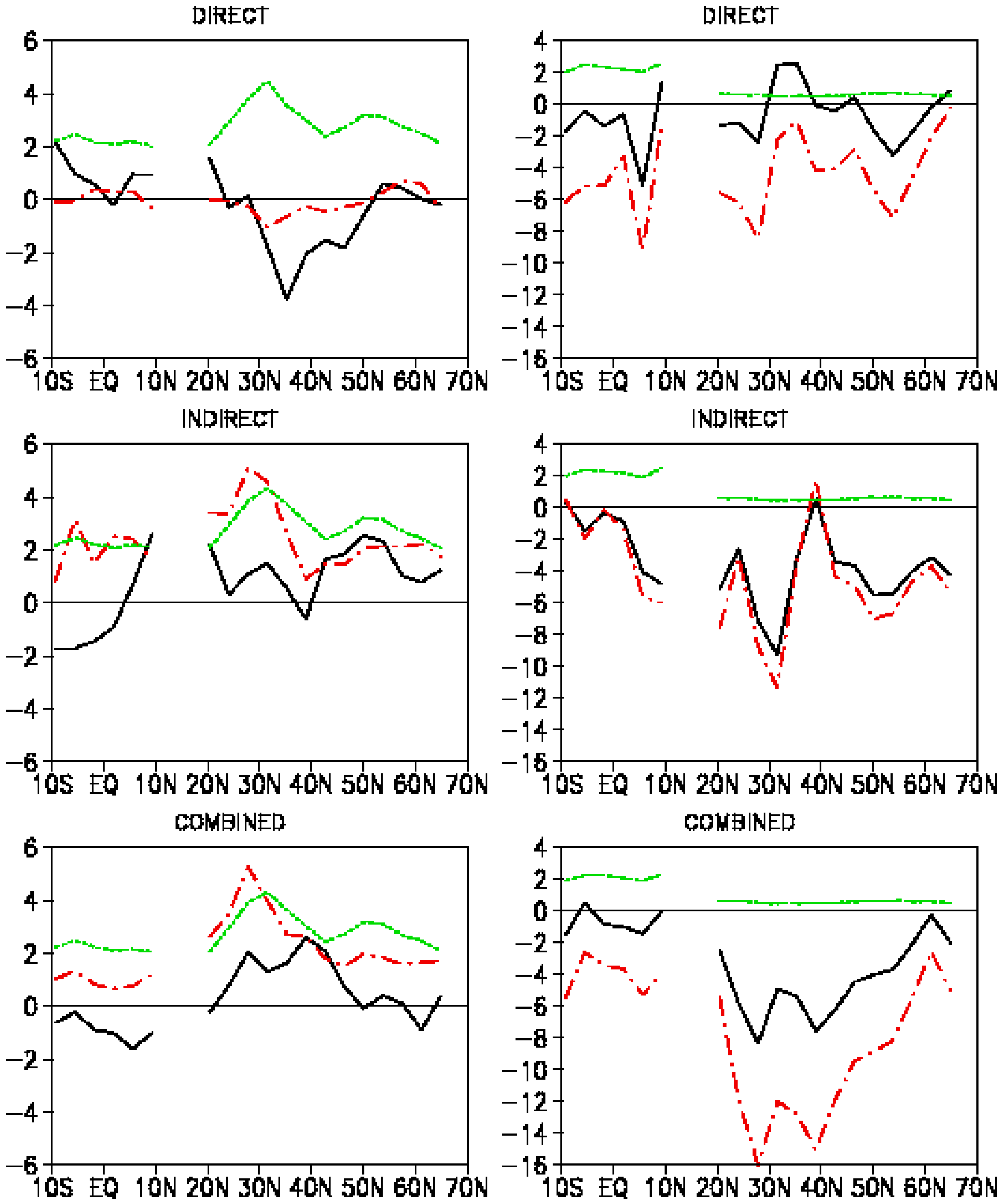
Experiment INDIRECT, r = 0.394

Exeriment COMBINED, r = 0.419

Annual zonal mean changes in BC burden [mg/m^2], cloud cover [%], LWP [0.1 g/m^2] (left),

ratio BC/sulfate burden, SW radiation [W/m^2] at TOA and surface (right)

[Lohmann and Feichter, 2001]



Annual global mean anthropogenic aerosol effects on water clouds

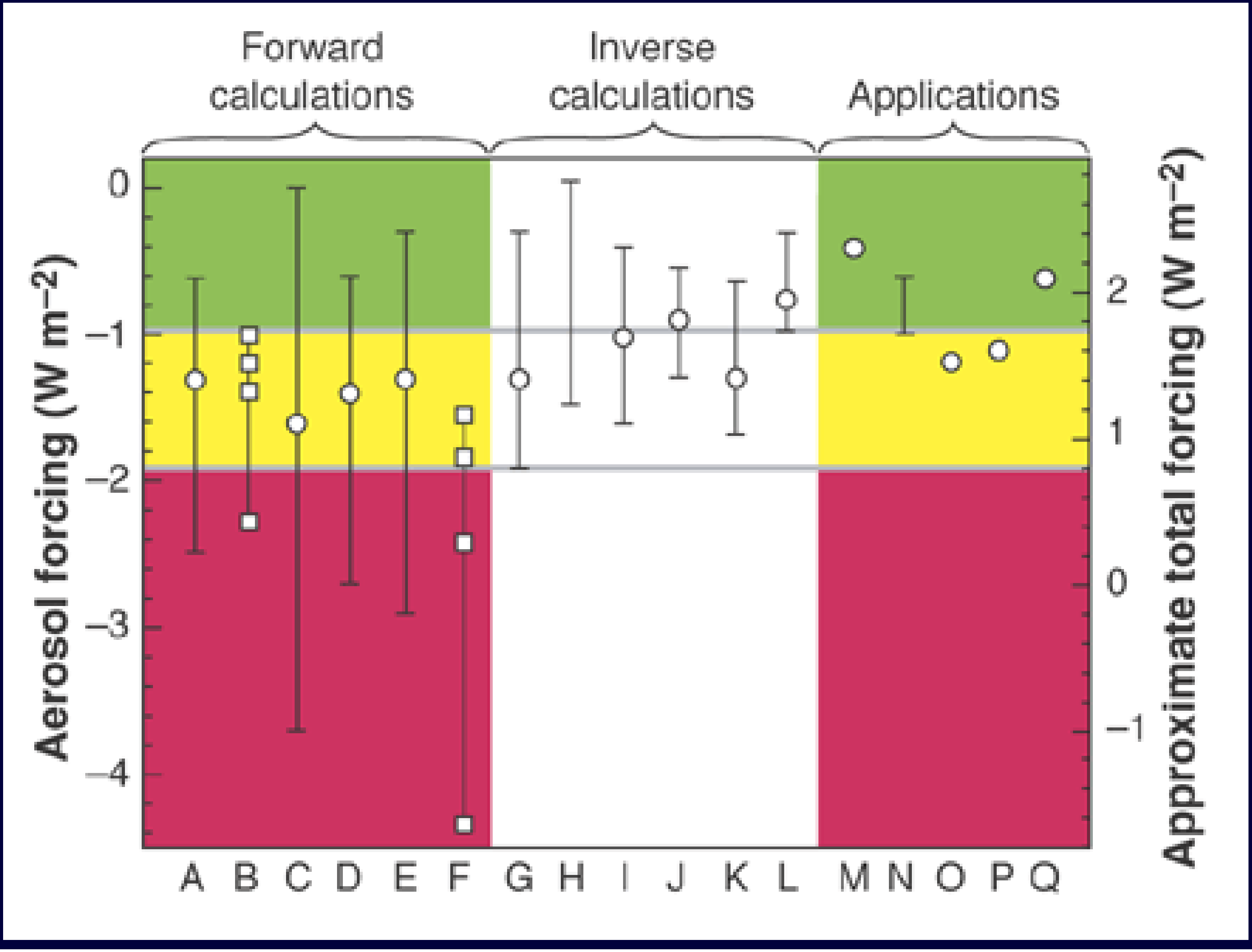
	COMBINED	INDIRECT	DIRECT
ΔAB (Tg)	2.6	2.6	2.5
ΔLWP (g/m^2)	9.5	10.1	-0.2
ΔTCC (%)	0.0	0.3	-0.2
ΔF_{SW}^{toa} (W/m^2)	-1.3	-1.4	-0.1
ΔF_{SW}^{sfc} (W/m^2)	-2.9	-1.8	-1.2

Conclusions: Semi-direct effect

- Black carbon can reduce the liquid water path considerably locally due to absorption of solar radiation, heating of the air and evaporation of cloud droplets
- However, the semi-direct effect is negligible as compared to the indirect aerosol effects on a global scale

Summary of Aerosol Forcing Estimates

[Anderson et al., 2003]

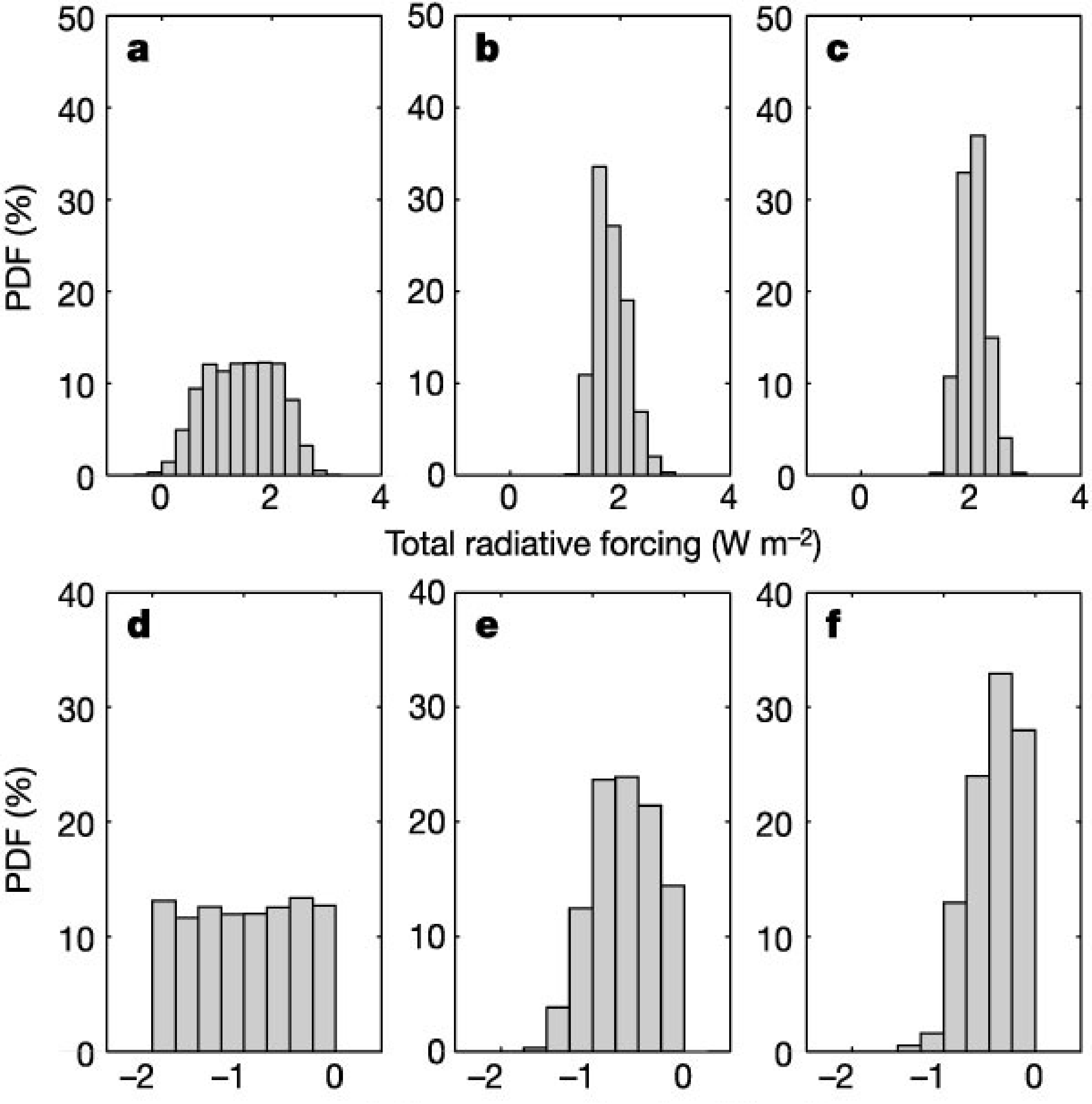


Example for Inverse Simulations

[Knutti et al., 2002]

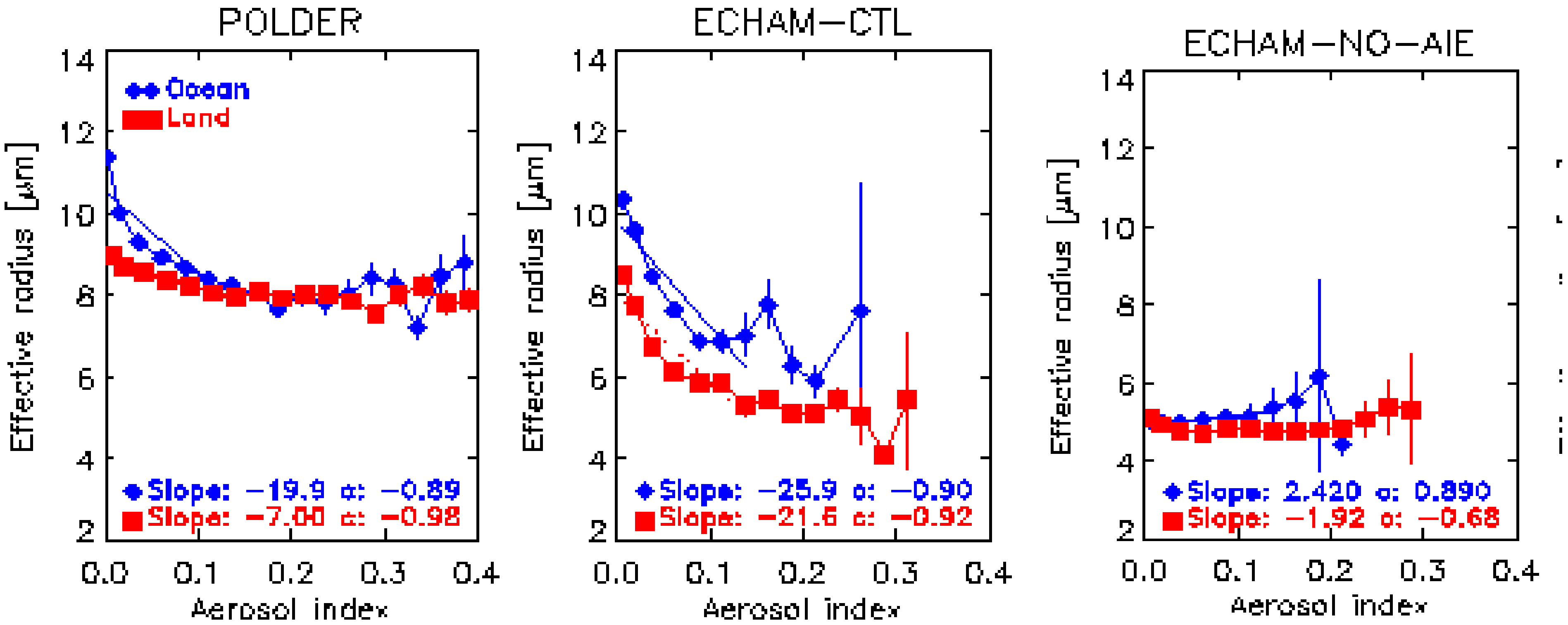
Probability density functions of the indirect effect estimated from past observations and a simple climate model if constrained by the IPCC climate sensitivity:

0 to –1.2 W/m²



Indirect aerosol effect: Slope of the cloud droplet radius as a function of the aerosol index

Lohmann and Lesins [2002]



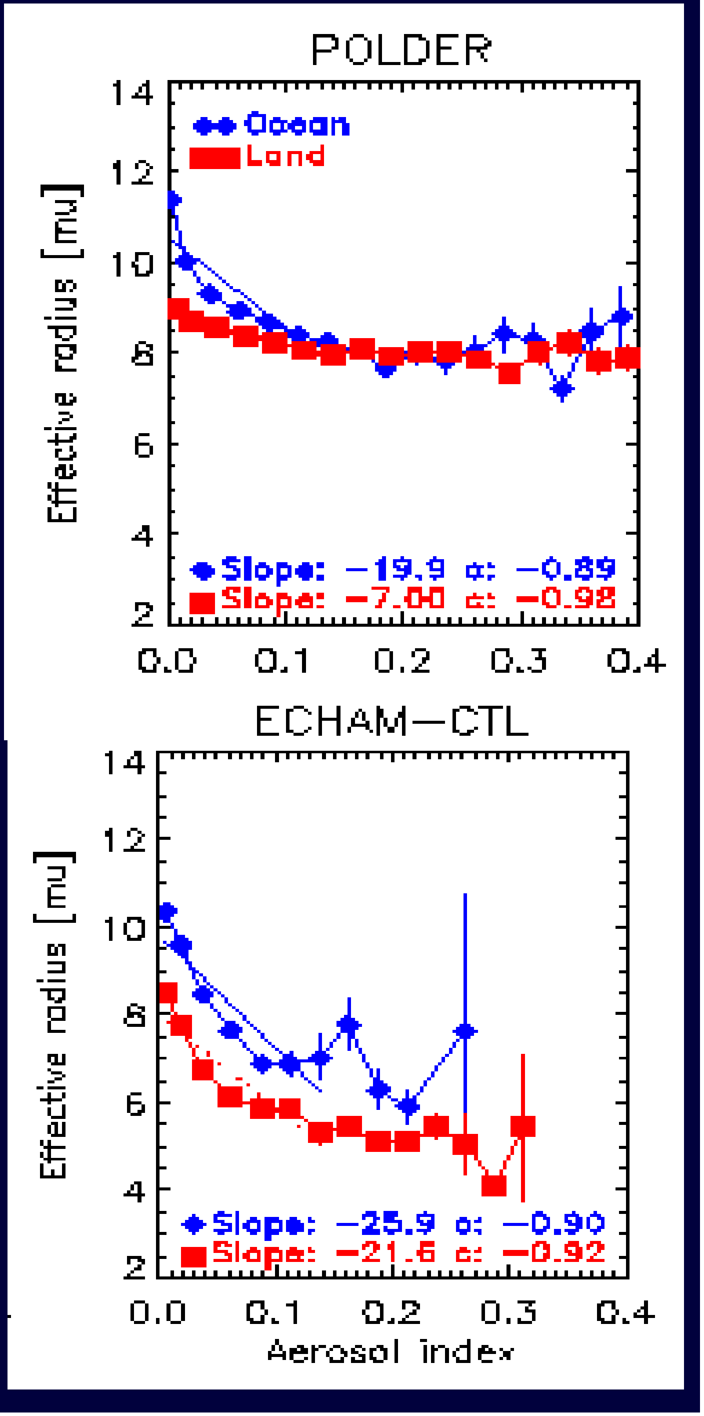
Polder data: Breon et al. [2002]

Change in net radiation at top of atmosphere (W m^{-2}) due to anthropogenic aerosol effects between pre-industrial and present-day times

	Original	Modified*
Ocean	-1.28	-0.98
Land	-1.62	-0.53
Global	-1.40	-0.85

*: after taking the difference in indirect aerosol effect from POLDER satellite data and ECHAM into account.

Lohmann and Lesins, Science [2002]

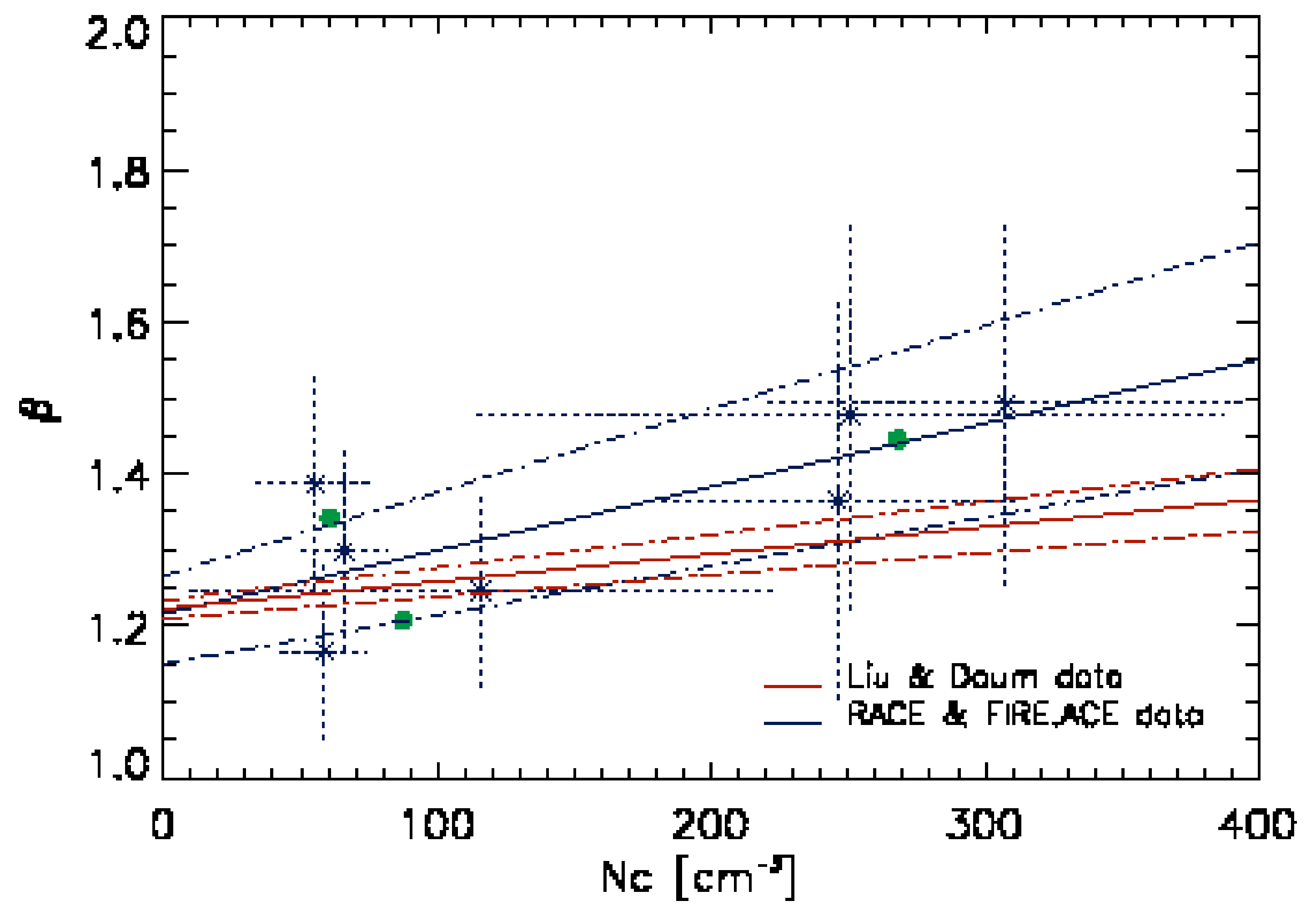


One explanation for discrepancy between ECHAM4 and POLDER: need to consider spectral dispersion β of cloud droplet size distribution

$$\beta=(1+2\varepsilon^2)^{2/3}/(1+\varepsilon^2)^{1/3}$$

ε=std.dev/ mean radius

$$r_e= \beta\,[3\,N / (4\pi\,\rho\,L)]^{1/3}$$

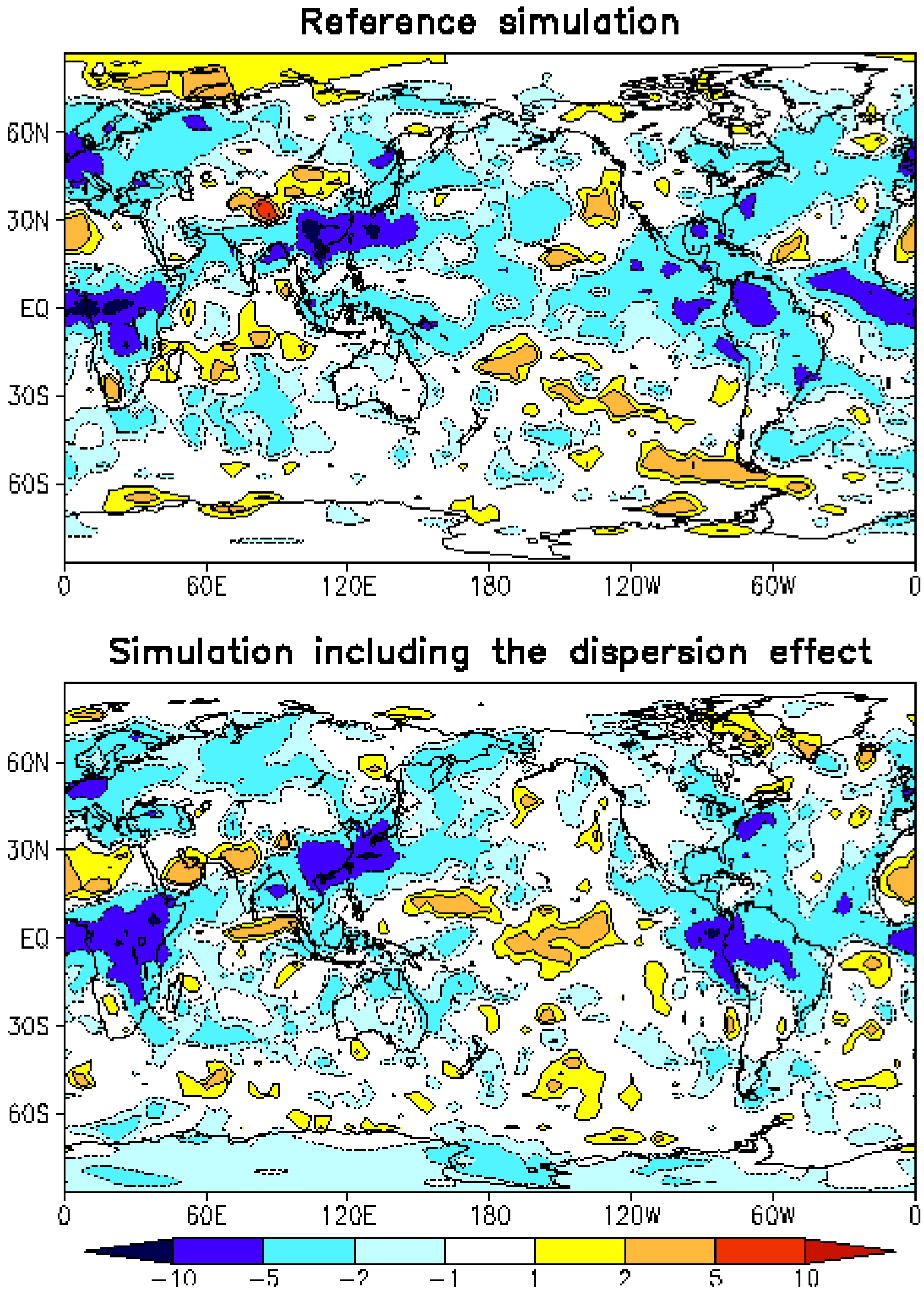


[Liu and Daum, 2002]

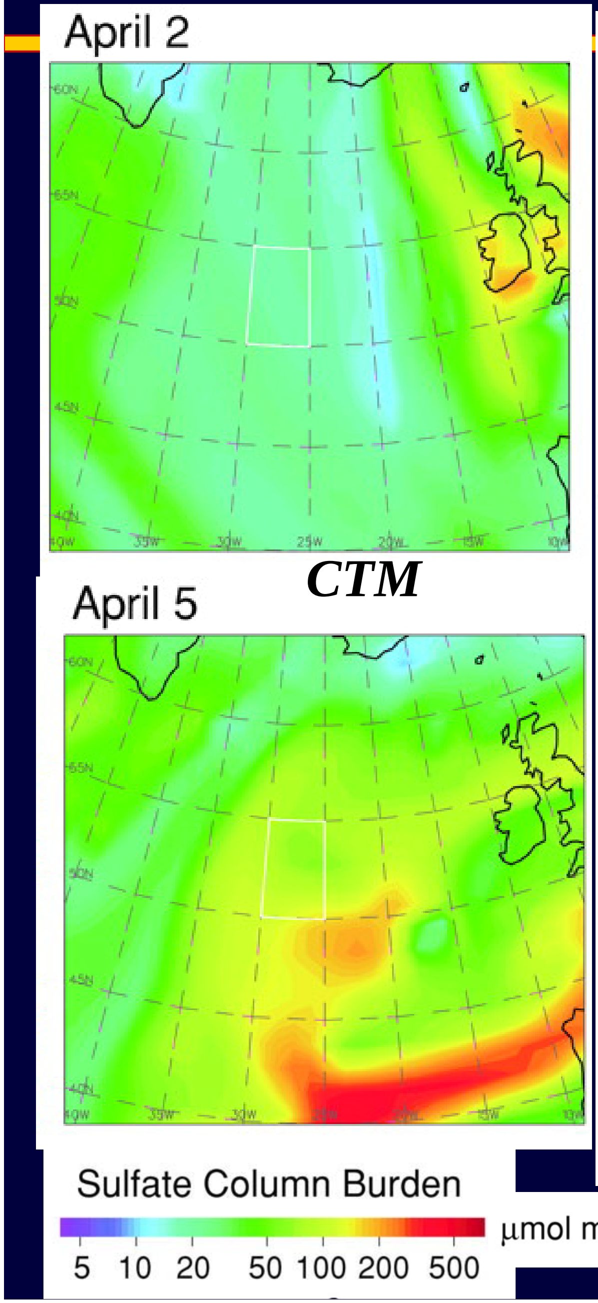
Global impact of the dispersion effect:

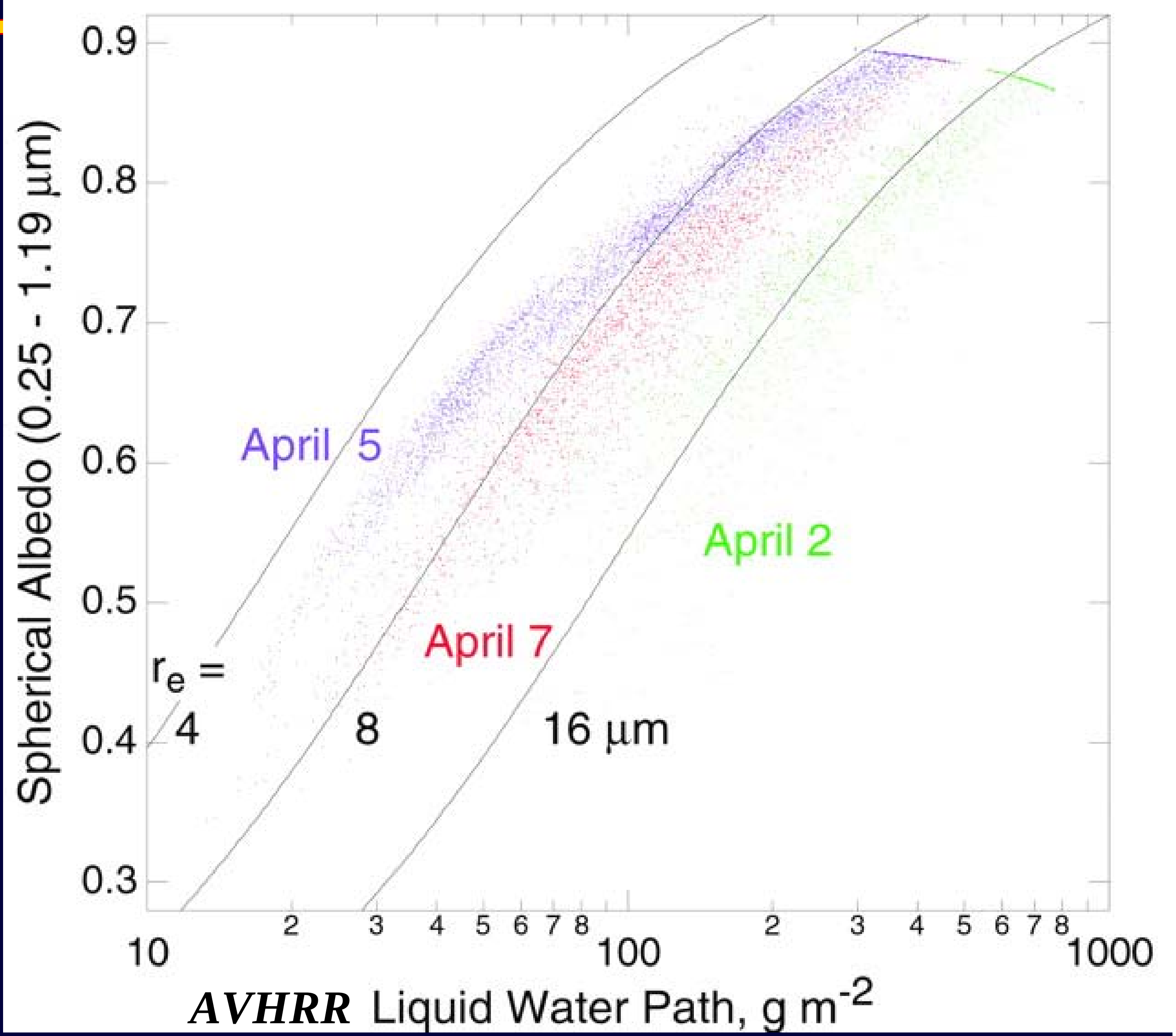
The anthropogenic indirect aerosol effect is reduced from –1.4 W/m² to –1.2 W/m²

[Peng and Lohmann, 2003]



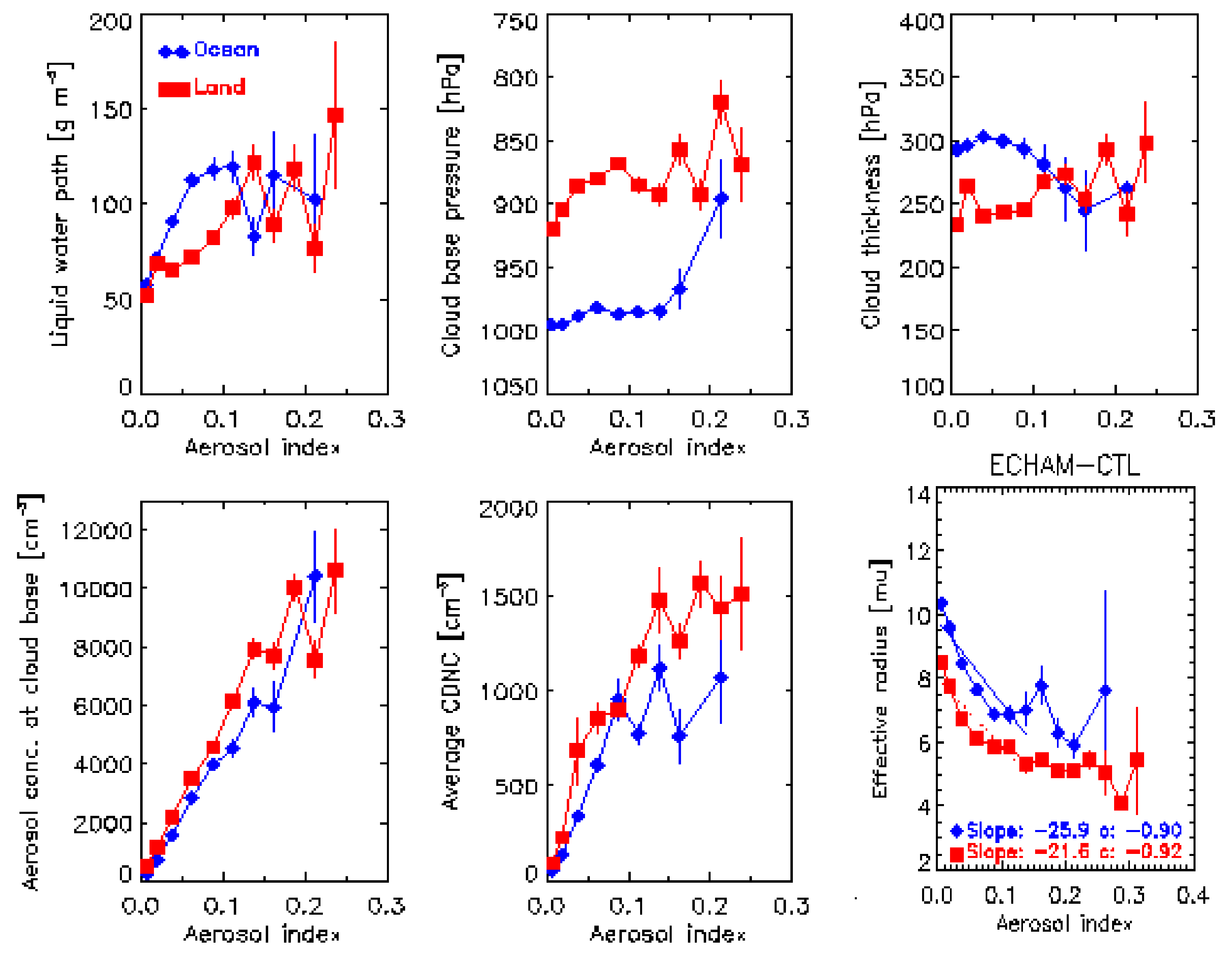
Aerosol indirect effect in the North Atlantic





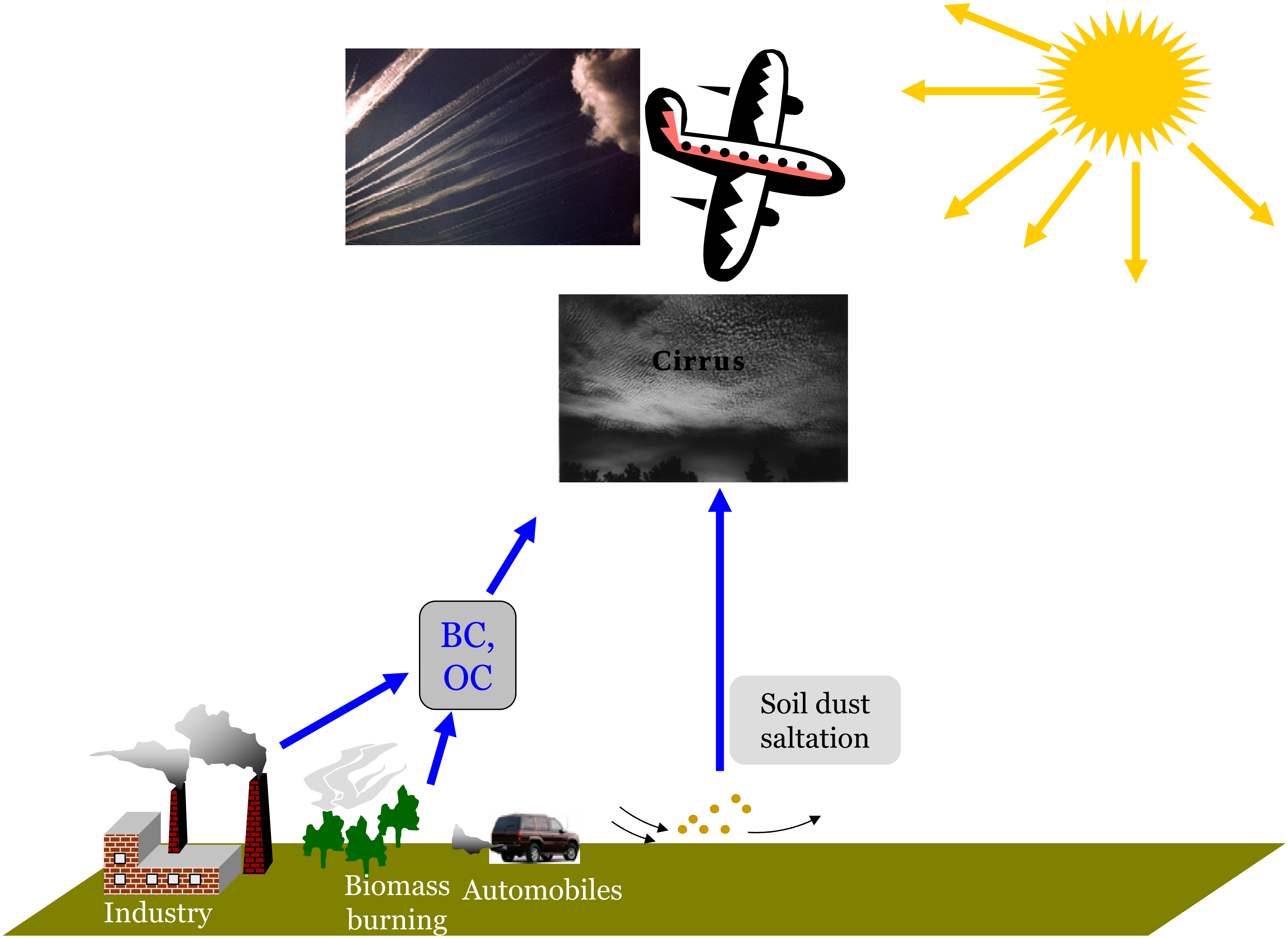
[Schwartz et al. 2002]

General differences between continental and maritime clouds *[Lohmann and Lesins, 2003]*



Conclusions to this Point:

- The *anthropogenic indirect aerosol effect* on the net radiation at the top of the atmosphere is reduced from –1.4 W m^{-2} to –0.85 W m^{-2} by combining ECHAM4 climate model and POLDER satellite data.
- This reduced estimate now agrees with past observations combined with a simple climate model of 0 to –1.2 W m^{-2} *[Knutti et al. 2002].*
- *Dispersion effect* explains 1/3 of this discrepancy.



Cirrus

BC,
OC

Soil dust
saltation

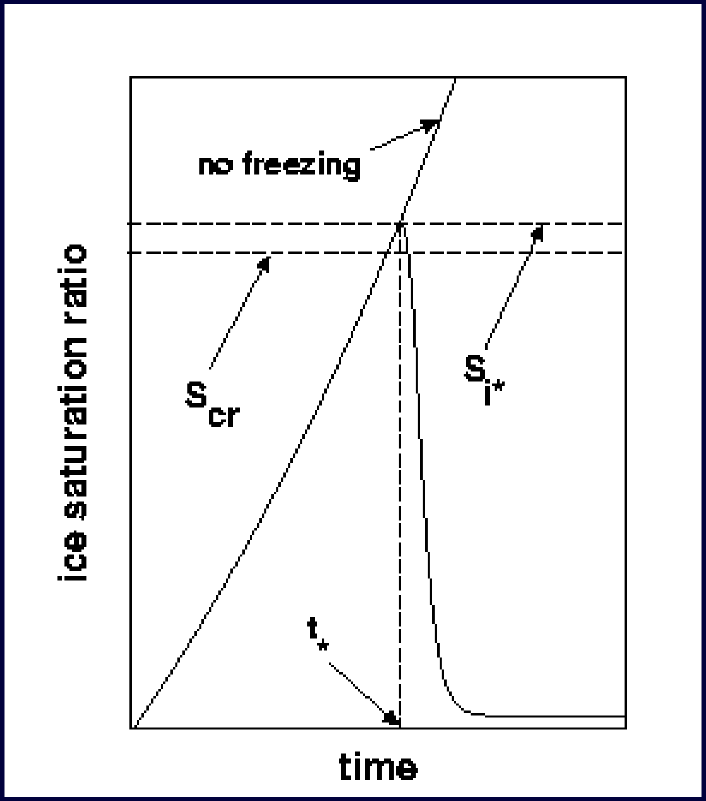
Industry

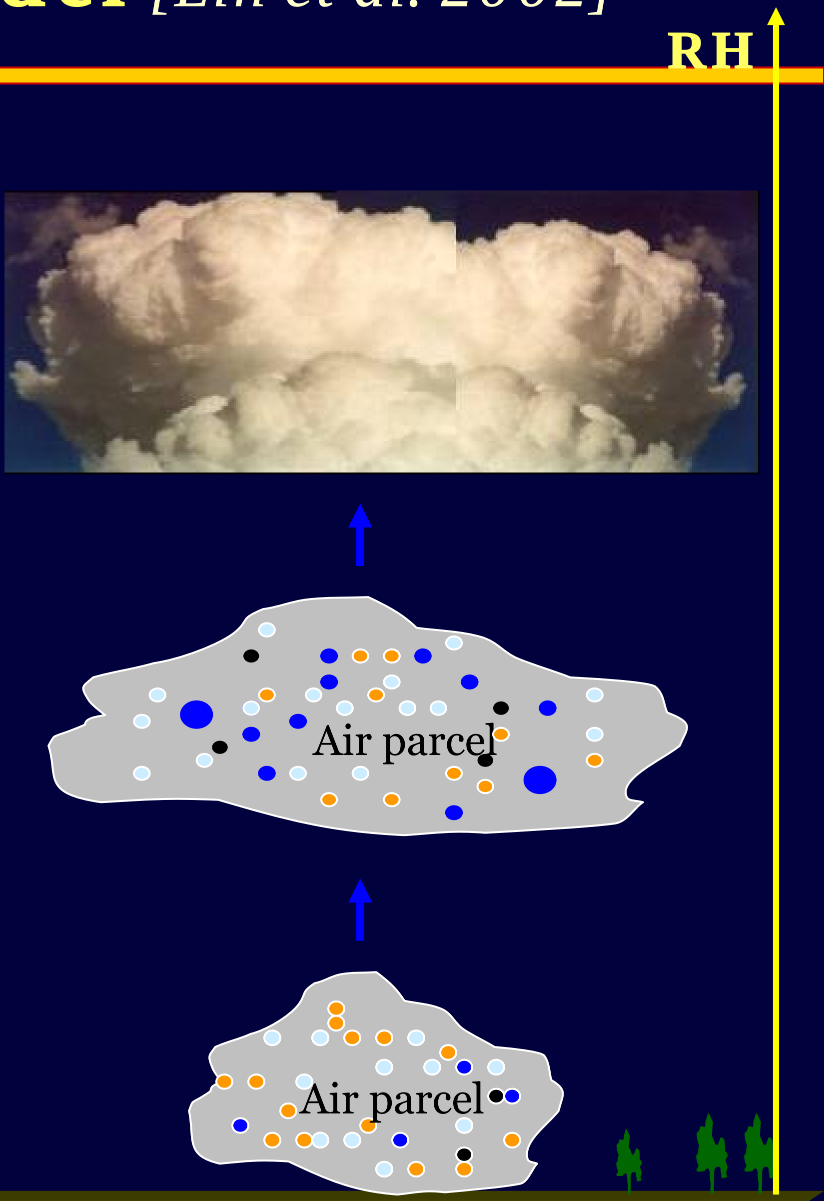
Biomass
burning

Automobiles

Adiabatic parcel model *[Lin et al. 2002]*

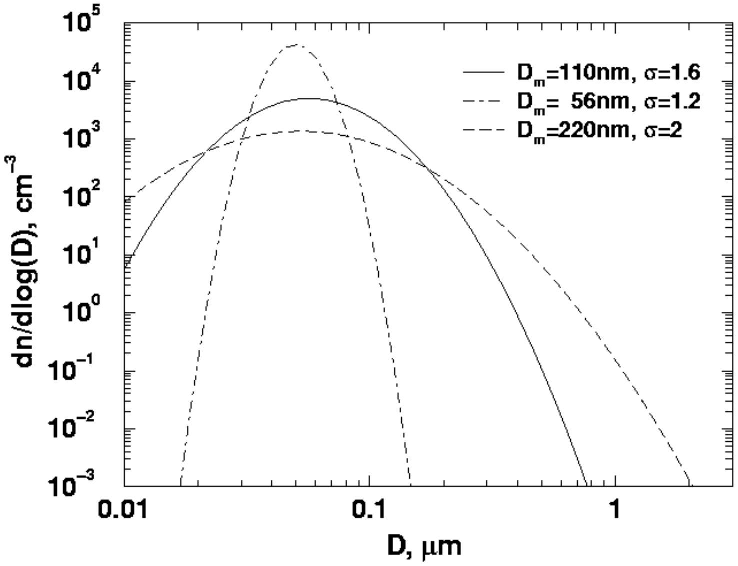
- **79 size bins for ice crystals**
- **56 size bins for aerosols**



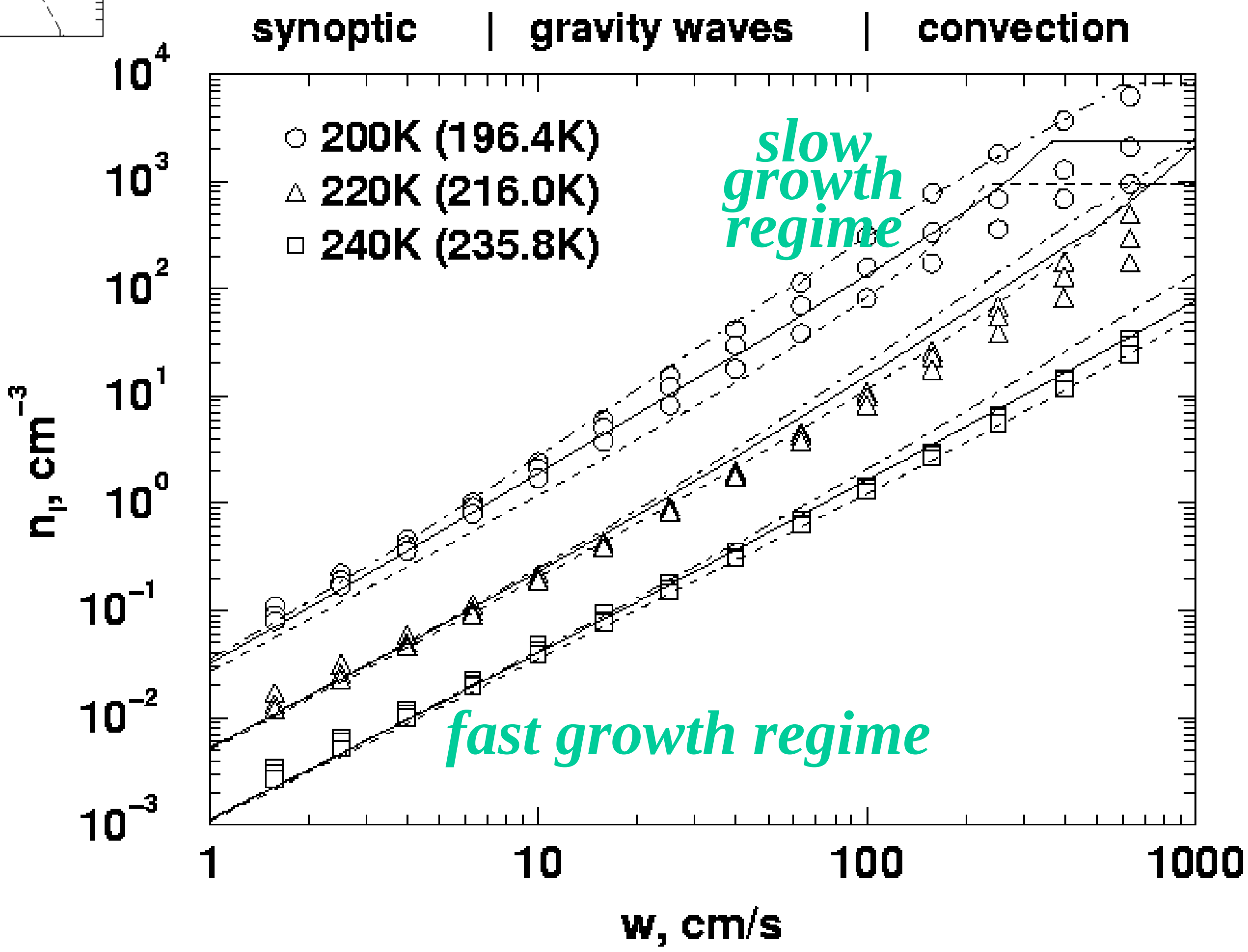


Homogeneous Freezing of Supercooled Aerosols

[Kärcher and Lohmann, 2002b]



Parcel model results (symbols), parameterization (lines)



Homogeneous Freezing of Supercooled Aerosols

[Kärcher and Lohmann, 2002a,b]

- Use an adiabatic parcel model to derive a parameterization of homogeneous freezing
- Obtain number of ice crystals n_i at critical supersaturation S_{cr}:

$$n_i = m_w/\rho_i\ [b_2/(2\pi\, b_1)]^{3/2}\ a_1\, S_{cr}/(a_2 + a_3\, S_{cr})\ w\, \tau^{-1/2}$$

w = updraft velocity,

τ = freezing time scale,

a_k, b_k = coefficients

- Note: No explicit dependence of nucleation rate on aerosol particle concentration n_a, but n_a serves as upper bound

Cloud Droplet vs. Ice Crystal Nucleation

- $n_d \sim n_a^{0.8}\, w^{3/10}\, n_{sat}^{-1/10}$ (typical for marine aerosols)
- $n_i \sim n_a^{0}\, w^{3/2}\, n_{sat}^{-1/2}$ ➔ weak aerosol effect for cirrus (valid for fast growth regime)
- $n_i \sim r_o^{-1}$ (slow growth regime)

- where: n_a = aerosol particle concentration, n_{sat} = water vapor saturation pressure over ice (number density), r_0 = aerosol or ice particle radius at time t_o

Which Aerosols Act as Ice Nuclei?

Liquid ammonium sulfate particles with solid kaolinite inclusions:

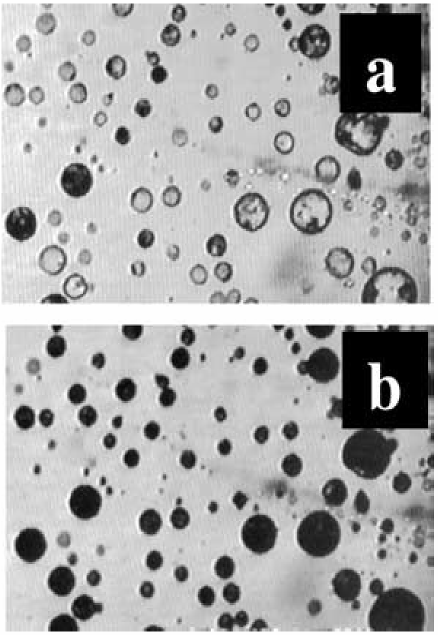
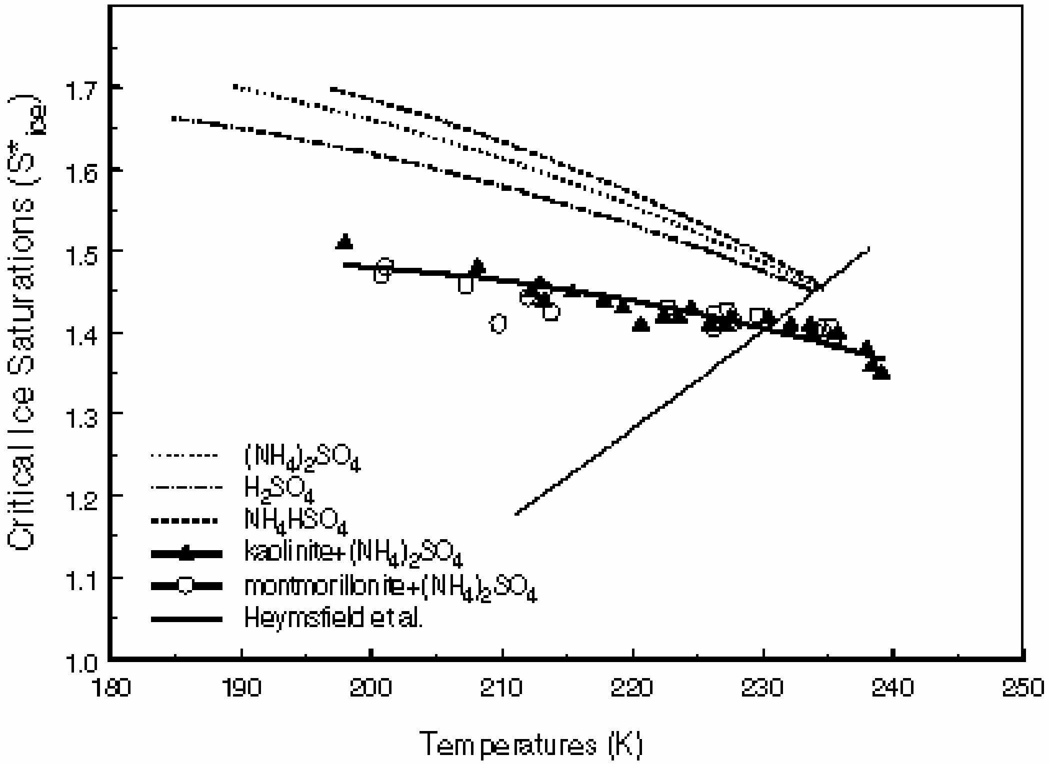


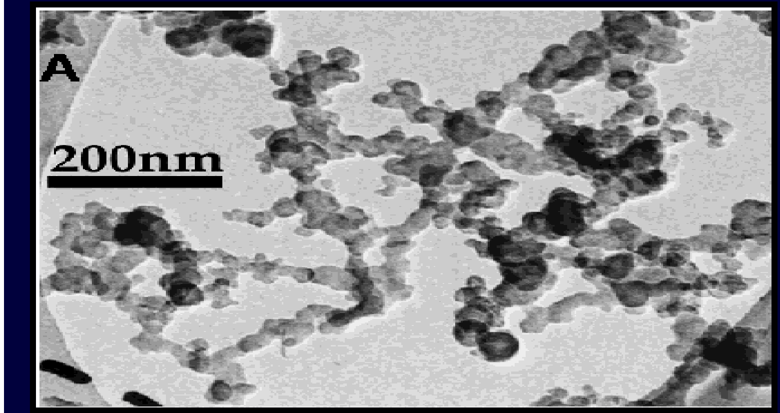
Figure 1. (a) Liquid ammonium sulfate particles with solid kaolinite inclusions. (b) Same particles with kaolinite inclusions, after freezing.



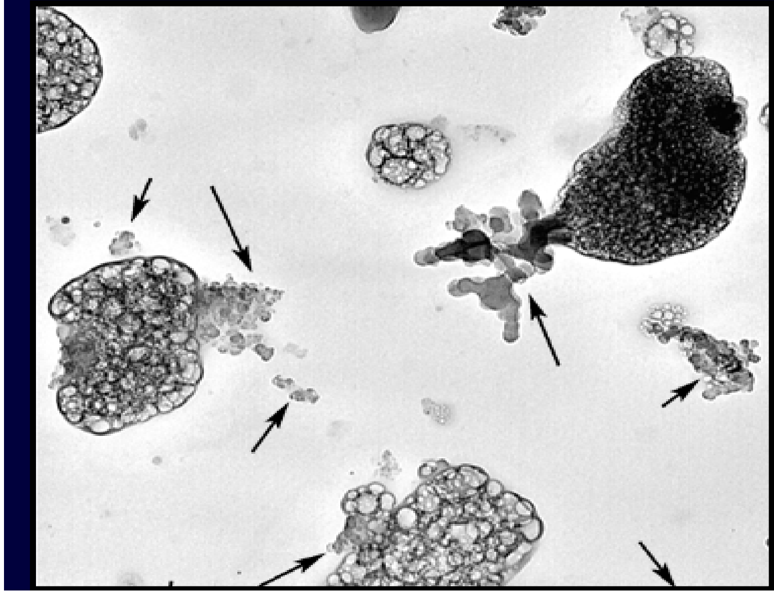
[Zuberi et al., 2002]

Which Aerosols Act as Ice Nuclei?

Soot:



Soot covered by sulfuric acid:



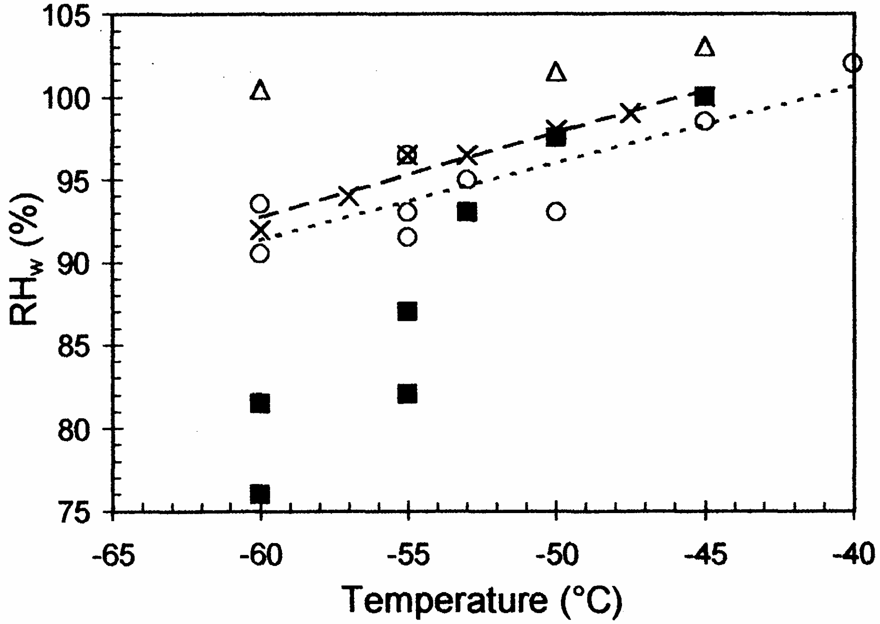


Figure 4. Summary of conditions for ice formation by 1% of soot particles subjected to different treatments. Linear regression fits to the data from experiments with untreated soot (o) and soot treated with an approximate monolayer of H_2SO_4 (×) are indicated, respectively, by the short-dashed ($r^2 = 0.95$) and long-dashed ($r^2 = 0.73$) lines. Experiments with multi-layer H_2SO_4 coverage are indicated by the filled square symbols. Experimental conditions for ice formation on small (<20 nm) pure sulfuric acid particles are given by triangle symbols.

[DeMott et al., 1999]

Heterogeneous Freezing of Supercooled Aerosols

[Water activity a_w ~ relative humidity]

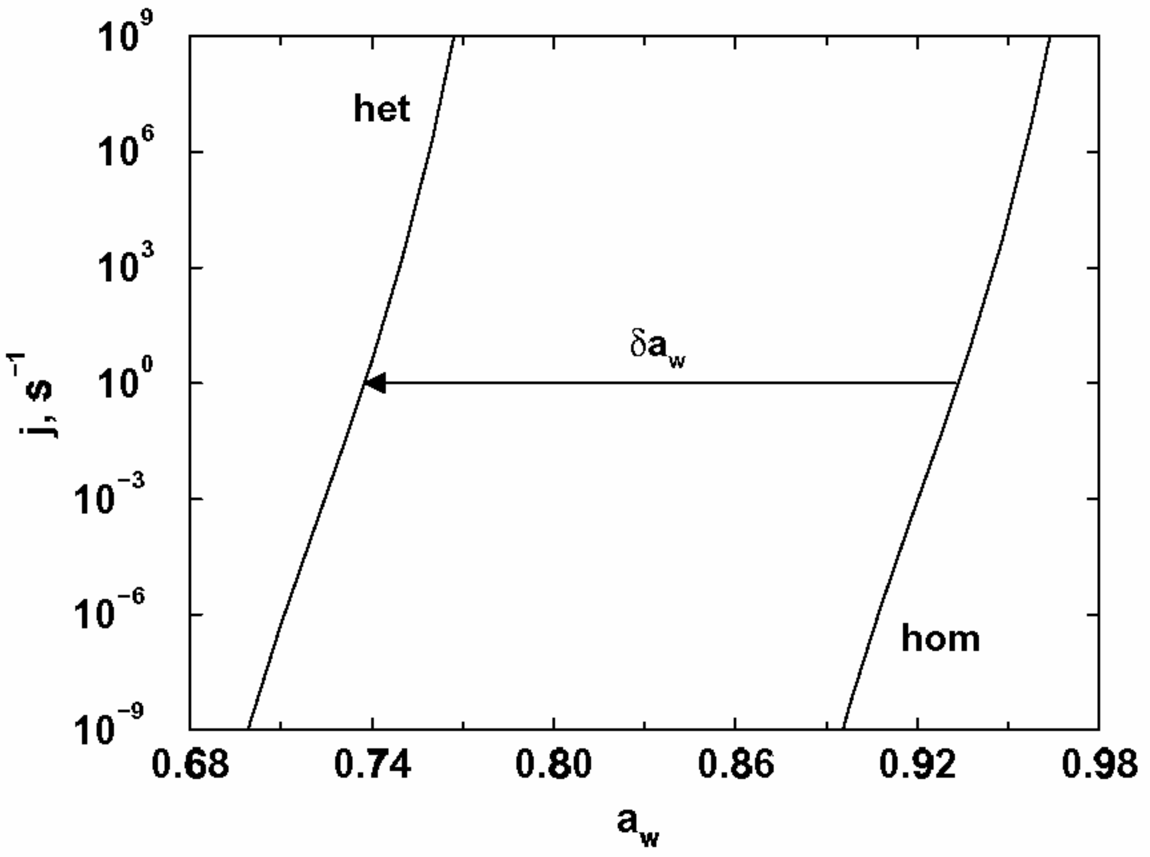


Figure 2. Nucleation rates per particle per second versus water activity at 220 K. Homogeneous (right curve) and heterogeneous (left curve) are shifted by the amount $\delta a_w \simeq 0.2$ resulting from $S_{\rm cr}^{\rm het} = 1.2$. The same particle that freezes homogeneously at $a_w \simeq 0.93$ would freeze heterogeneously at $a_w \simeq 0.73$ under these conditions.

Heterogeneous Freezing of Supercooled Aerosols

[Kärcher and Lohmann, 2003]

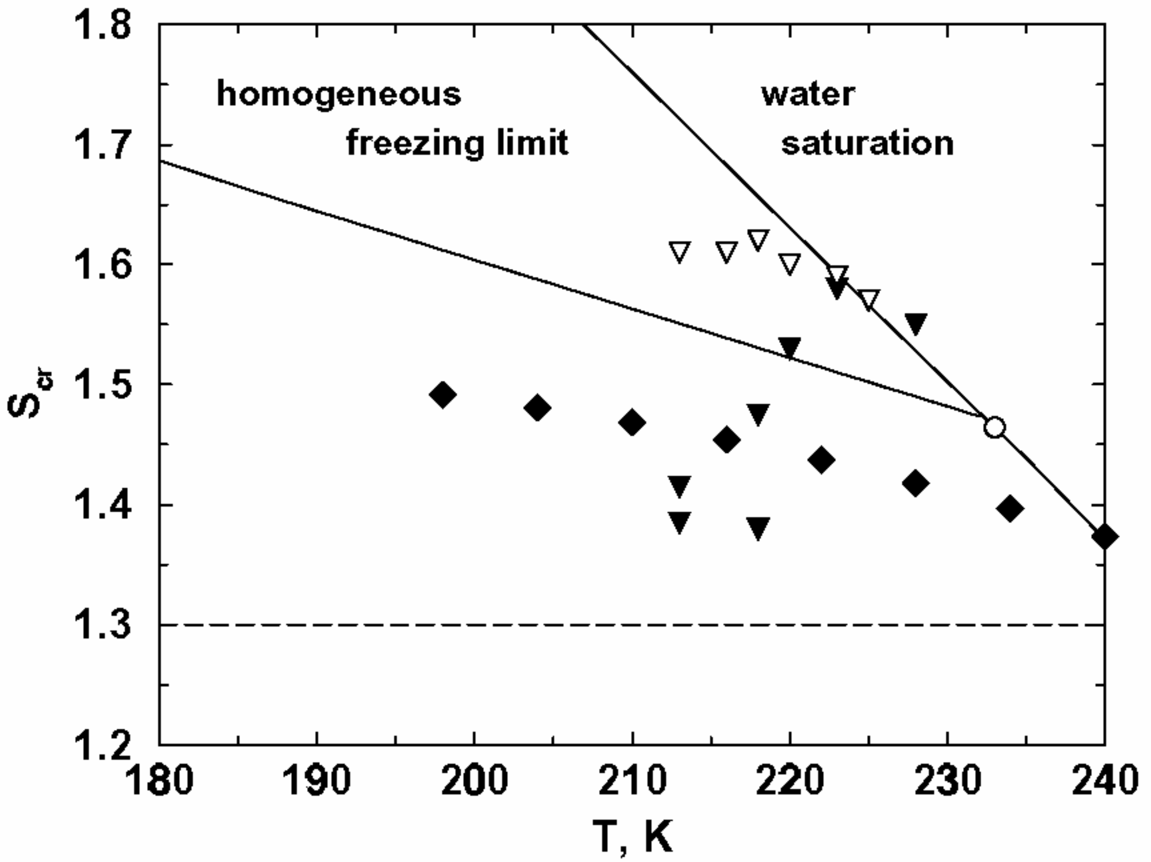


Figure 3. Freezing threshold saturation ratios over ice versus temperature for homogeneous freezing (upper left solid curve) from (5) and for a constant value of 1.3 (dashed line). The upper right solid curve shows the ice saturation ratios where liquid water would be saturated. Homogeneous freezing at temperatures to the right of the open circle occurs at water saturation. Also shown are experimental data for black carbon (BC) particles from DeMott et al. [1999] (filled/open triangles for multilayer/monolayer coverage with H_2SO_4) and for ammonium sulfate particles with mineral dust immersions from Zuberi et al. [2002] (diamonds). The latter authors have fitted their data to a shifted water activity.

Ice Crystal Concentration for Heterogeneous Versus Homogeneous Freezing of Supercooled Aerosols

[Kärcher and Lohmann, 2003]

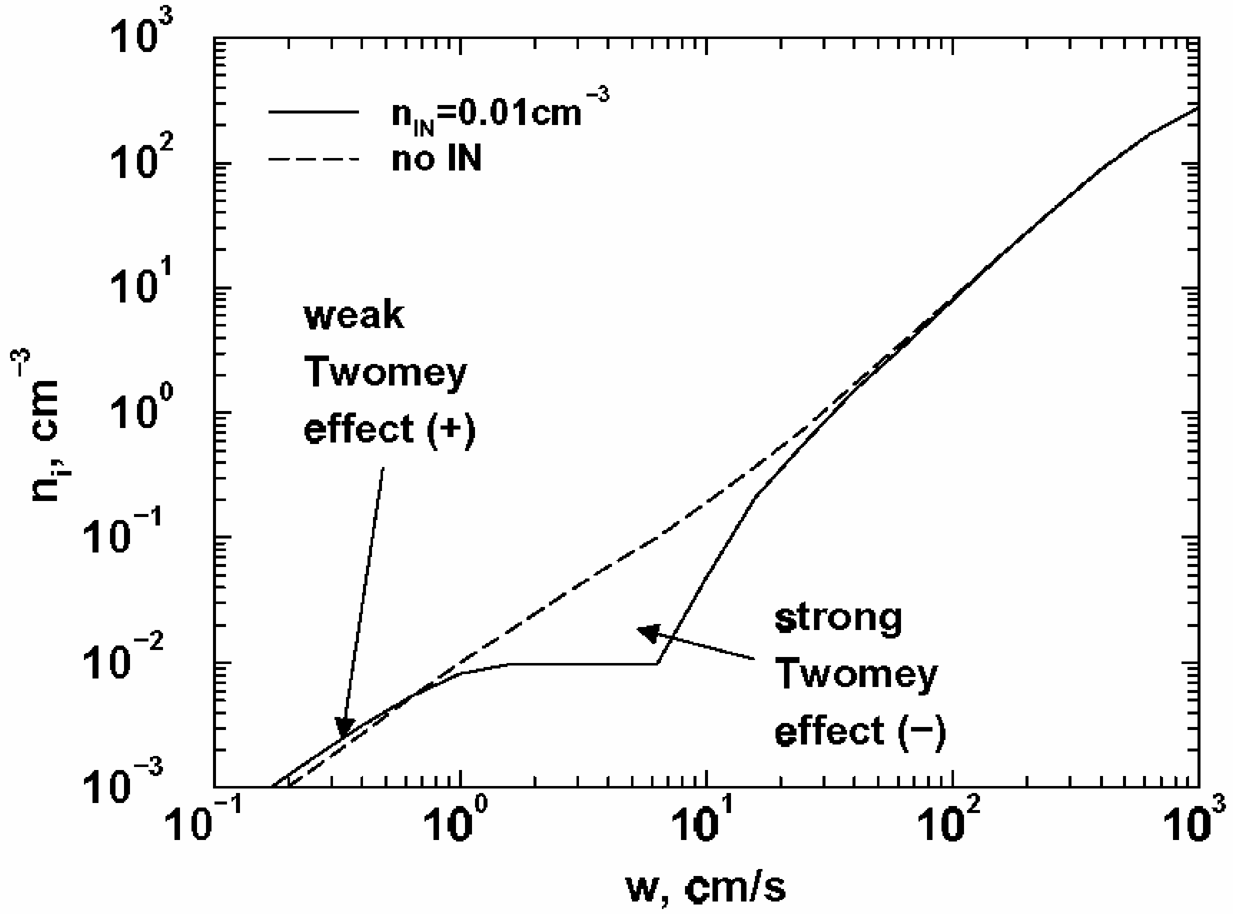


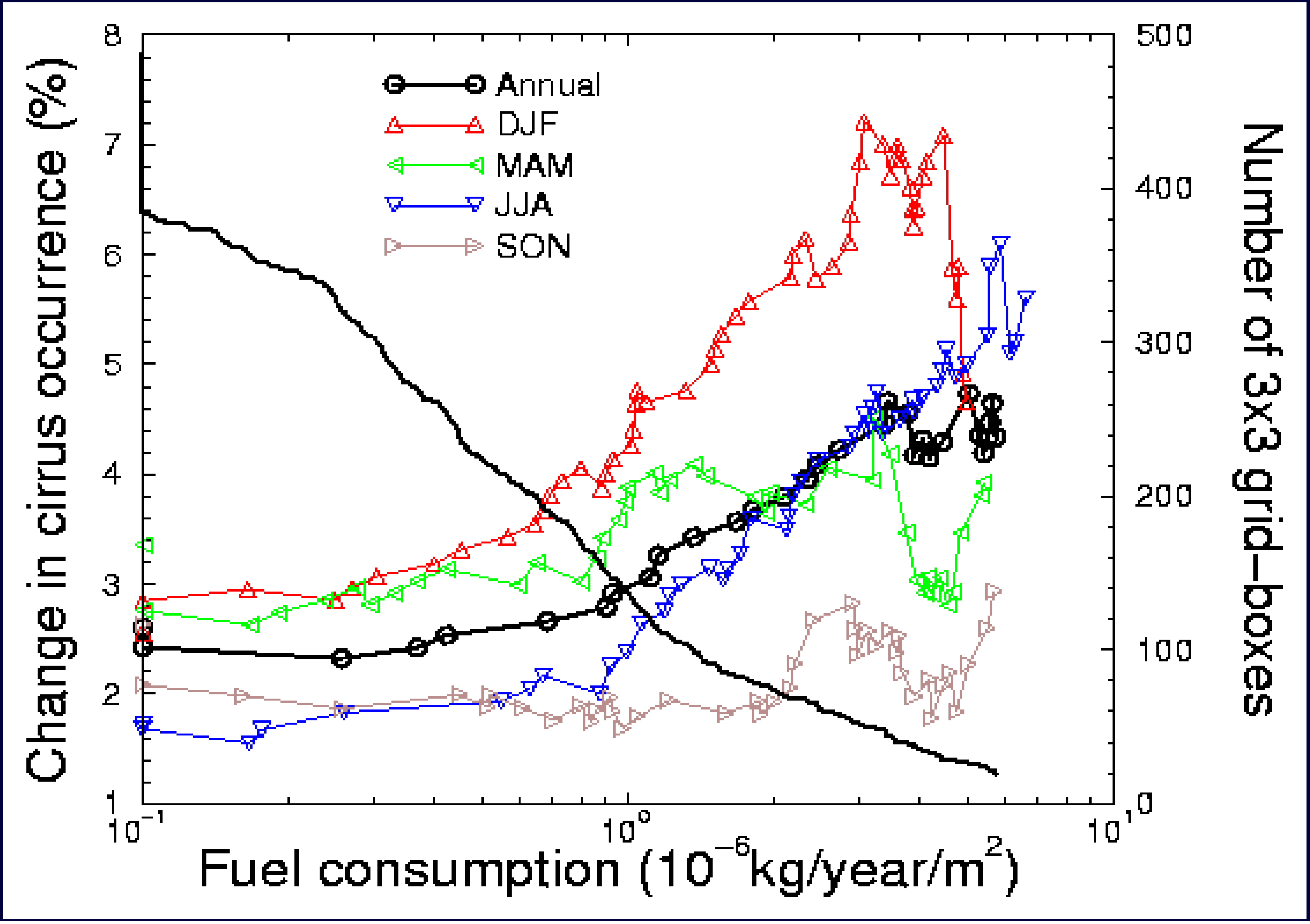
Figure 9. Number density of ice particles versus vertical velocity from APSC simulations starting at 225 K and ice saturation. In the model, 400 cm^{-3} homogeneous nuclei are present with 0.01 cm^{-3} (solid curve) and without (dashed curve) heterogeneous IN with $S_{\mathrm{cr}}^{\mathrm{het}} = 1.3$.

Conclusions to this Point:

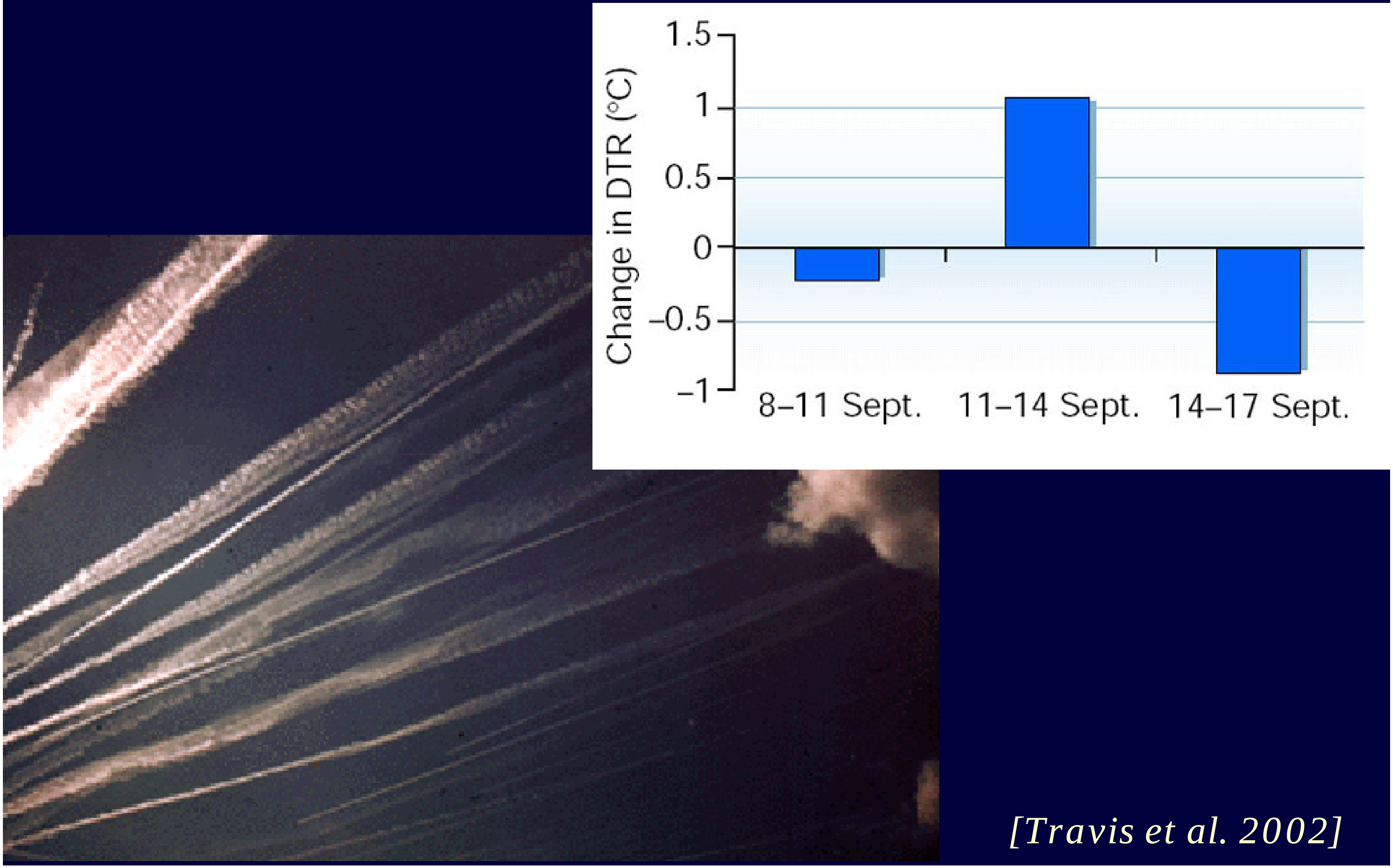
- If only one type of less potent ice nuclei (IN) with freezing saturation ratios above 1.3-1.4 triggers cirrus formation, cloud properties are not very susceptible to changes of IN properties, as in the case of homogeneous freezing.
- However, a much stronger indirect aerosol effect on cirrus clouds is possible if several IN types with distinct freezing thresholds compete during the freezing process, most likely leading to a suppression of the ice crystal concentration.

Air Traffic may Increase Cirrus Cloudiness

[Boucher, 1999]

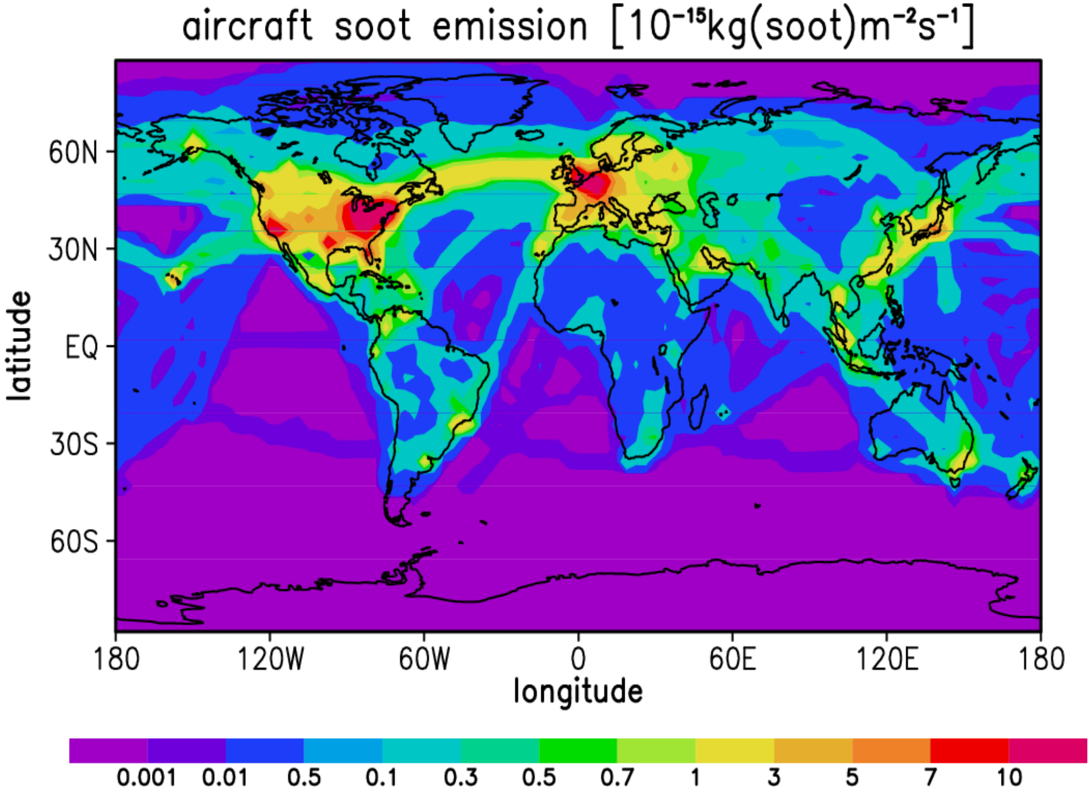


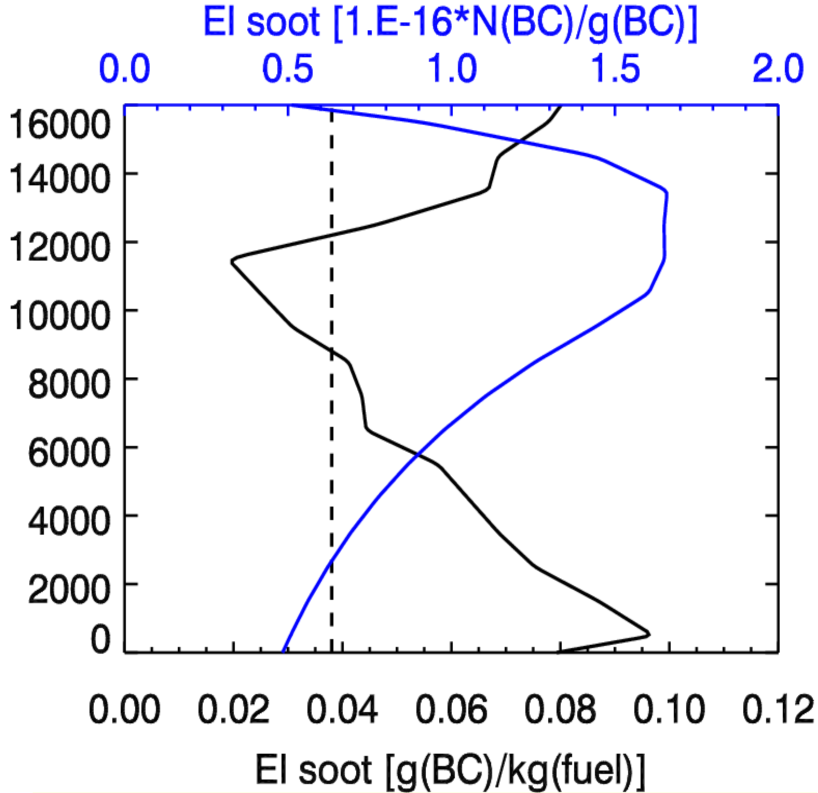
Contrails Reduce Diurnal Temperature Range



[Travis et al. 2002]

Global Simulation of Aircraft Soot Emissions with ECHAM4 *[Hendricks et al. 2003]*





Soot emissions (1992)

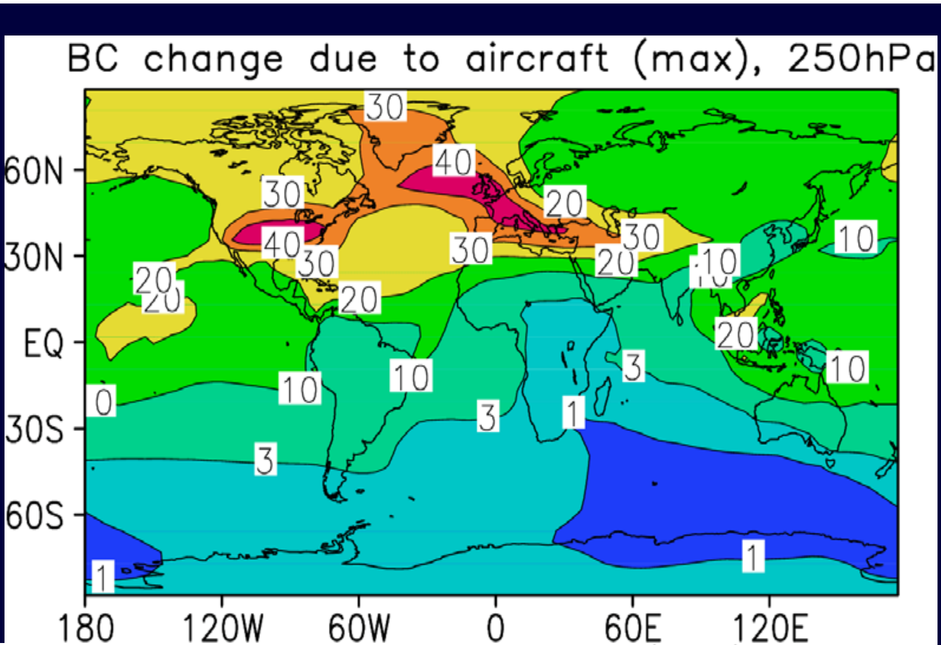
Soot emission index (1992)
[Mass, Particle number]

Source: Schmitt und Brunner [1997]

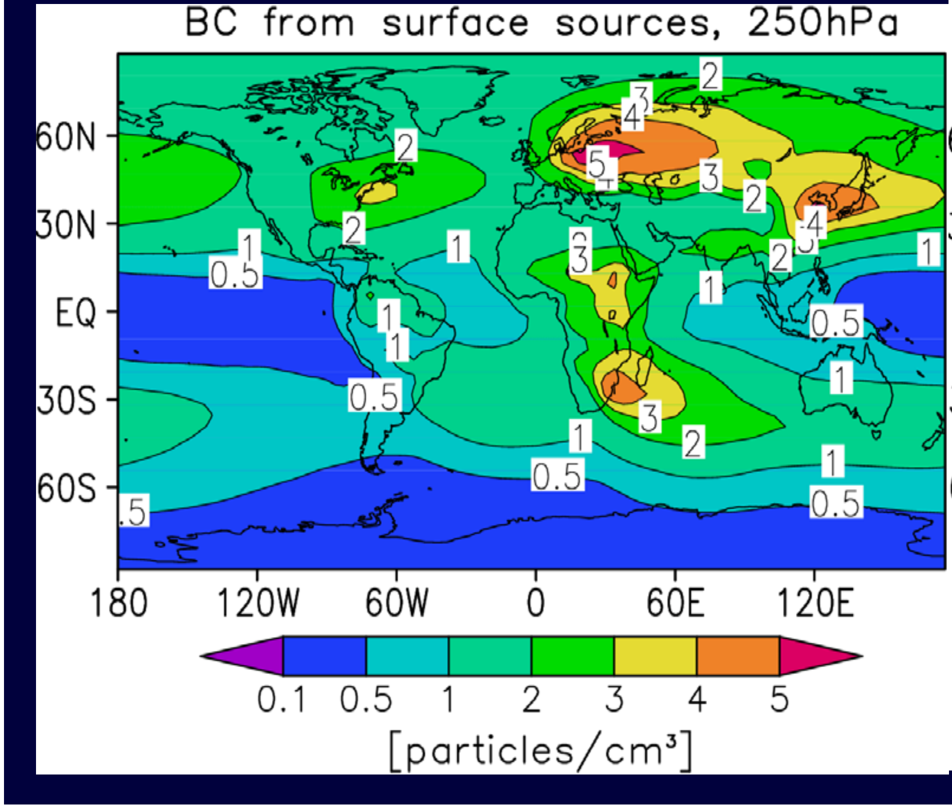
Source: Döpelheuer [2002]

Global simulation of aircraft soot emissions with ECHAM

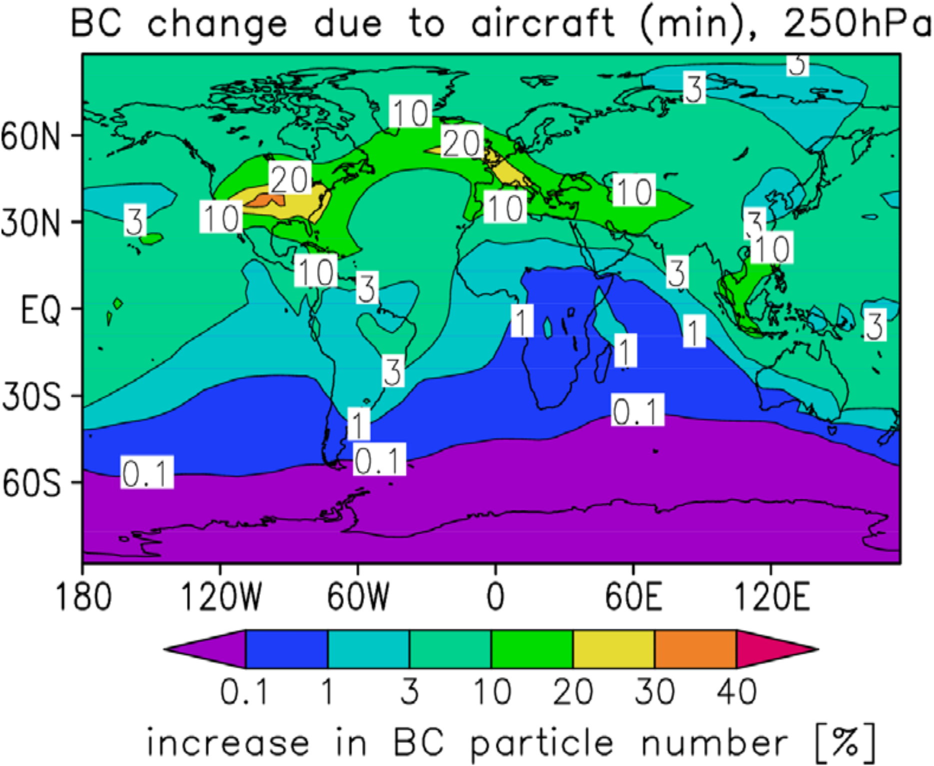
[Hendricks et al. 2003]



BC change due to aircraft (max), 250hPa

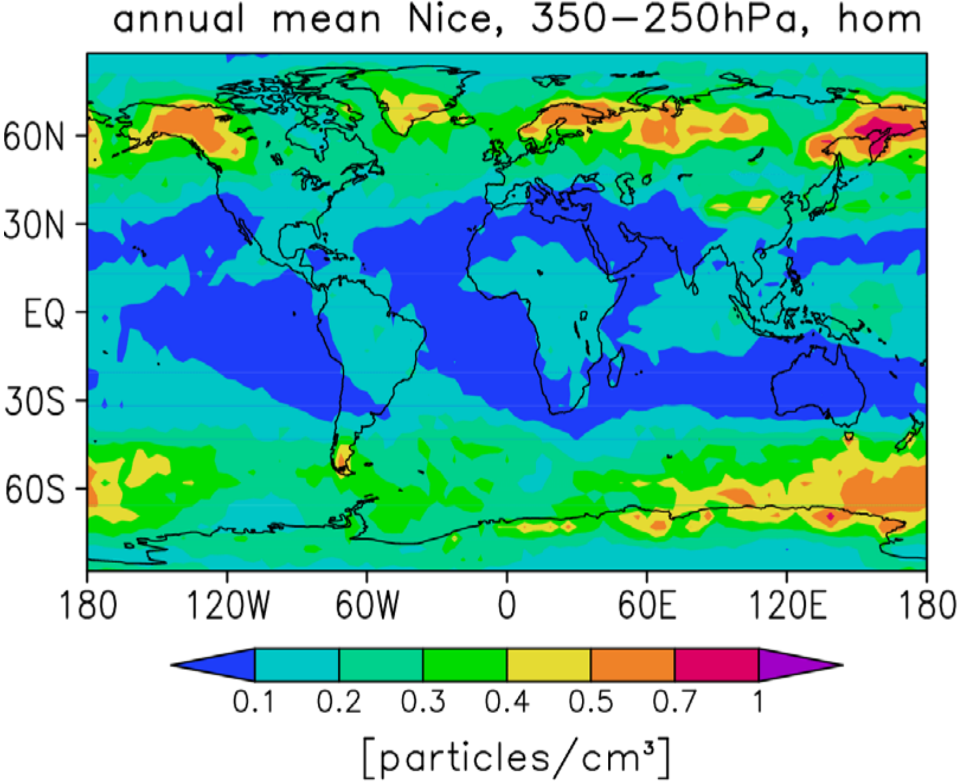


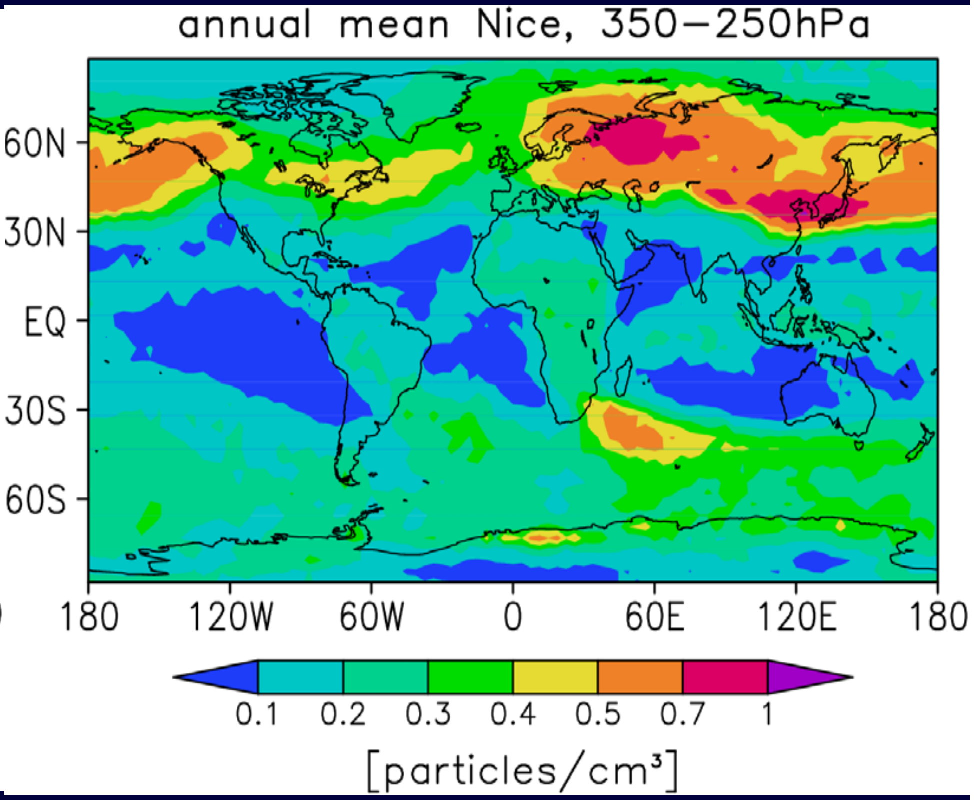
BC from surface sources, 250hPa



BC change due to aircraft (min), 250hPa

Global Simulation of Aircraft Soot Emissions with ECHAM4 *[Hendricks et al. 2003]*





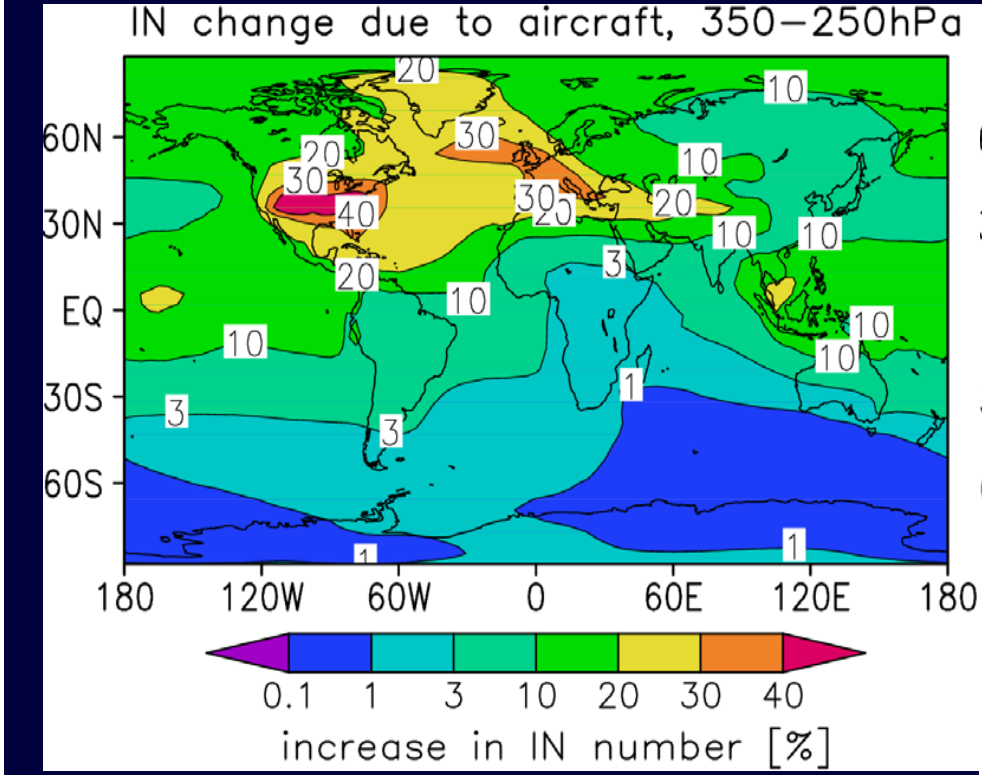
Homogeneous nucleation:
Lohmann and Kärcher [2002]

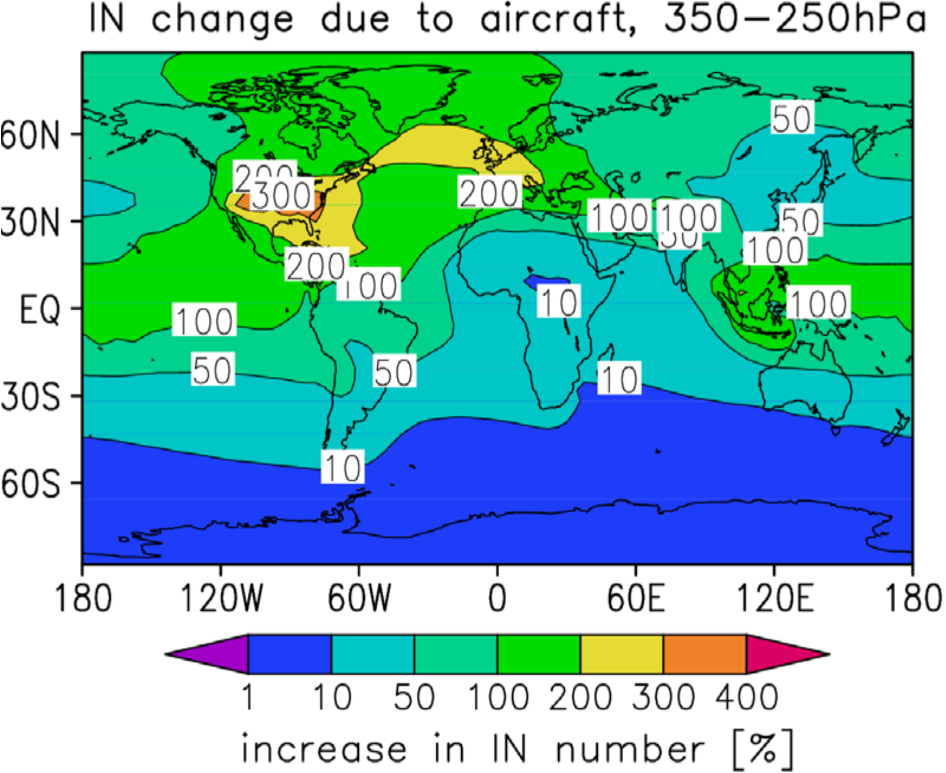
Heterogeneous nucleation:

$$dN_{ice}/dt = \begin{cases} f(IN_{het}), & IN_{het} \geq IN_{crit} \\ dN_{ice}/dt_{\,hom}, & IN_{het} < IN_{crit} \end{cases}$$

$IN_{het} = N_{BC} + N_{dust}$
$IN_{crit} = 0.5/cm^3$

Global Simulation of Aircraft Soot Emissions with ECHAM4 *[Hendricks et al. 2003]*





Experiment 1:

$IN_{het} = N_{dust} + N_{BC}$ (surface)

Experiment 2:

$IN_{het} = IN_{het}$ (Exp 1) + N_{BC}(aircraft)

Experiment 3:

$IN_{het} = N_{dust}$

Experiment 4:

$IN_{het} = IN_{het}$ (Exp 3) + N_{BC}(aircraft)

Conclusions to this Point:

- Aircraft soot emissions may contribute significantly to heterogeneous ice nuclei on the Northern Hemisphere

Conclusions:

Aerosols affect liquid clouds by decreasing cloud droplet size

-> inhibition of drizzle formation

-> more reflection of solar radiation

-> evaporation of cloud droplets in case of soot

Aerosols may affect ice clouds by

-> freezing seems to occur at colder temperatures

-> inhibition of riming

-> more contrails