Title: Importance of anthropogenic soot aerosols for clouds and climate

The burning of fossil fuels and biofuels due to human activities has greatly increased the amount of particular matter in the atmosphere. The major aerosol components are mineral dust, sea salt, sulfates, nitrates, black carbon (also termed soot) and particulate organic matter (POM). The natural aerosol species, mineral dust and sea salt, dominate the mass concentration in the atmosphere. On average they contribute 39 mg/m^2 and 13 mg/m^2 whereas the anthropogenic components, sulfate, POM and black carbon only contribute 3.9, 3.3 and 0.4 mg/m^2 to the annual global average as deduced from 20 different global models (Kinne et al., 2005). So far, nitrate is not included in most models, because of its semi-volatile nature.

Optically, mineral dust and sea salt are less important because of their larger size. Thus, mineral dust and sea salt each contribute only as much to the aerosol optical as sulfate does (25%). Black carbon, which contributes only 3% to the optical depth, is the main aerosol type that absorbs solar radiation and can lead to a warming of the surrounding air. This warming can prevent cloud formation because the atmosphere becomes more stable or even lead to an evaporation of cloud droplets (e.g., Koren et al., 2004). This called semi-direct effect thus counteracts some of the negative aerosol forcings from scattering aerosols, such as sea salt and sulfate, at the top-of-the atmosphere (e.g., Lohmann and Feichter, 2005).

Aerosols also act as centers for cloud droplets and ice crystals, thereby changing cloud properties. If more aerosols compete for the uptake of water vapor, the resulting cloud droplets do not grow as large. More smaller cloud droplets have a larger surface area than fewer larger cloud droplets for the same amount of cloud water. Thus, a polluted cloud reflects more solar radiation back to space, resulting in a negative radiative forcing at TOA (cloud albedo effect). In addition, these more numerous but smaller cloud droplets collide less efficiently with each other, which reduces the precipitation efficiency of polluted clouds and prolongs their lifetime. It also implies more scattering of solar radiation back to space, thus reinforcing the cloud albedo effect.

On the contrary, if a fraction of the hydrophilic black carbon acts as a contact ice nucleus at temperatures between 0°C and -35°C, then increases in aerosol concentration from pre-industrial times to present-day pose a ``glaciation indirect aerosol effect'' on clouds (Lohmann, 2002). Here increases in contact ice nuclei in the present-day climate result in more frequent glaciation of supercooled clouds and increase the amount of precipitation via the ice phase. This reduces the cloud cover and the cloud optical depth of mid-level clouds in mid- and high latitudes and results in more absorption of solar radiation within the Earth-atmosphere system. Therefore, this effect can at least partly offset the aerosol cloud lifetime effect.

New parameterizations of contact freezing and immersion freezing in mixed-phase clouds for black carbon and mineral dust assumed to be composed of either kaolinite (simulation KAO) or montmorillonite (simulation MON) are introduced into the ECHAM4 general circulation model (Lohmann and Diehl, 2005). The effectiveness of black carbon and dust as ice nuclei as a function of temperature is derived from a compilation of laboratory studies. This is the first time that freezing parameterizations take the chemical composition of ice nuclei into account. The rather subtle differences between these simulations in the present-day climate have rather large implications for the anthropogenic indirect aerosol effect. The decrease in net radiation at the top-of-the-atmosphere from pre-industrial to present-day times varies from 1 W m^{-2} to 2.1 W m^{-2} depending on whether dust is assumed to be composed of kaolinite or montmorillonite. In simulation KAO, black carbon has a higher relevancy as an ice nucleus than in simulation MON, because kaolinite is not freezing as effectively as montmorillonite. In simulation KAO, the addition of anthropogenic black carbon aerosols results in a larger ice water path, a slightly higher precipitation rate and a reduced total cloud cover. On the contrary, in simulation MON the increase in ice water path is much smaller and globally the decrease in precipitation is dominated by the reduction in warm-phase precipitation due to the indirect cloud lifetime effect.

References:
Kinne S., et al., 2005: An AeroCom initial assessment – optical properties in aerosol component modules of global models, *Atmos. Chem. Phys. Disc*., submitted.
Koren, I., Y.J. Kaufman, L.A. Remer, and J.V. Martins: 2004, Measurements of the effect of smoke aerosol on inhibition of cloud formation. Science 303, 1342-1345.
Lohmann, U. and K. Diehl, Sensitivity studies of the importance of dust nuclei for the indirect aerosol effect on mixed-phase clouds, J. Atmos. Sci., in press, 2005.
Lohmann, U. and J. Feichter, 2005: Global indirect aerosol effects: A review. *Atmos. Chem. Phys.*, **5,** 715-737.
Lohmann, U., A glaciation indirect aerosol effect caused by soot aerosols, Geophys. Res. Lett., 29, doi: 10.1029/2001GL014357, 2002.

Importance of anthropogenic soot aerosols for clouds and climate

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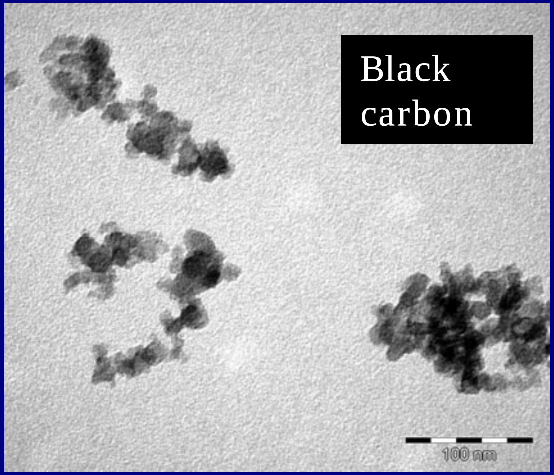

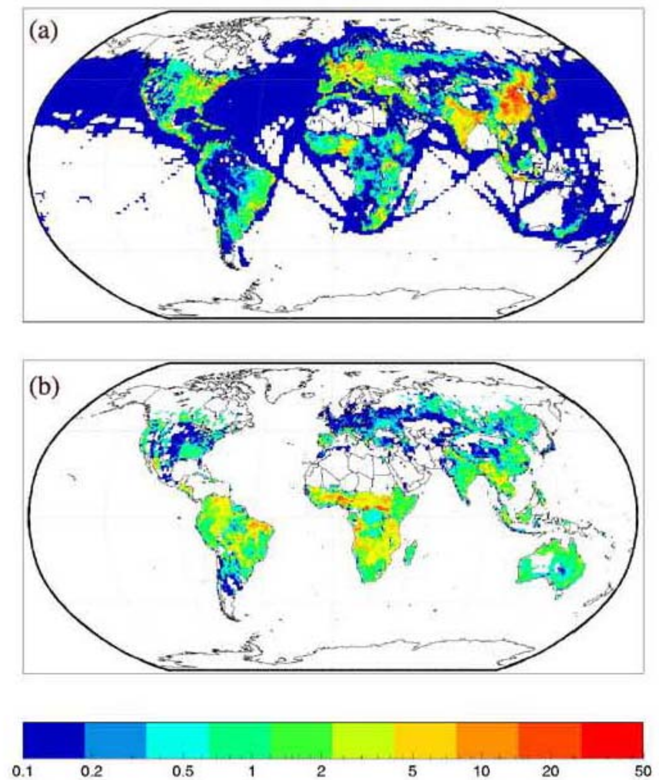

Figure 2. Emissions of black carbon aerosol. (a) "Contained" combustion, based on 1996 activity data. (b) Open burning, annual average. The color coding is an approximately logarithmic scale. Units are $ng/m^2/s$ ($1\ ng/m^2/s \sim 32\ kg/km^2/yr$).

Bond et al., JGR, 2004

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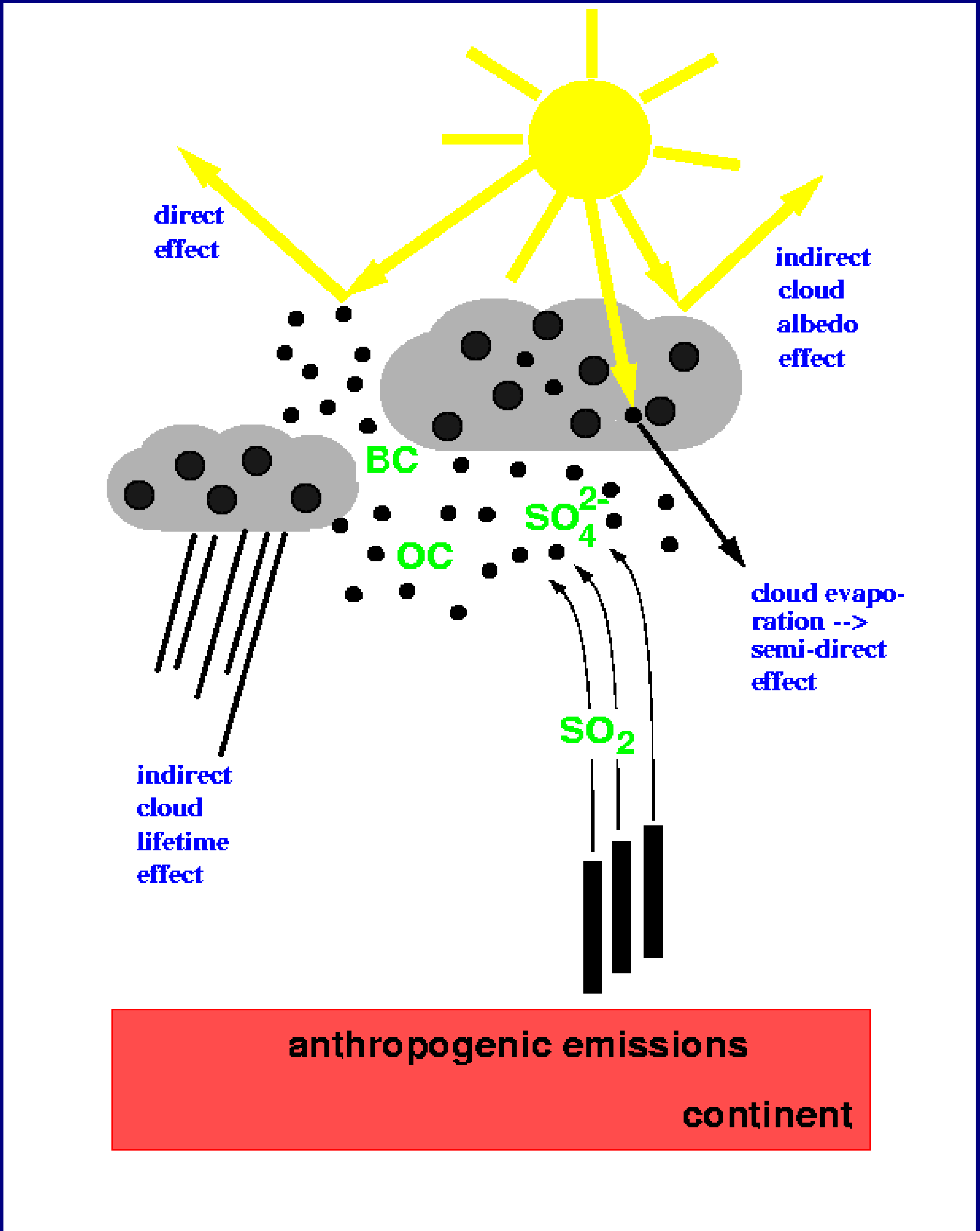
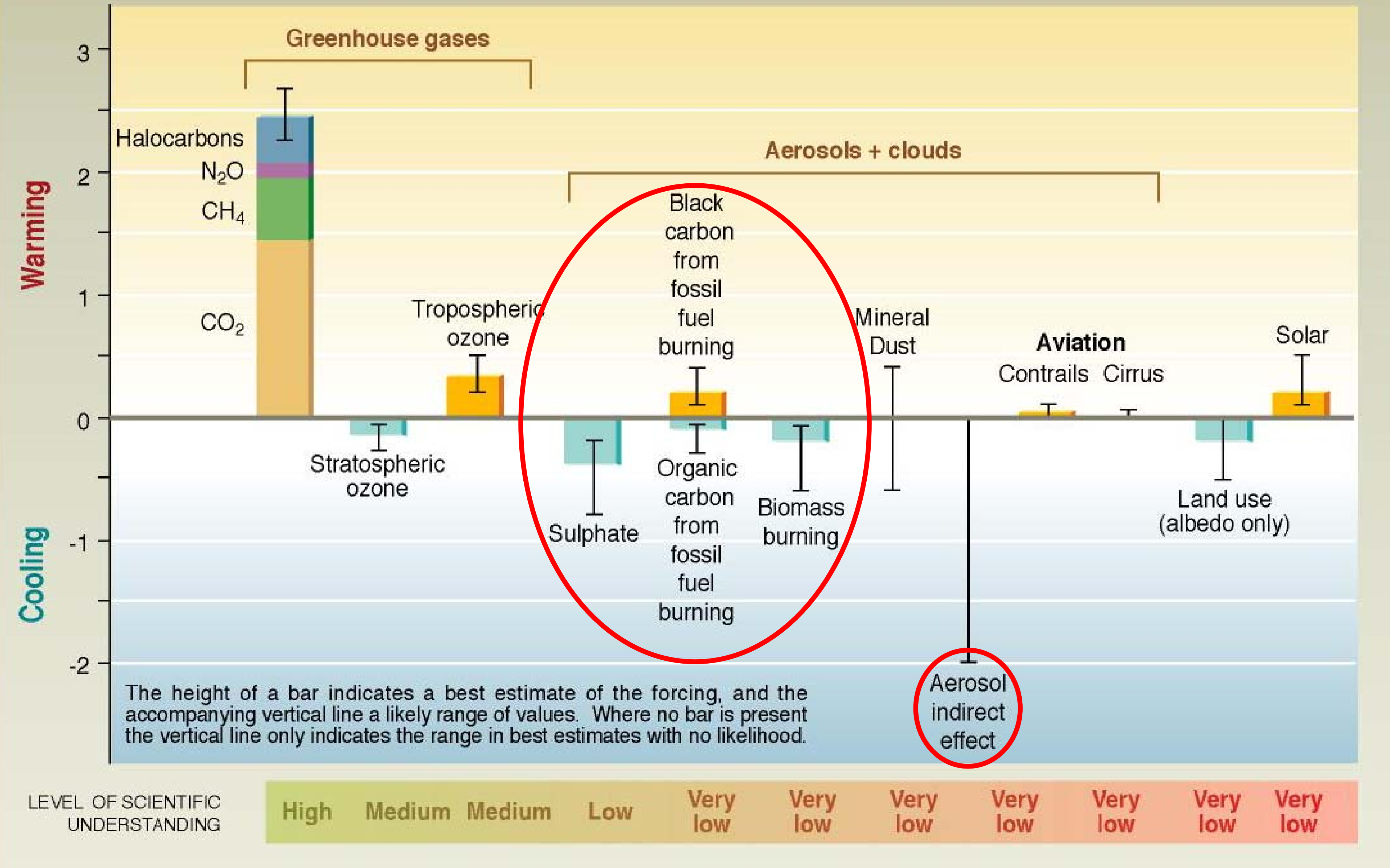
Swiss Federal Institute of Technology Zurich

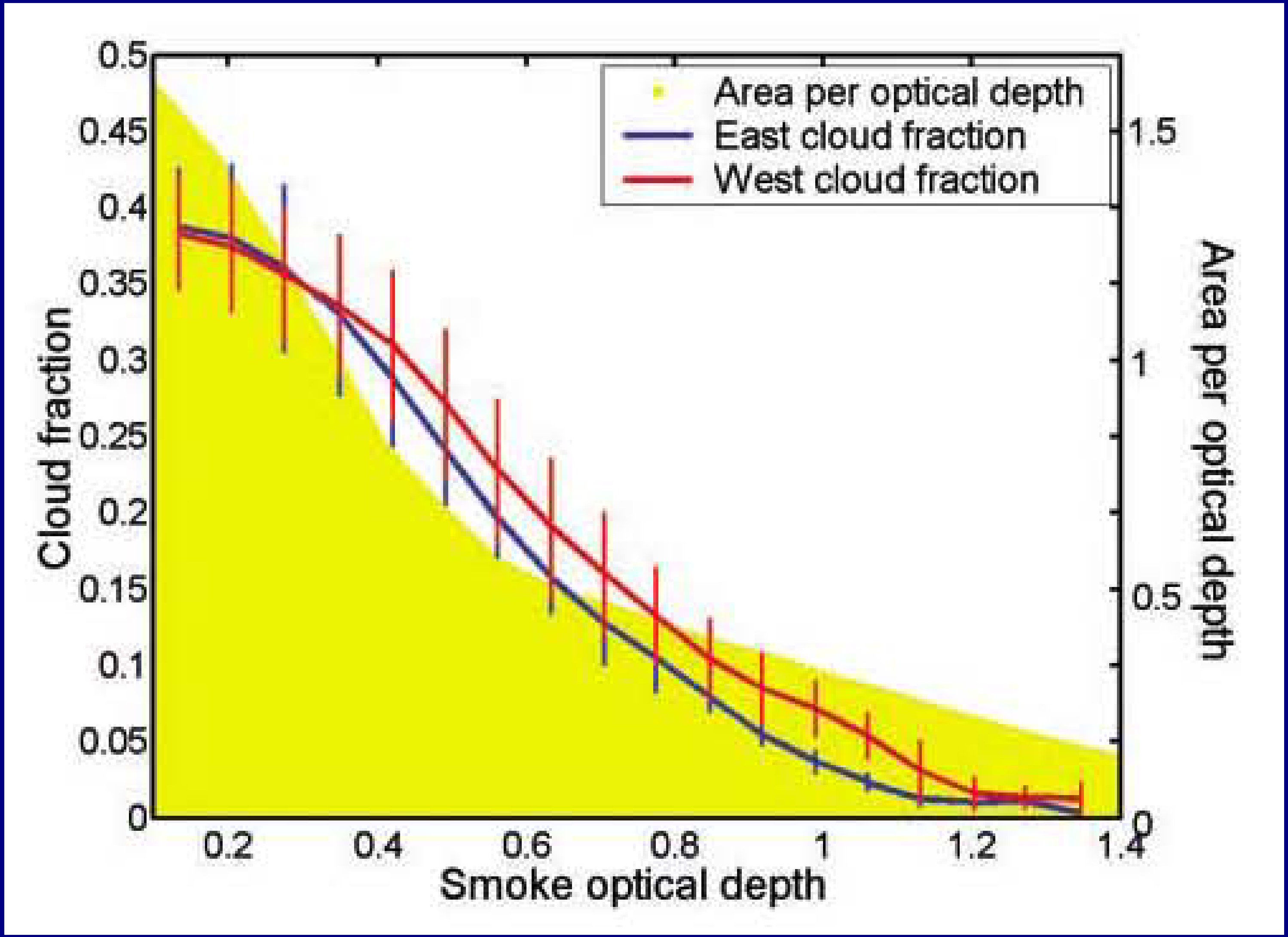
Overview of the different aerosol effects



Top-of-the-atmosphere global-mean radiative forcing (W m^{-2}) for 2000 relative to 1750 *[IPCC, 2001]*



Semi-direct effect:
Reduction of cloudiness due to soot?

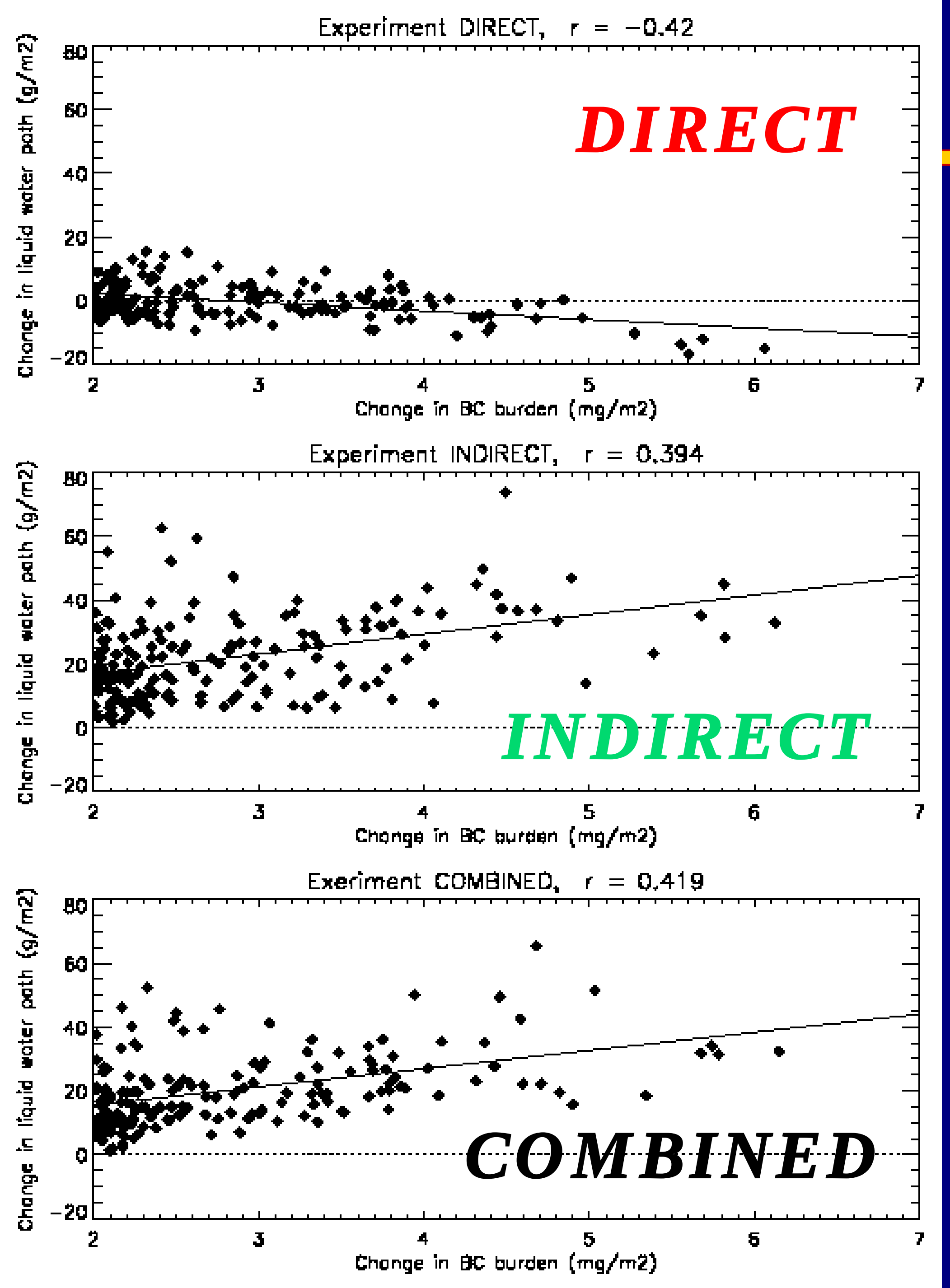


Koren et al., Science, 2004

Semi-direct effect

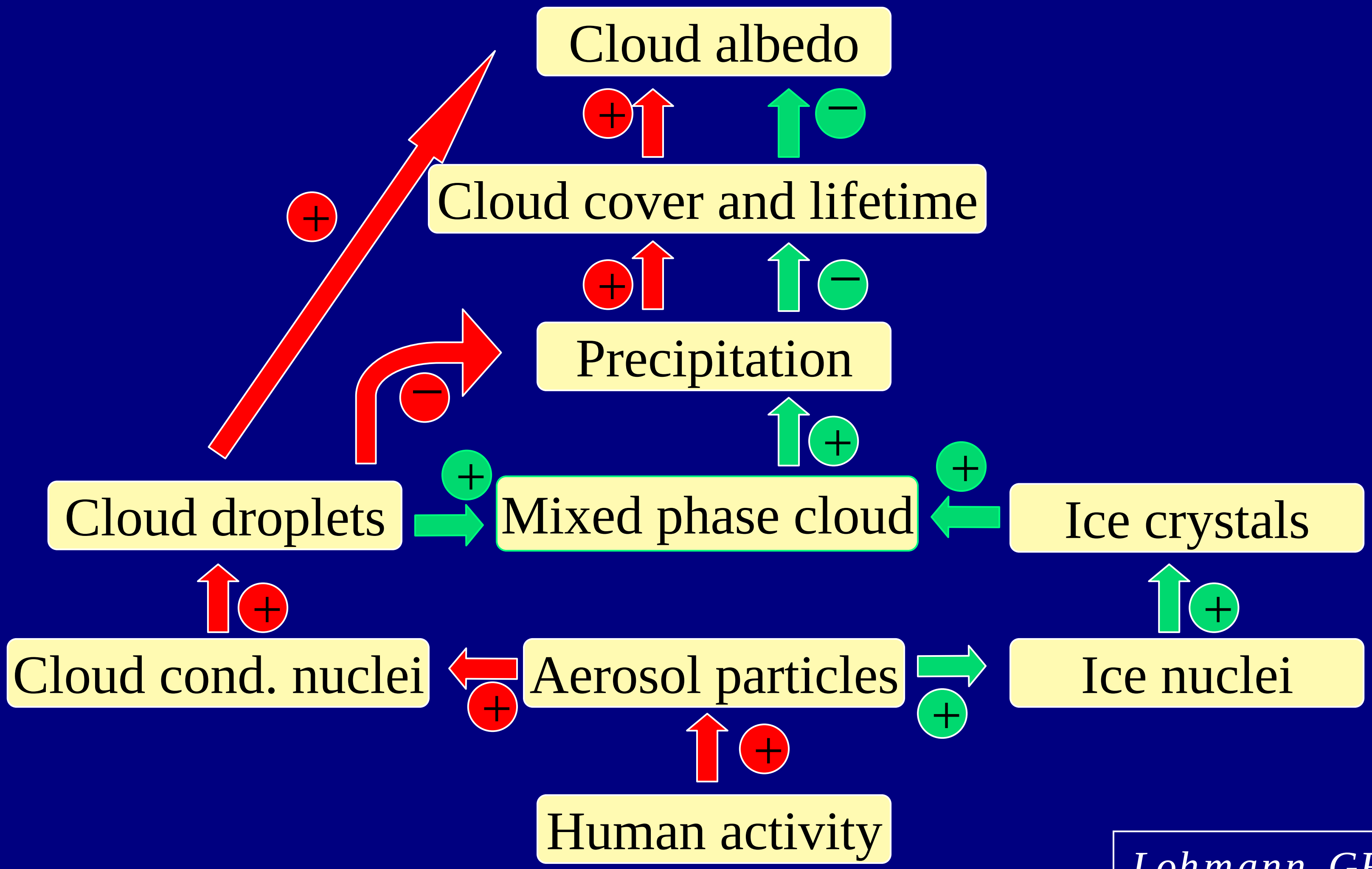
Change in liquid water path with increasing soot burden for the experiments DIRECT, INDIRECT and COMBINED

global mean change in shortwave radiation at the top-of-the atmosphere:
Direct: -0.1 W/m^2
Indirect: -1.4 W/m^2
Combined: -1.3 W/m^2



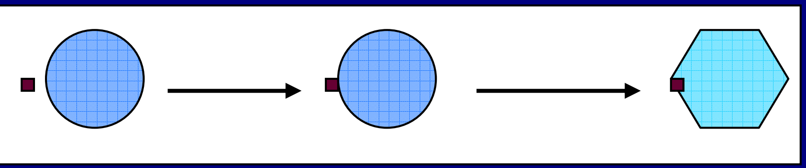
Lohmann & Feichter, GRL, 2001

Pathways of the Traditional Warm Indirect Aerosol Effect and the Glaciation Indirect Aerosol Effect

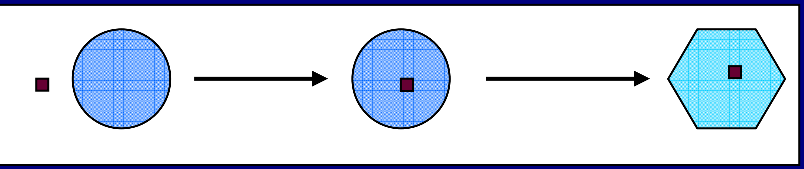


Lohmann, GRL, 2002

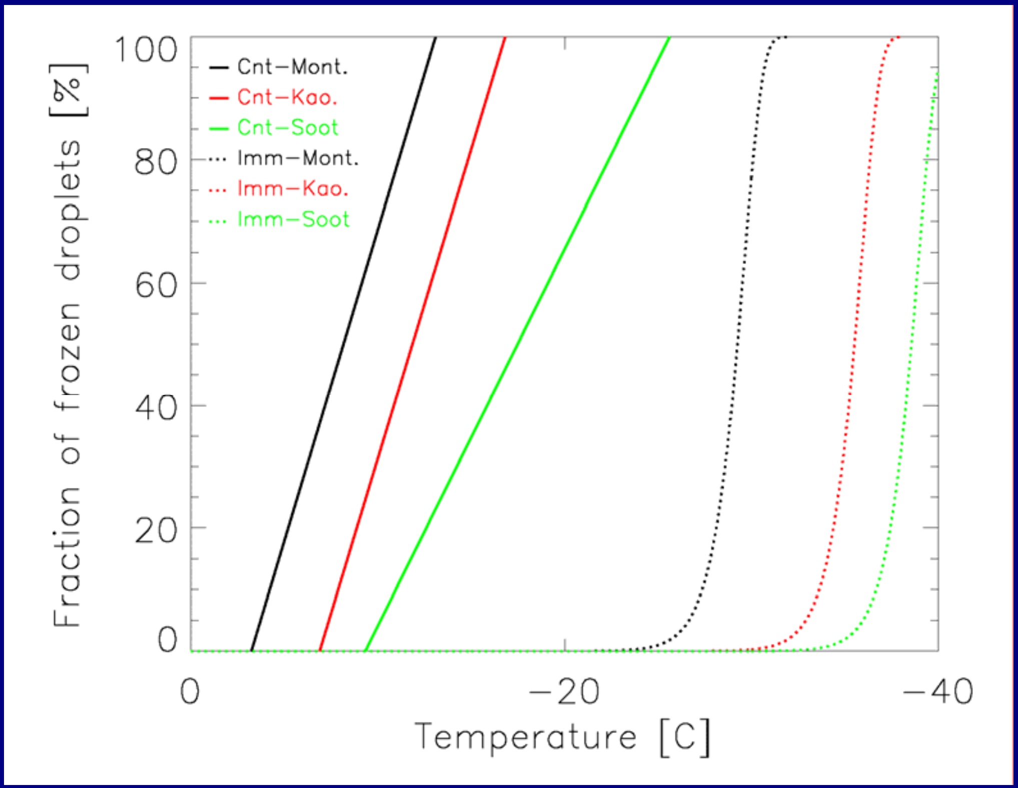
Experimental design *[Lohmann and Diehl, JAS, 2005, in press]*



[contact freezing]



[immersion freezing]

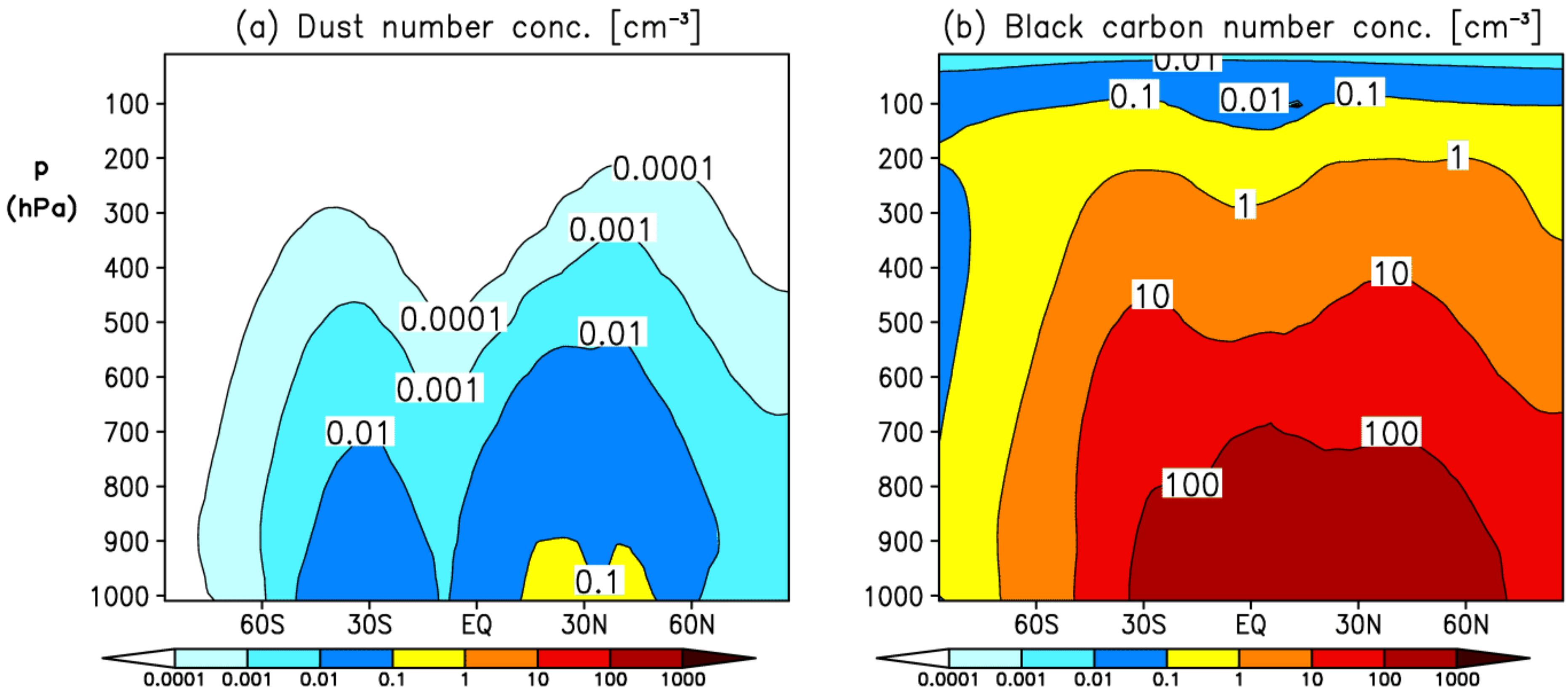


Apply these temperature dependent freezing characteristics of black carbon and dust in the ECHAM4 climate model:

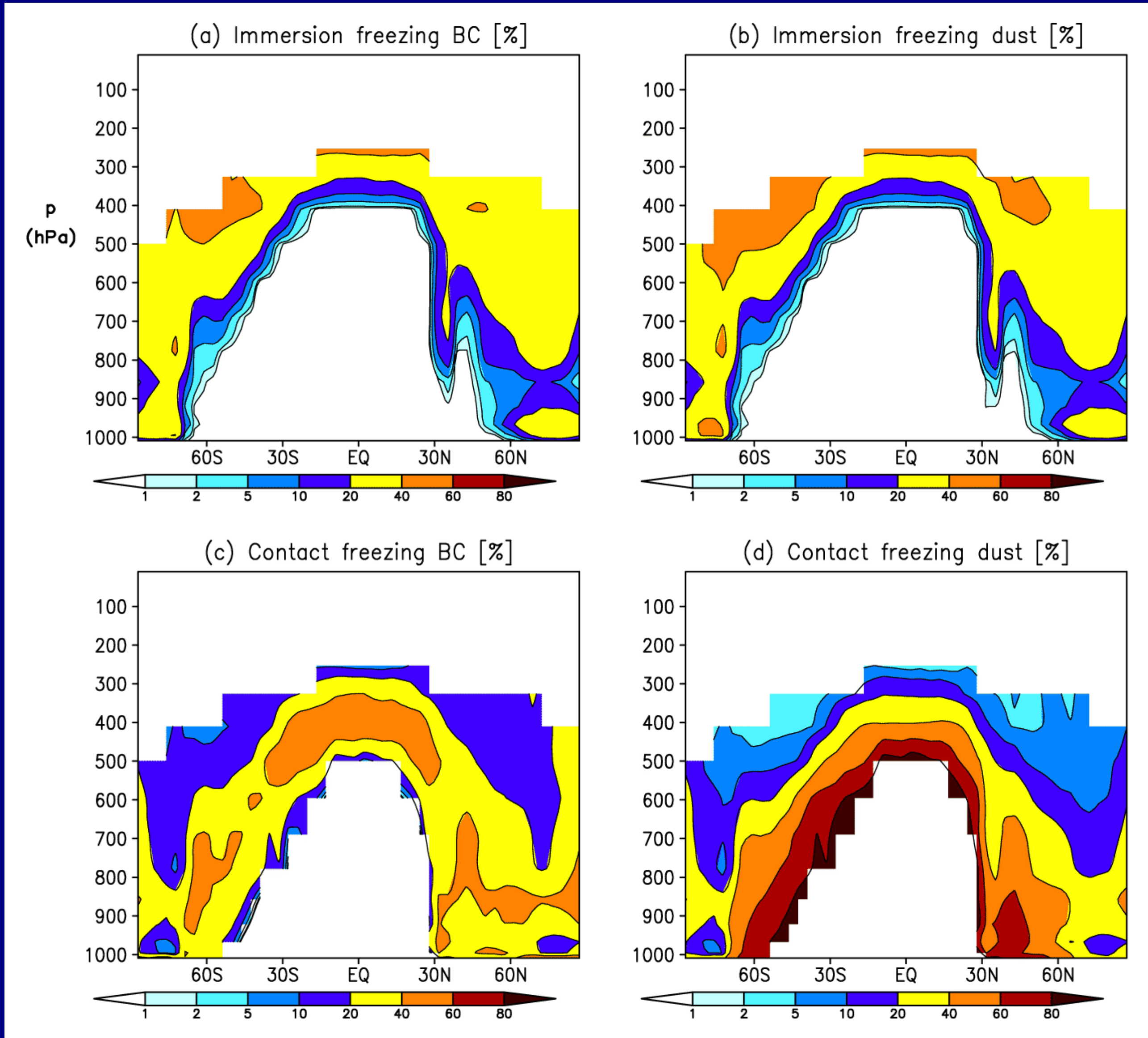
* Simulation KAO: treat dust as kaolinite
* Simulation MON: treat dust as montmorillonite
* 10-year simulations with present-day and pre-industrial aerosol emissions

Zonal 10-year means of the dust and black carbon aerosol concentration from simulation ECHAM-KAO

[Lohmann and Diehl, JAS, 2005, in press]



Zonal 10-year means of the percentage of freezing caused by immersion vs. contact freezing of black carbon vs. dust from simulation ECHAM-KAO

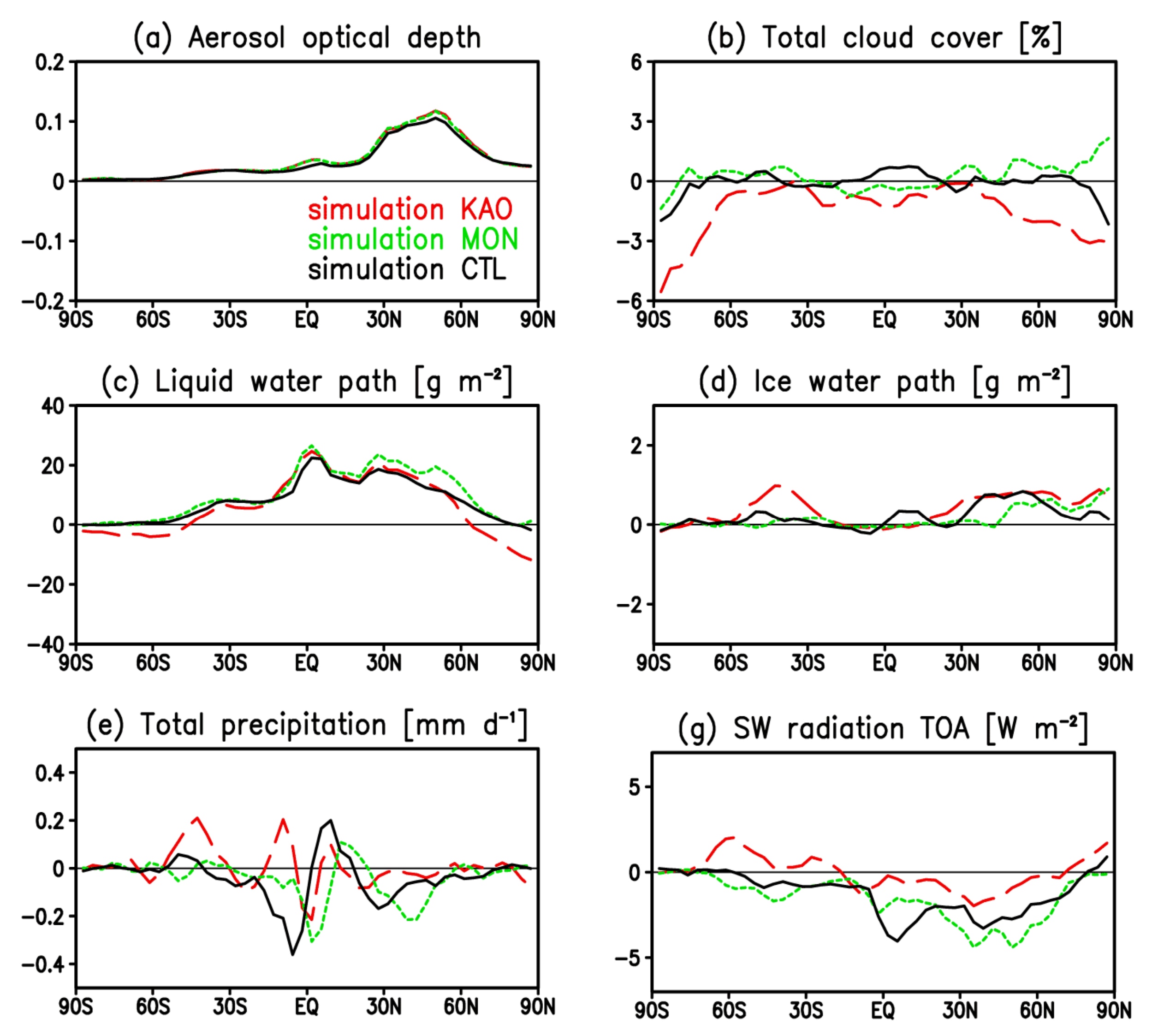


Anthropogenic indirect aerosol effect:

Simulation MON:
dust freezes effectively
→ warm indirect aerosol effect dominates
→ precip decreases

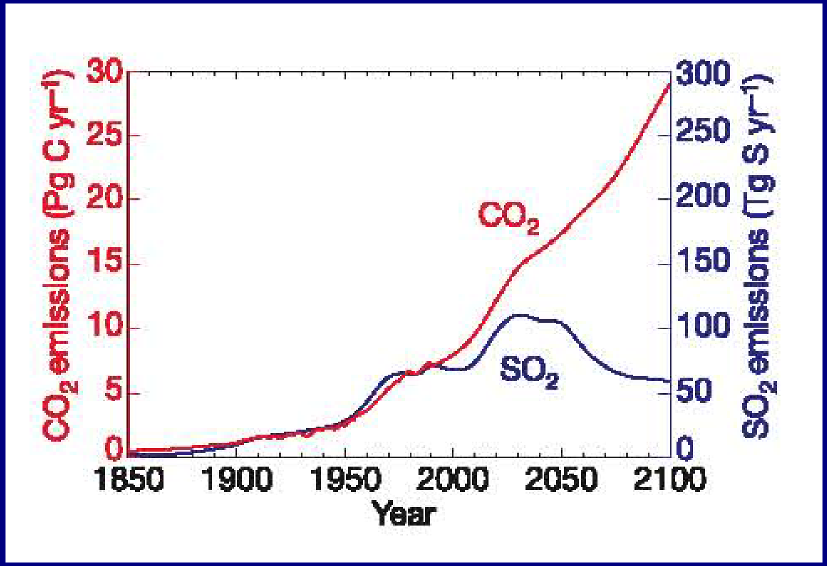
Simulation KAO:
dust does not freeze effectively
→ glaciation indirect aerosol effect of soot as ice nuclei dominates
→ precip increases

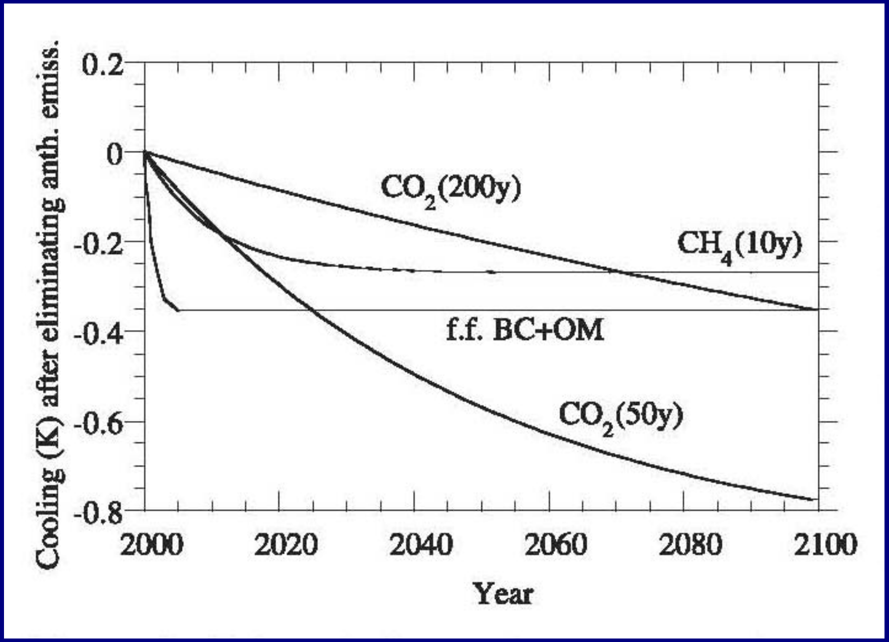
global mean change in SW radiation at TOA:
CTL: -1.6 W/m^2
MON: -1.8 W/m^2
KAO: -0.2 W/m^2



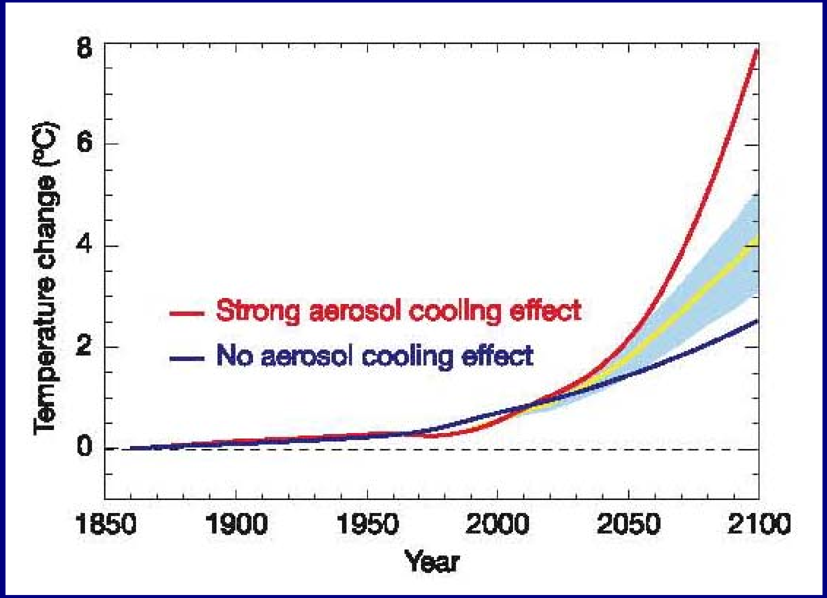
Lohmann and Diehl, JAS, 2005, in press.

Difference between abs0rbing aerosols (soot) and scattering aerosols (sulfate)



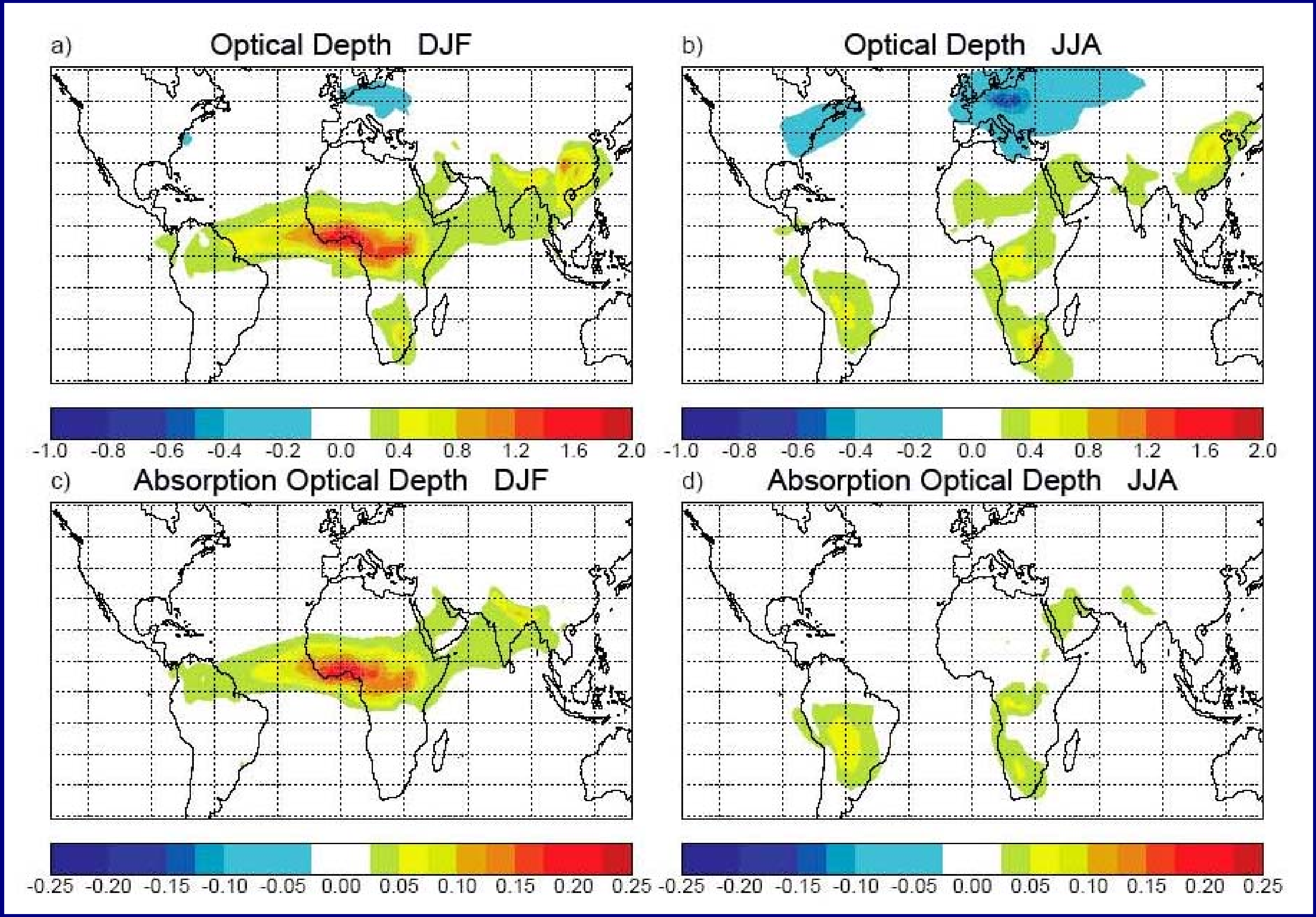


Jacobson, JGR, 2001



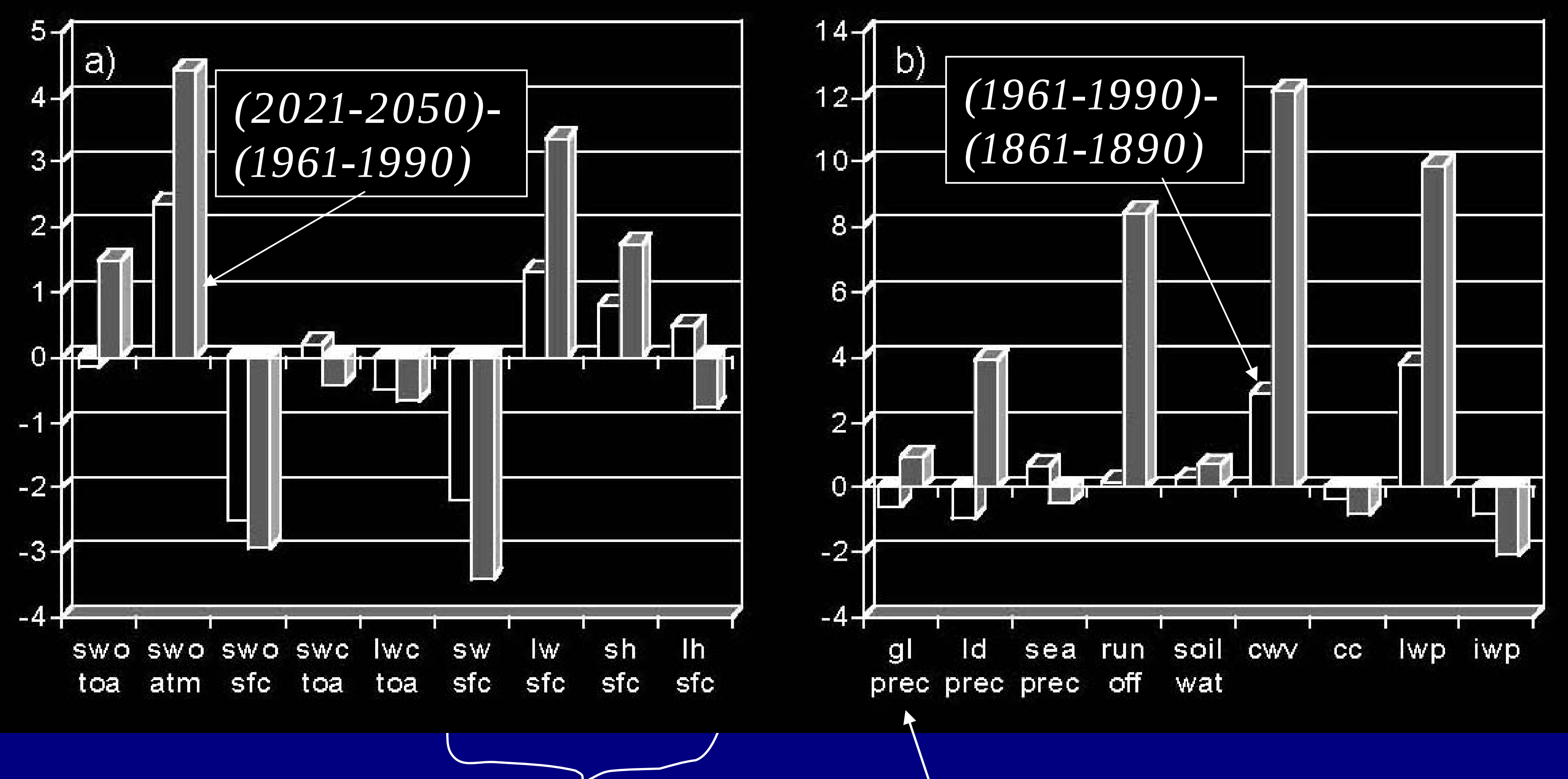
Andreae et al., Nature, 2005

Changes in aerosol optical depth between (2021-2050) - (1961-1990)



Roeckner et al., JC, subm. 2005

Changes in the energy and water budget



Surface energy budget (W/m^2)

global mean precip (%)

Roeckner et al., JC, subm. 2005

Conclusions

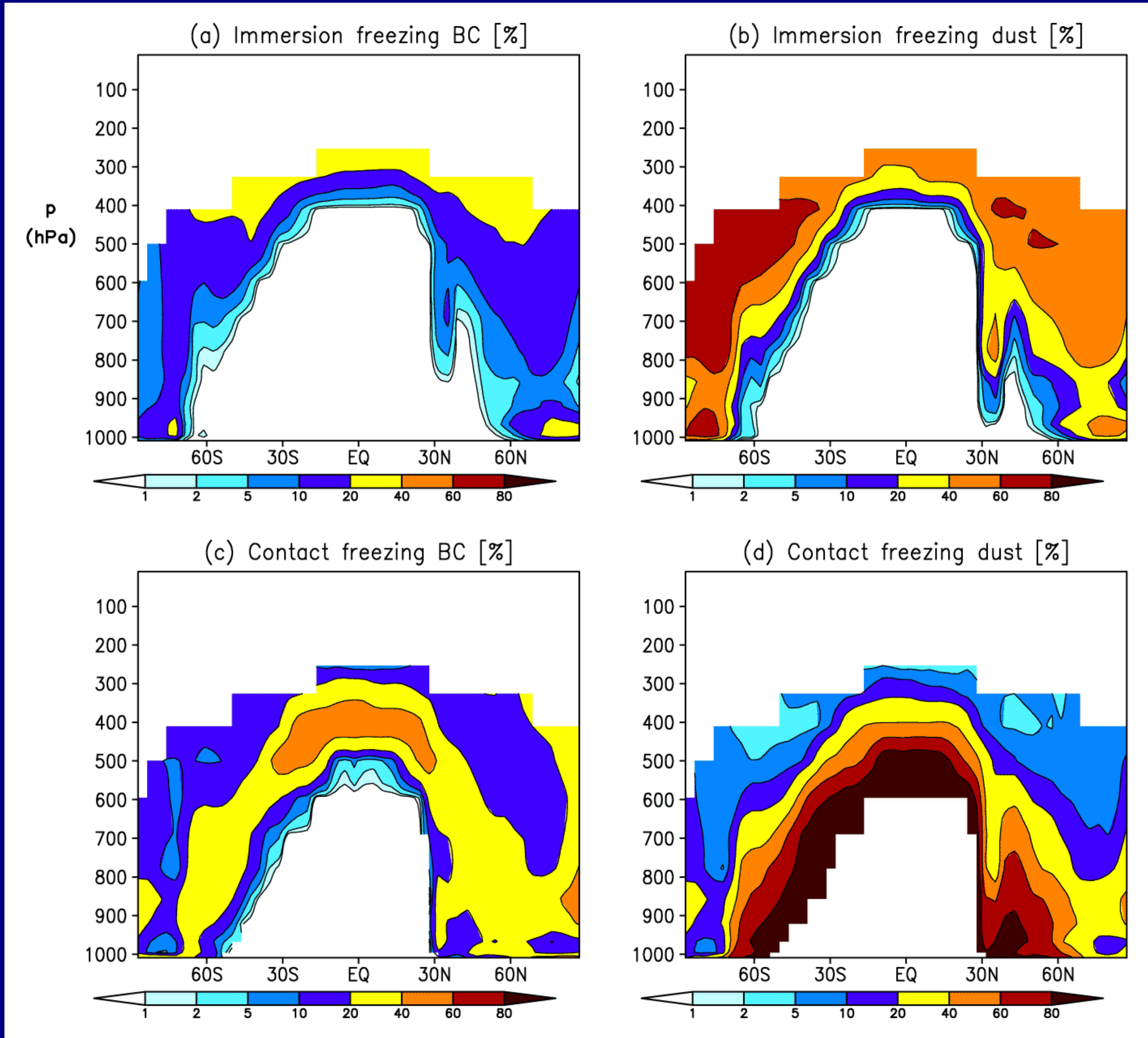
- → Light absorbing particles (e.g., soot) need to be distinguished from scattering aerosols (e.g., sulfates).

→ Soot contributes to a warming and has adverse health effects, i.e. a reduction of soot is necessary.

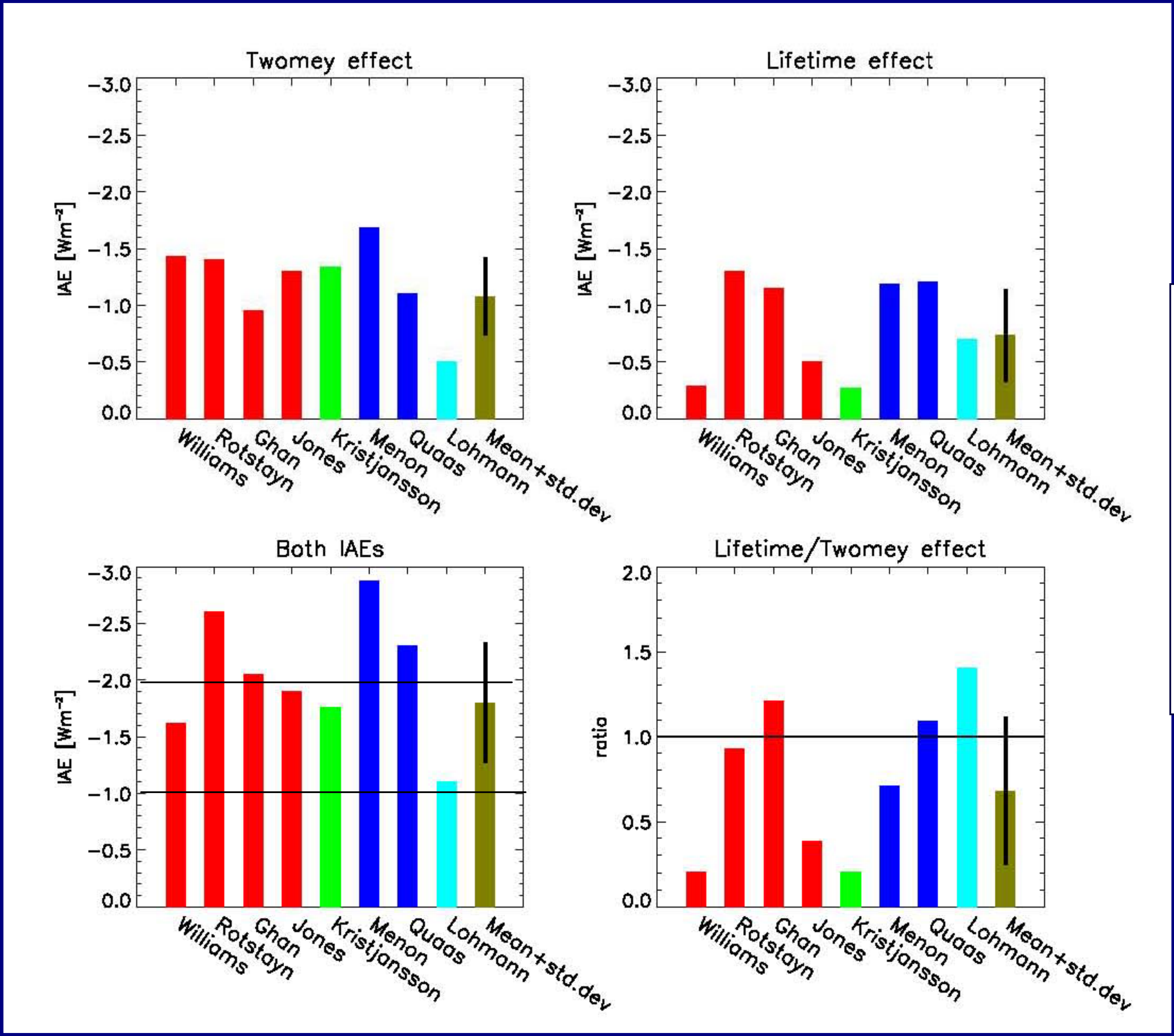
→ Sulfates contribute to acid rain and therefore should be reduced as well.

→ If aerosols in general are reduced in future, it has positive effects for air quality and human health, whereas the climatic implications are not known yet.

Zonal 10-year means of the percentage of freezing caused by immersion vs. contact freezing of black carbon vs. dust from simulation ECHAM-MON



Global mean indirect aerosol effect (Twomey vs. lifetime) from different climate models



Sulfate

Soot (BC) and sulfate

Organic aerosols (OC) and sulfate

BC, OC and sulfate

Lohmann and Feichter, ACP, 2005