

Clouds and Aerosols

(1) How do aerosols, clouds and precipitation interact?

(2) Why is it relevant?

(3) How do we model and observe these interactions?

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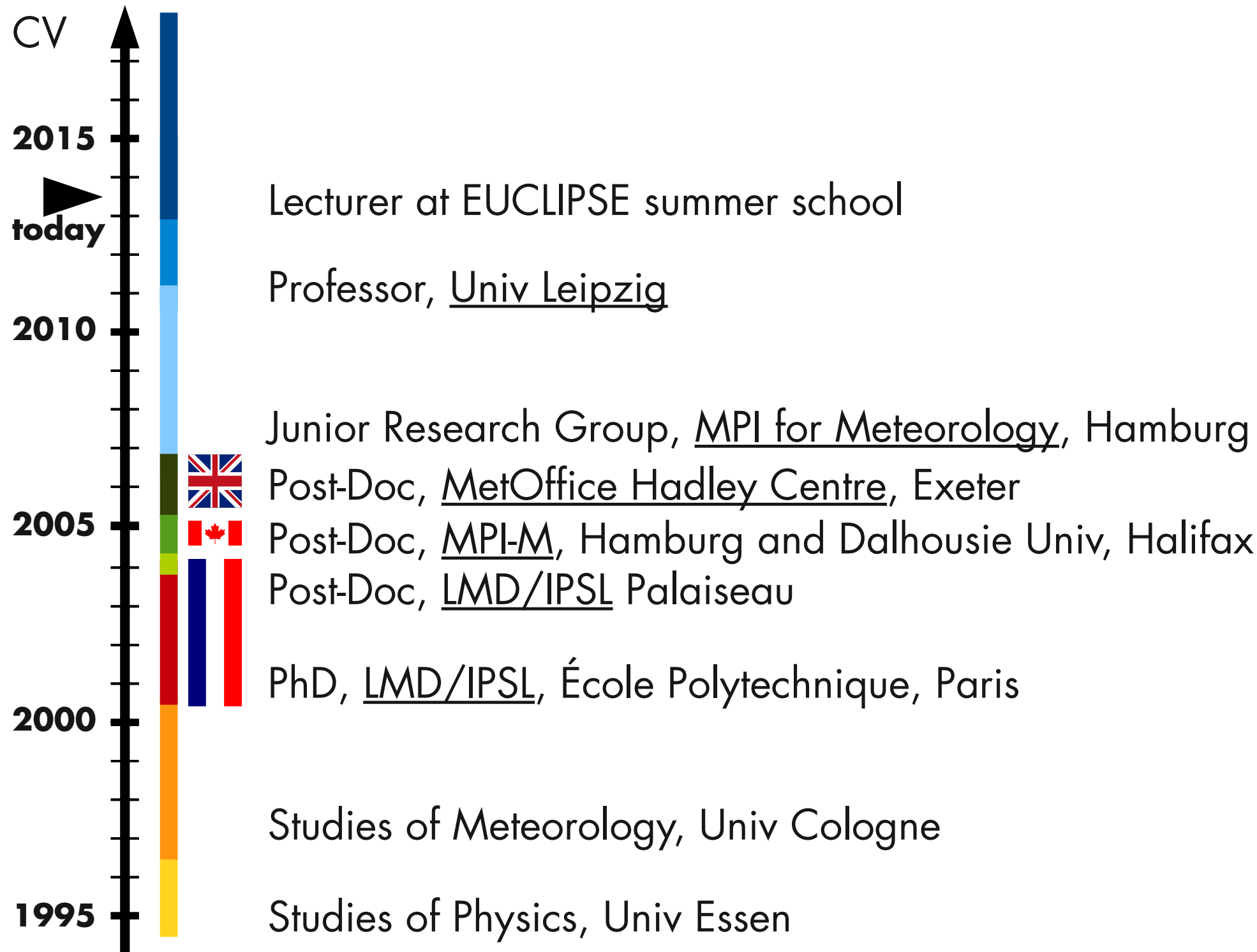
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1. Aerosols

Definition: Solid or liquid particles suspended in a gas

Here: All particles in the atmosphere that are not cloud or precipitation particles

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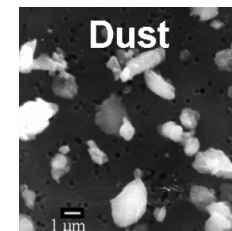
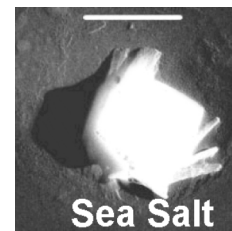
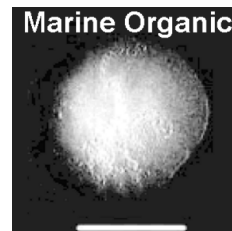
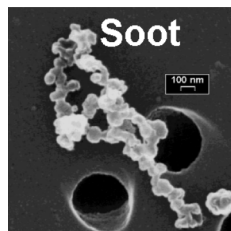
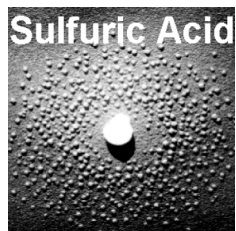
1.1 Chemical characterisation

Species

Mineral dust	(DU)
Sea salt	(SS)
Sulfate	(SO ₄)
Soot, Black carbon	(BC)
Organic matter	(OM)
Nitrate	(NH ₄)

Source (partly **anthropogenic**)

windblown, mostly from deserts (also: **roads, agriculture**)
windblown
combustion, biogeochemistry, volcanos
combustion (fossil fuel, biomass, wildfires)
combustion, biogeochemistry
air chemistry, fertilisers, combustion



1. Aerosols

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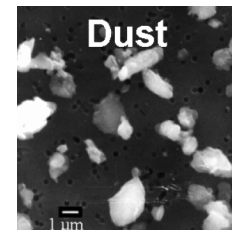
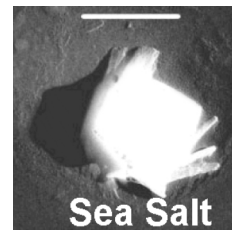
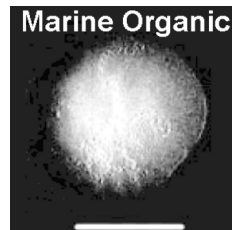
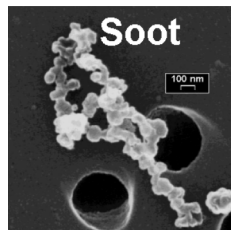
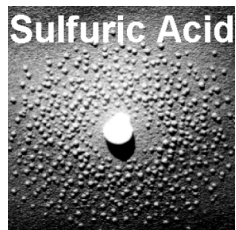
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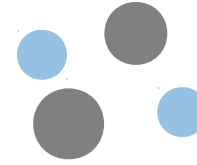


▶ **Primary** aerosols (emitted in form of particles)

▶ **Secondary** aerosols (emitted as gas, transformed in the atmosphere to particles)

1.2 Mixing state

External mixture: Pure chemical composition per particle

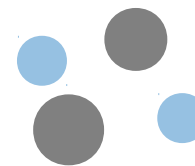


Internal mixture: A particle consists of different chemical components



1.2 Mixing state

External mixture: Pure chemical composition per particle



Internal mixture: A particle consists of different chemical components

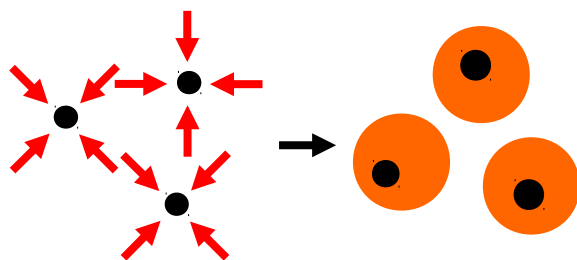


Interactions of particles:

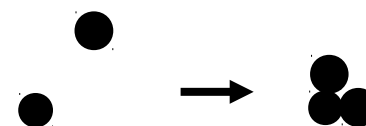
- (i) microphysical (by collision and coalescence, as in clouds, see Hanna Pawlowska's talk)
- (ii) chemical (e.g., condensation of SO_2 onto dust or black carbon)

processing of aerosols also called "aging"

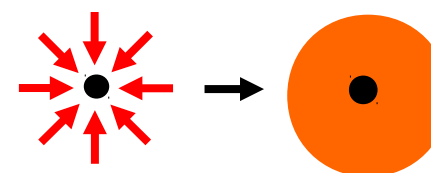
Sulfuric + Carbonaceous



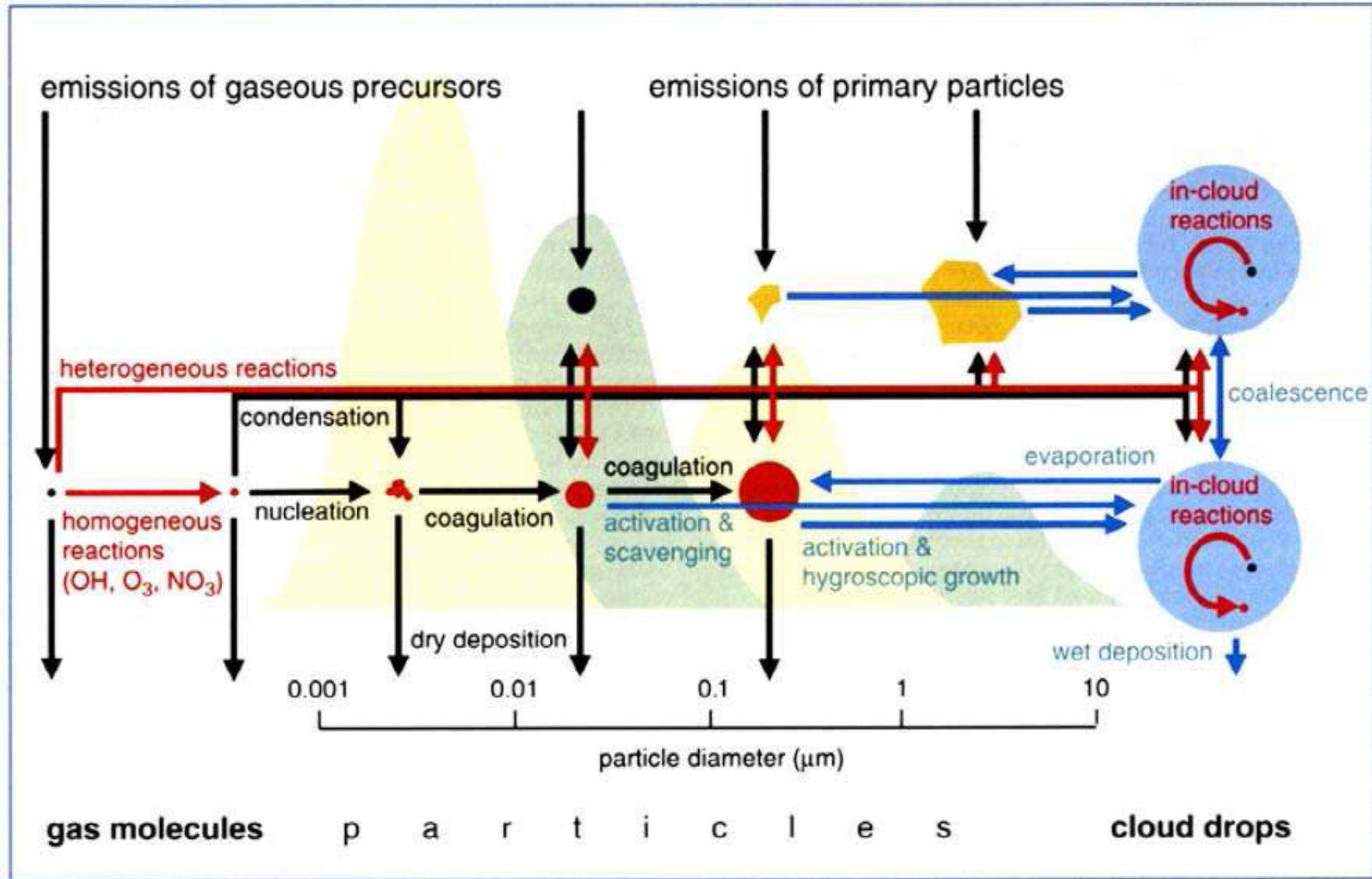
Carbonaceous



Sulfuric

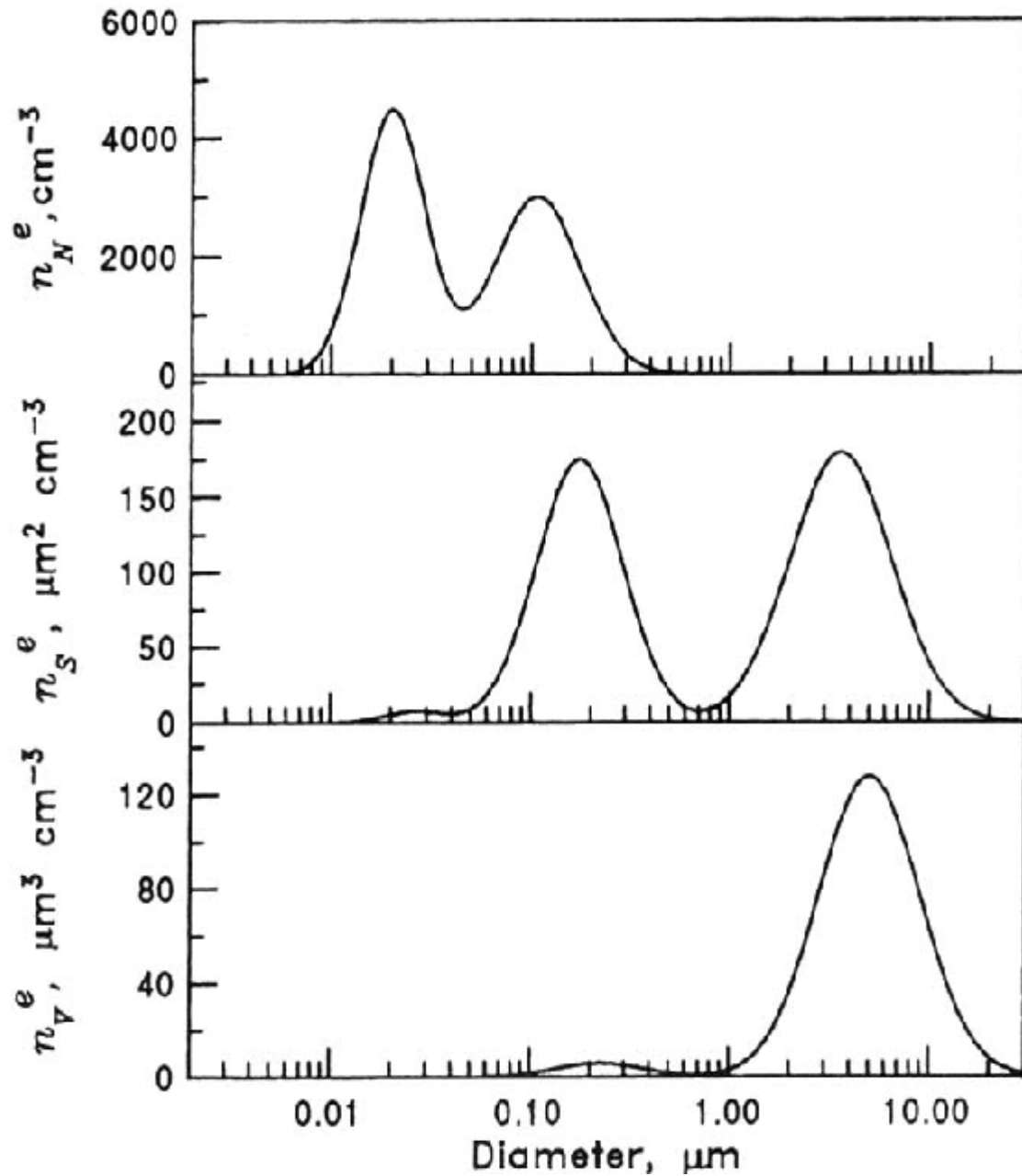


1.2 Size characterisation



1 Å = 0.1 nm. H₂ Molecule ~ 1.4 Å diameter

1.4 Size distributions



Log-normal size distributions

typical: 2-3 different "modes"

by number

→ cloud interaction

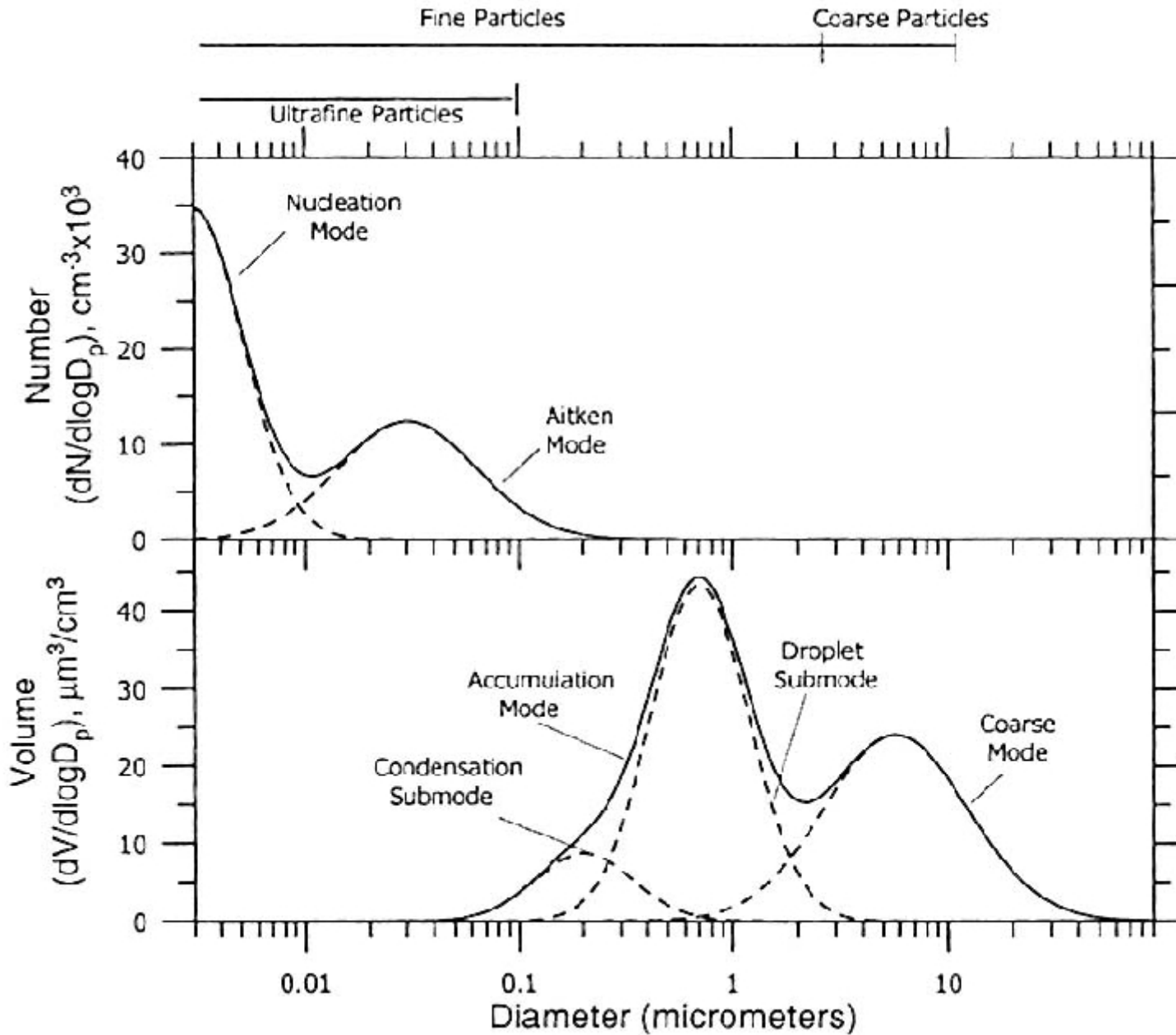
by surface

→ scattering

by volume

→ mass

1.4 Size distributions



Simplified descriptions

< 1 μm fine mode
> 1 μm coarse mode

PM10:
Mass of particles with
diameters > 10 μm

PM2.5
PM1

1.5 Continuity equation

In general necessary: one continuity equation per aerosol type and size class

May be simplified by considering just a few aerosol types and/or a few aerosol size classes (e.g., modes, or bulk representation)

$$\frac{D}{Dt}q_a = \sum S_a$$

q_a – aerosol mass concentration

S_a – aerosol sources/sinks

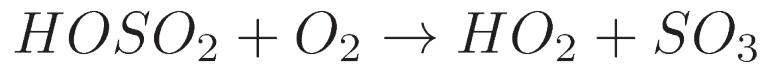
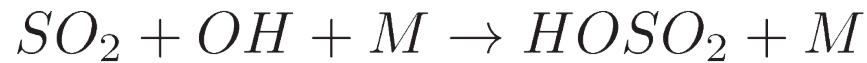
sources: emission (at surface)
chemical and microphysical transformation

sinks: dry deposition
wet deposition
chemical and microphysical transformation

1.6 Chemical transformations of sulfur dioxide to sulfate

Sulfur is emitted by combustion of fossil and biofuel in form of SO_2

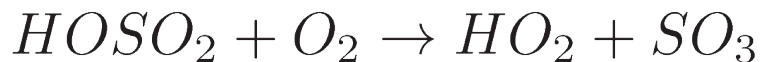
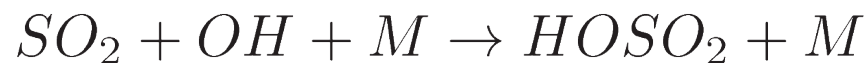
Oxidation in the air by OH radical:



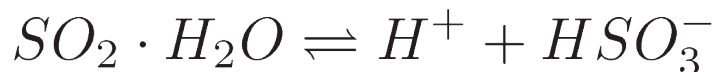
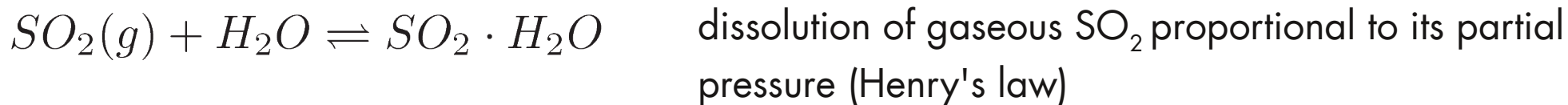
1.6 Chemical transformations of sulfur dioxide to sulfate

Sulfur is emitted by combustion of fossil and biofuel in form of SO_2

Oxidation in the air by OH radical:



Oxidation within cloud and precipitation particles (aqueous chemistry):



Aqueous production of sulfate usually much more efficient.

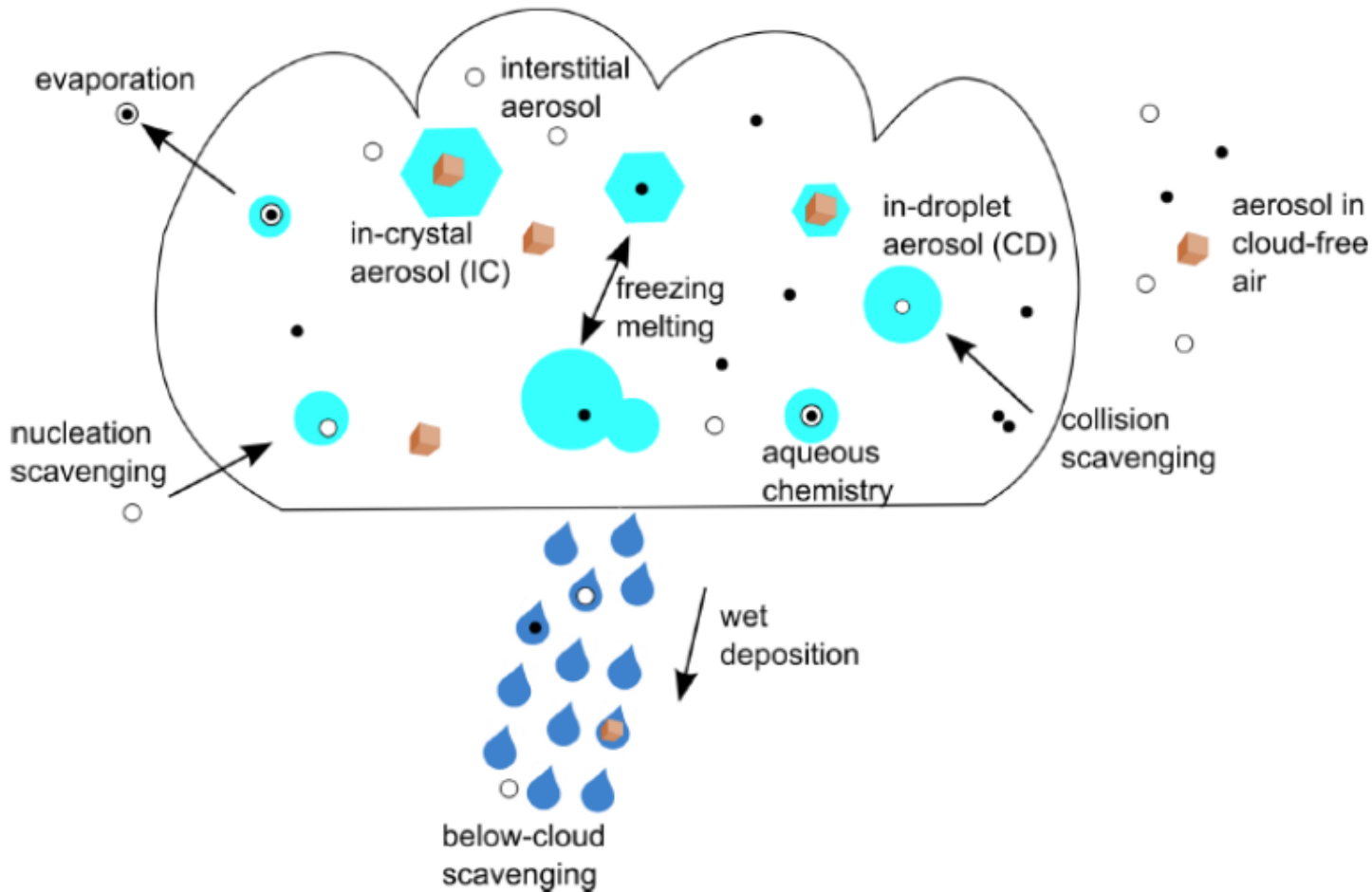
1.7 Cloud processing of aerosols

(i) aqueous chemistry (see example above)

(ii) coagulation inside cloud droplets:

→ particles within cloud droplets / ice crystals from one internally mixed particle

→ usually much better cloud condensation nuclei capabilities



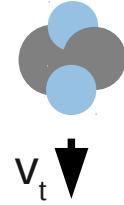
1.8 Aerosol sinks

1. Dry deposition

Sedimentation velocity (see Hanna Pawlowska's talk)

→ Stokes regime, $v_T \sim r_a^2$

since aerosol particles are small, fall velocity usually relatively slow



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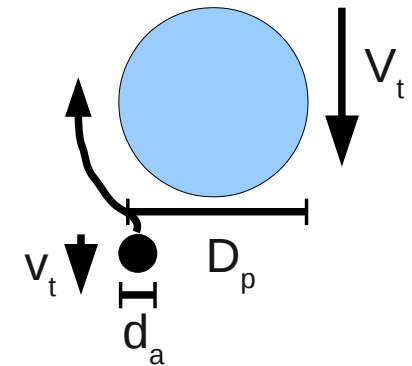
→ Stokes regime, $v_T \sim r_a^2$

since aerosol particles are small, fall velocity usually relatively slow



2. Wet deposition

- (i) Aerosols inside cloud droplets / ice crystals
 - the condensation/ice nuclei and/or aerosols taken up by collision/coalescence
 - Deposition as rain forms in clouds
- (ii) Aerosols below precipitating clouds
 - taken up by collision/coalescence



Where present, wet deposition is much more effective.

1.9 Aerosol lifetime

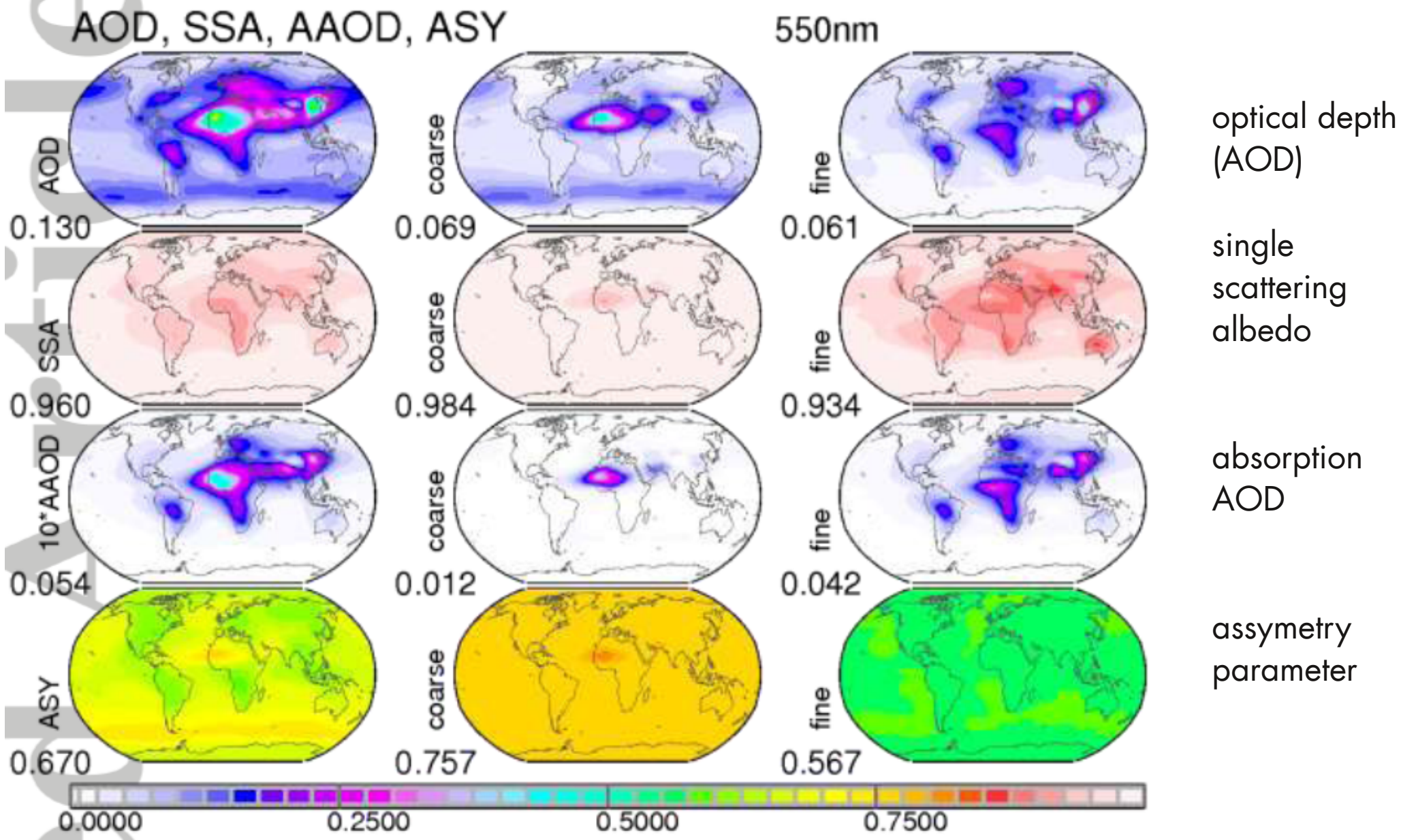
For a stationary situation, $\frac{D}{Dt}q_a = 0$, in the global mean, the average aerosol lifetime is:

$$\tau_a = \frac{M_a}{S_a}$$

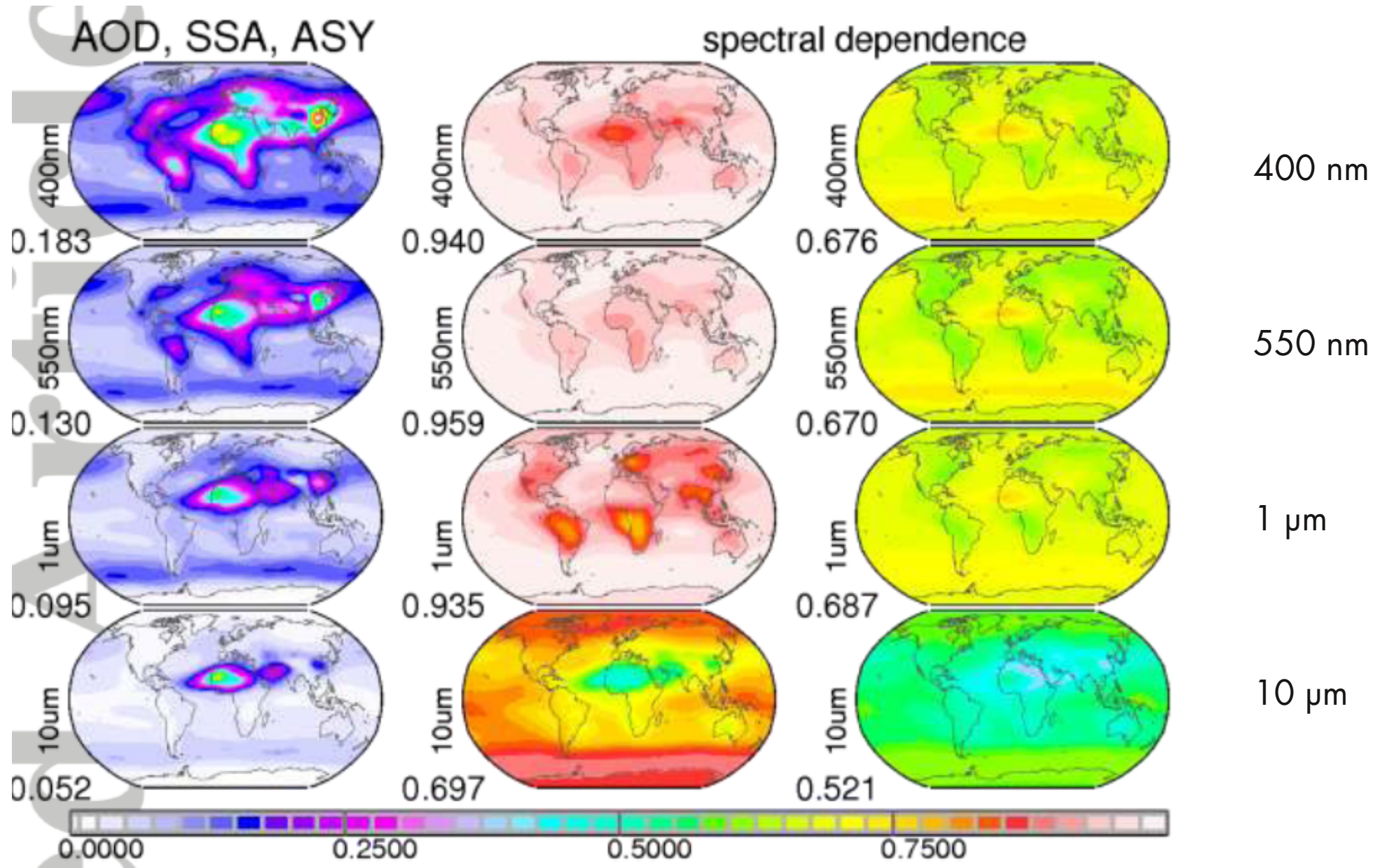
the total atmospheric mass divided by the average source strength.

For sulfate, $M_a \sim 4 \text{ Mt}$, $S_a \sim 200 \text{ Mt yr}^{-1} \rightarrow \tau_a \sim 1 \text{ week}$

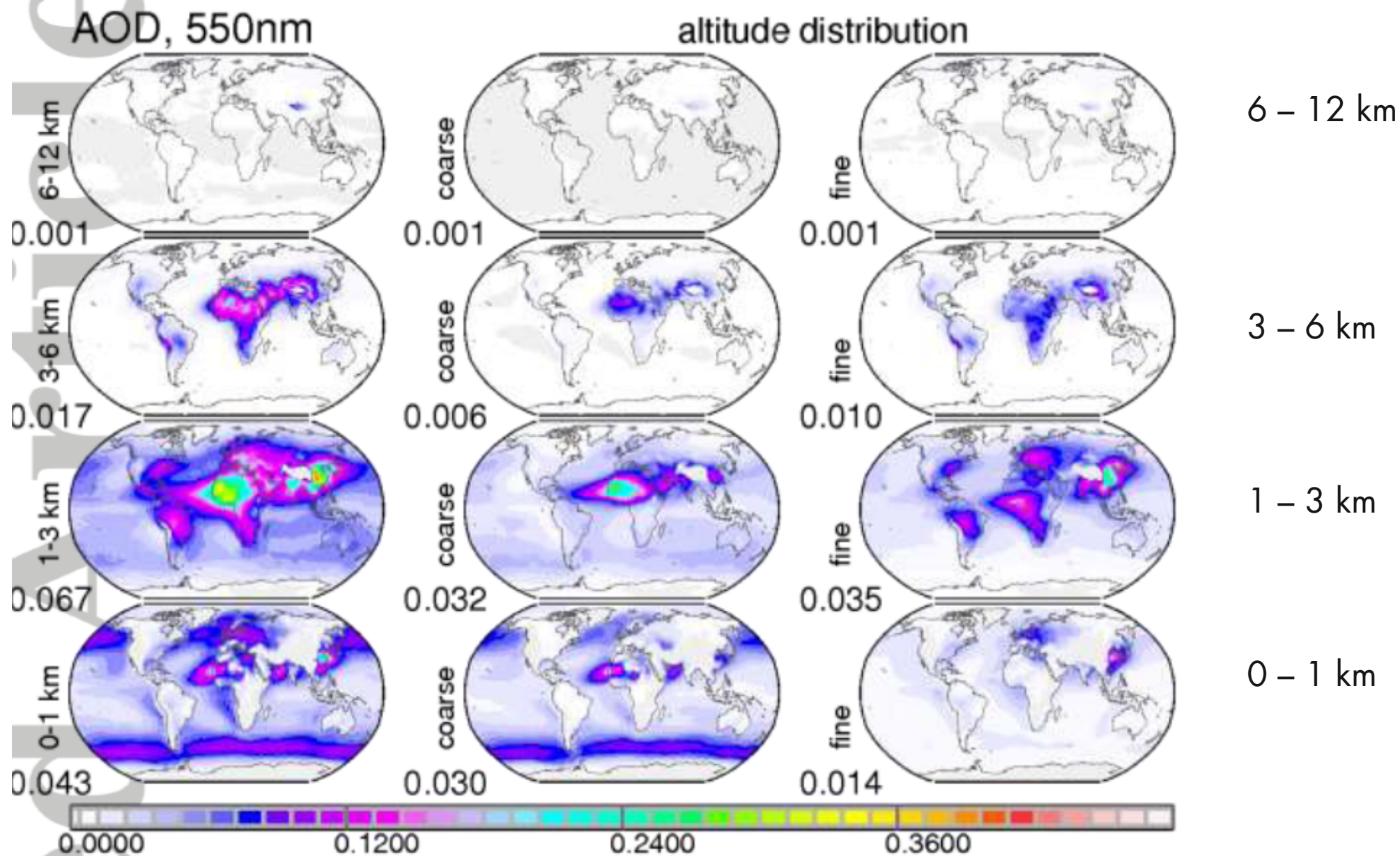
1.10 Aerosol distributions: Kinne climatology



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1.1.1 Light scattering and absorption: direct and semi-direct effects



→ aerosol particles scatter sunlight
(appear white from above)

1.11 Light scattering and absorption: direct and semi-direct effects



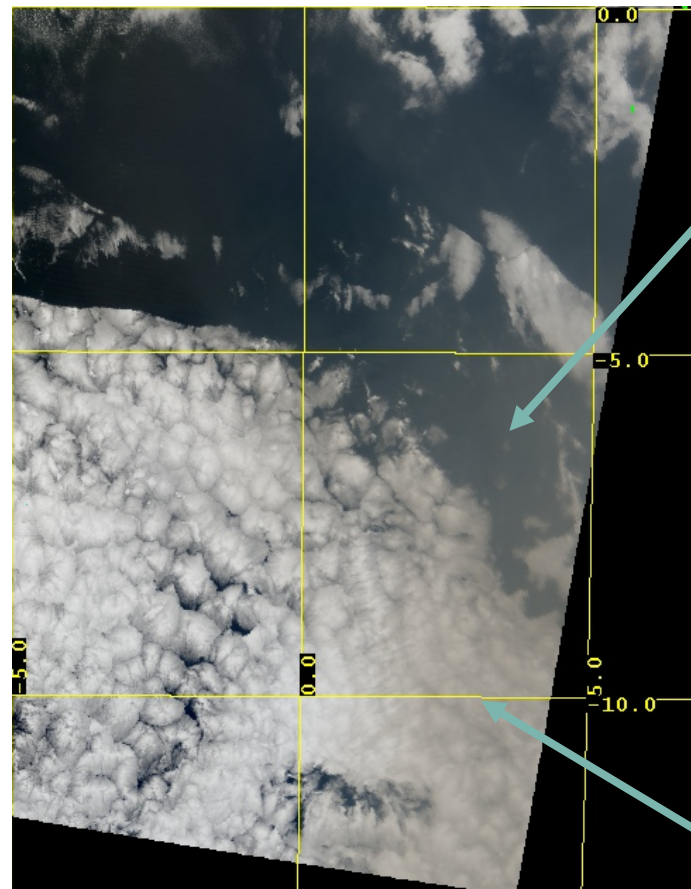
→ aerosol particles scatter sunlight
(appear white from above)

→ some also absorb sunlight
(appear black)



1.11 Light scattering and absorption: direct and semi-direct effects

- Scattering acts to cool system (top-of-atmosphere) and surface no effect within the atmosphere
- Absorption acts to warm the system cools the surface warms the atmosphere



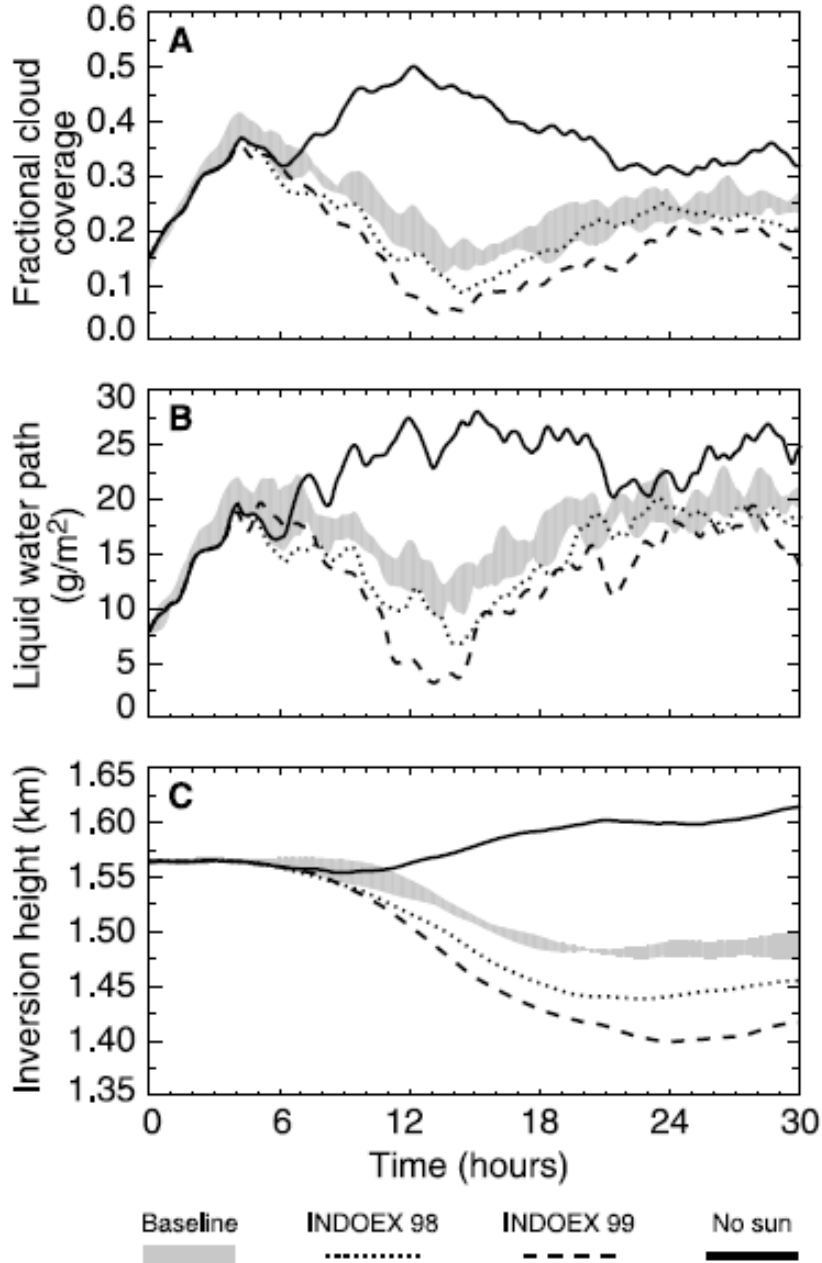
Albedo enhancement



Albedo reduction

MODIS 13 August 2006

1.12 Semi-direct effect?



Large-eddy simulations

- Aerosol absorption heats the upper boundary layer and reduces inversion and subsequently cloudiness
- could be opposite if aerosol layer is above clouds

2. No particles - no fog



Youtube "No particles no fog"
<http://www.youtube.com/watch?v=EneDwu0HrVg>

2.1 Köhler equation

(see lecture by Hanna Pawlowska)

$$S(r, T, B) \equiv \frac{p_{v,\text{sat,drop}}}{p_{v,\text{sat}}} = \exp \left(\frac{A(T)}{r} - \frac{B}{r^3} \right)$$

curvature effect (Kelvin term) solution effect (Raoult term)

in thermodynamic equilibrium

S – saturation ratio

(saturation vapour pressure above solution droplet vs. saturation vapour pressure over flat water surface)

↔ relative humidity

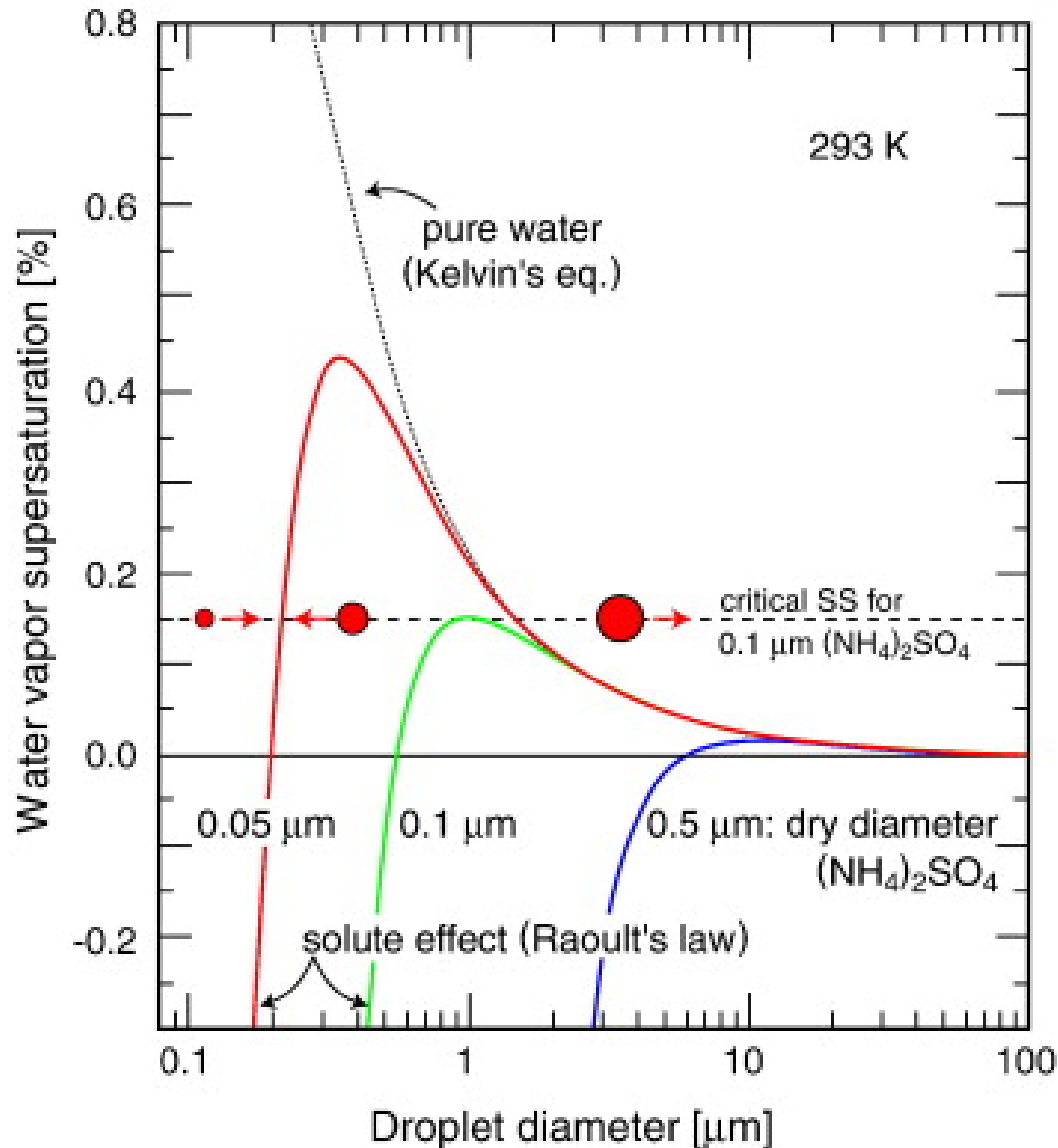
r – haze droplet radius

B – aerosol term

→ dependent on aerosol mass $\sim r_a^3$

→ dependent on solubility.

2.1 Köhler curve



- (i) Haze particles stable at low humidities
- (ii) Cloud droplets do not nucleate at 100% relative humidity
- (iii) Aerosol particle size very important
- (iv) Solubility relevant

2.2 Some results on basic aerosol-cloud interactions



true-colour image of dissipating cumulus



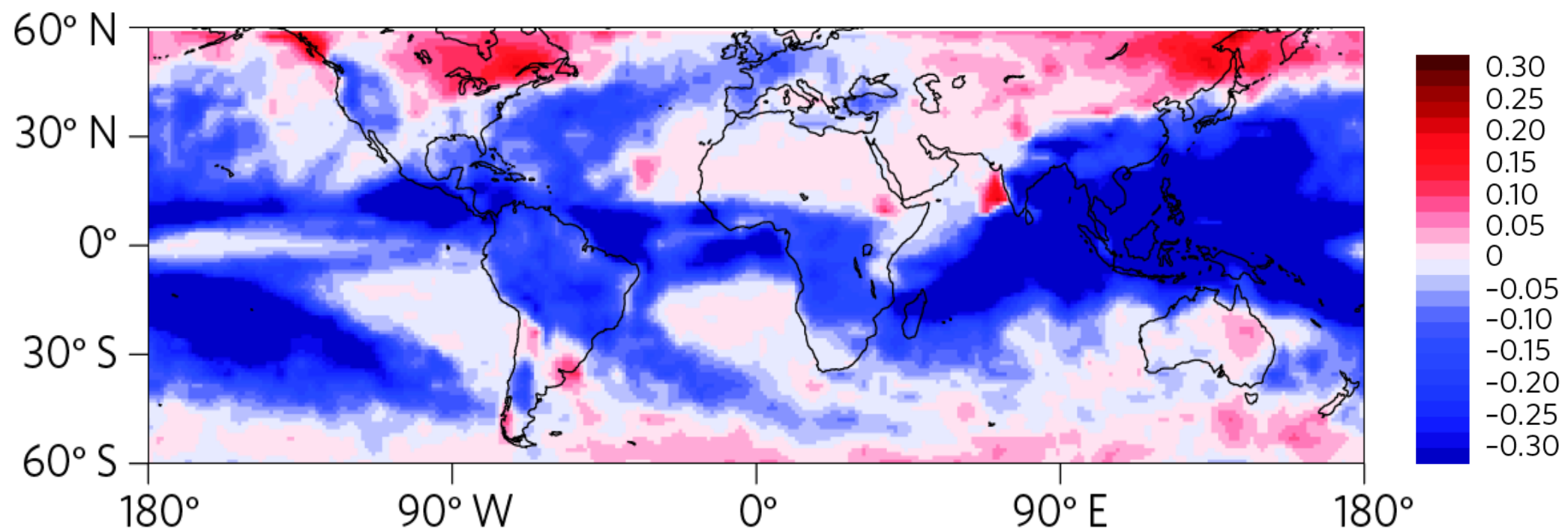
background gradients (molecular scattering) removed



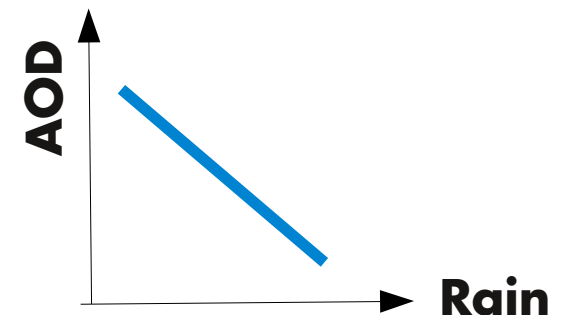
cloud masked and increased sensitivity

2.2 Some results on basic aerosol-cloud interactions

Statistical relationship between aerosol optical depth (logarithm of solar radiation dampening) and precipitation intensity

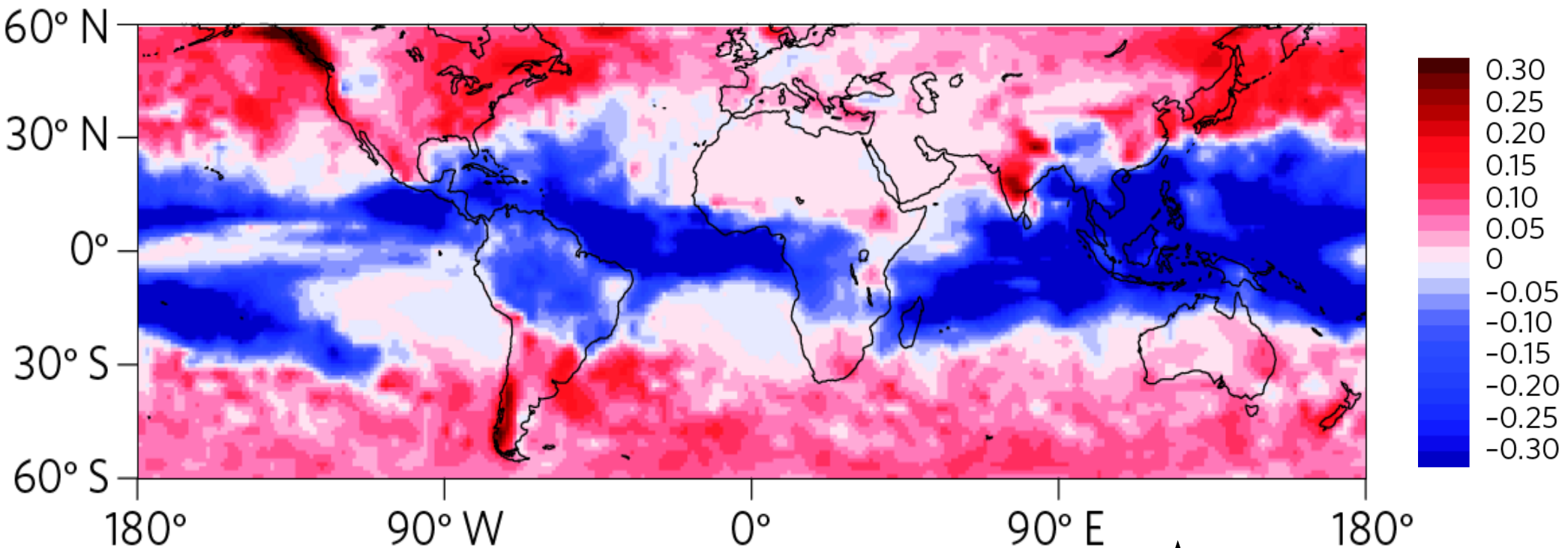


At each grid-point for one season of data difference between upper and lower tercile in aerosol optical depth (**dry** aerosol) in rain intensity [mm hour^{-1}]

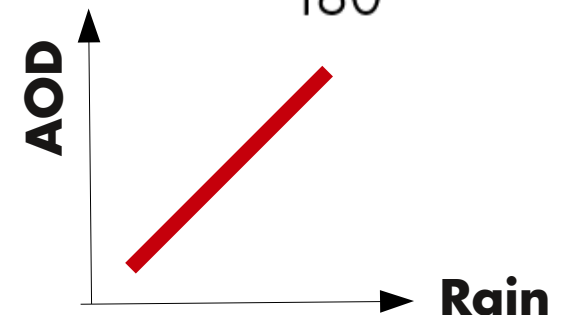


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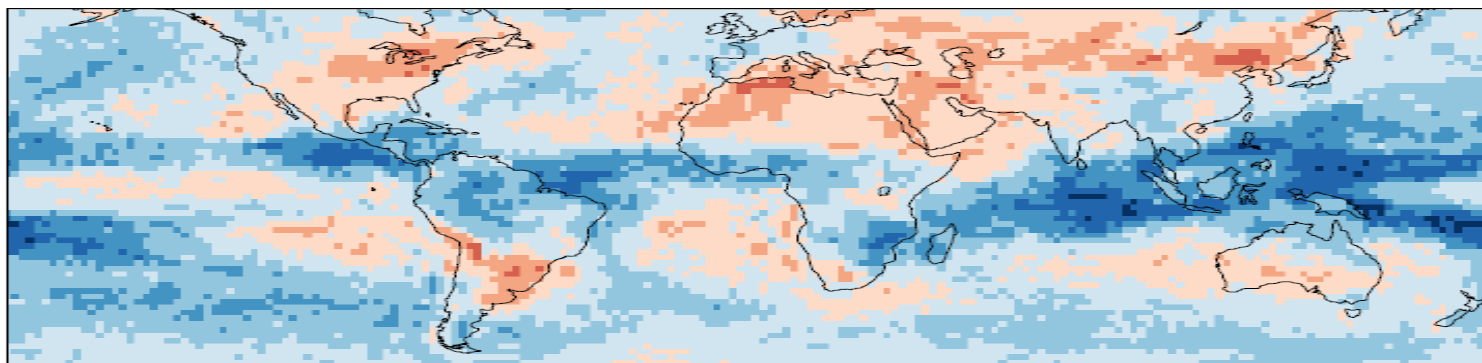
Statistical relationship between aerosol optical depth (logarithm of solar radiation dampening) and precipitation intensity



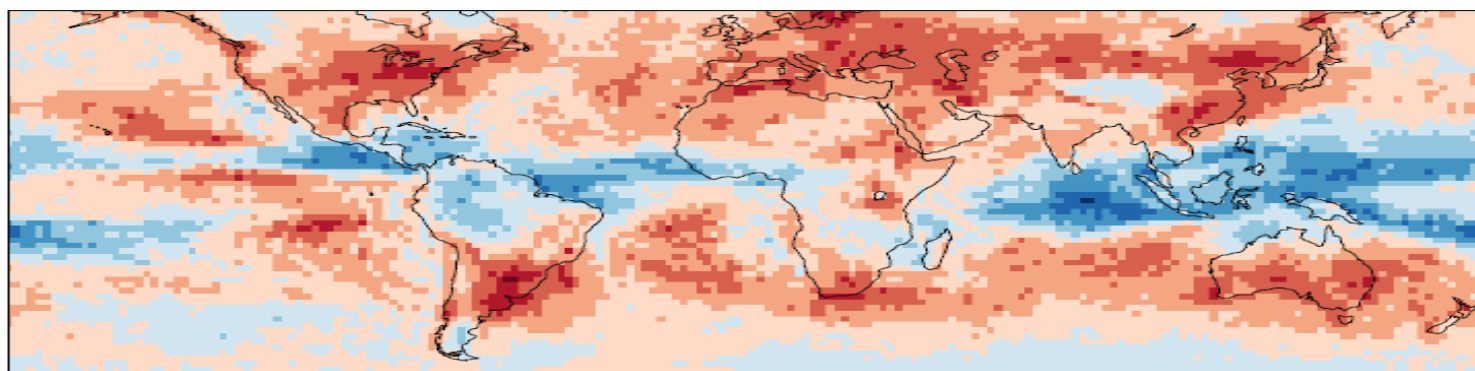
At each grid-point for one season of data difference between upper and lower tercile in aerosol optical depth (**wet** aerosol) in rain intensity [mm hour^{-1}]



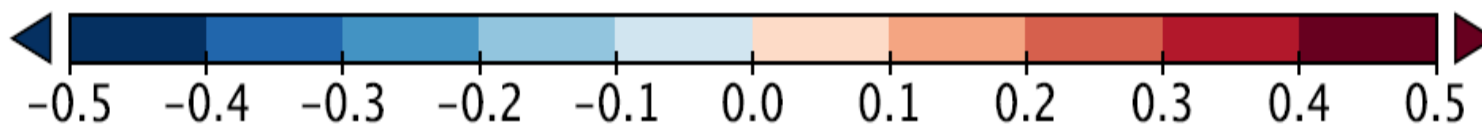
Statistical relationship between AOD and cloud cover



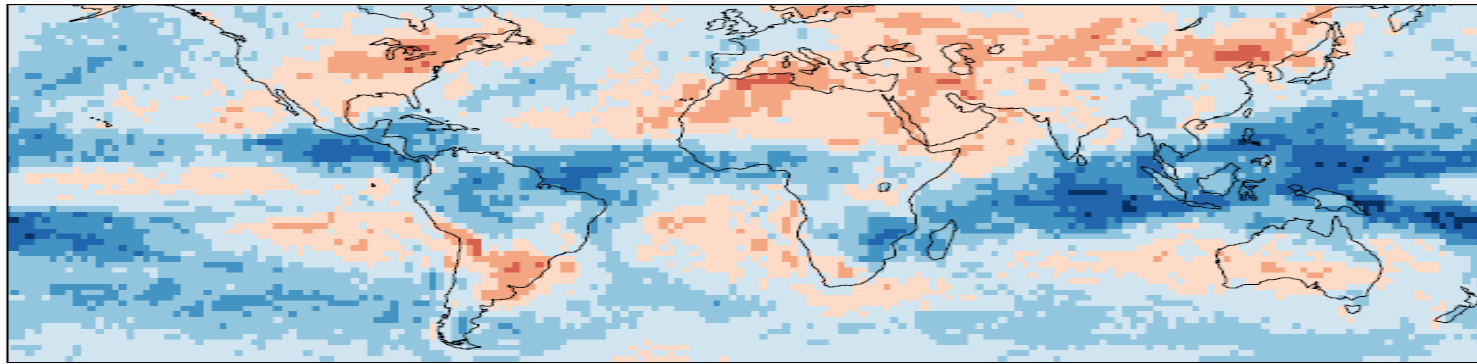
Dry AOD



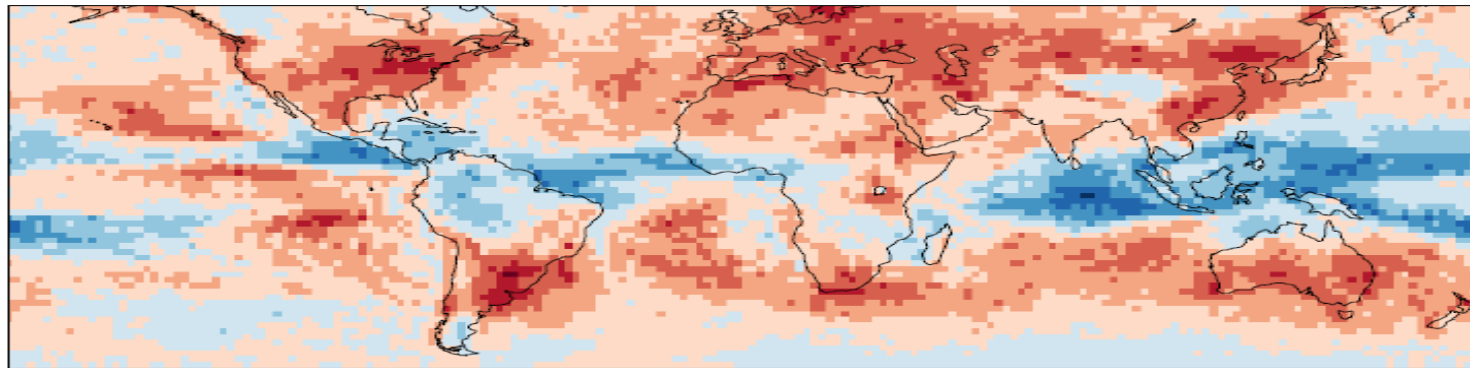
Wet AOD



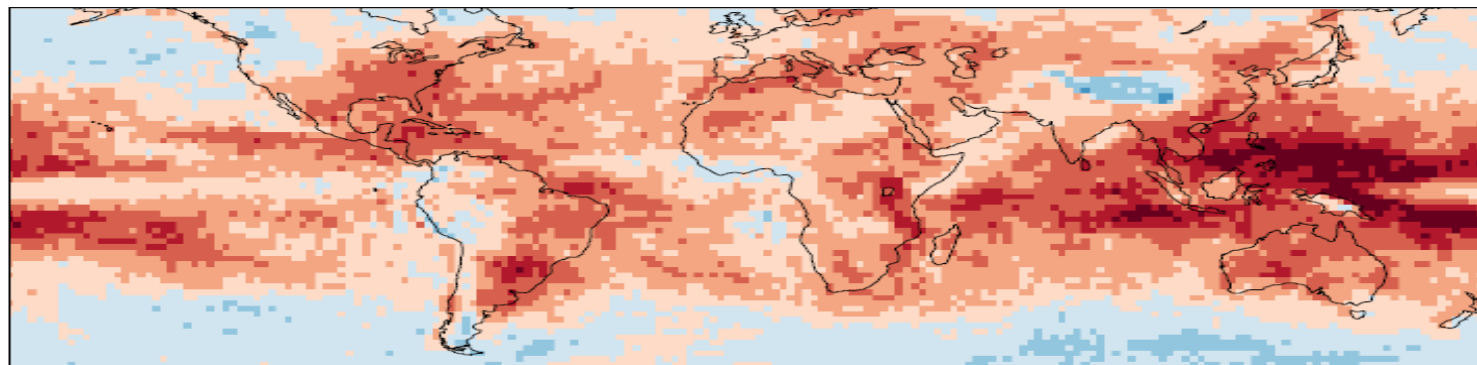
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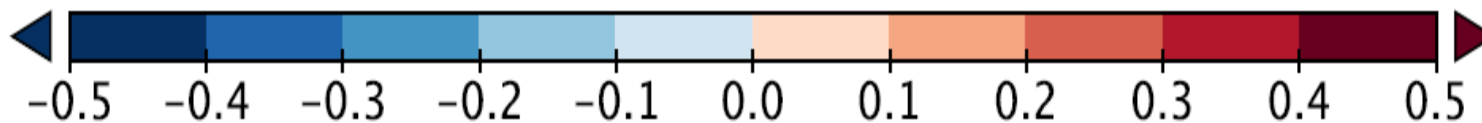
Dry AOD



Wet AOD

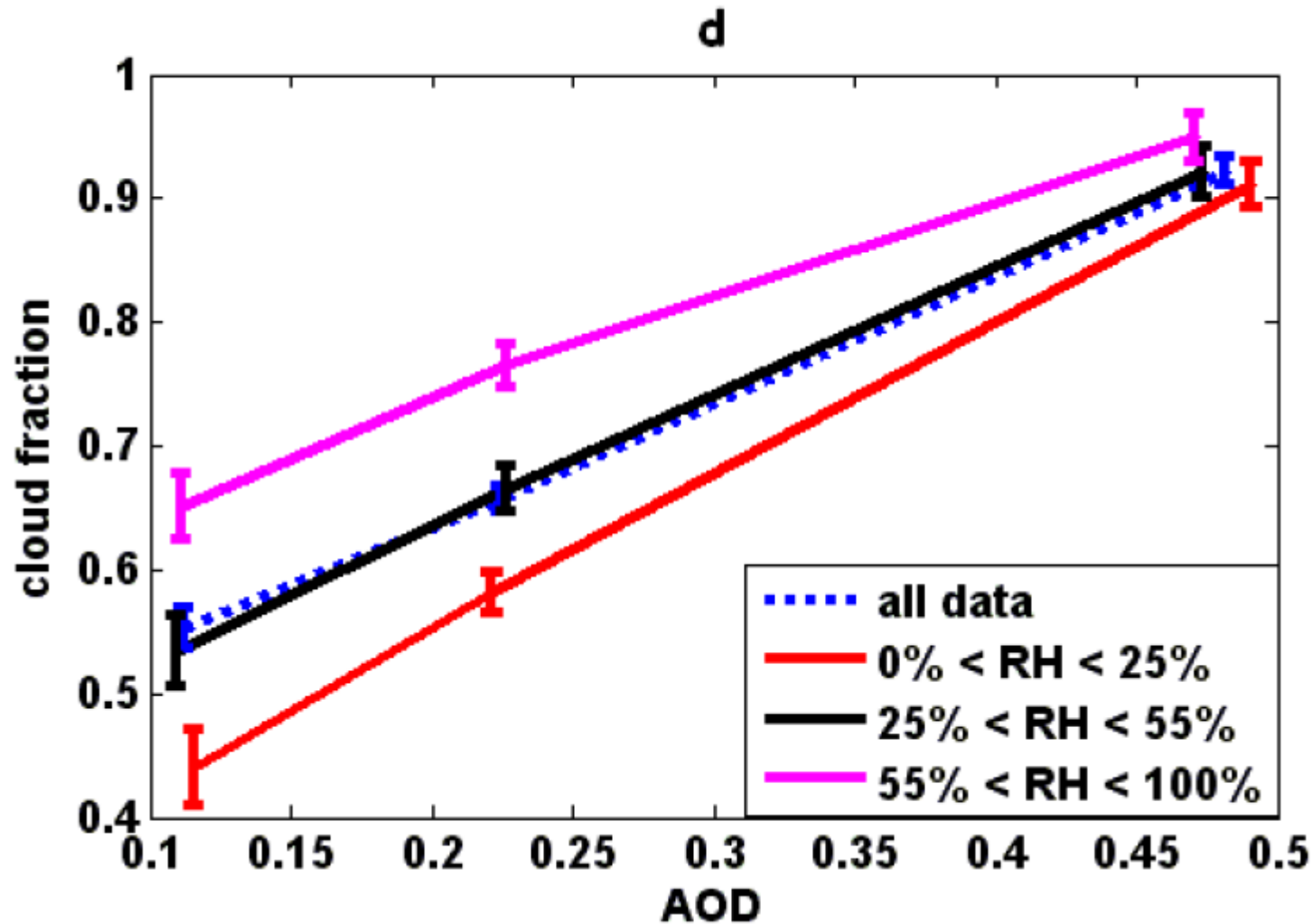


No scavenging by
convective
precipitation



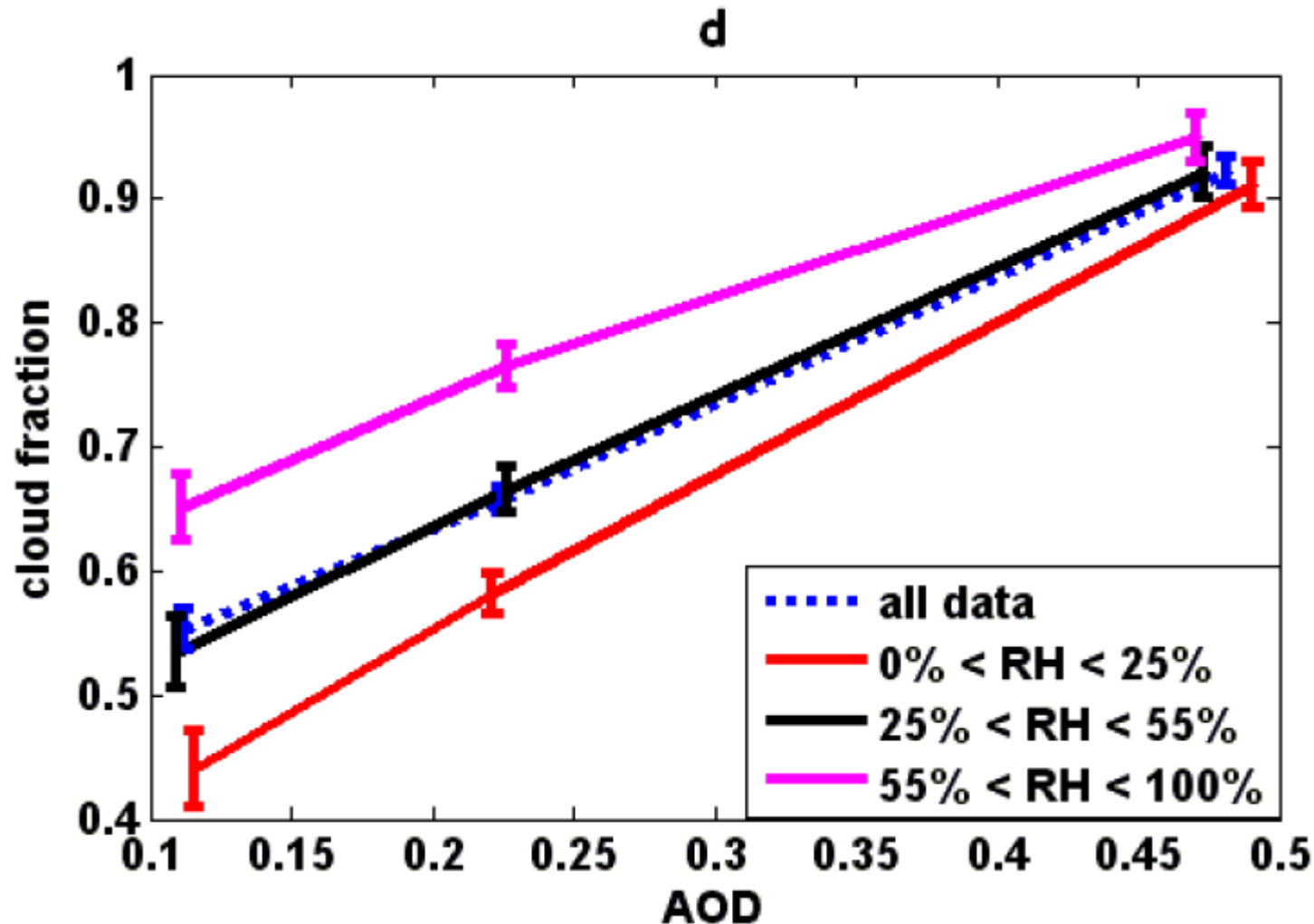
2.2 Some results on basic aerosol-cloud interactions

MODIS cloud fraction as a function of AOD for classes of re-analysis relative humidity at 350 hPa



2.2 Some results on basic aerosol-cloud interactions

MODIS cloud fraction as a function of AOD for classes of re-analysis relative humidity at 350 hPa



→ even for $RH < 25\%$ cloud fractions of up to 90%.

→ **subgrid-scale variability of humidity** essential (strong non-linearity of swelling)

→ see Poster by Vera Schemann and Matthias Brück

2.3 Radiation sensitivity: First aerosol indirect effect

See H el ene Chepfer's lecture:

$$\tau = \frac{3}{2} \frac{L}{\rho_w r_e} \quad (\text{vertically homogeneous droplet size distribution})$$

with $r_e = \beta r_v$ and $N_d = \int_0^\infty n(r) dr$ the total cloud droplet number concentration

$$r_e = \beta \sqrt[3]{\frac{3}{4\pi} \frac{q_l}{\rho_w} \frac{1}{N_d}}$$

$\tau \propto \sqrt[3]{N_d}$ optical depth a cube-root function of droplet concentration, N_d .

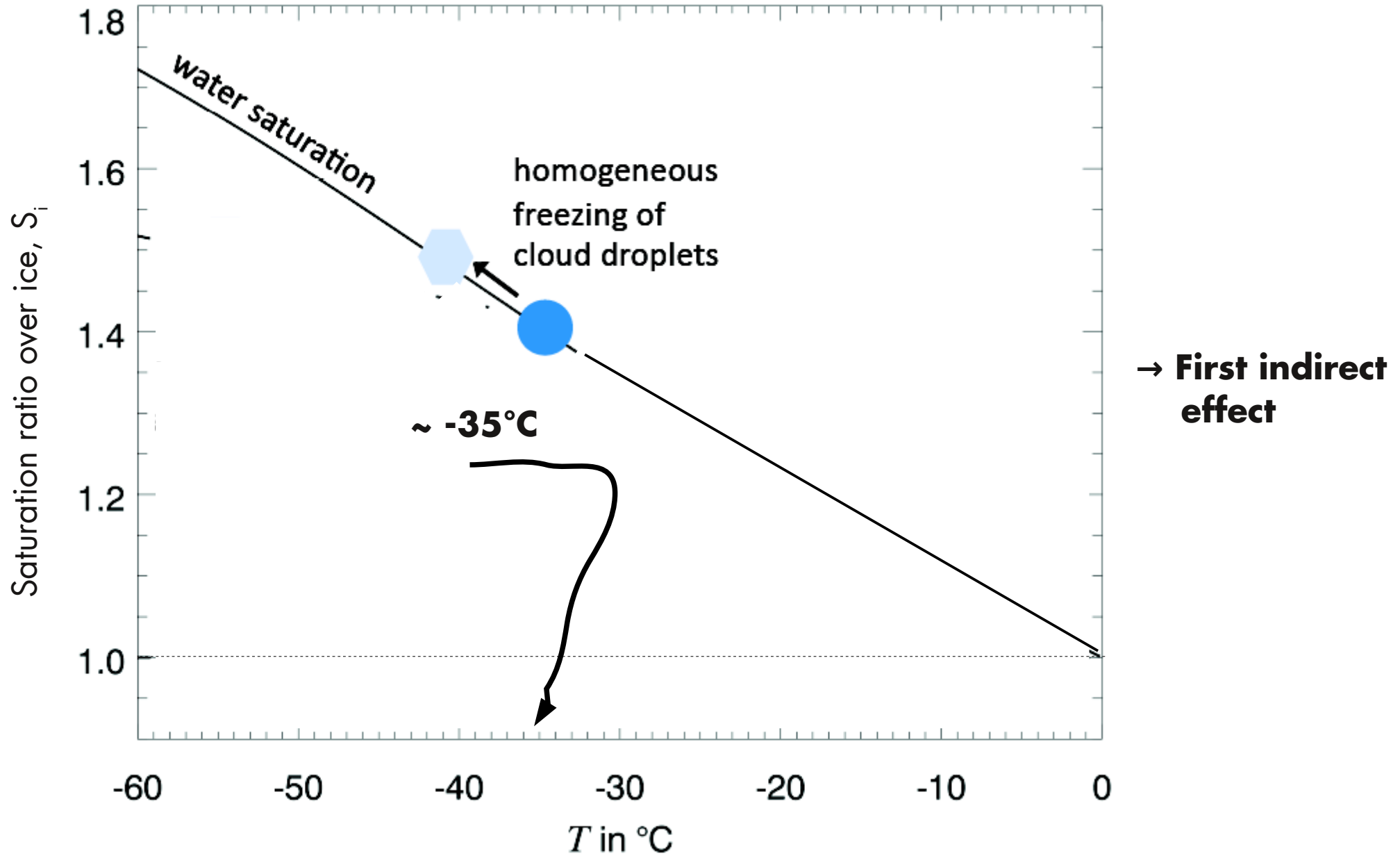
In two-stream approximation for the cloud albedo, α :

$$\Delta \ln \alpha = (1 - \alpha) \Delta \ln \tau$$

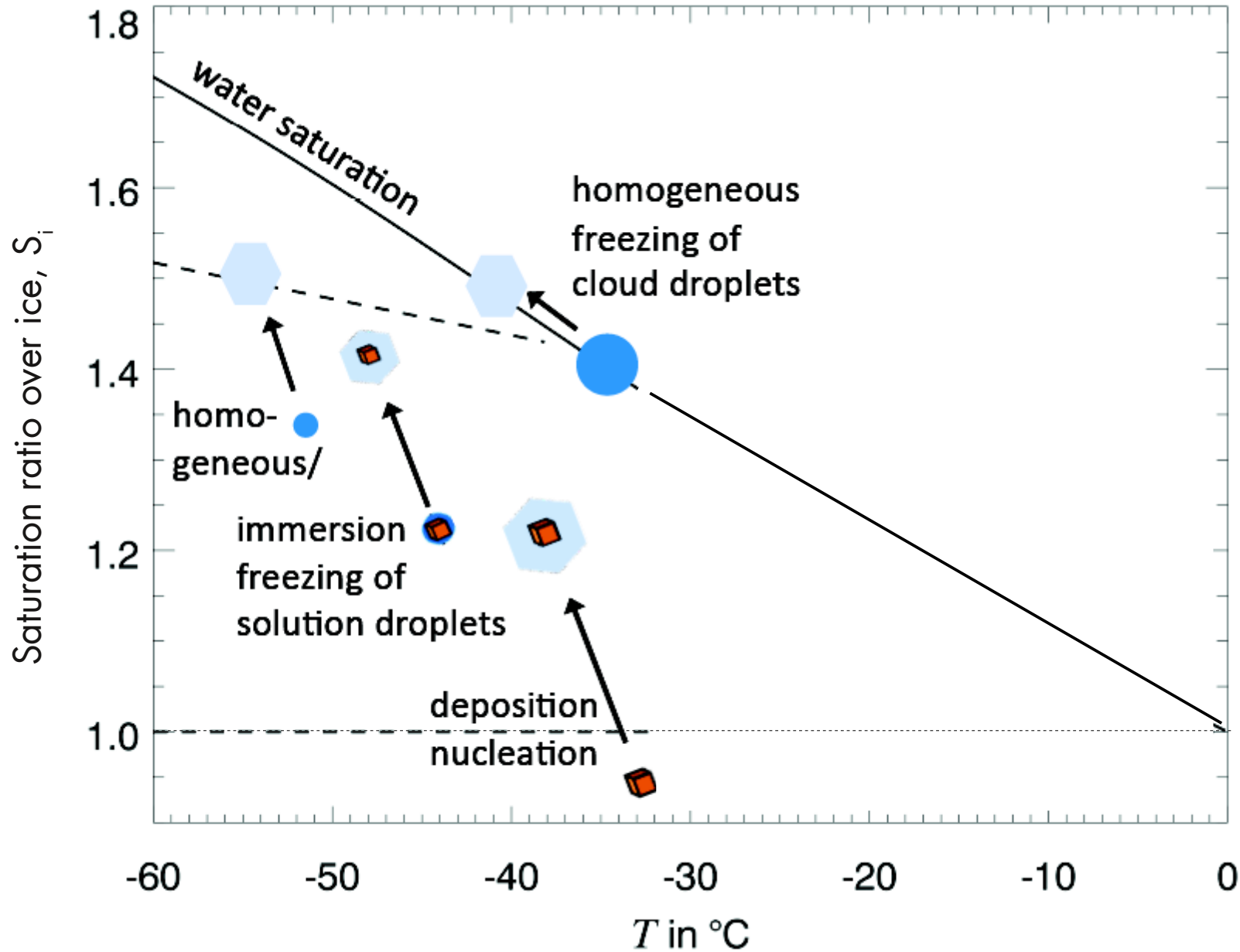
For constant droplet spectrum shape and liquid water path:

$$\frac{\Delta \ln \alpha}{\Delta \ln N_d} = \frac{1 - \alpha}{3} \quad \rightarrow \text{first aerosol indirect effect (Twomey effect)}$$

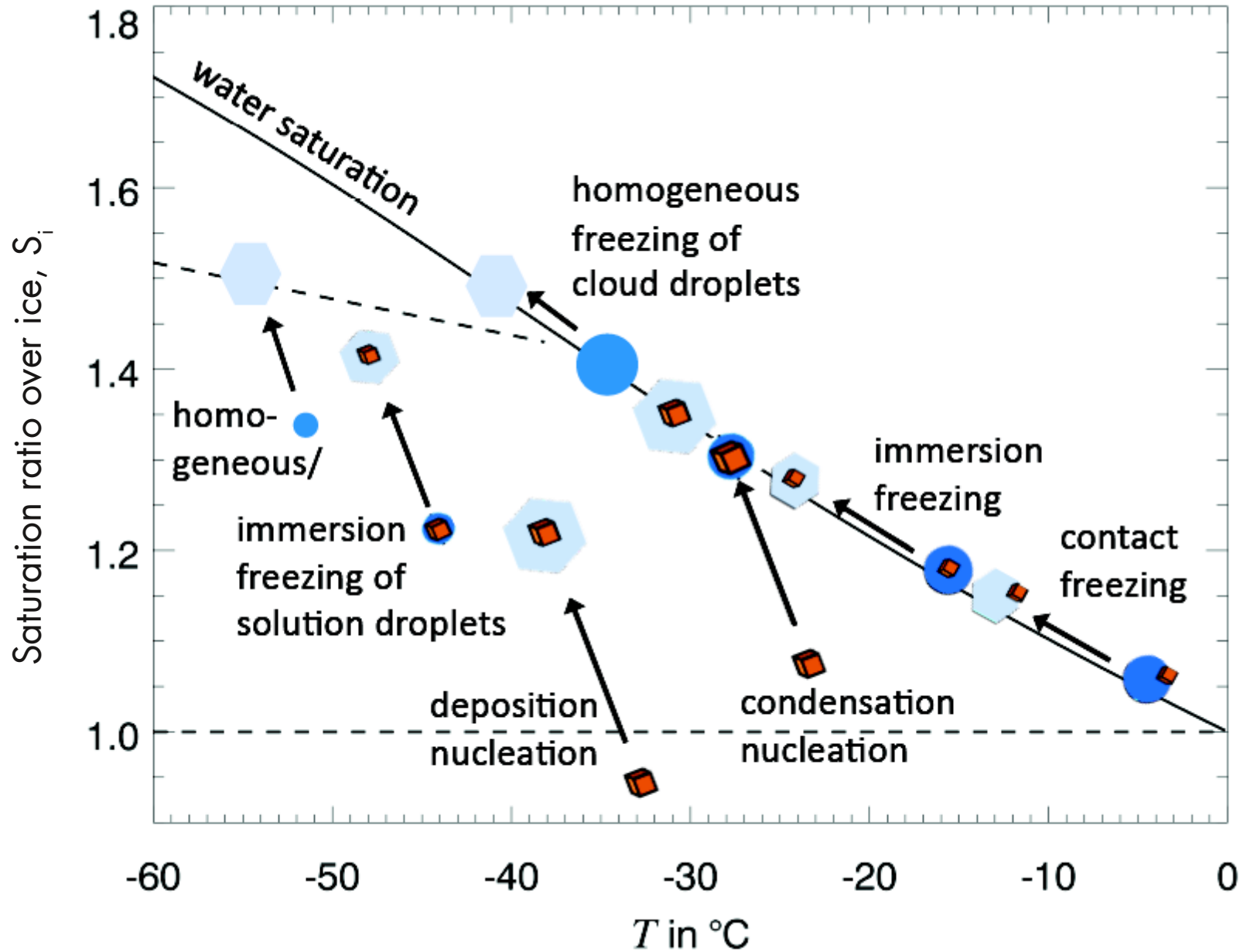
2.4 Ice crystals



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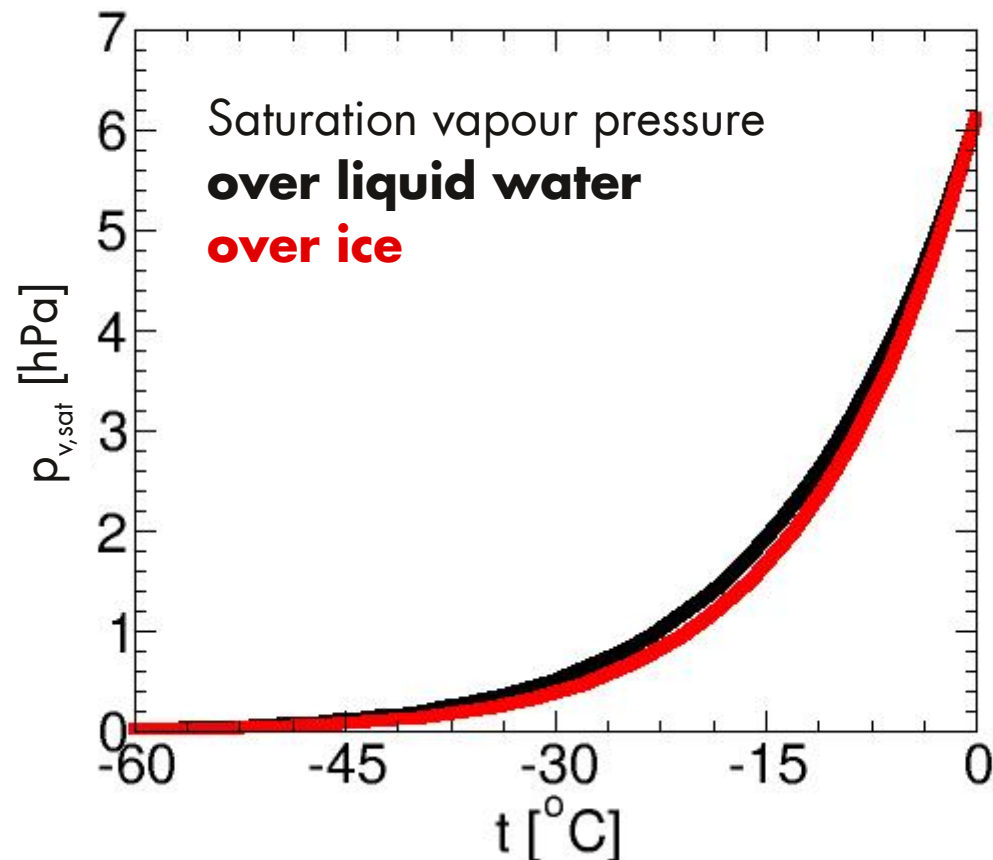


**heterogeneous
freezing of
cloud droplets**

→ effect ?

2.5 Mixed-phase clouds

- (i) Freezing of drops is a function of droplet size:
 - In larger drops the probability of formation of an ice germ is larger
- (ii) Ice crystals grow at the expense of liquid droplets:
 - Bergeron-Findeisen process



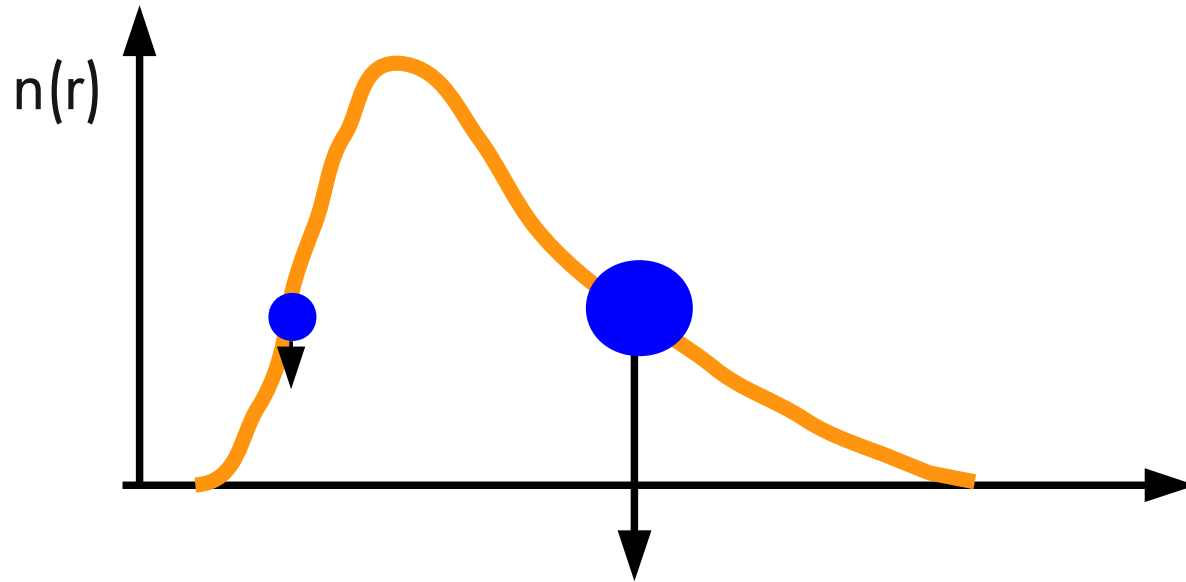
2.6 Glaciation and de-activation effects

- (i) first indirect effect (homogeneous & deposition freezing)
- (ii) Bergeron-Findeisen process:
 - if IN lead to early onset of freezing, the entire cloud might glaciate
 - precipitation → reduced lifetime
- (iii) De-activation of IN:
 - if sulfate condenses onto dust particles, they may not serve as IN any more
 - longer lifetime ?

2.7 Microphysics

Terminal fall velocity depends on particle radii

→ particles with different sizes fall at different speeds

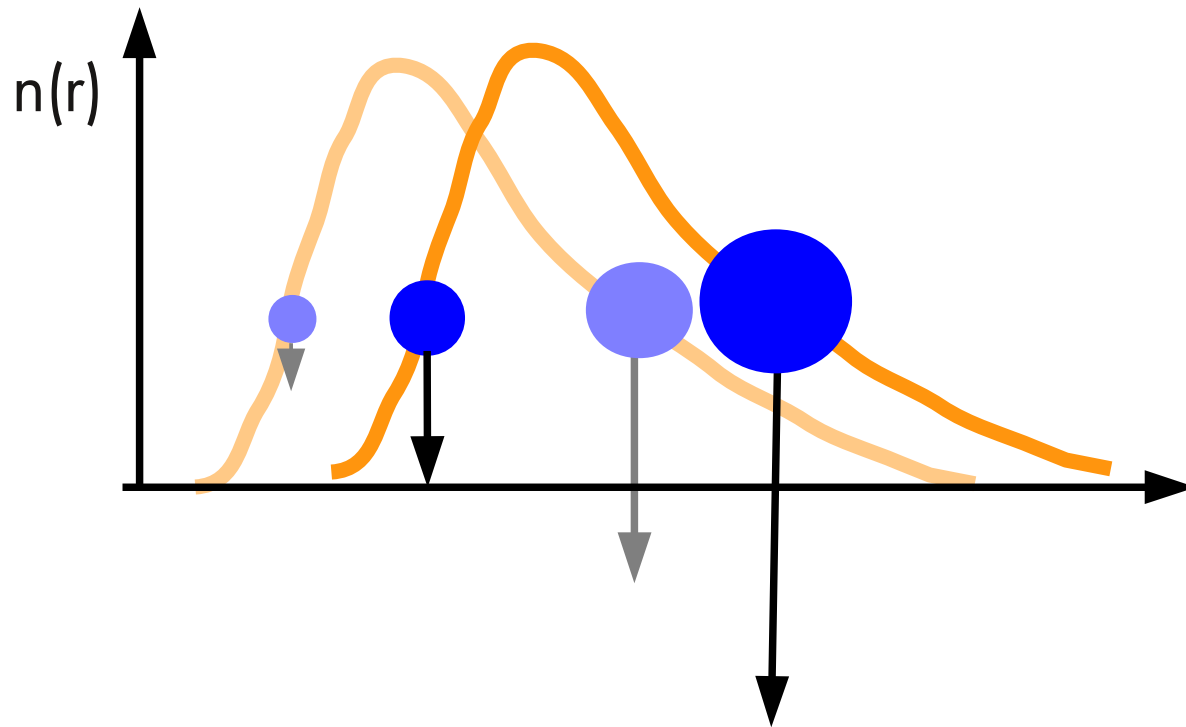


2.7 Microphysics

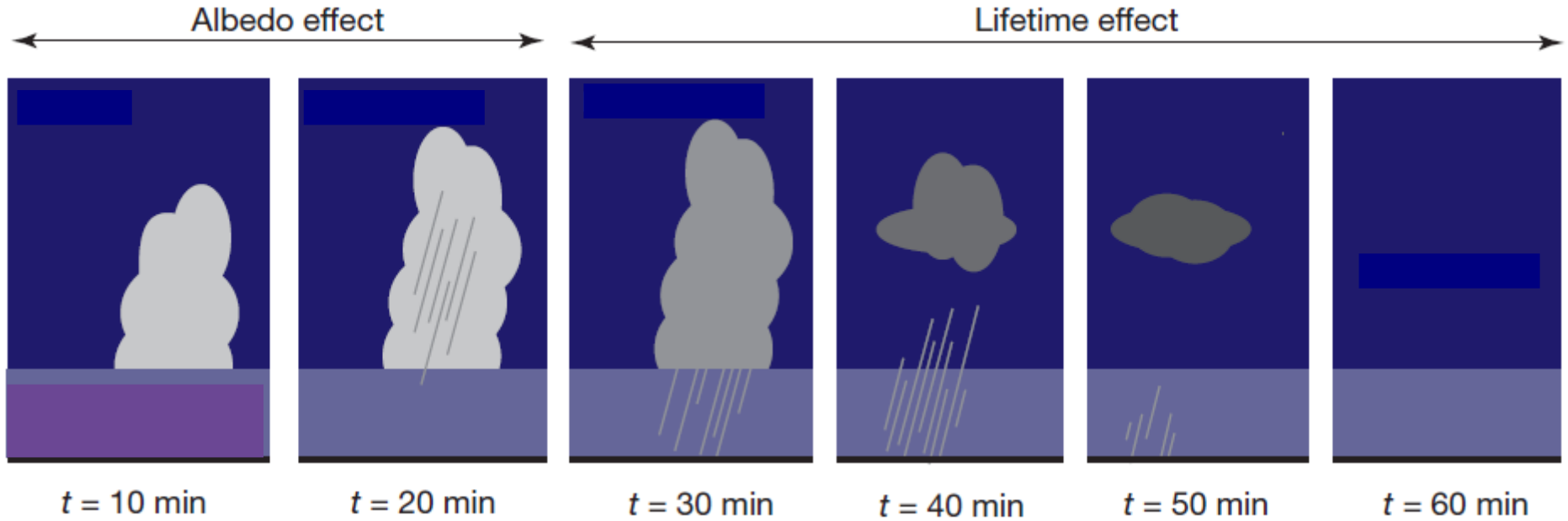
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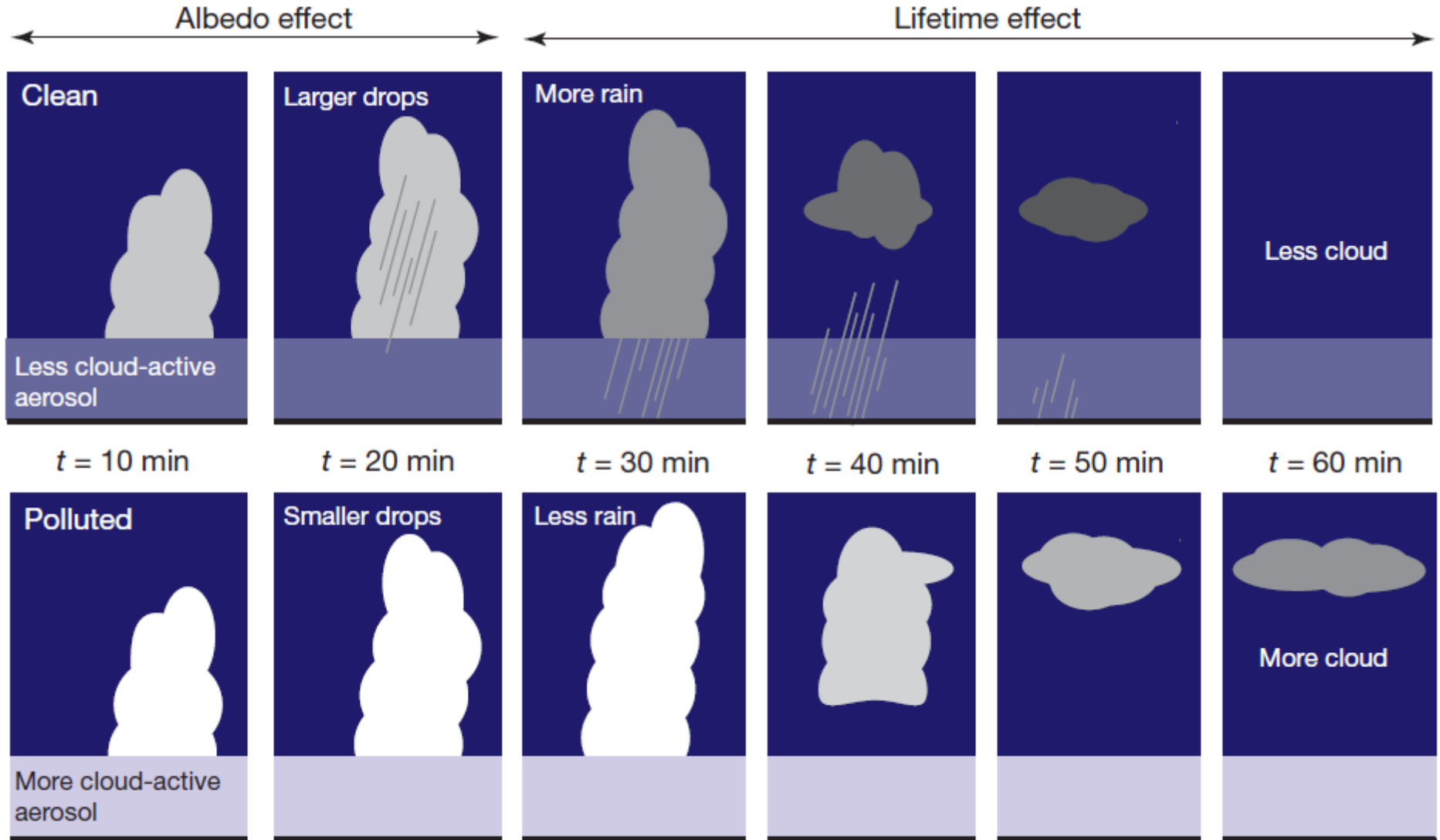
→ shift in droplet size spectrum affects collision/coalescence process



2.8 Cloud lifetime effect



2.8 Cloud lifetime effect



→ larger cloud liquid water path
→ larger cloud fraction

2.9 Water budget consideration

- Total water (water vapour, cloud water, precipitation water) is conserved.
- Changes in accumulated precipitation (at a large scale over long times) possible only if evaporation fluxes change

$$\langle P \rangle = \int_{\text{area}} \int_{t=0}^{\tau} p(P) I(P) dt dA$$

A – Area

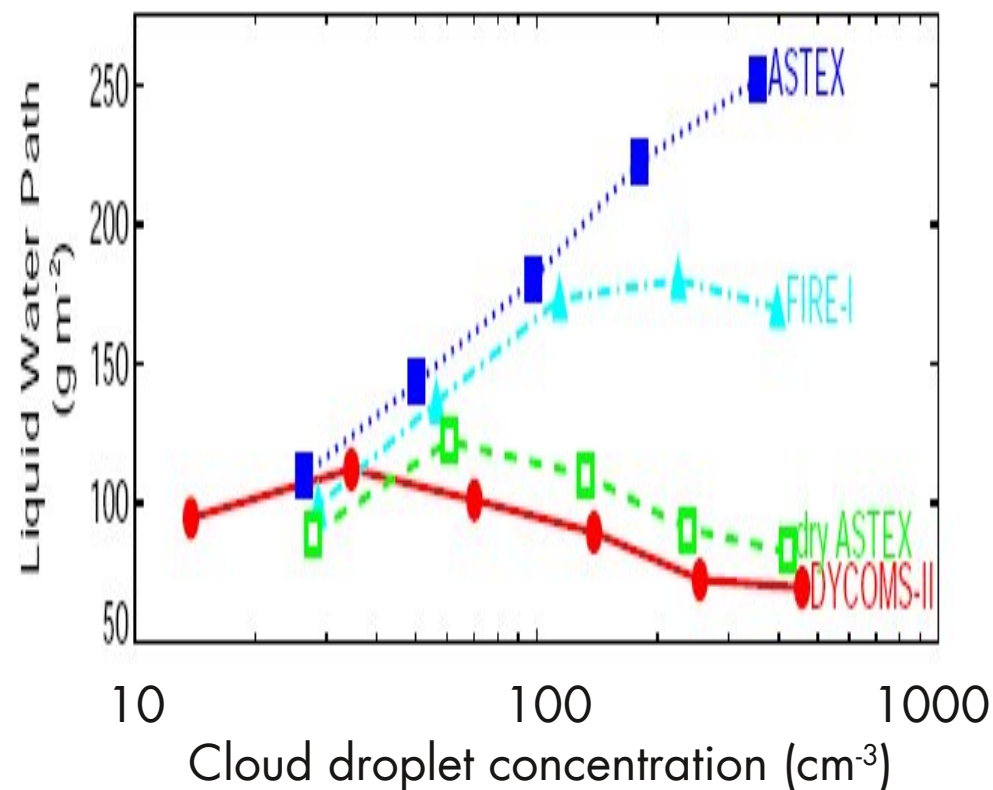
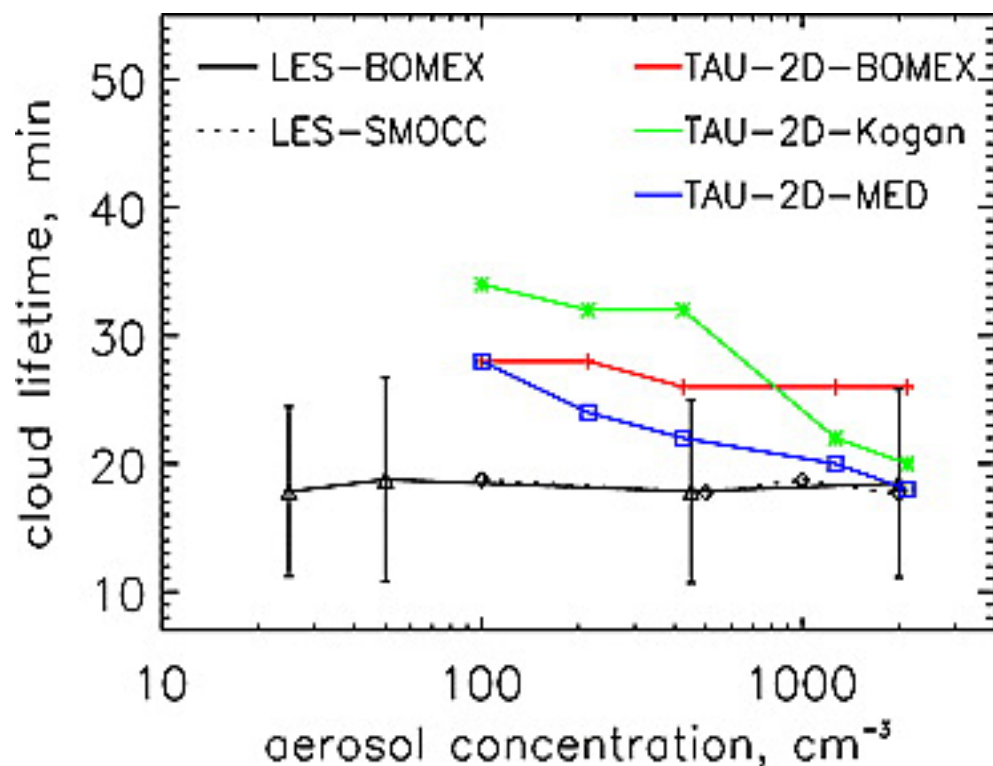
t – time

p(P) – probability of occurrence of precipitation

I(P) – intensity of precipitation

- Precipitation characteristics [p(P) and I(P)] not constrained by this.

2.10 Feedbacks, "dampening"

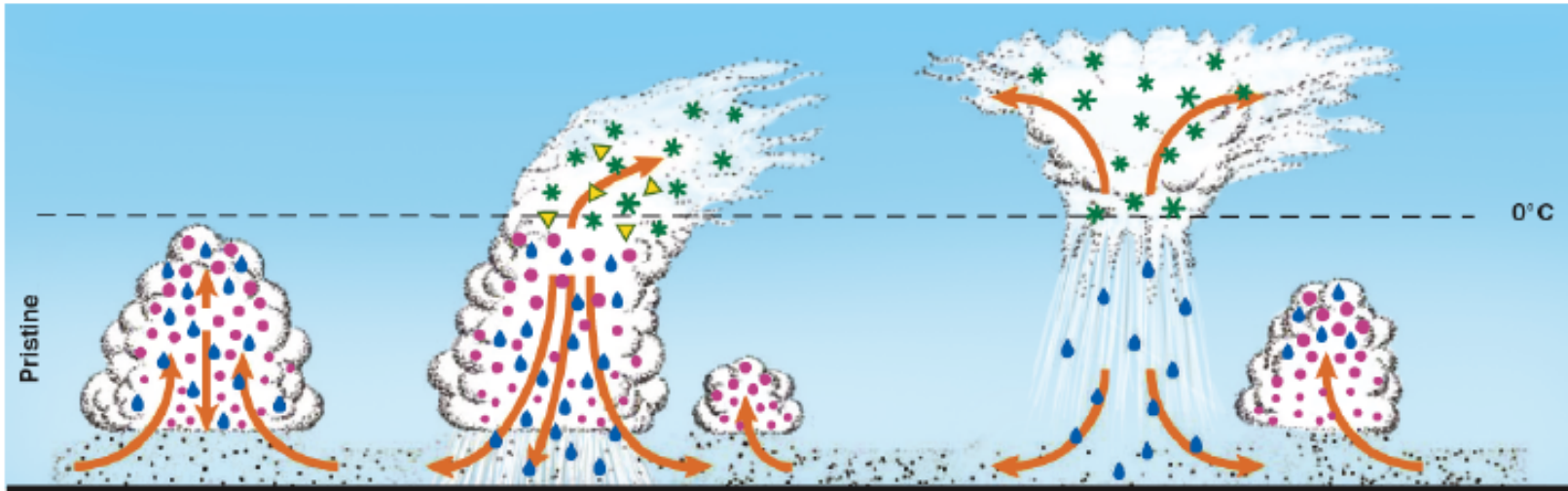


Large-eddy simulations for different cases

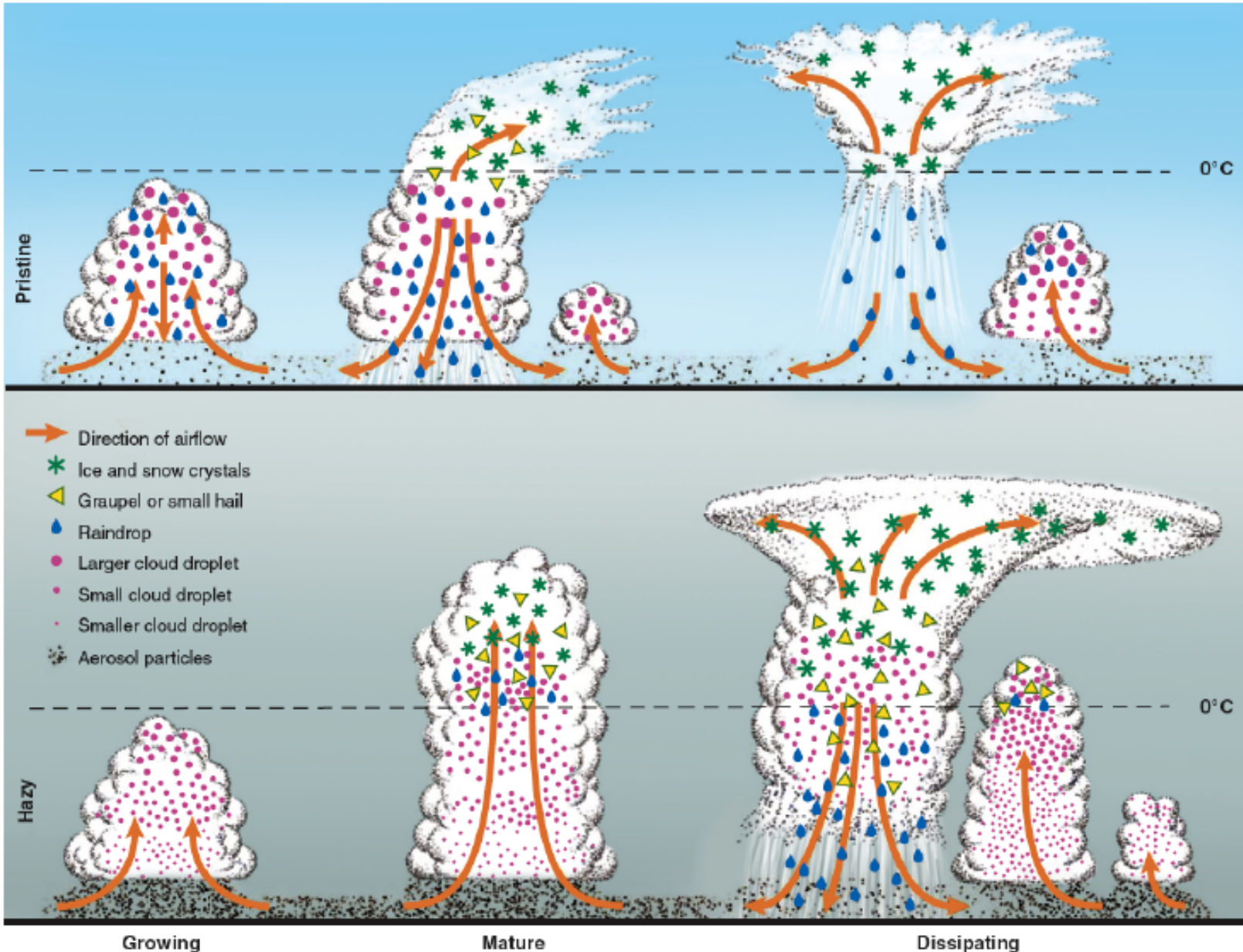
→ often shorter rather than longer cloud lifetimes due to faster evaporation of smaller droplets

→ liquid water path response also different for different cloud regimes

2.11 Freezing effect: Invigoration



2.11 Freezing effect: Invigoration

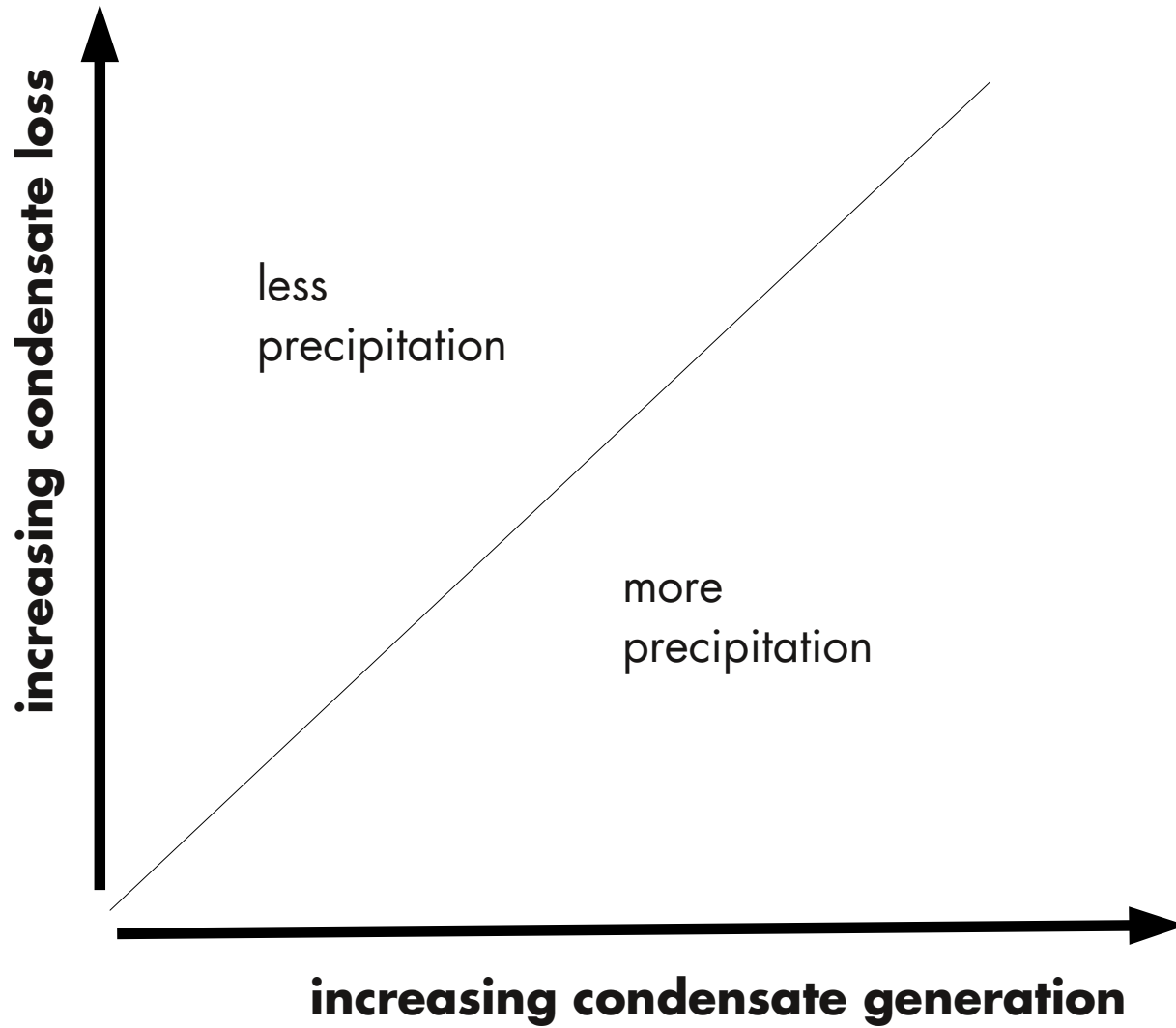


(i) delayed precipitation
→ more liquid water brought to freezing level

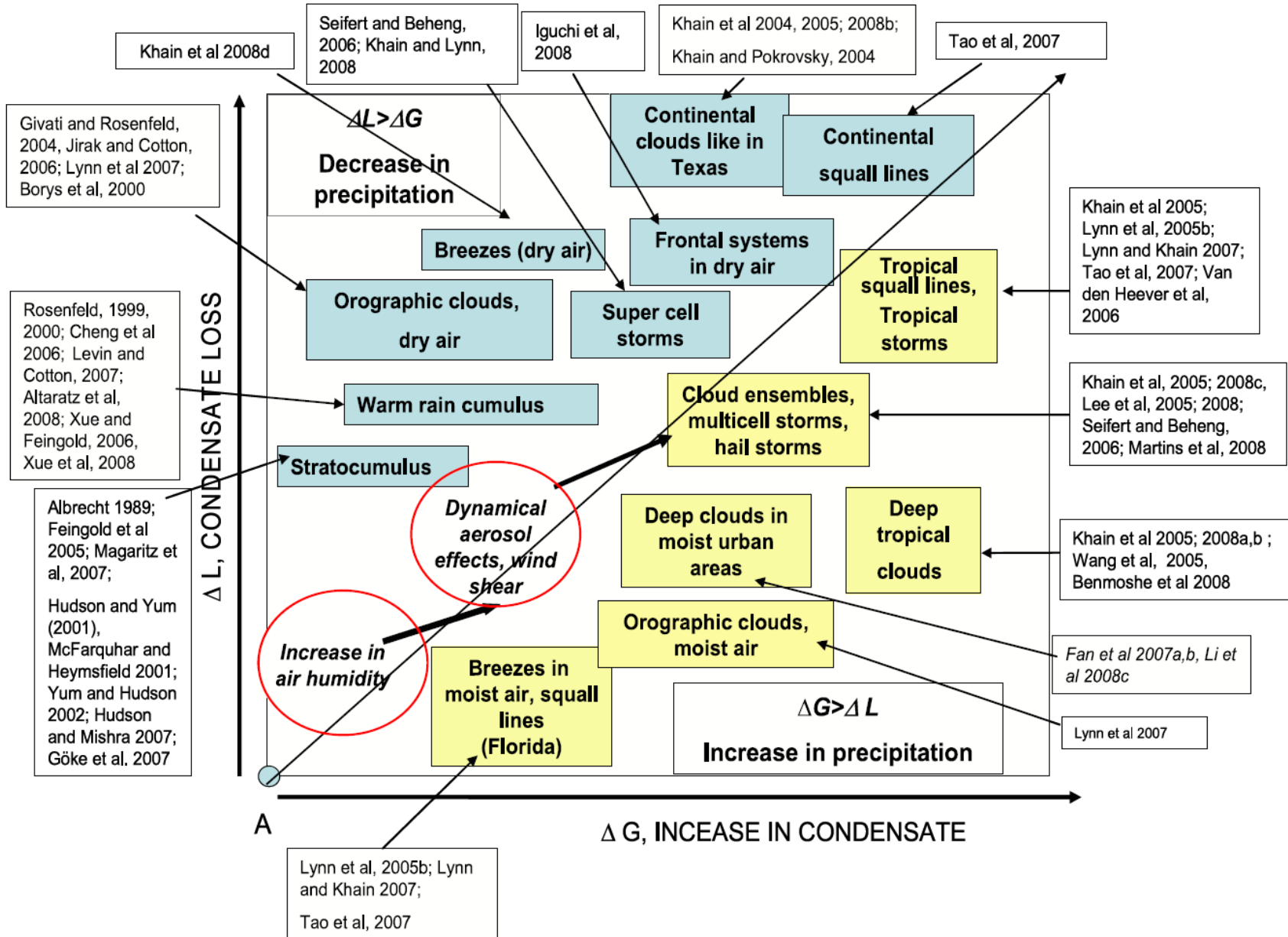
(ii) smaller droplets freeze later (higher)
→ grow more
→ more liquid water freezes

→ more latent heat release

2.12 Results from small scales on condensate



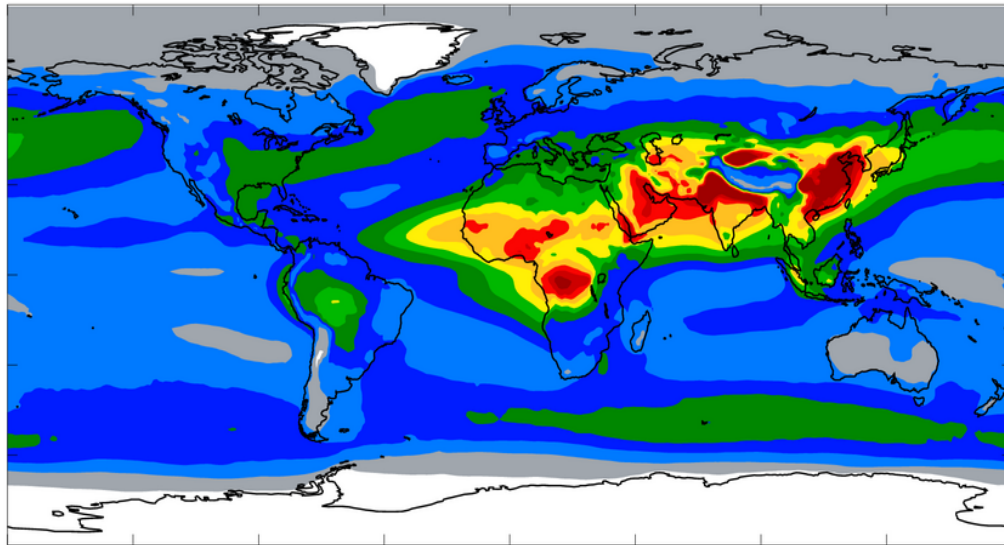
2.12 Results from small scales on condensate



3. Anthropogenic aerosols

3.1 Monitoring Atmospheric Composition and Climate: MACC reanalysis

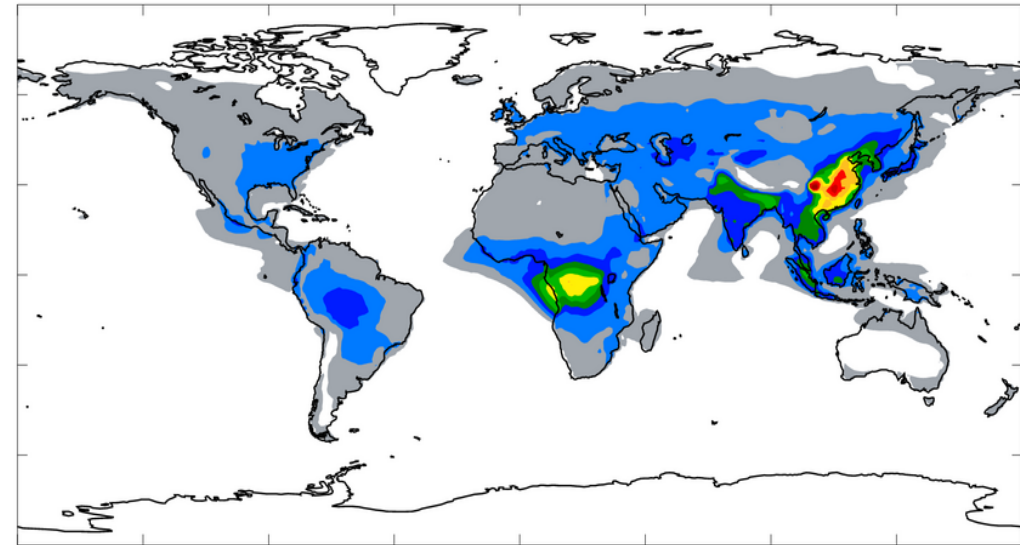
(see Poster Karoline Block)



Mean: 0.173



Total Aerosol optical depth (2003-05)
Global annual mean: 0.173

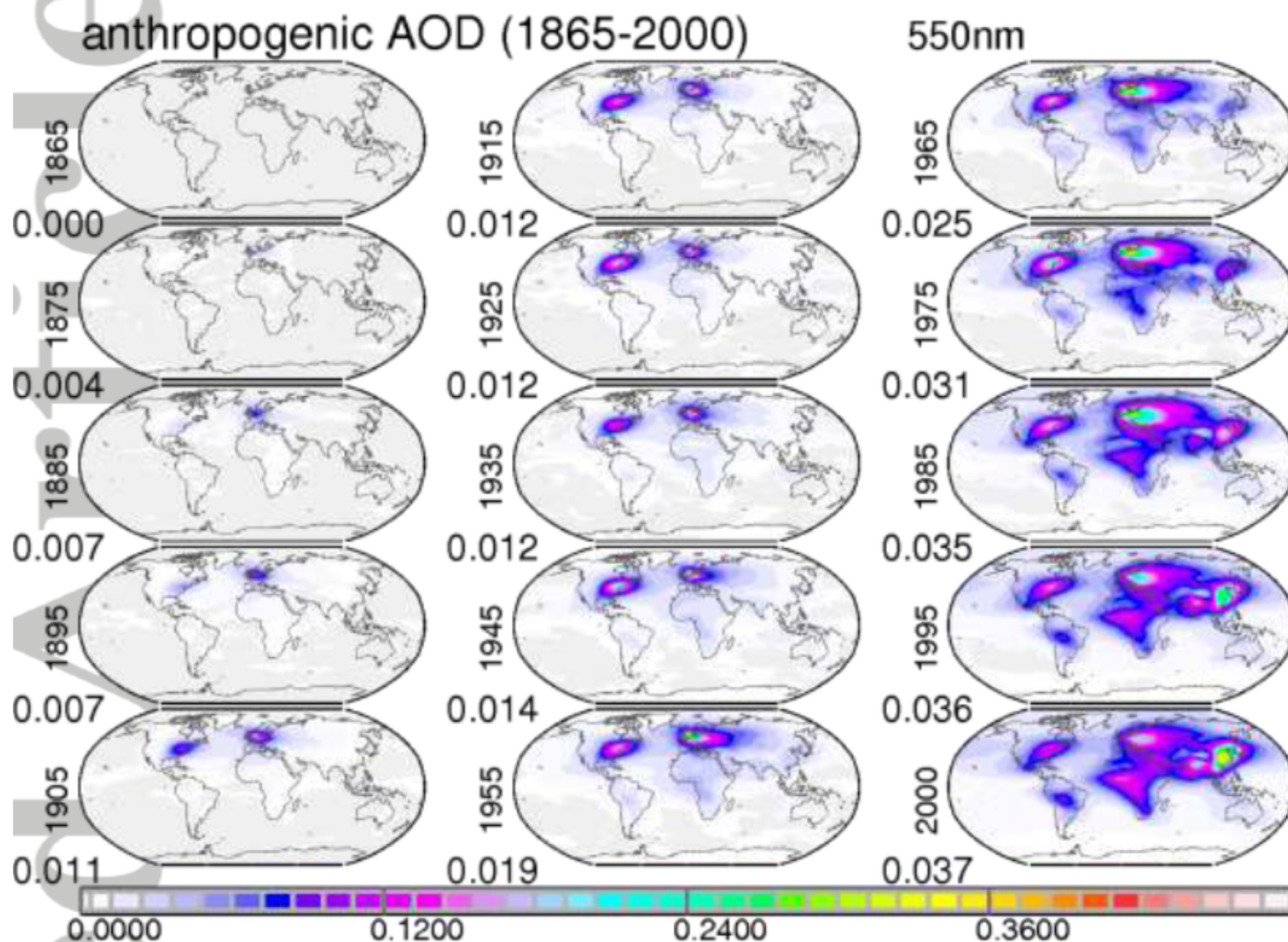


Mean: 0.047

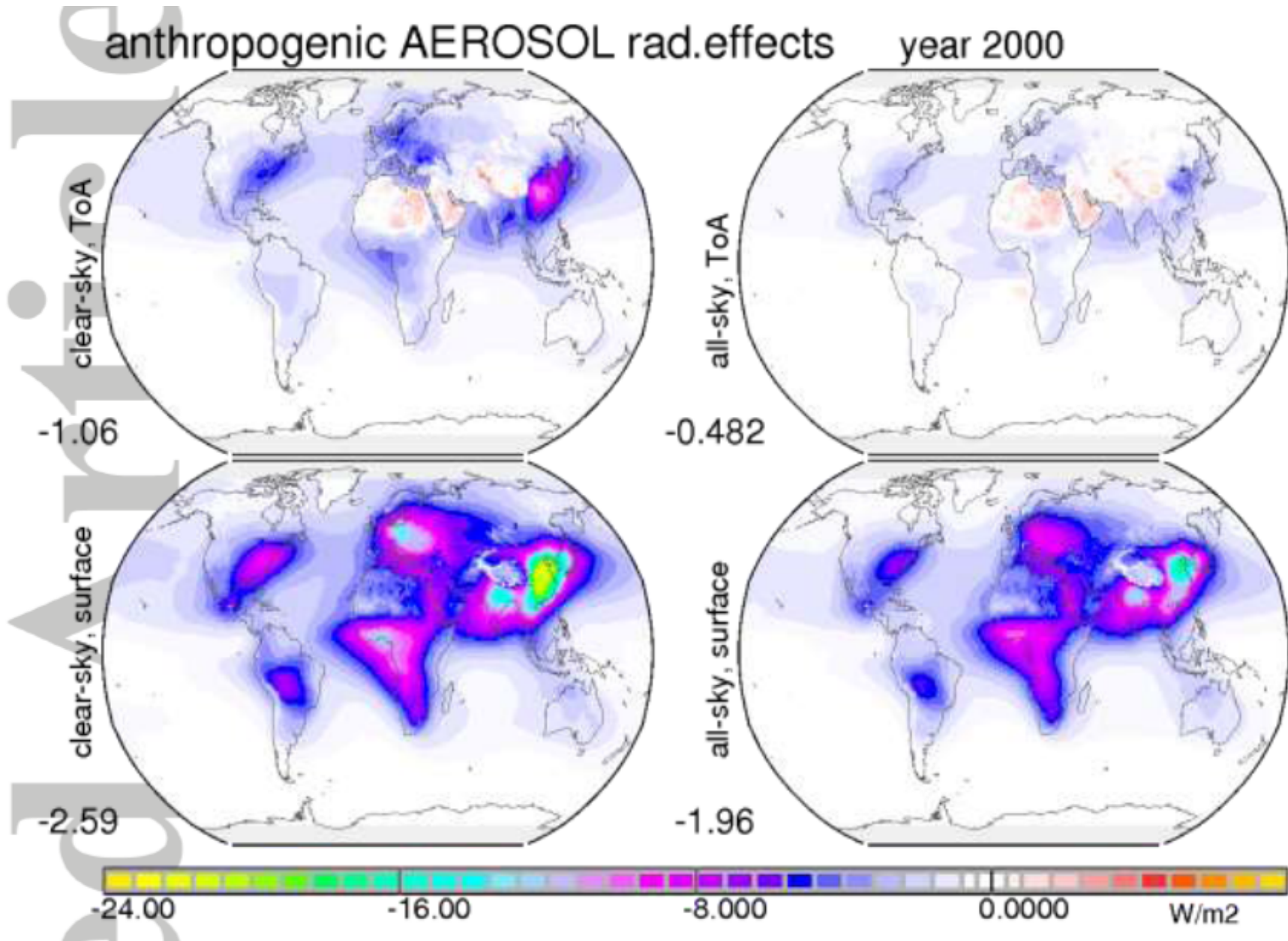


Anthropogenic AOD:
Global annual mean: 0.047
(~ 30%, large regional variability)

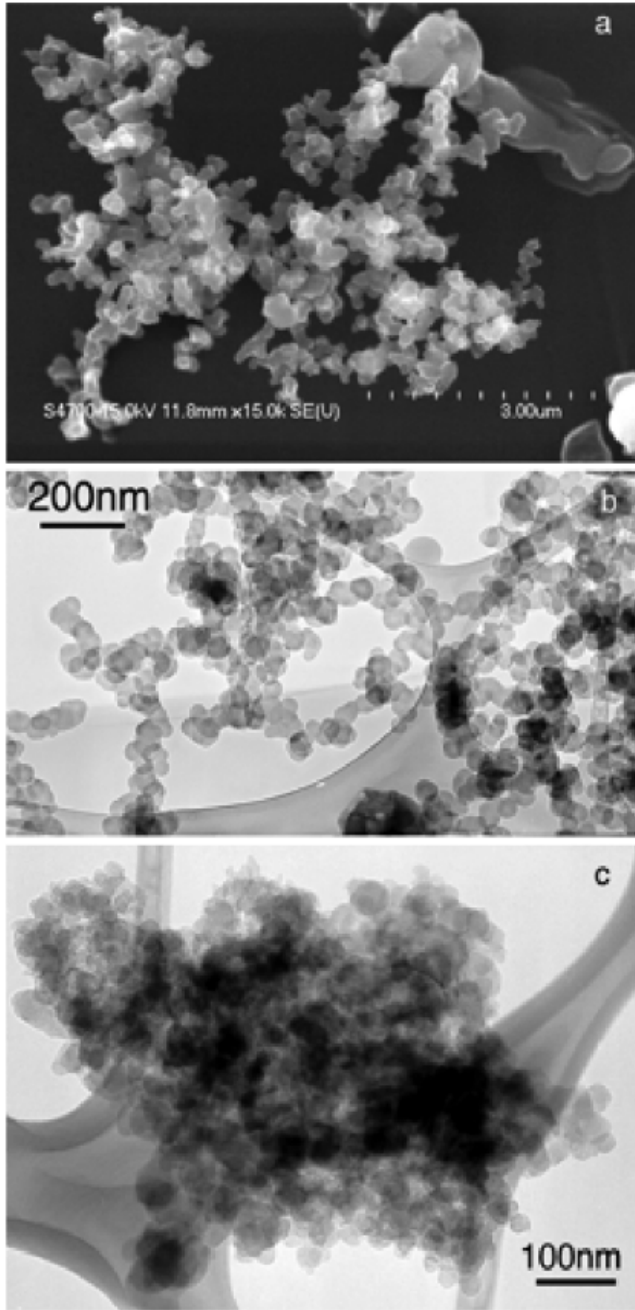
3.2 Anthropogenic aerosols: Kinne climatology



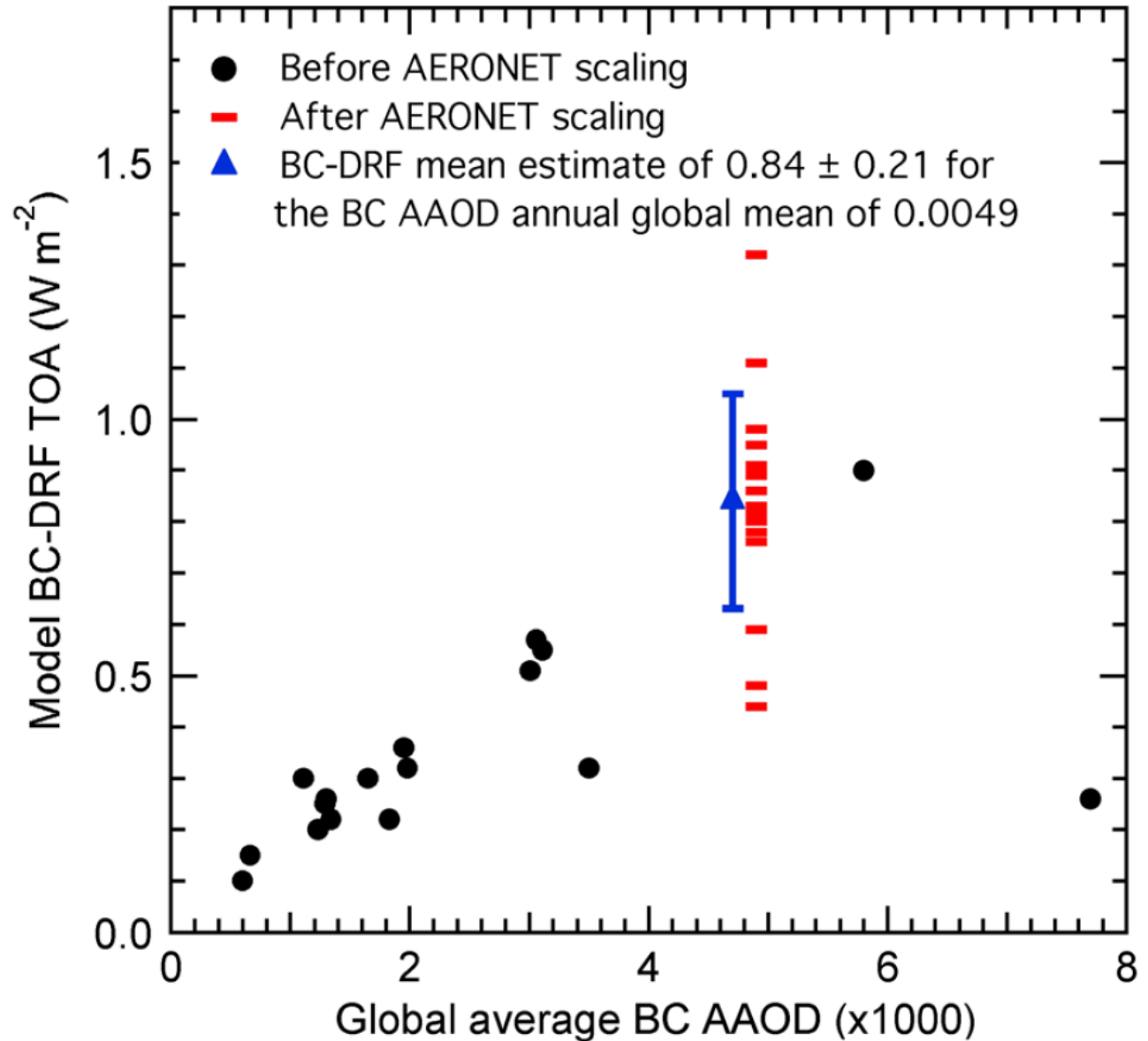
3.2 Anthropogenic aerosols: Kinne climatology



3.3 Anthropogenic direct effect: Soot



AeroCom model results for BC direct forcing in the industrial era



3.4 Anthropogenic aerosol indirect radiative forcing

Forcing Radiation

Cloud
particles

Aerosol
perturbation

$$\Delta F_{\text{aie}} = \frac{dR}{d \ln N_c} \cdot \frac{d \ln N_c}{d \ln \alpha} \cdot \Delta \ln \alpha_{\text{ant}}$$

3.4 Anthropogenic aerosol indirect radiative forcing

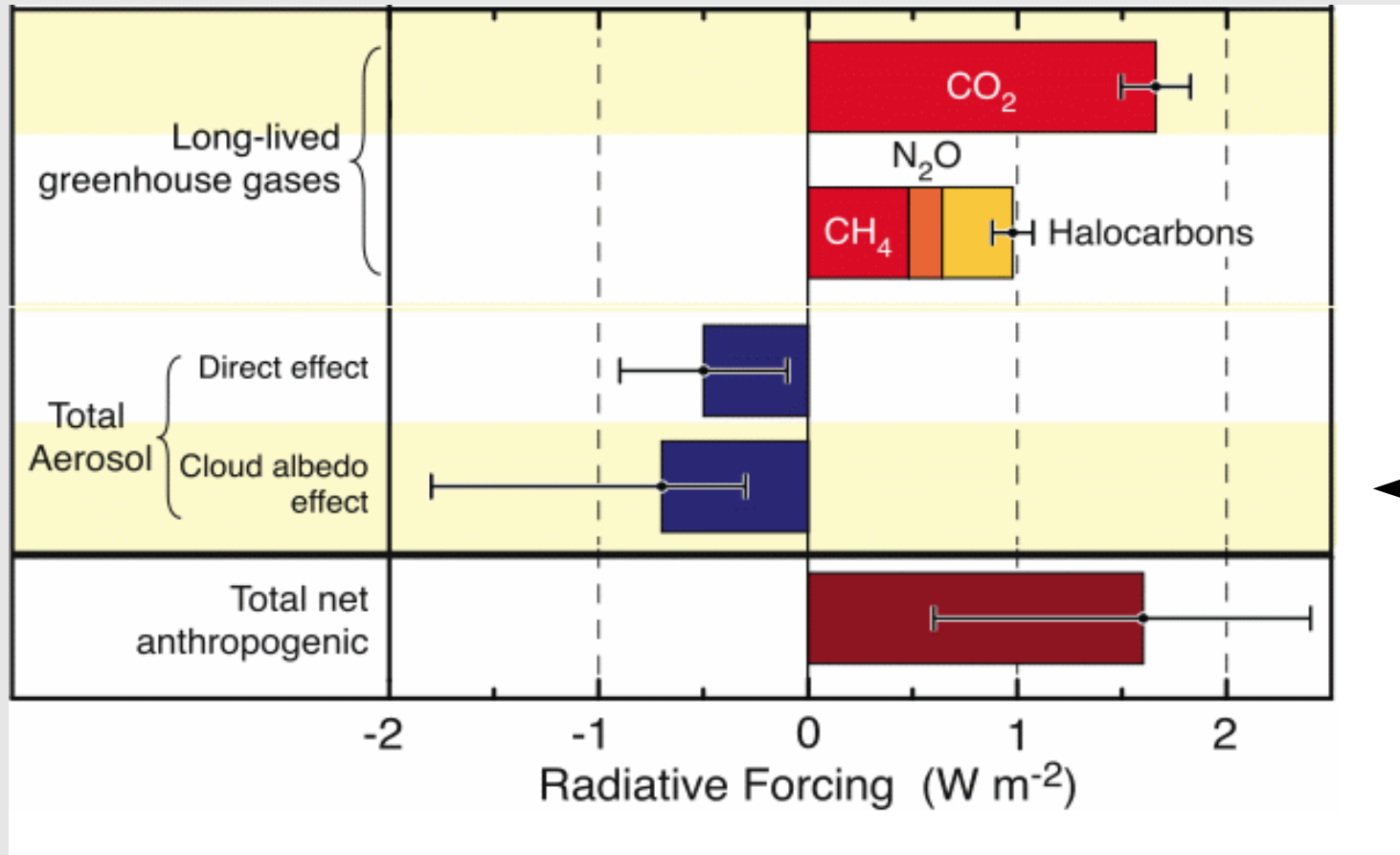
Forcing Radiation Cloud particles Aerosol perturbation

$$\Delta F_{\text{aie}} = \frac{dR}{d \ln N_c} \cdot \frac{d \ln N_c}{d \ln \alpha} \cdot \Delta \ln \alpha_{\text{ant}}$$

$$= \underbrace{\frac{\partial R}{\partial \ln N_c}}_{\text{First indirect effect}} + \underbrace{\frac{\partial R}{\partial f} \frac{\partial f}{\partial \ln N_c} + \frac{\partial R}{\partial L} \frac{\partial L}{\partial \ln N_c} + \frac{\partial R}{\partial T_{\text{top}}} \frac{\partial T_{\text{top}}}{\partial \ln N_c}}_{\text{Second indirect effect}}$$

Cloud fraction f , Water path L ,
Top temperature T_{top}

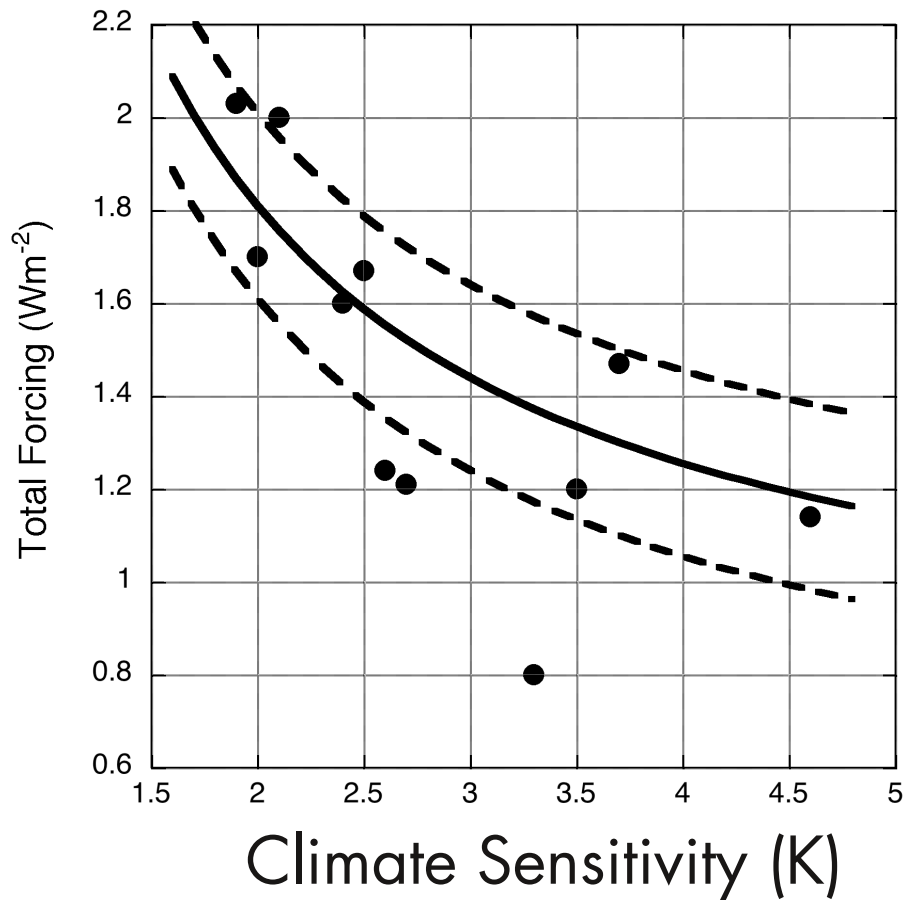
3.4 Anthropogenic aerosol forcing: Quantitative estimates



4. Importance for climate

4.1 Link to climate sensitivity

Spread in global warming due to CO₂ doubling

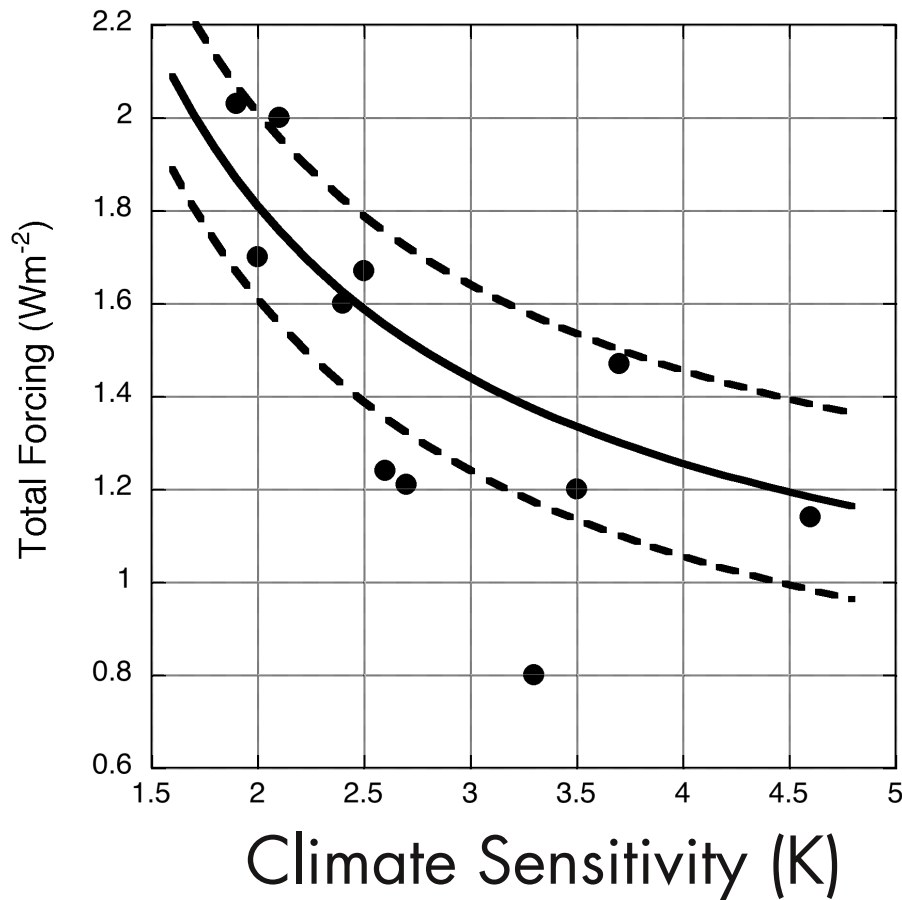


In CMIP3 climate models:

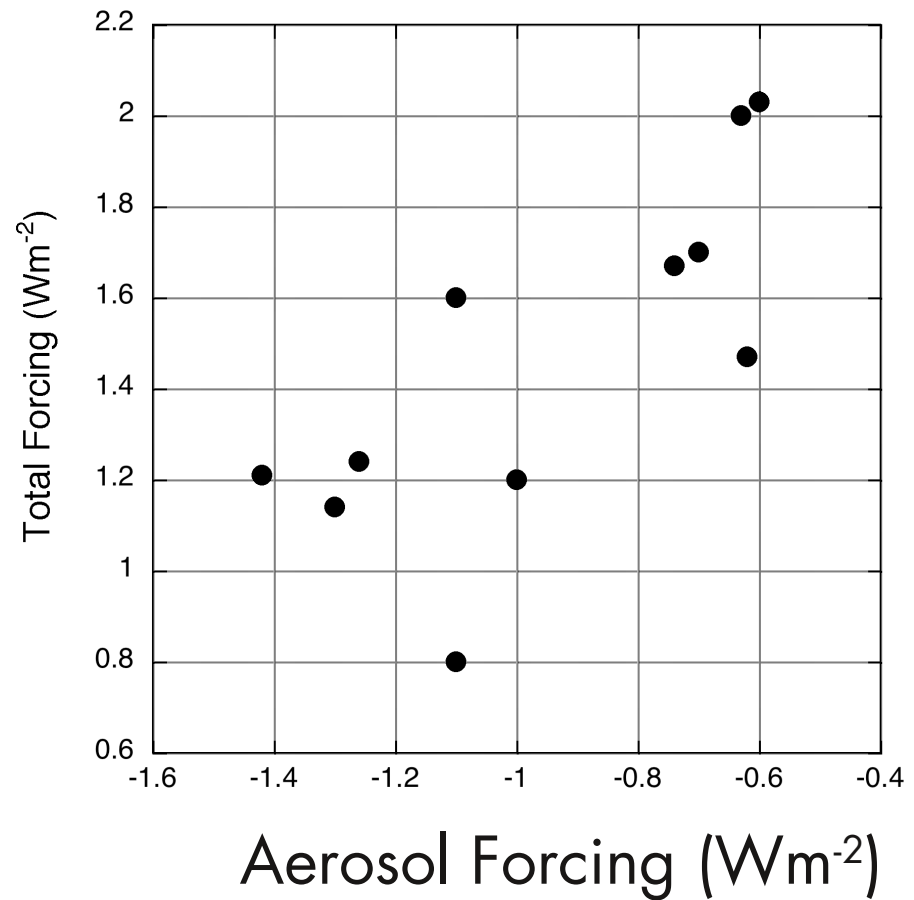
Climate sensitivity uncertainty

4.1 Link to climate sensitivity

Spread in global warming due to CO₂ doubling



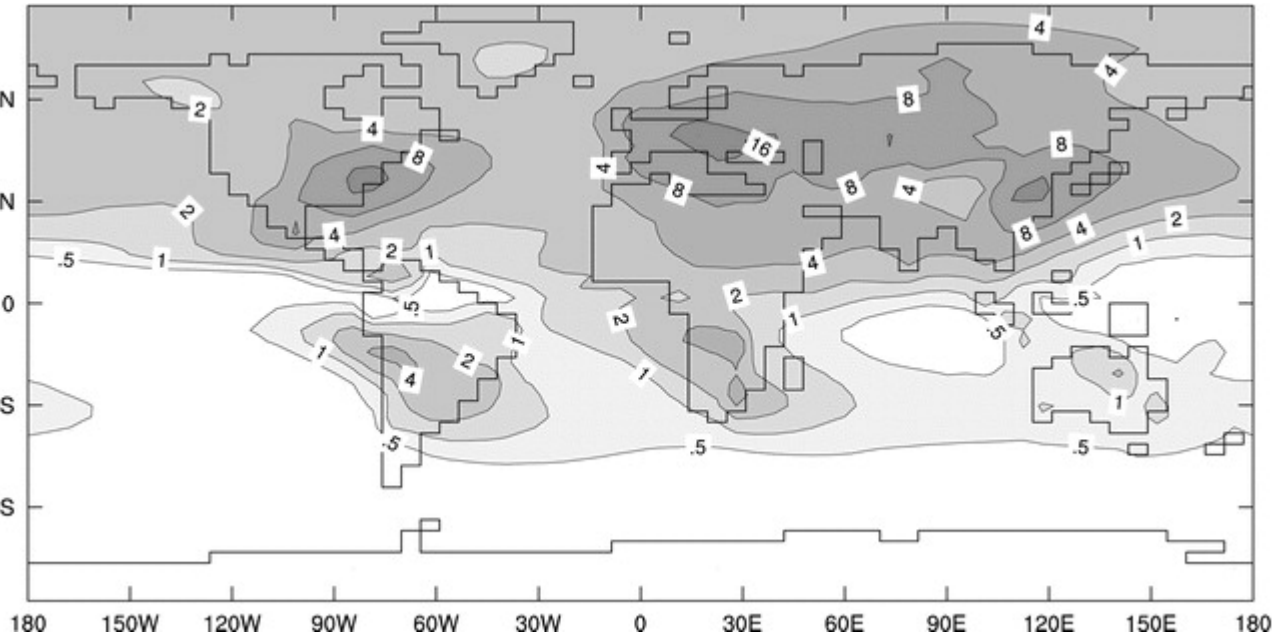
Spread in aerosol forcing



In CMIP3 climate models:

Climate sensitivity uncertainty ↔ Aerosol forcing uncertainty

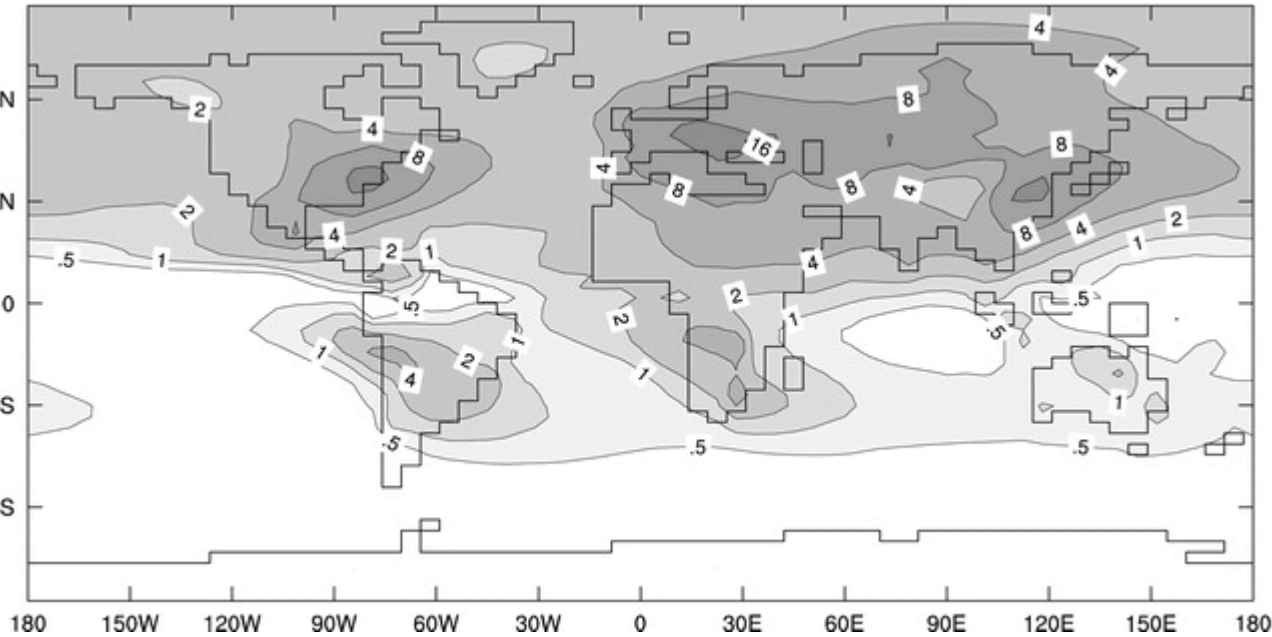
4.2 Effects on dynamics: Sahel drying



Simulated change in sulfate aerosol burden [mg m^{-2}], annual mean peak at 16 mg m^{-2}

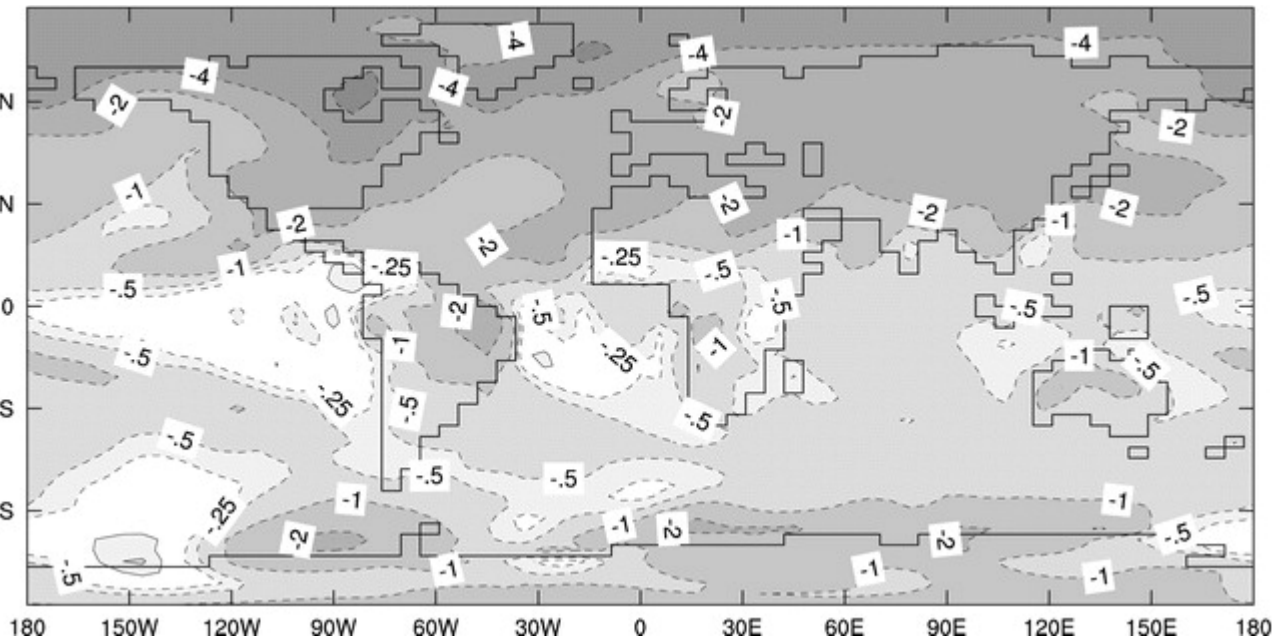
Strong forcing: -1.8 W m^{-2} in global mean

4.2 Effects on dynamics: Sahel drying



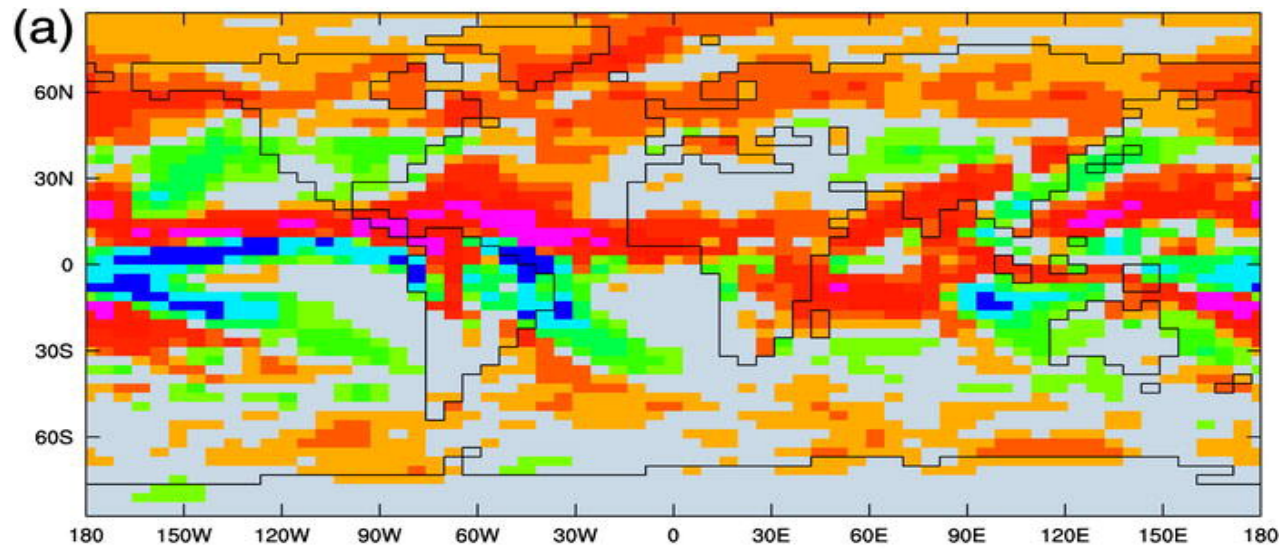
Simulated change in sulfate aerosol burden [mg m⁻²], annual mean peak at 16 mg m⁻²

Strong forcing: -1.8 W m⁻² in global mean



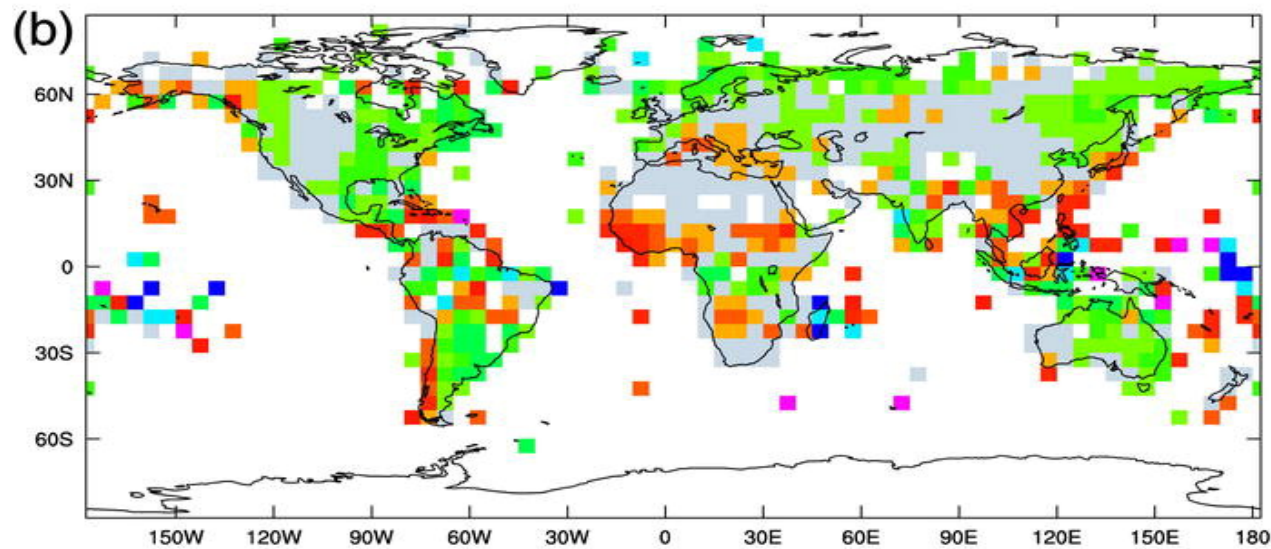
Simulated corresponding surface temperature change [K]
Peak at -8 K, -2 K widespread over Northern hemisphere mid- to high latitudes

4.2 Effects on dynamics: Sahel drying



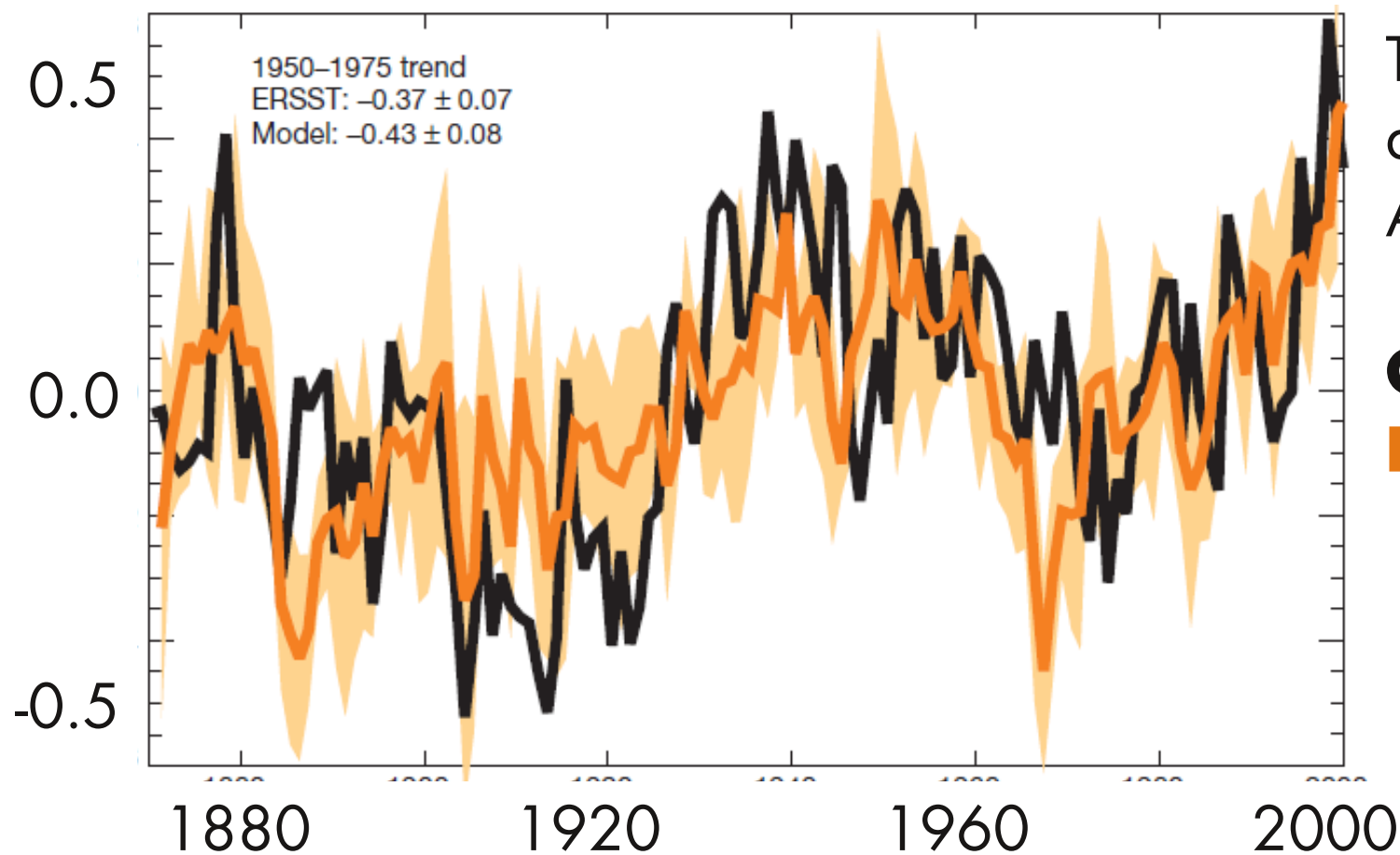
Trends in annual-mean precipitation

Top: Simulated
Bottom: Observed



-2 -1 -0.5 -0.1 0.1 0.5 1 2 mm day⁻¹ century⁻¹

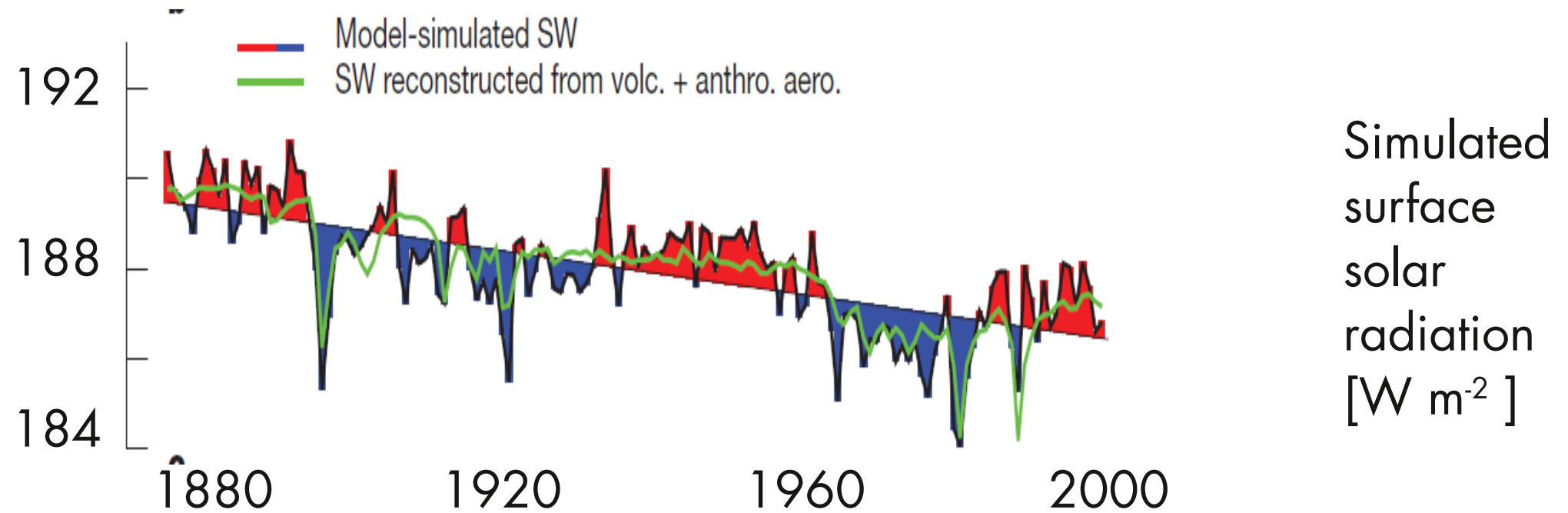
4.3 Effects on dynamics: North Atlantic Oscillation



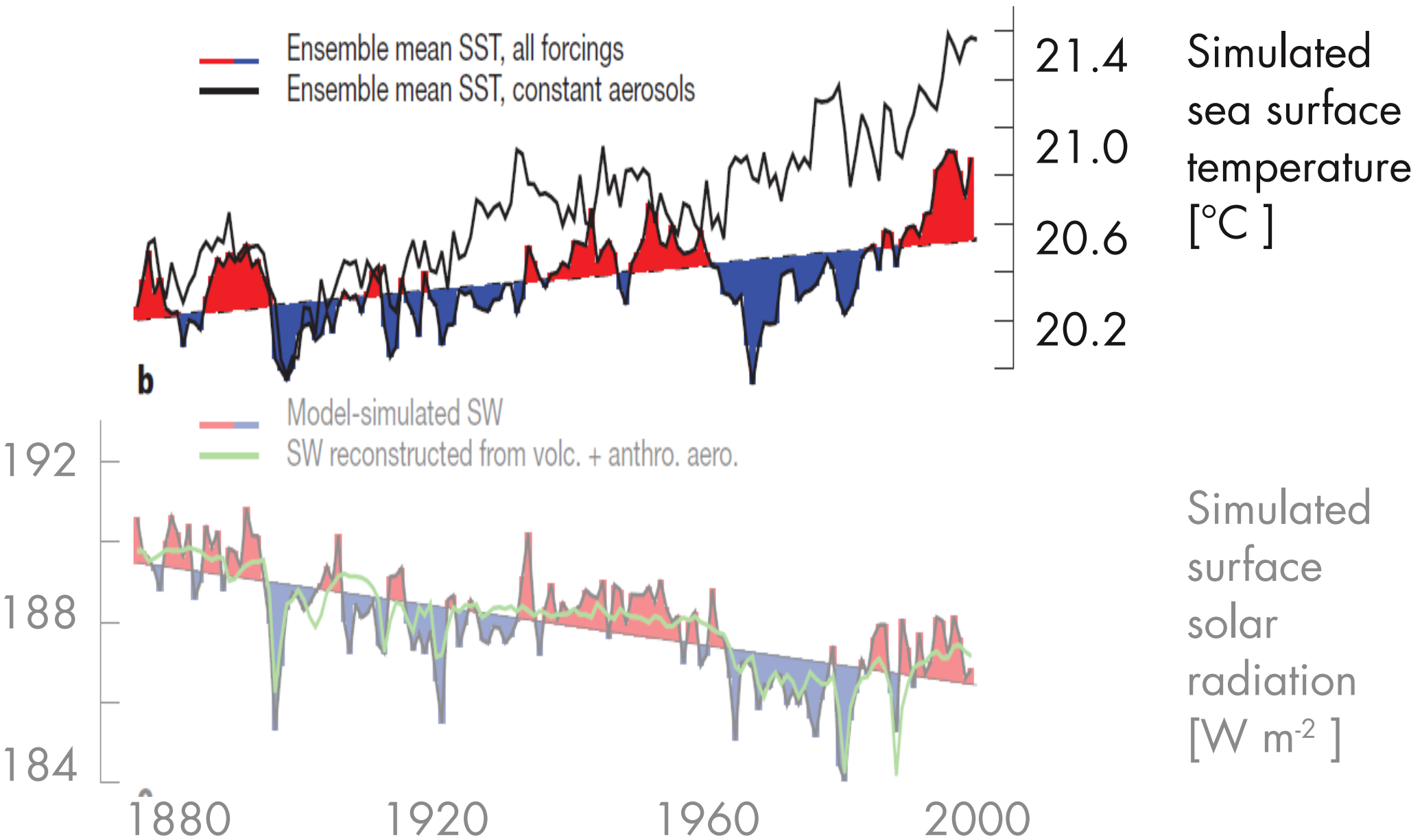
Temperature anomalies North Atlantic Ocean [K]

Observations
HadGEM2-ES

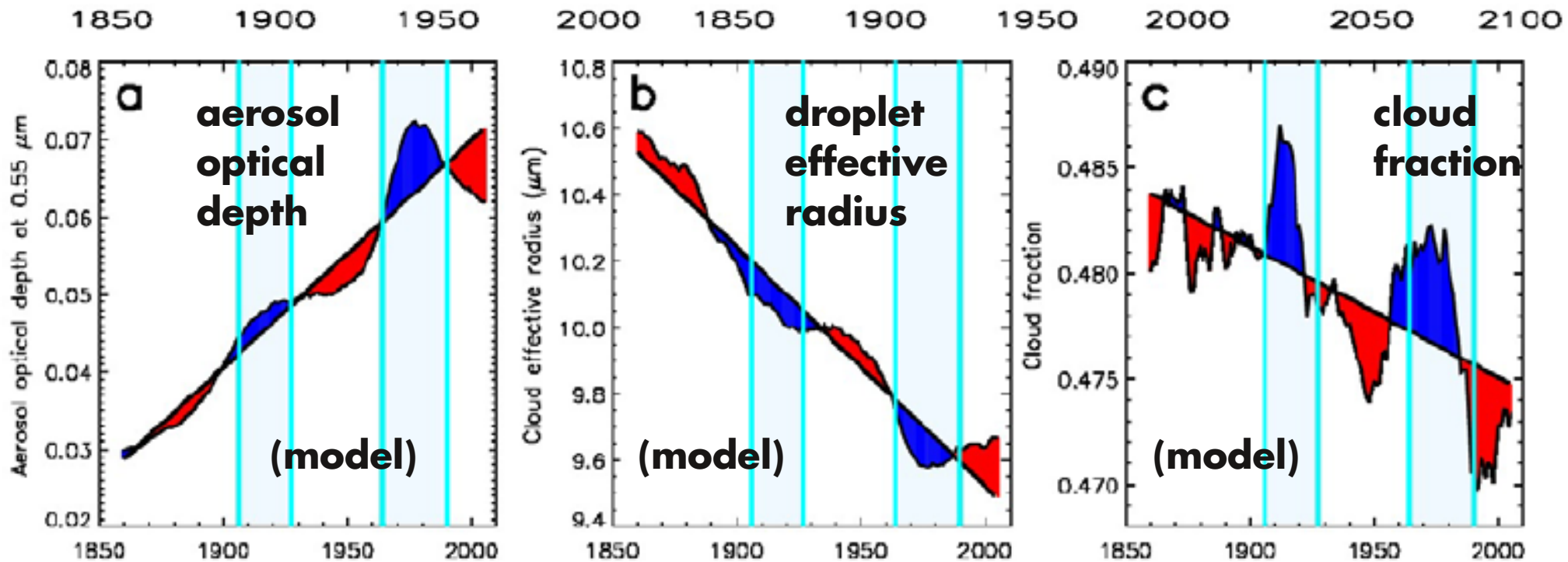
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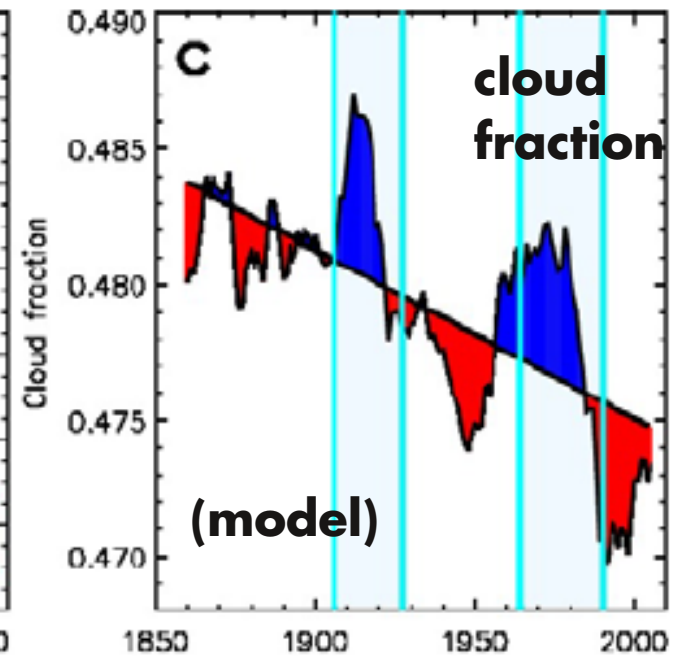
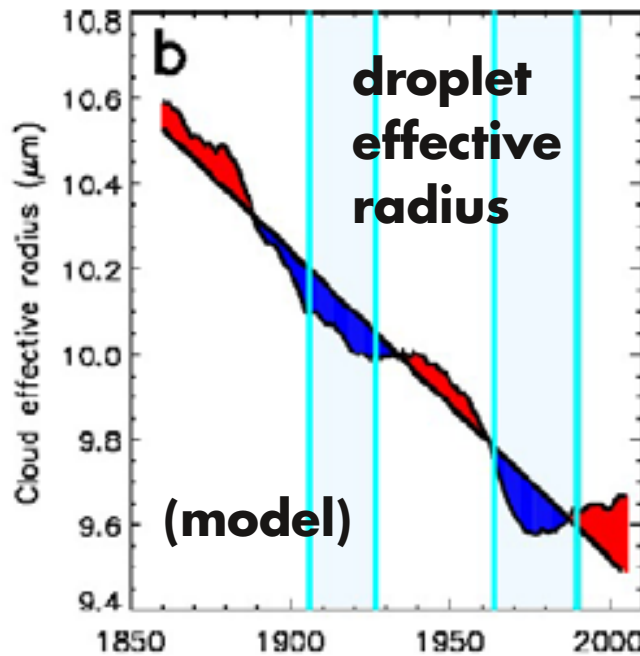
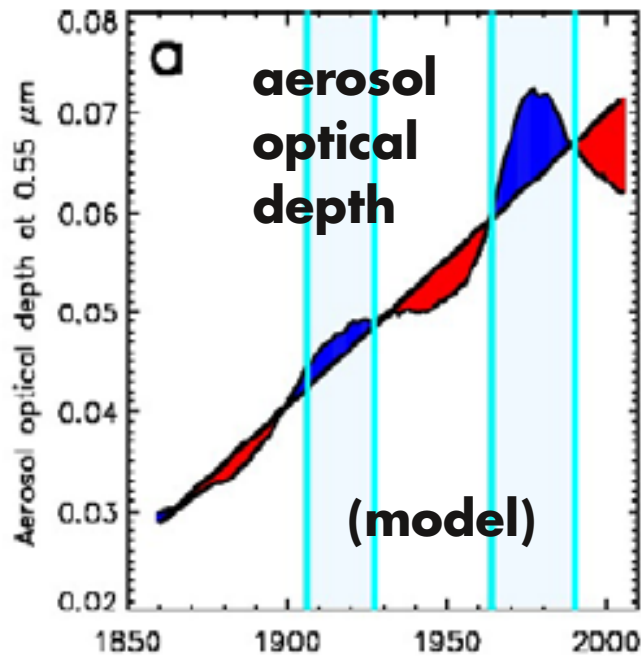
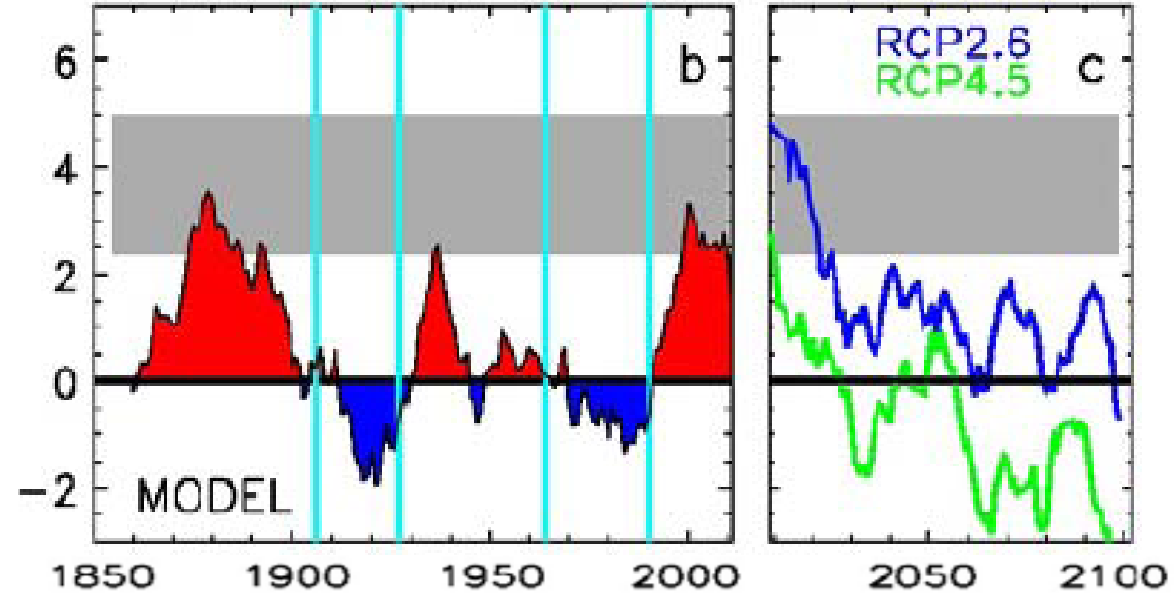


4.4 Effects on dynamics: Hurricanes

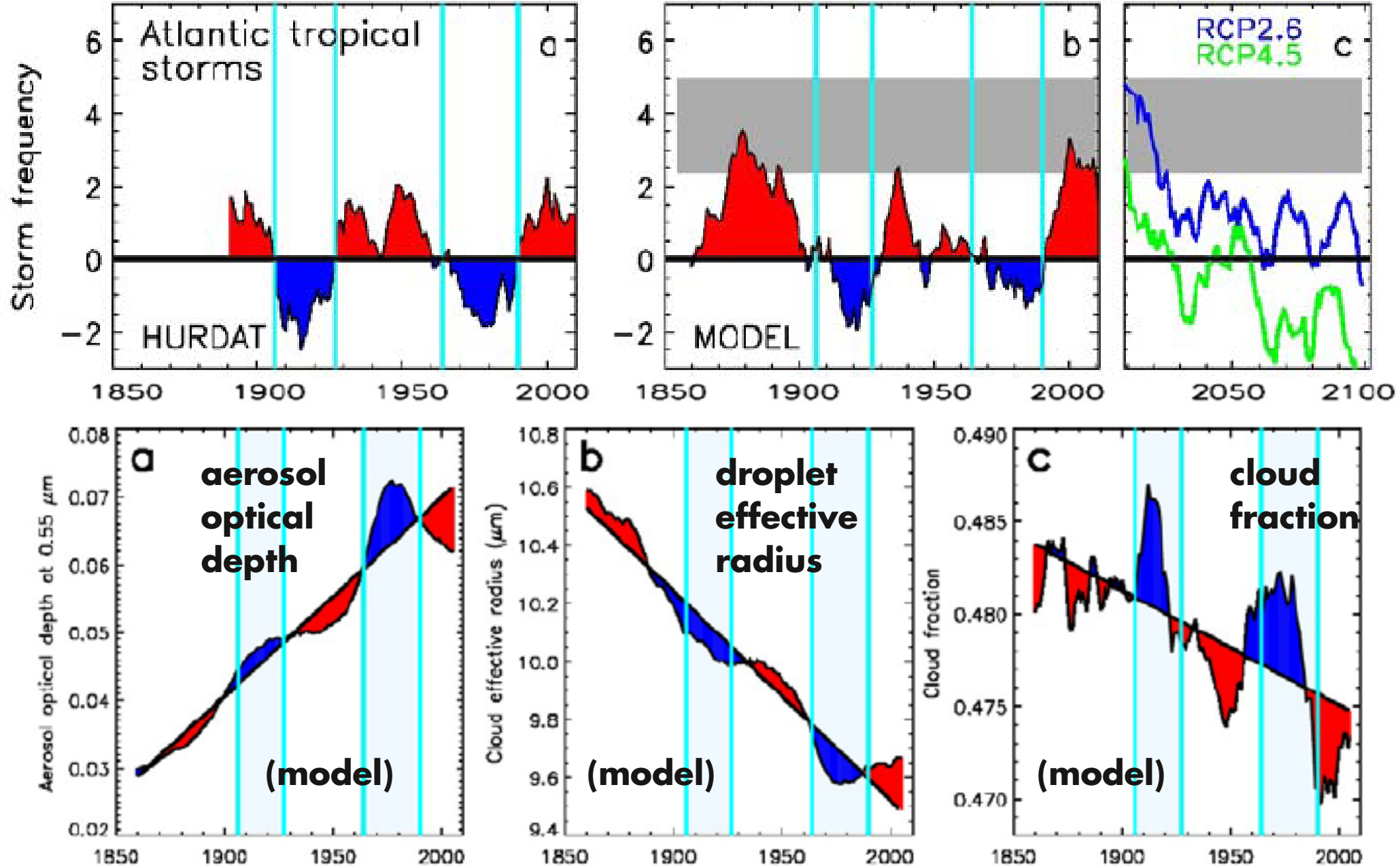


4.4 Effects on dynamics: Hurricanes

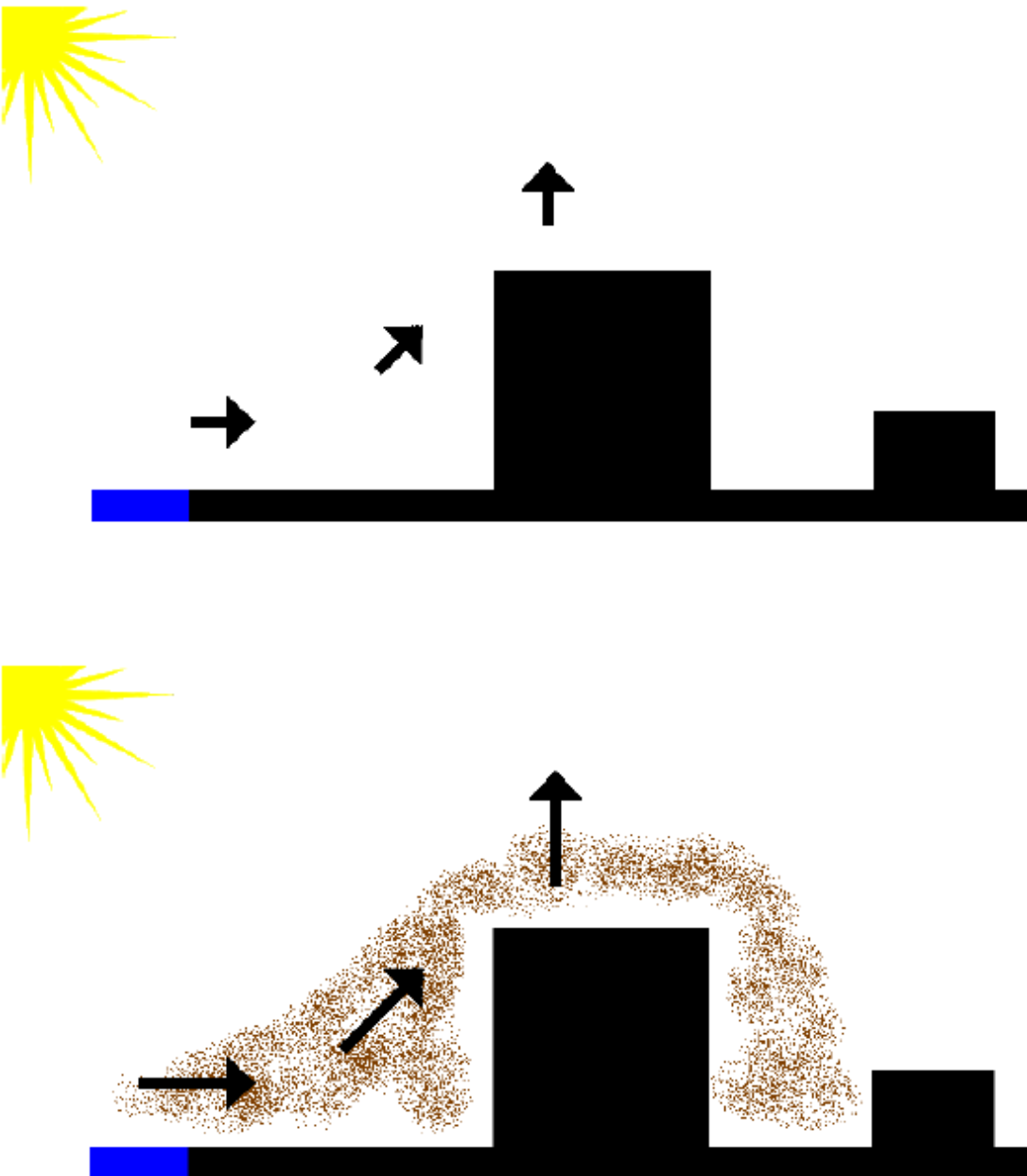
Hurricane index
frequency of occurrence



4.4 Effects on dynamics: Hurricanes

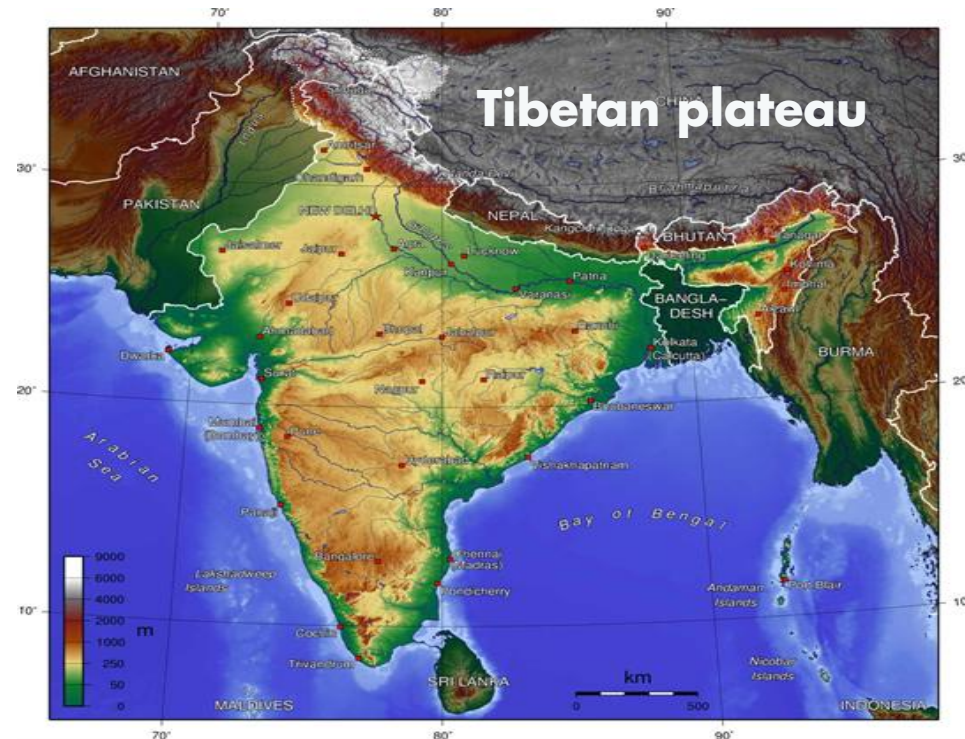


4.5 Effects on dynamics: Monsoon



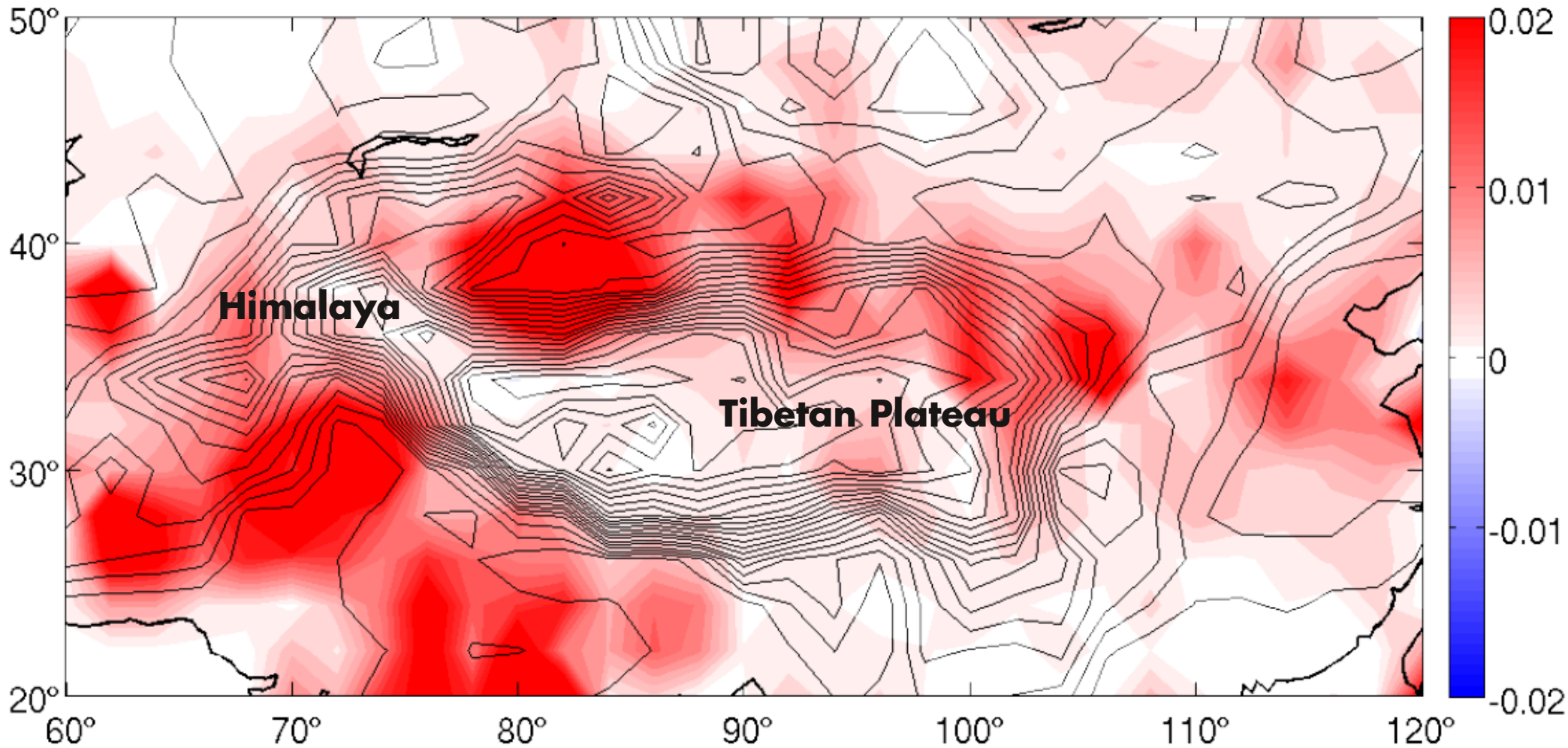
“Elevated heat pump”-
hypothesis

Absorption of sunlight over
Tibetan Plateau enforces
monsoon circulation?

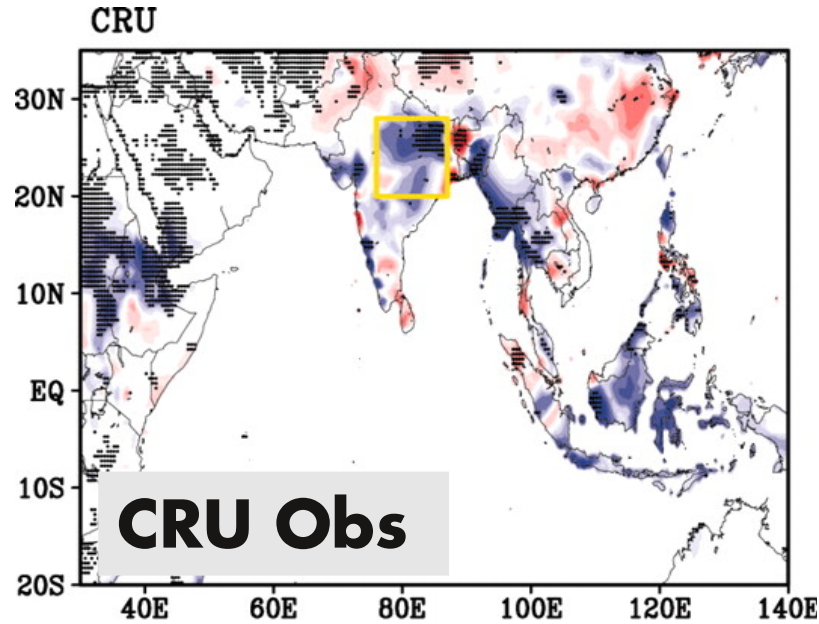


4.5 Effects on dynamics: Monsoon

Heating rates [K day⁻¹] by aerosol
(CALIPSO lidar satellite data and radiative transfer modelling)

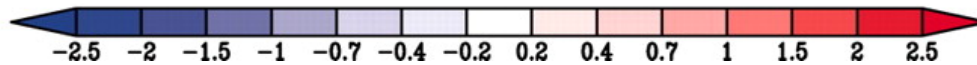


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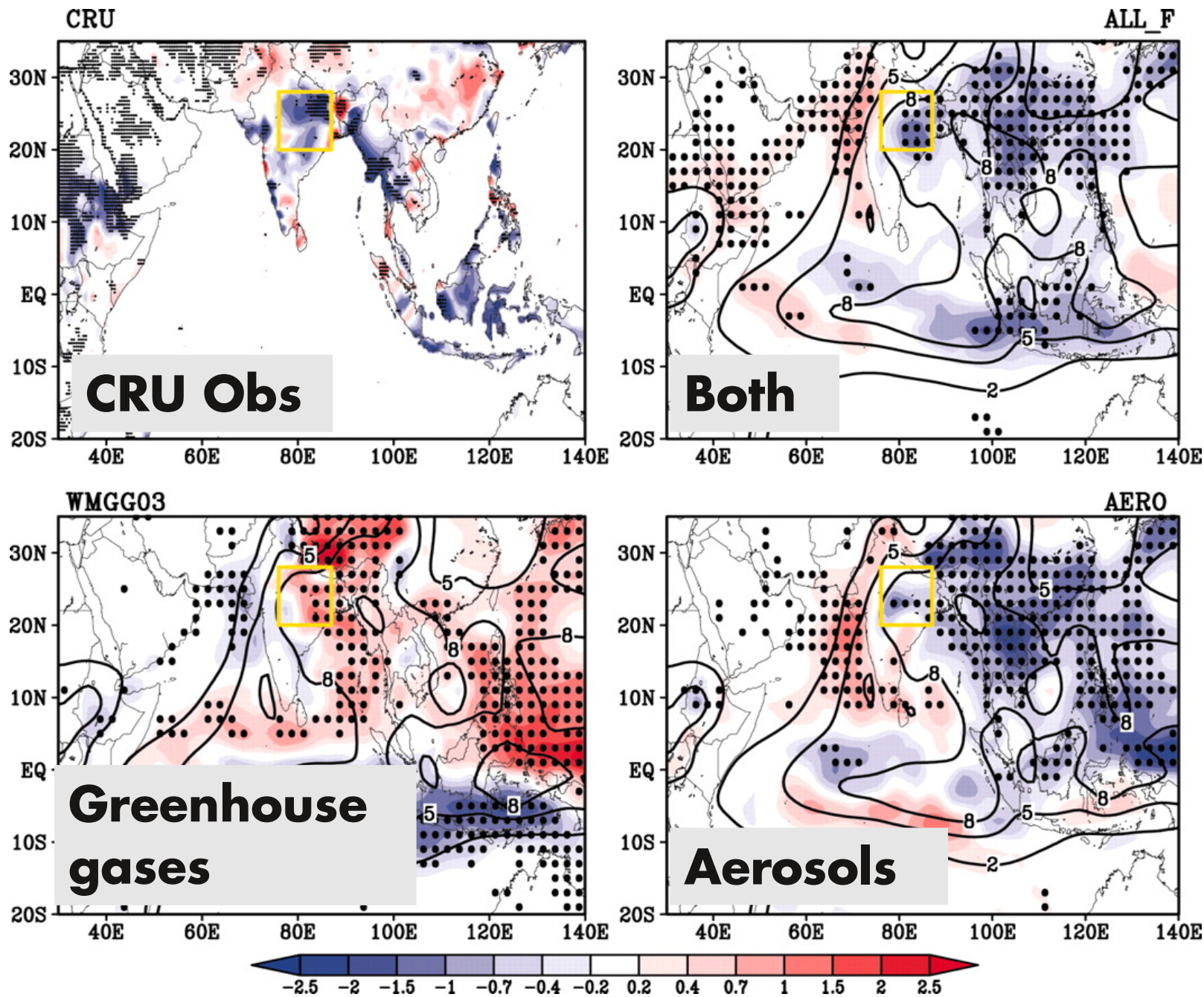


Linear trends
1950-1999 in
June-September
precipitation

[$\text{mm day}^{-1} (50 \text{ yr})^{-1}$]



4.5 Effects on dynamics: Monsoon



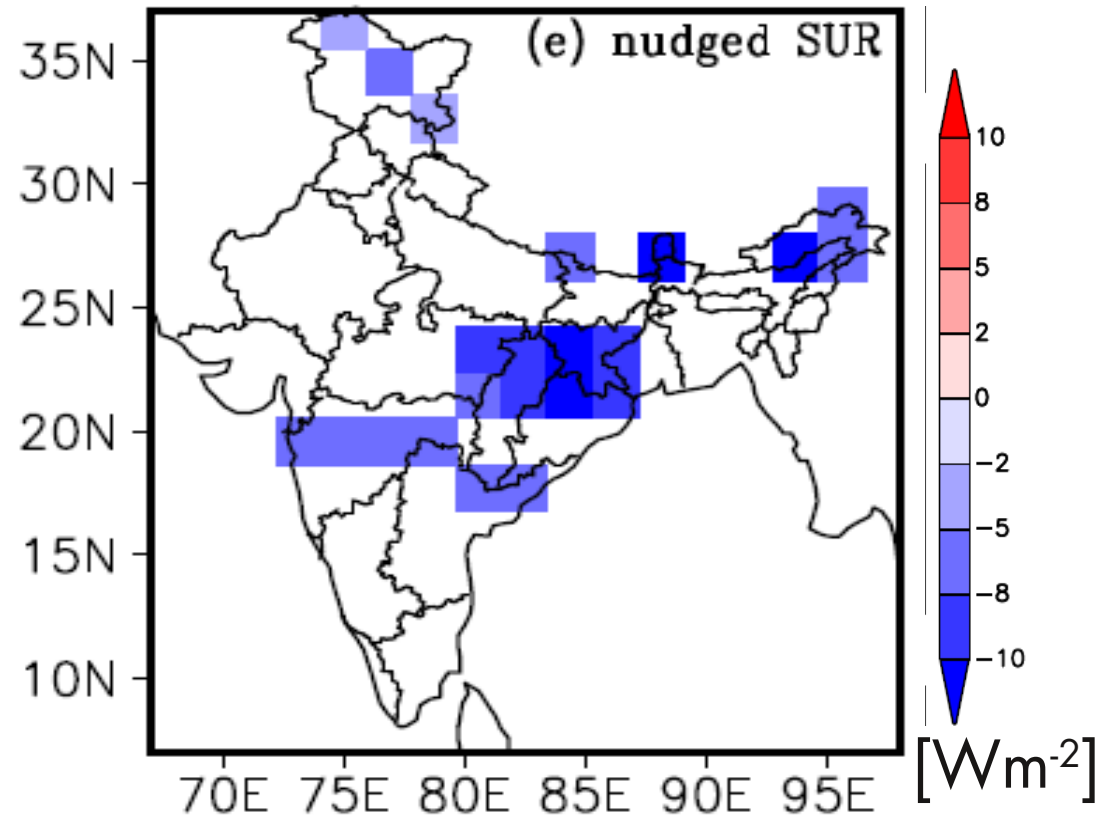
Linear trends
1950-1999 in
June-September
precipitation

$[\text{mm day}^{-1} (50 \text{ yr})^{-1}]$

Slowdown of
meridional
overturning
compensates
hemispherical
energy imbalance

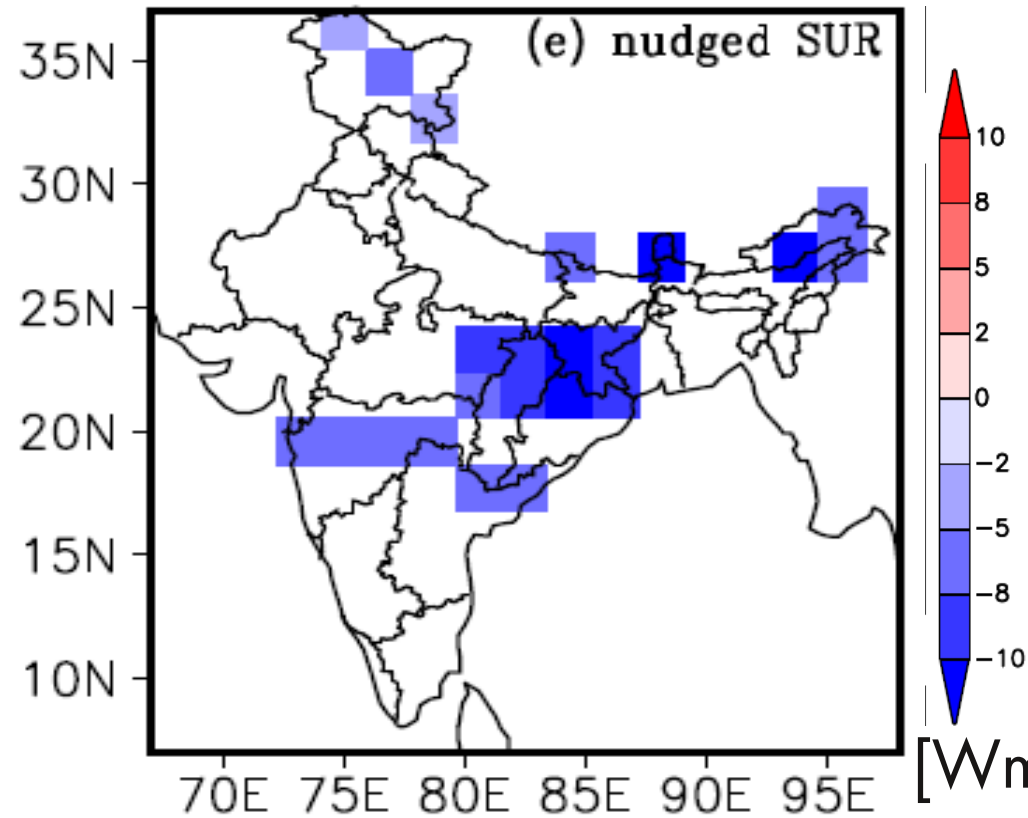
4.5 Effects on dynamics: Monsoon

Surface radiative forcing due to anthropogenic aerosol

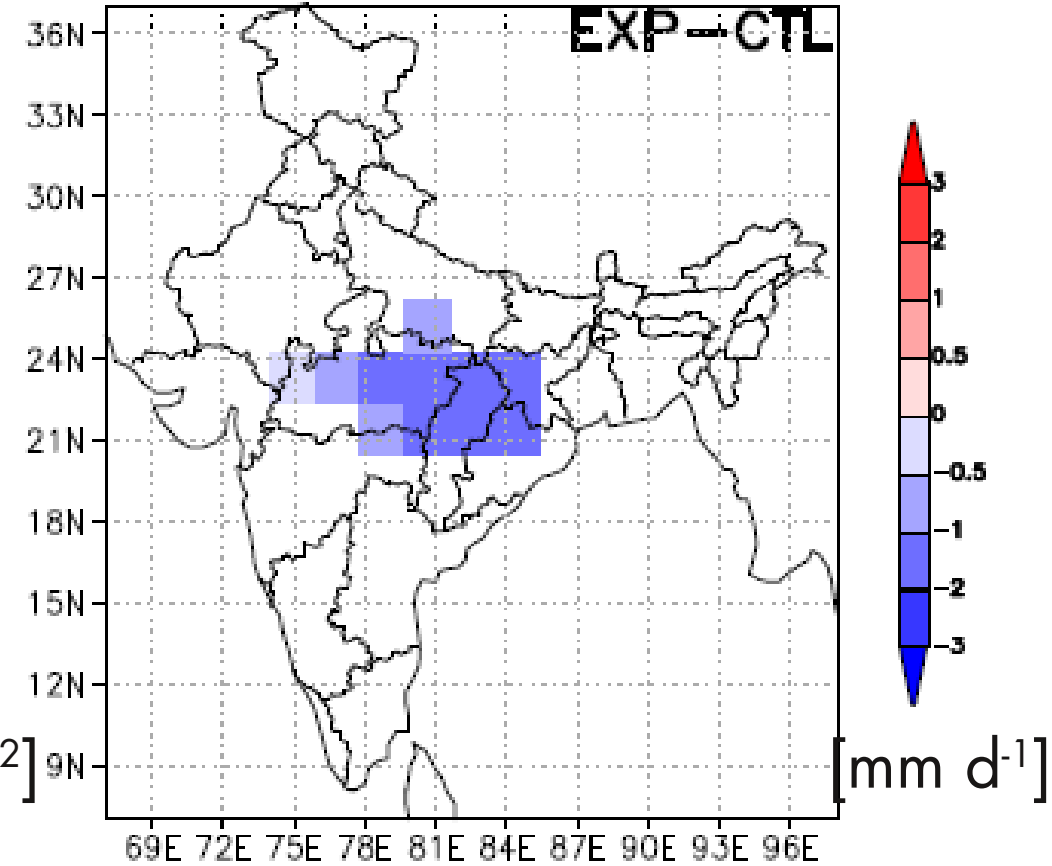


4.5 Effects on dynamics: Monsoon

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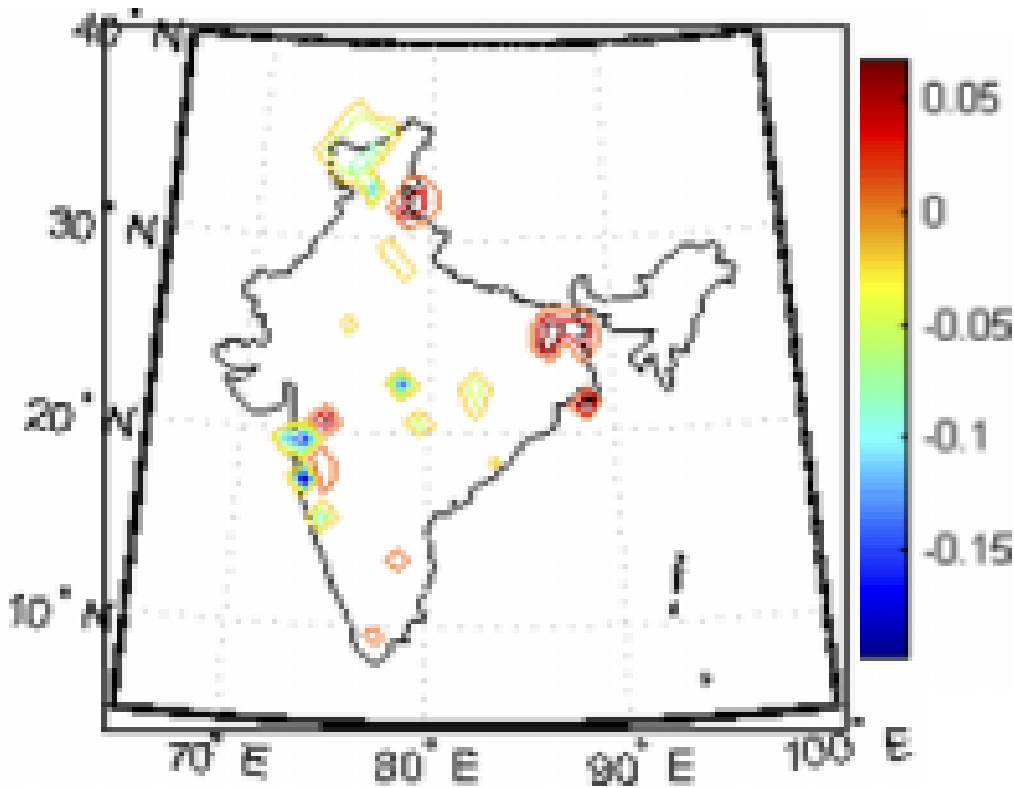


Monsoon precipitation change due to anthropogenic aerosol

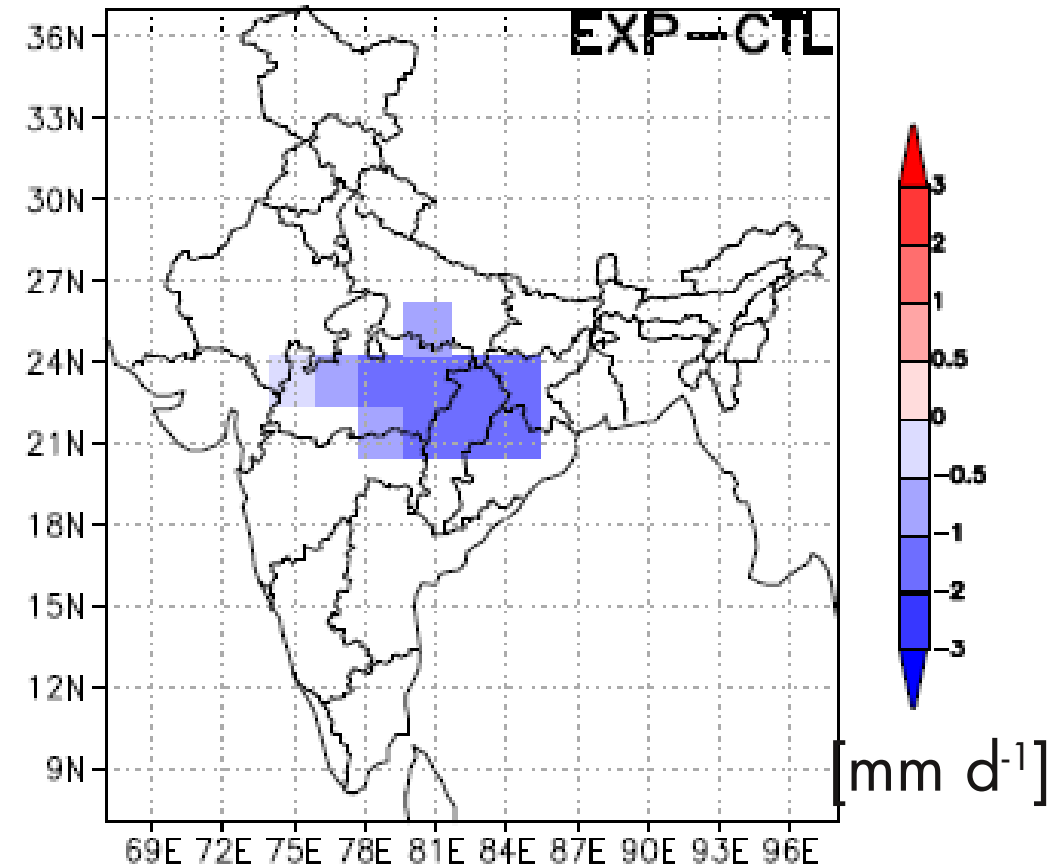


4.5 Effects on dynamics: Monsoon

Observed precipitation trend
since 1950 [mm yr^{-1}]



Monsoon precipitation change
due to anthropogenic aerosol

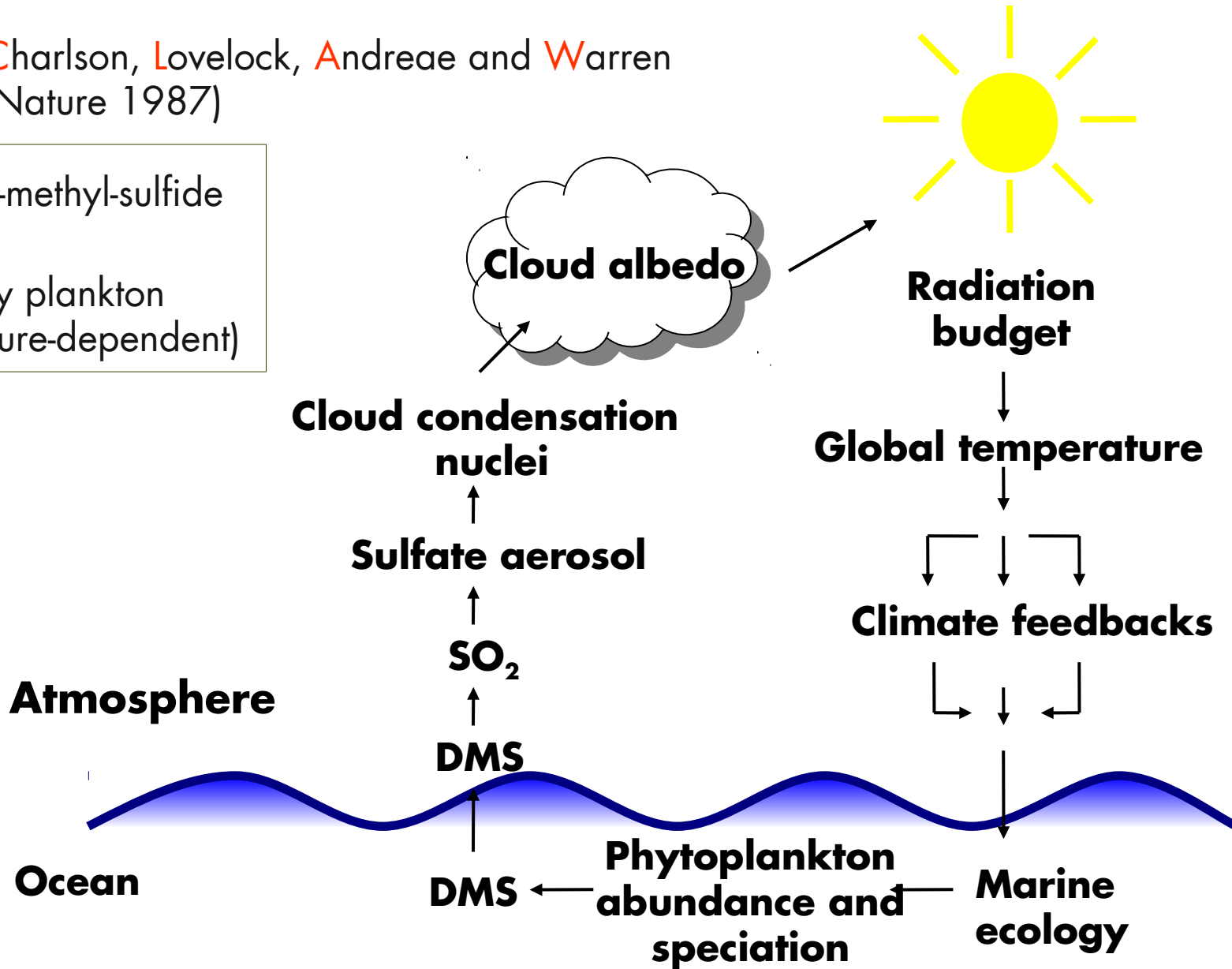


4.6 Stabilising climate feedbacks

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CLAW : Charlson, Lovelock, Andreae and Warren
(Nature 1987)

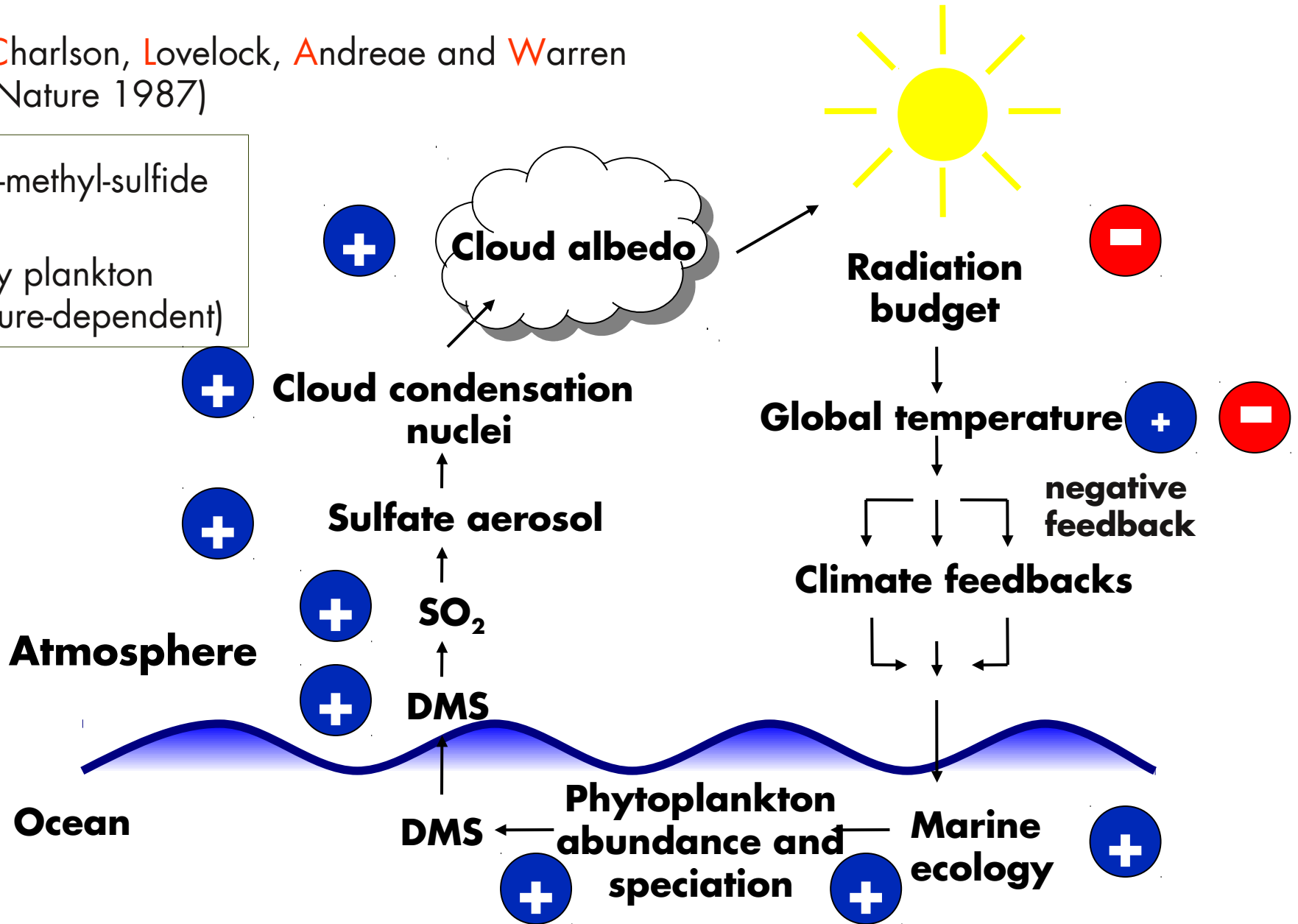
DMS – di-methyl-sulfide
 $S(CH_3)_2$
emitted by plankton
(temperature-dependent)



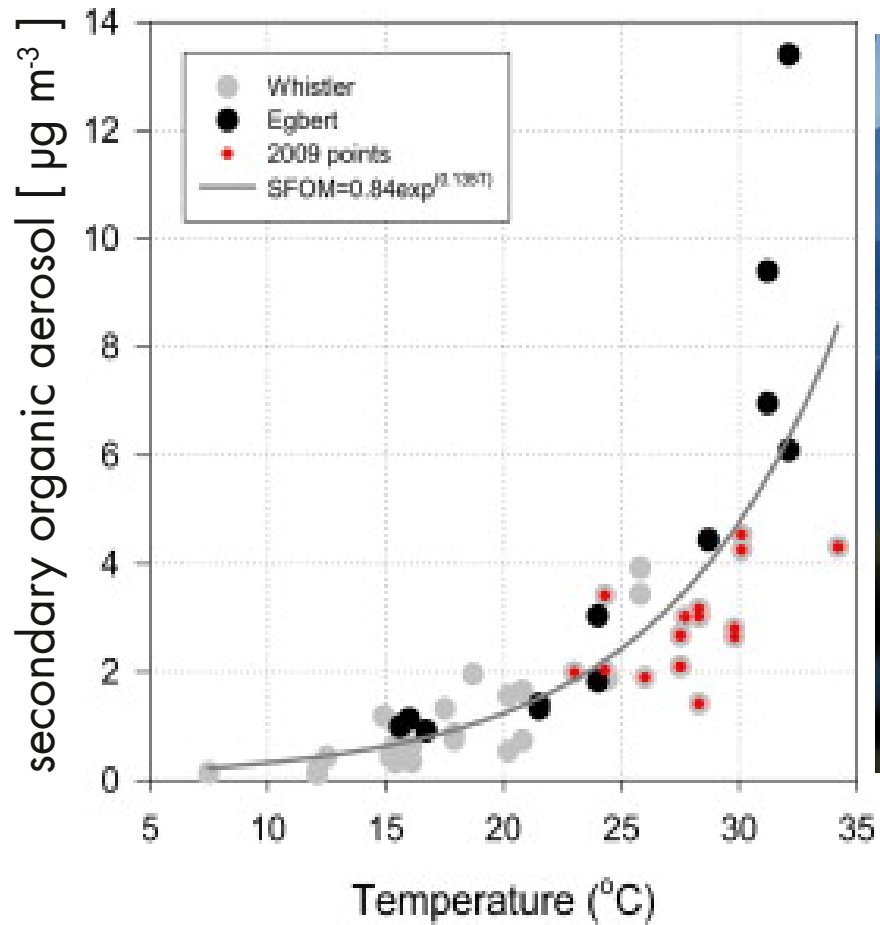
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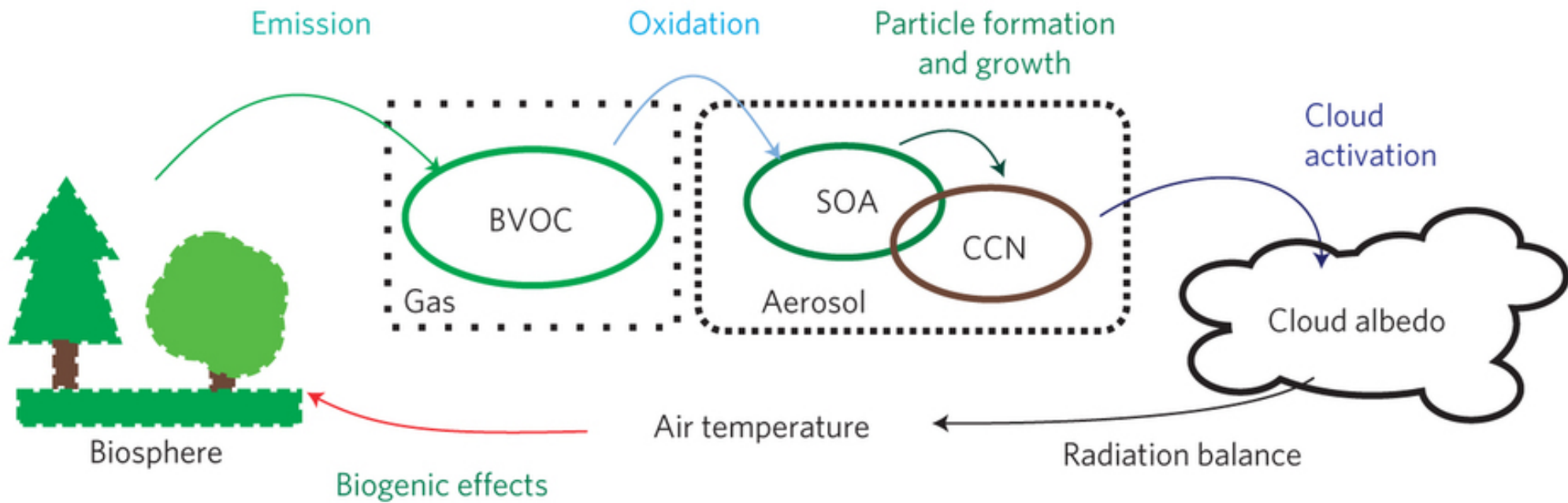


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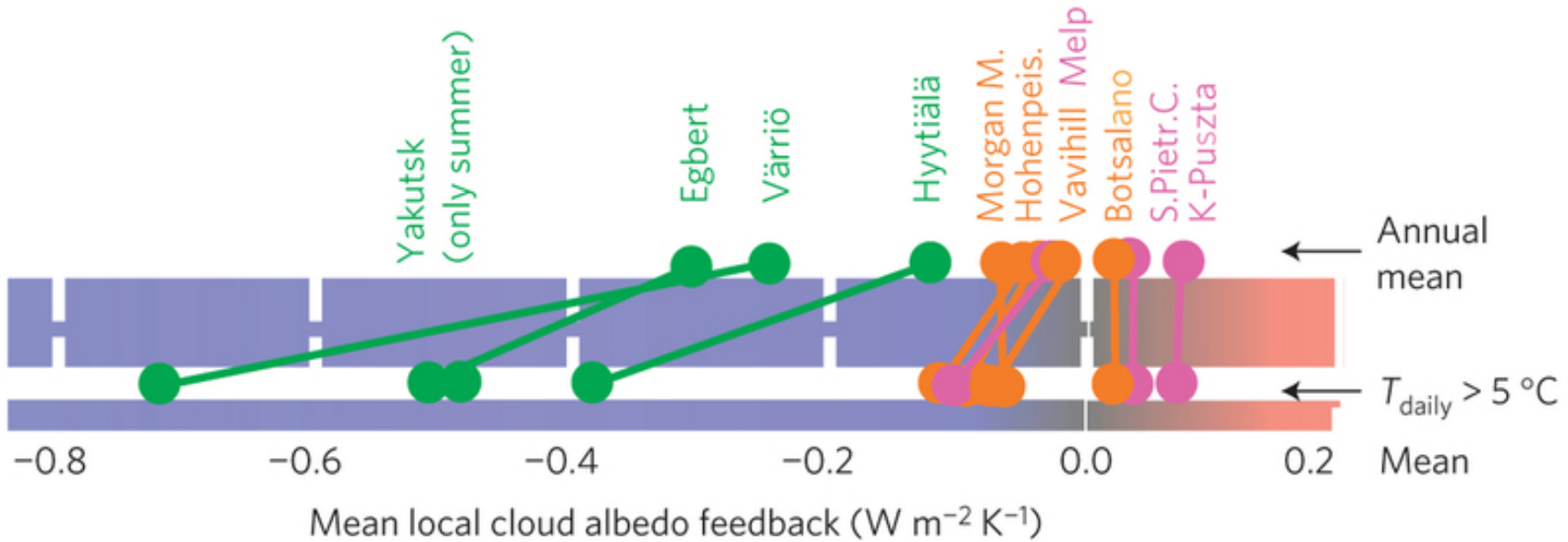
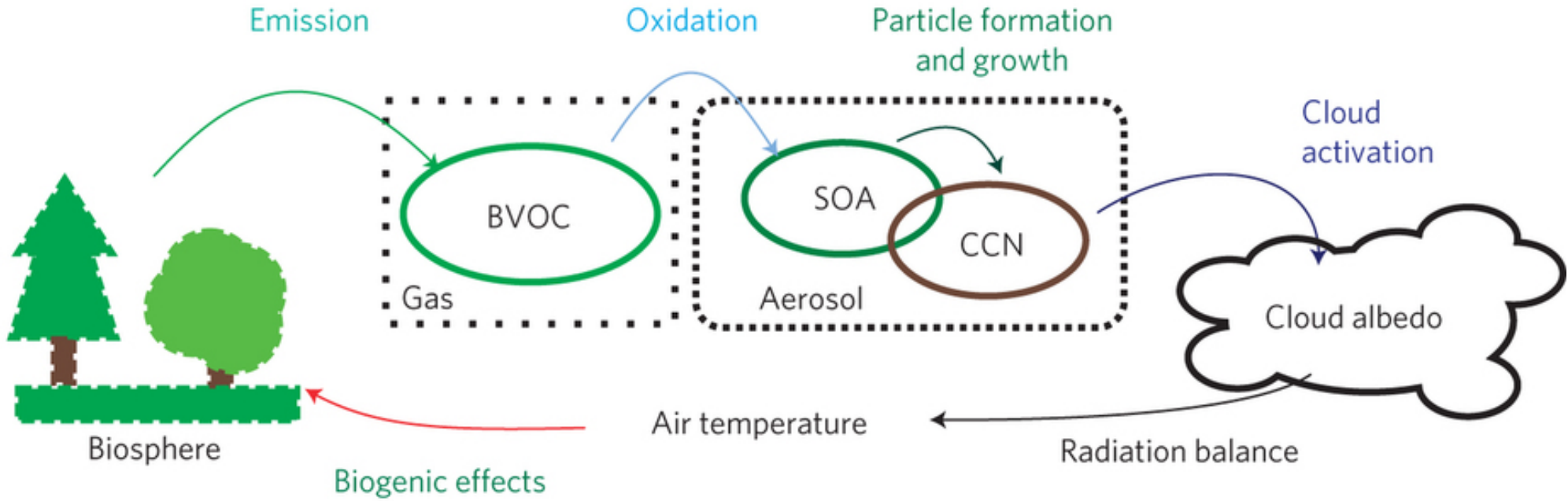
4.6 Stabilising climate feedbacks

a

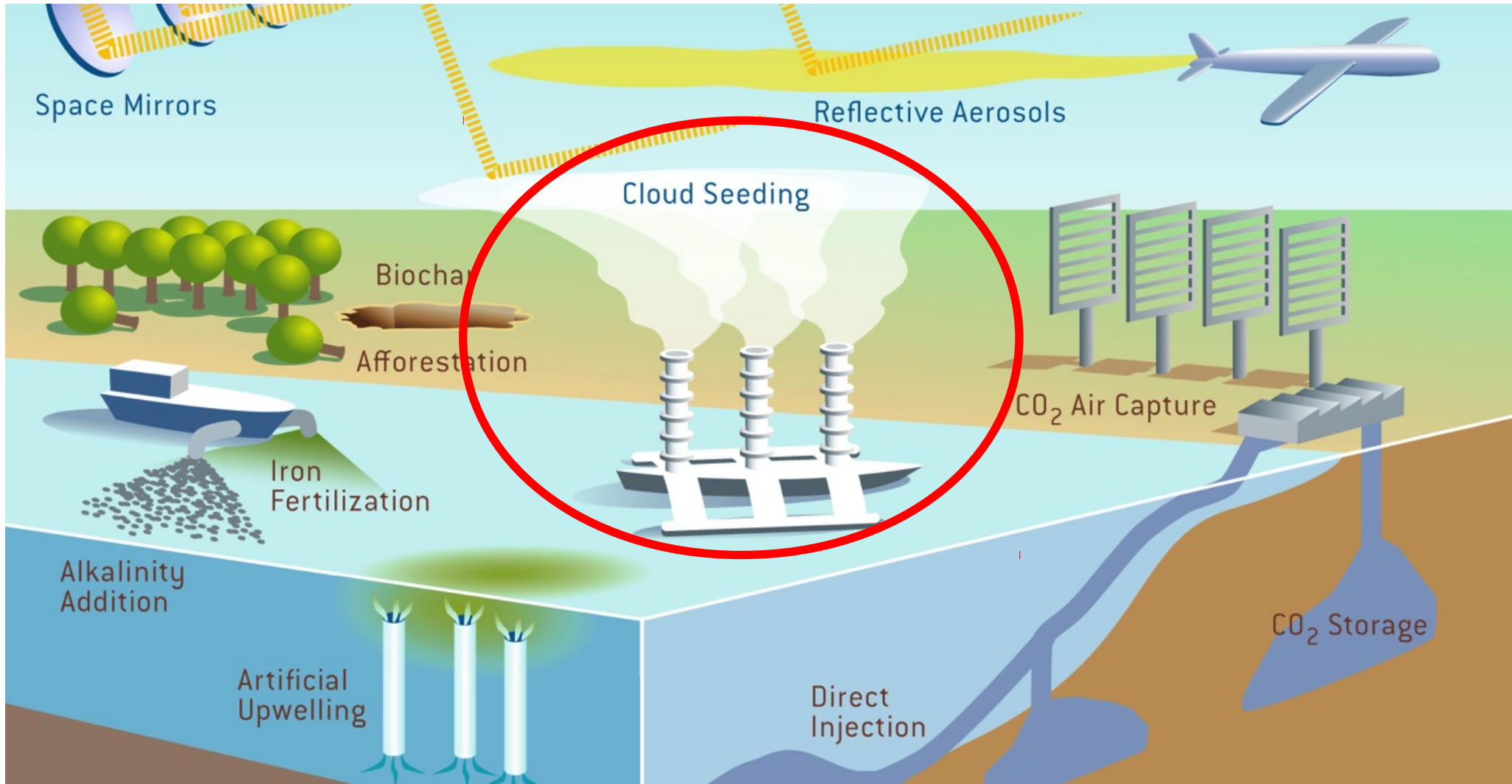


4.6 Stabilising climate feedbacks

a



4.7 Geoengineering



4.8 Co-benefits of improving air quality and mitigating climate change

- Soot bad for health
- BC warms climate by absorption of sunlight
- But: co-emitted SO₂ cools, BC might have (cooling) indirect effect on clouds

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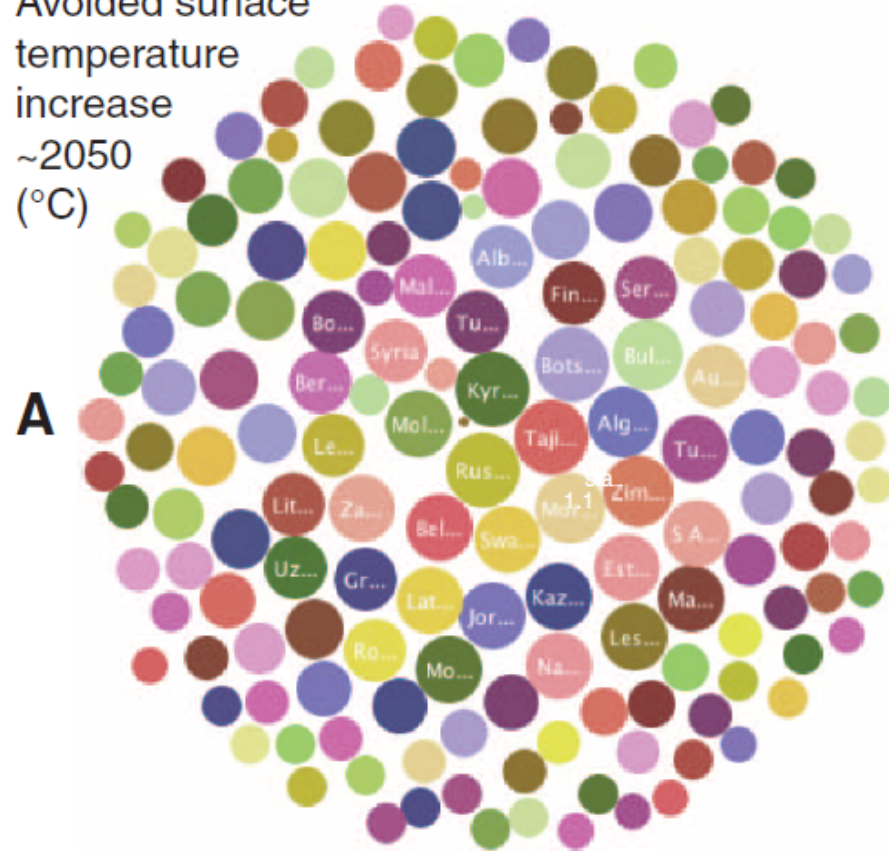
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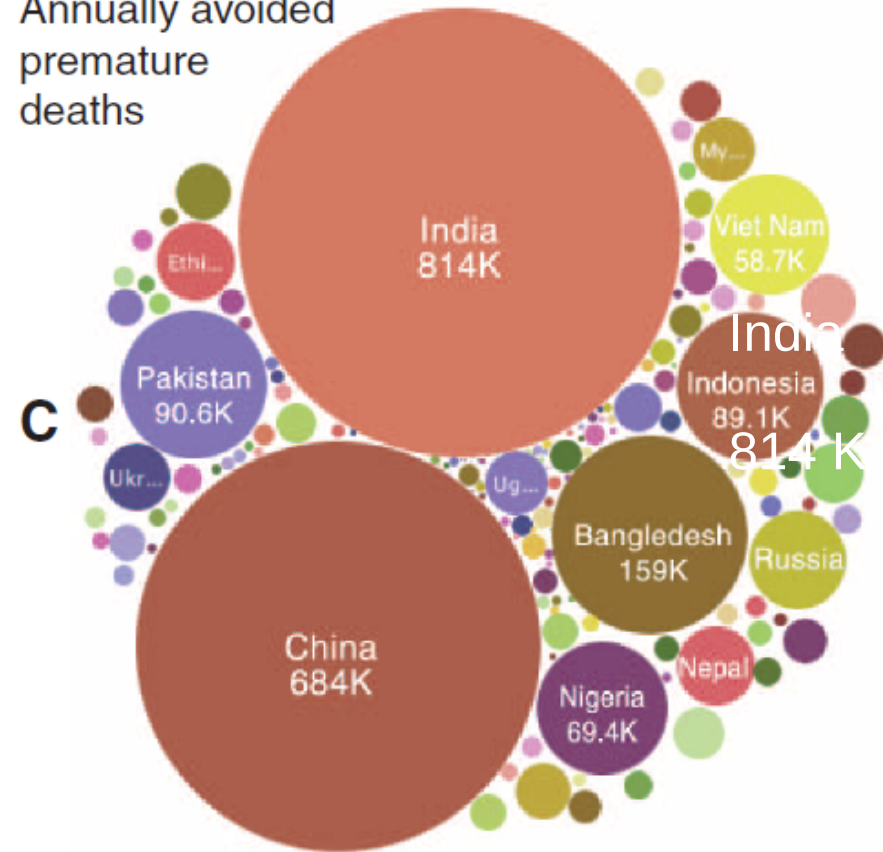
If maximum-feasible policies are implemented to cut methane and black carbon:

Avoided surface temperature increase ~2050 (°C)



0.4 K less warming by 2050

Annually avoided premature deaths



1.7M less premature deaths globally

Conclusions 1/2

■ **Effects of clouds and precipitation on aerosols**

- aqueous chemistry and cloud processing enhances aerosol
- in-cloud / below-cloud scavenging efficiently removes aerosol
- aerosol grows to haze in the vicinity of clouds

■ **Effects of aerosols on clouds and precipitation**

- additional aerosol enhances cloud particle concentrations enhancing cloud albedo (first indirect effect)
- alter cloud microphysical processes (precipitation formation, mixed-phase processes, latent heating; second indirect effect)
- subsequently change environment and dynamics (buffering?)
- absorption may alter cloud characteristics (semi-direct effect)

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Conclusions 2/2

■ Anthropogenic radiative forcings

- models show substantial direct and indirect effects
(-0.4 Wm^{-2} plus -0.9 Wm^{-2})

■ Quantitative understanding essential

- for climate sensitivity
- for weather in Africa, Europe, America, India
drought NAO Hurricanes Monsoon
- stabilising feedbacks
- geoengineering
- climate mitigation and air quality improvement policies

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■ Do you have an AA battery for me?