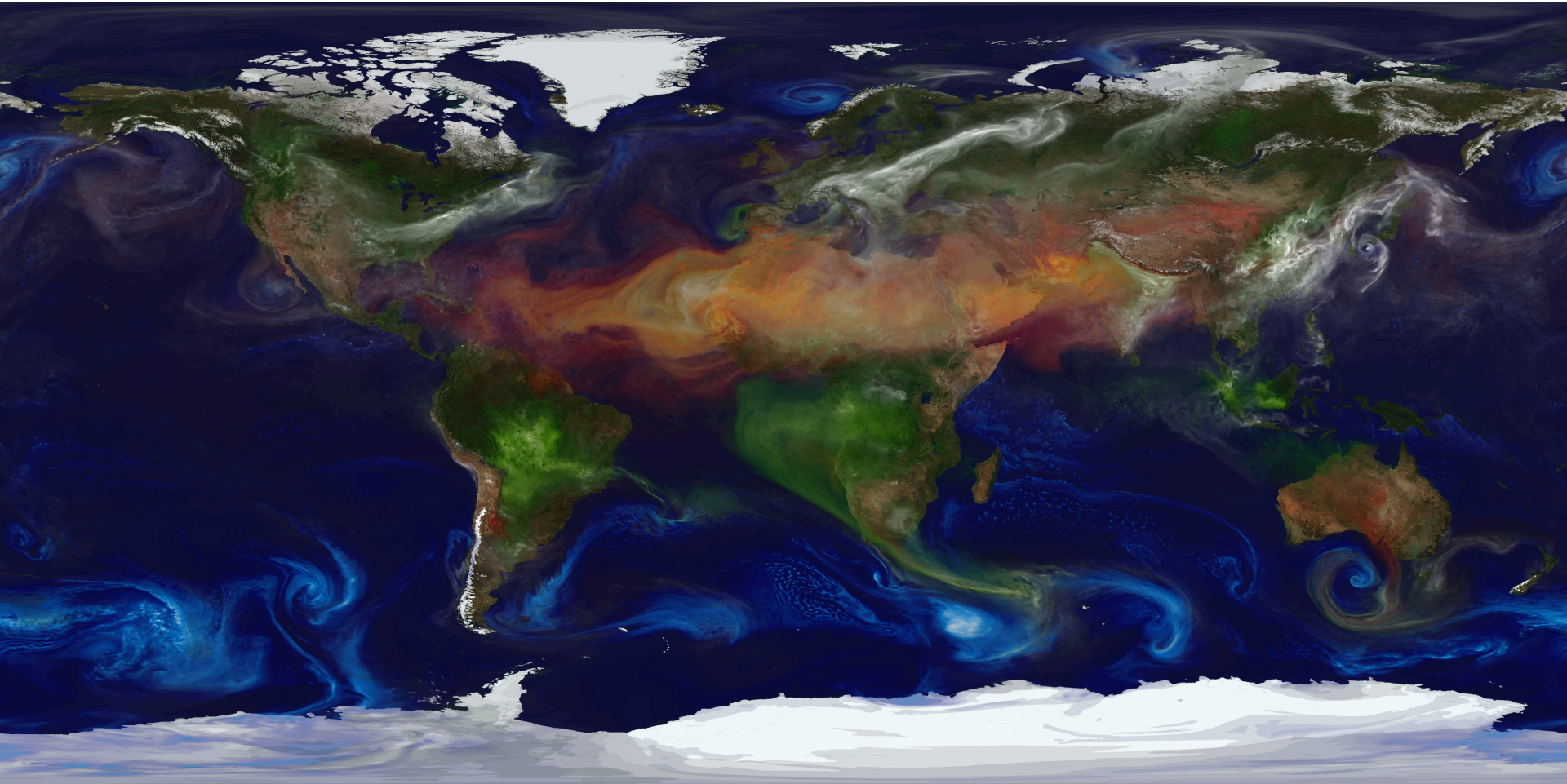



ARM Summer Training and Science Applications: Aerosol Radiative Forcing in Clear and Cloudy Skies



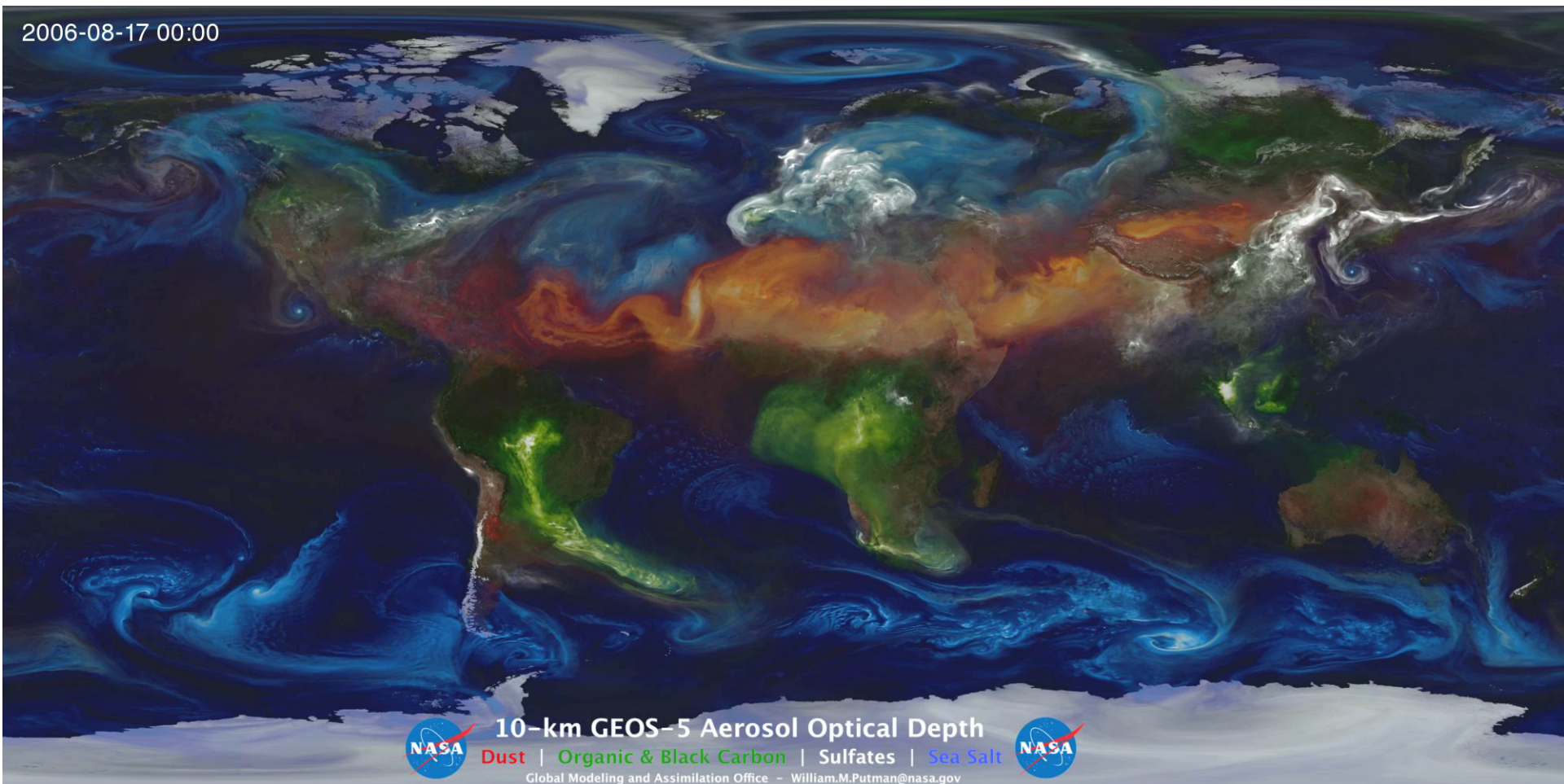
Allison McComiskey, NOAA ESRL



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GEOS-Chem Aerosol Optical Depth by Type

2006-08-17 00:00



 dust

 organic &
black carbon

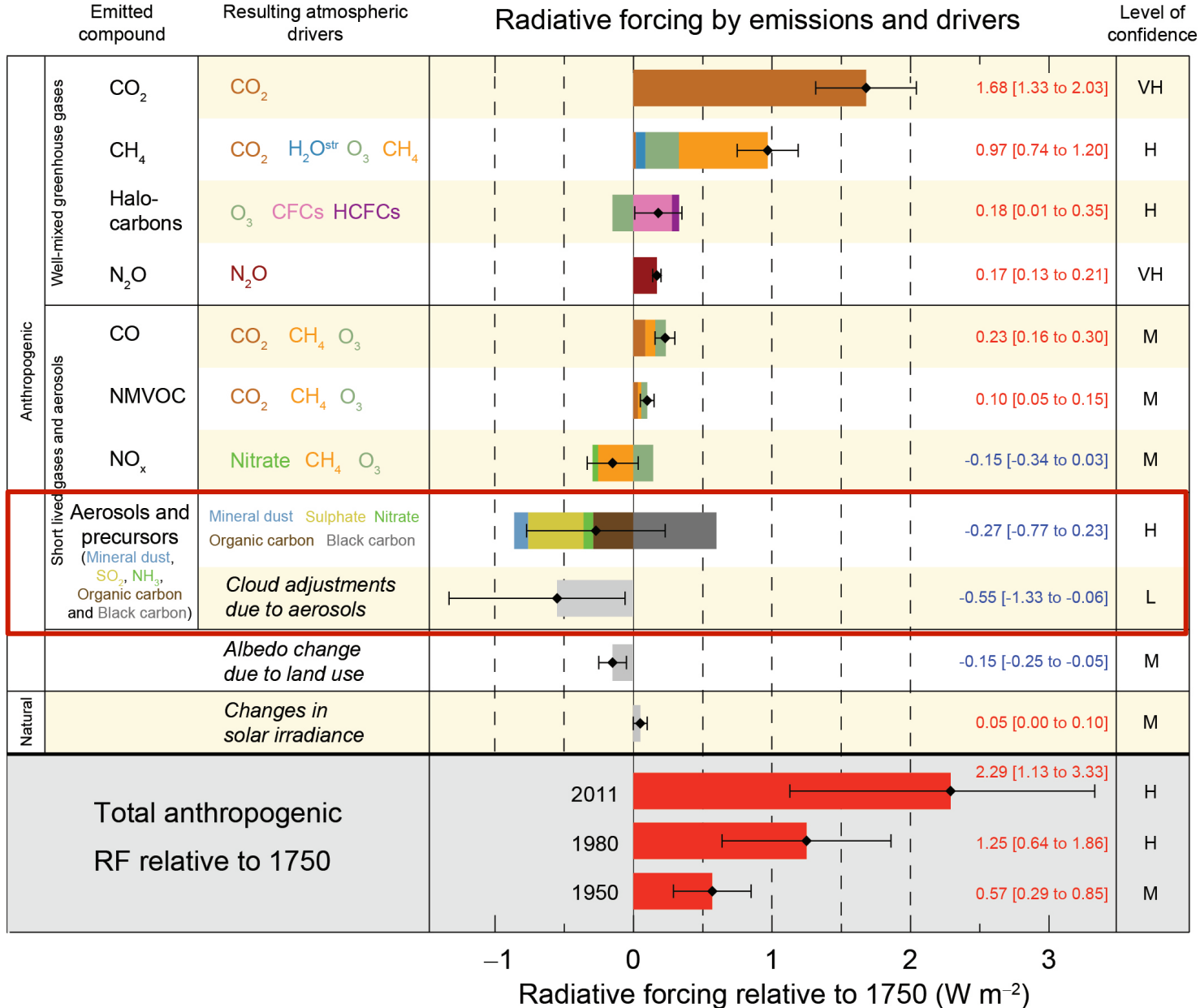
 sulfate

 sea salt

Rationale for Studying Aerosol

- 
- ✧ Important in biogeochemical cycles (e.g. Sulfer cycle)
 - ✧ Direct Effects on Visibility
 - ✧ Effects on Human Health
 - ✧ Water Cycle: Effects on Clouds and Precipitation Formation
 - ✧ ***Radiative Forcing of Climate***

IPCC AR5 Radiative Forcing



uncertainty in aerosol RF
 1 W m^{-2}
 1.27 W m^{-2}

total RF
 2.29 W m^{-2}

Aerosol Radiative Forcing

net irradiance without
anthro aerosol

TOA forcing
global climate

$$\Delta T_s = \lambda F$$



$$RF = (f_a \downarrow - f_a \uparrow) - (f_0 \downarrow - f_0 \uparrow) \text{ W m}^{-2}$$

Direct

net irradiance with
natural + anthro aerosol

Indirect

-0.27 [-0.77 to 0.23] W m⁻²

-0.55 [-1.33 to -0.06] W m⁻²

**direct
radiative
forcing**

- concentration
- optical properties
- spatial distribution

scattering

absorption

transmission

surface albedo

sign and magnitude dependent on
coupled surface-atmosphere system

SRF forcing
regional cooling

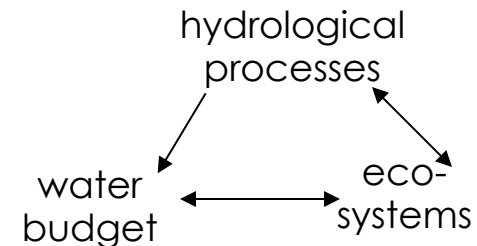
**cloud-albedo effect
(first indirect / Twomey)**

microphysical processes

- number concentration
- drop size

Secondary: fraction

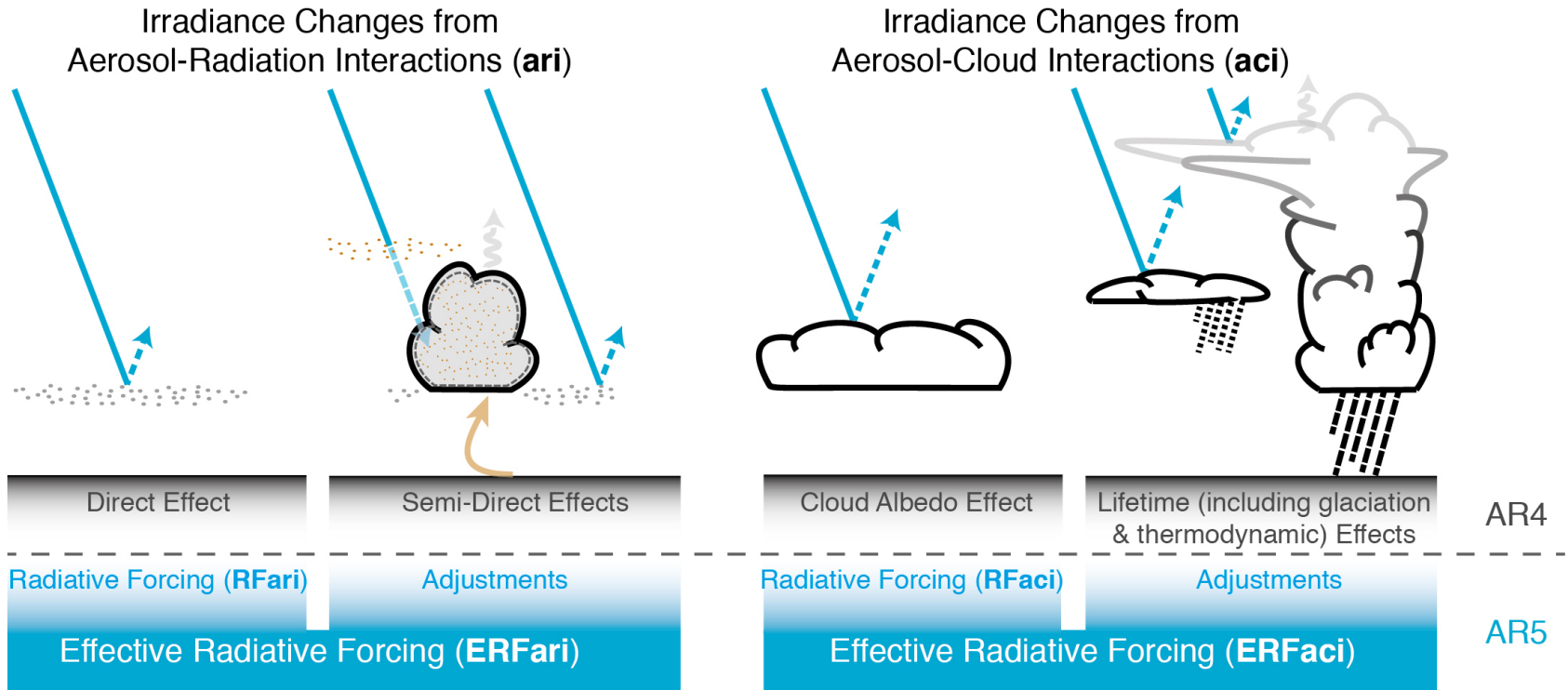
- precipitation (second indirect)
- evaporation (semi-direct)
- lifetime



Aerosol Radiative Forcing according to IPCC AR5

Aerosol Direct Effect – aerosol radiative forcing in clear skies

Aerosol Indirect Effect – aerosol radiative forcing in cloudy skies



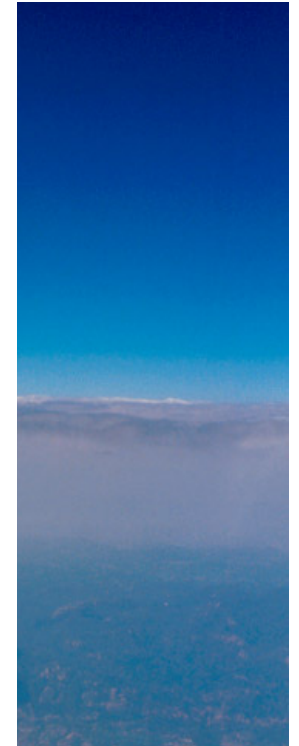
What role does aerosol play in the climate system?

Aerosol Direct Effect – aerosol radiative forcing in clear skies

Aerosol Indirect Effect – aerosol radiative forcing in cloudy skies



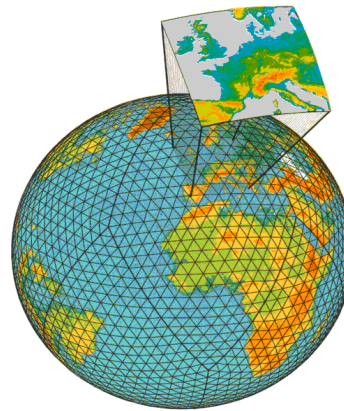
What about 'transition zones'?
the integrated aerosol cloud
system?
ERF ARI+ACI



Top of the Atmosphere RF

Atmospheric RF

Surface RF

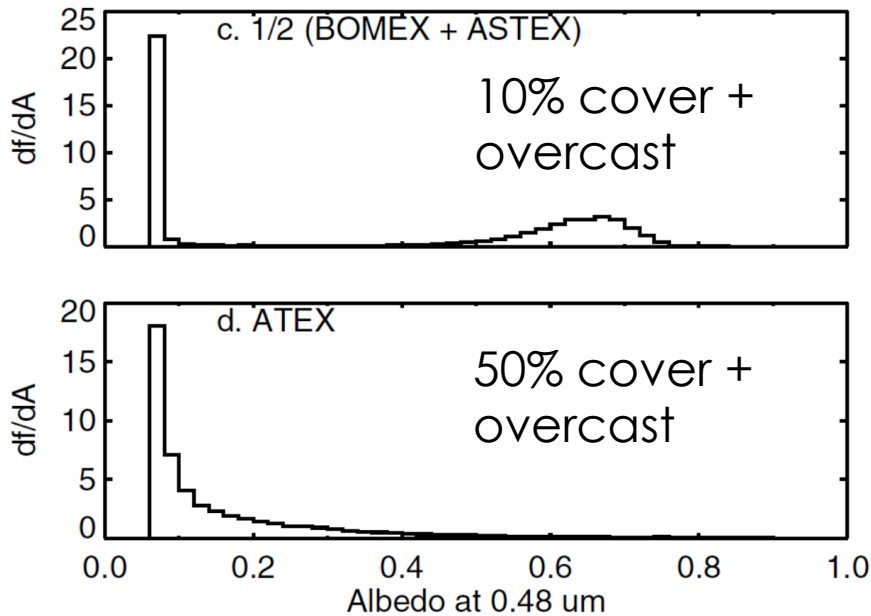


Global – Regional – Local
Annual – Diurnal - Instantaneous

TOA Global Annual Average does not tell the whole aerosol story!

The cloudy-clear transition zone (a.k.a. 'twilight zone', 'halos')

LES

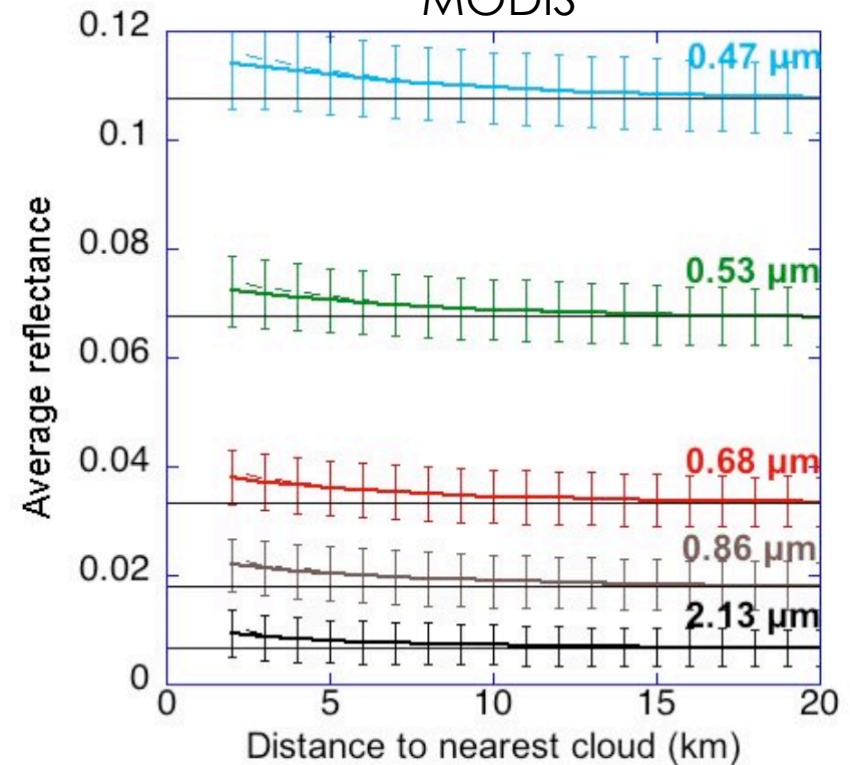


Charlson et al 2007

humidified aerosol between clouds

What is a cloud anyway?

MODIS



Varnai and Marshak 2009

Aerosol Direct Radiative Forcing



solar geometry

$$RF = (f_a \downarrow - f_a \uparrow) - (f_0 \downarrow - f_0 \uparrow) \text{ W m}^{-2}$$

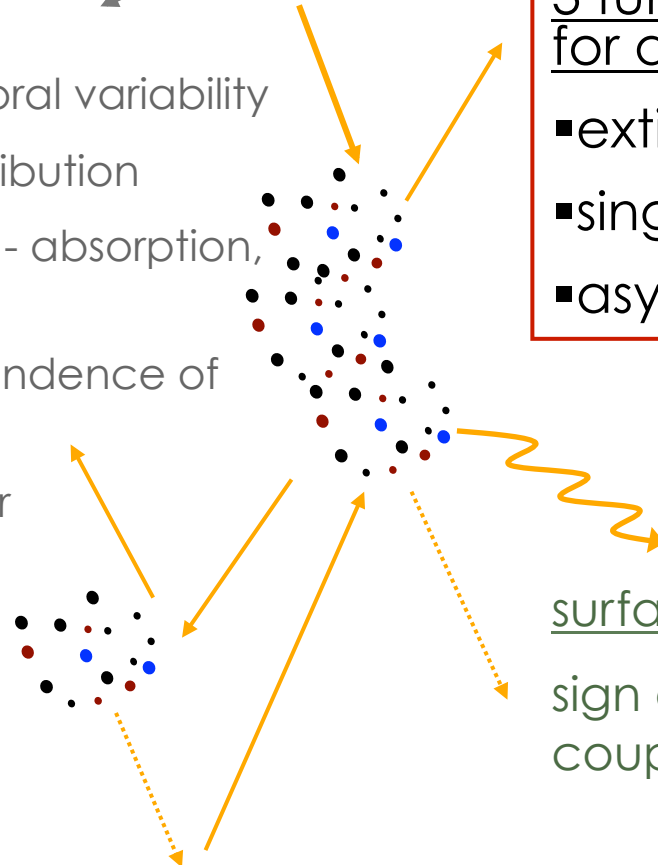
-0.27 [-0.77 to 0.23] W m⁻²

Uncertainties:

- spatial and temporal variability
 - vertical distribution
- optical properties - absorption, mixing state
- wavelength dependence of optical properties
- measurement error

3 fundamental optical properties for aerosol radiative forcing

- extinction/optical depth
- single scattering albedo
- asymmetry parameter



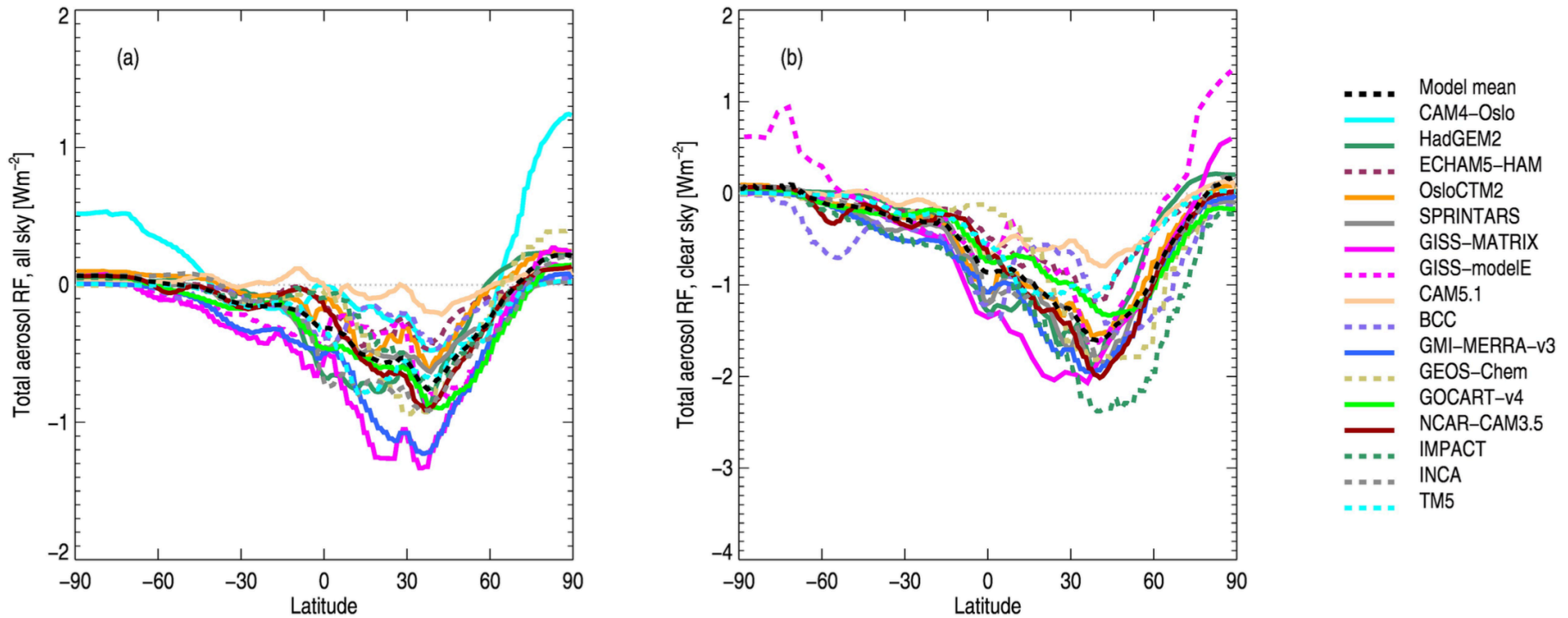
surface albedo

sign and magnitude dependent on coupled surface-atmosphere system

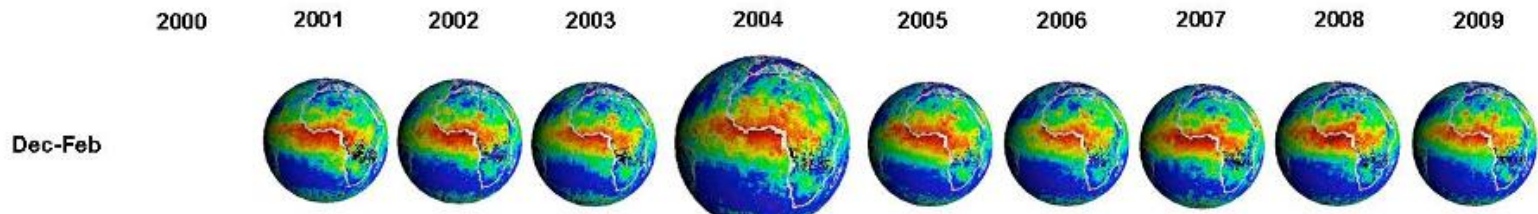
SRF forcing
regional cooling

What are uncertainties for estimating aerosol radiative forcing?

The AeroCom Project: Aerosol comparisons between observations and models



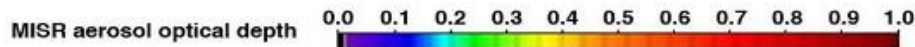
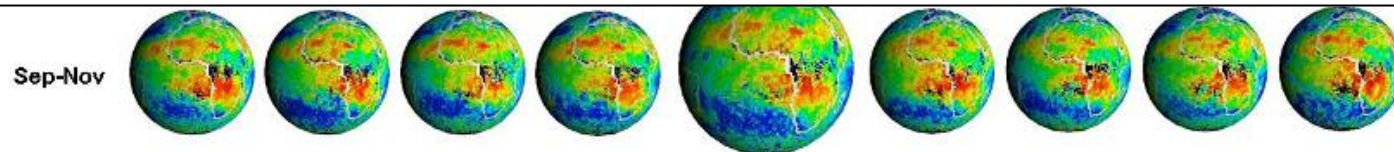
Ten Years of Seasonally Averaged Mid-visible Aerosol Optical Depth from **MISR**



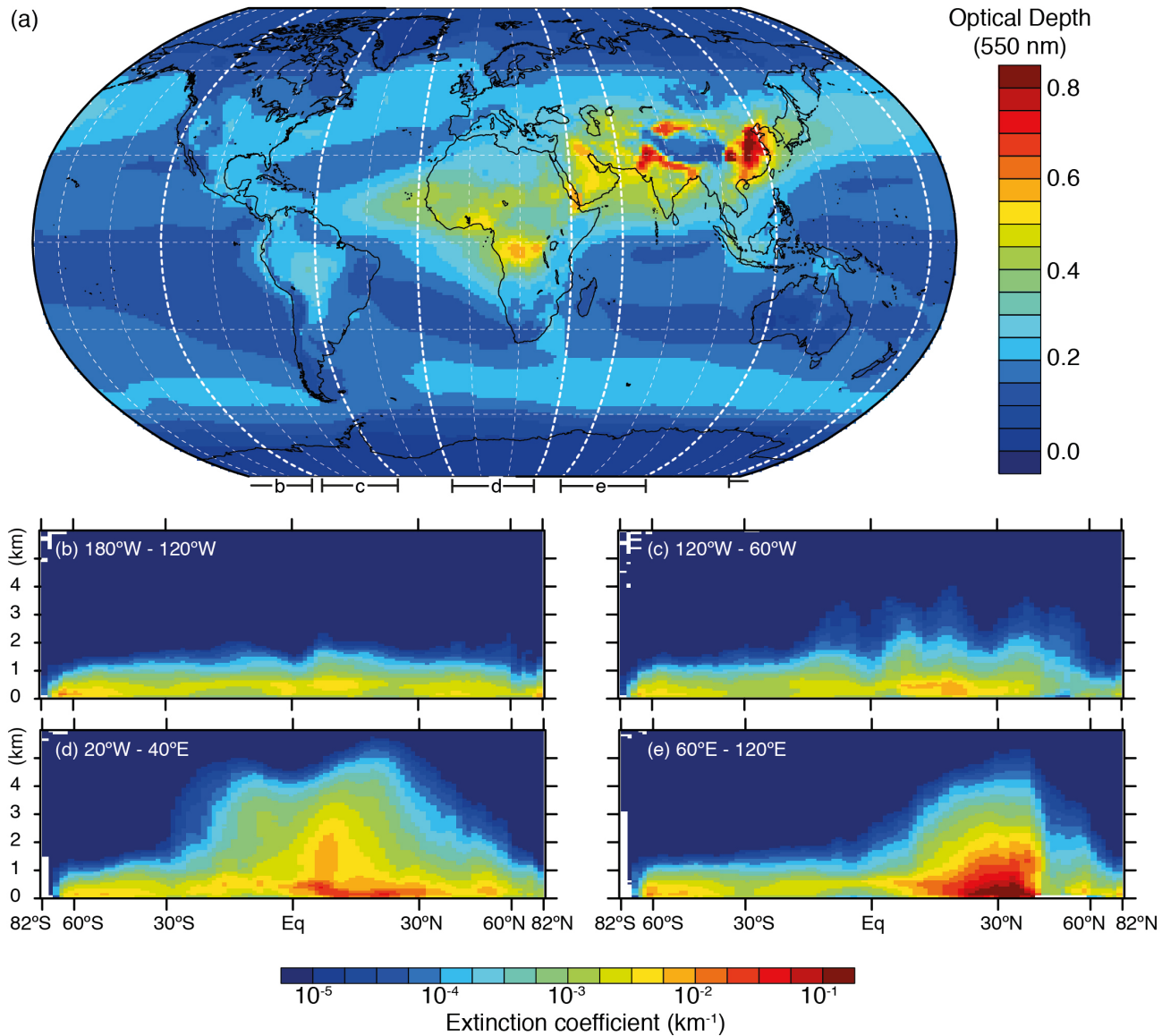
concentrations and distributions are remarkably seasonally consistent at a given location – why?

emissions and transport (meteorology) patterns are seasonally consistent

but what happens if emissions and circulation patterns change?



Aerosol optical depth horizontal and vertical distribution 8-year average: IPCC AR5



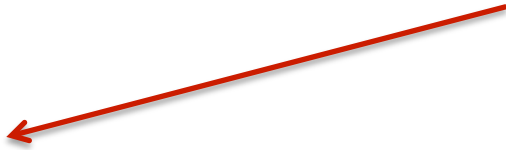
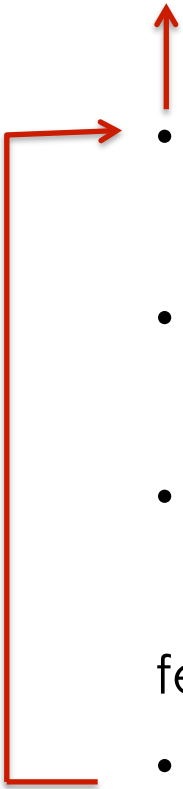
What are uncertainties for estimating aerosol radiative forcing?

spatial distribution (horizontal and vertical) of aerosol amount and properties

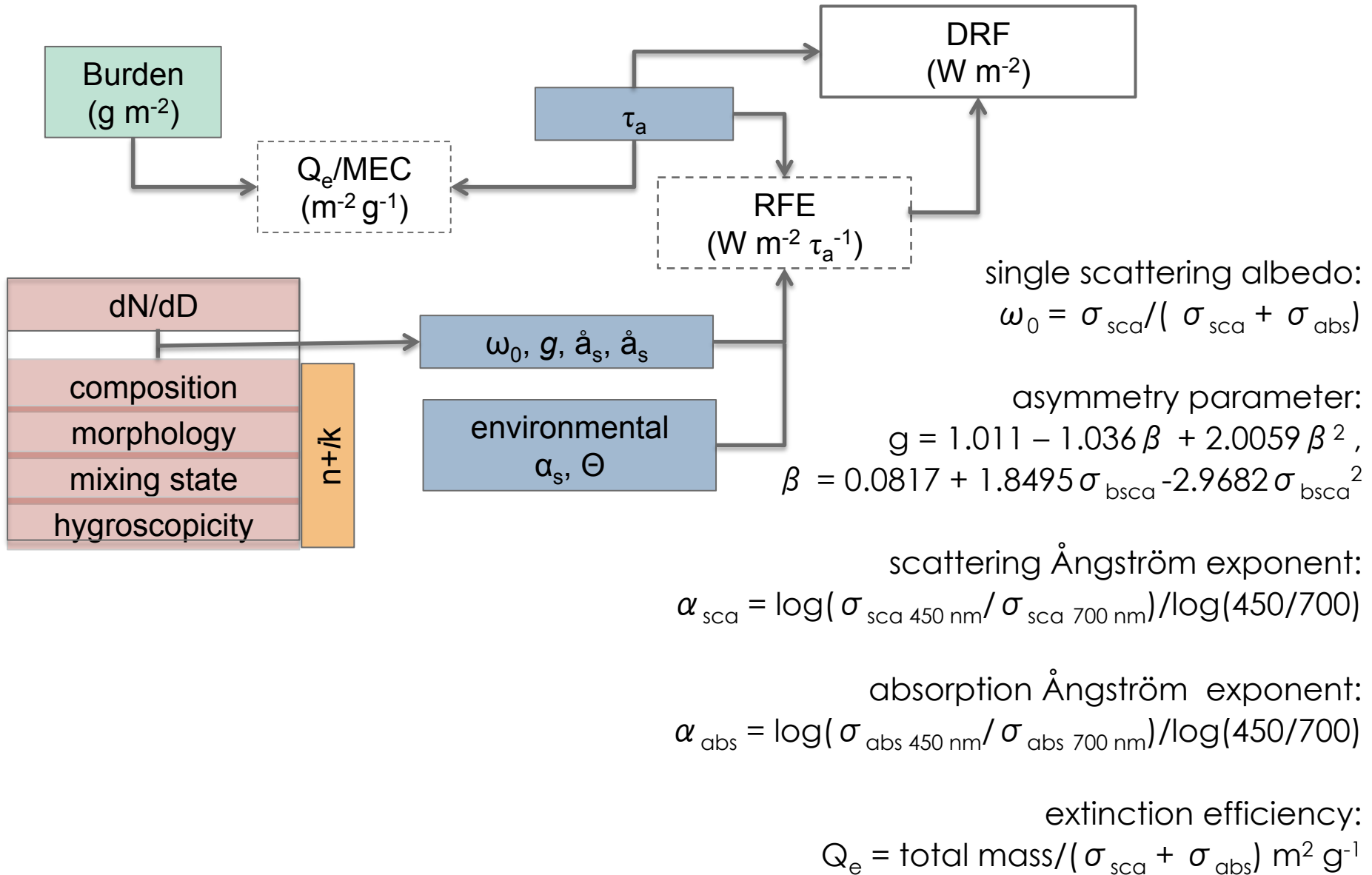
- aerosol formation processes: emissions to burden
- composition & size distribution to optical properties
- optical properties to radiative fluxes

feedbacks

- aerosol heating and cooling effects on circulation, cloud development, temperature



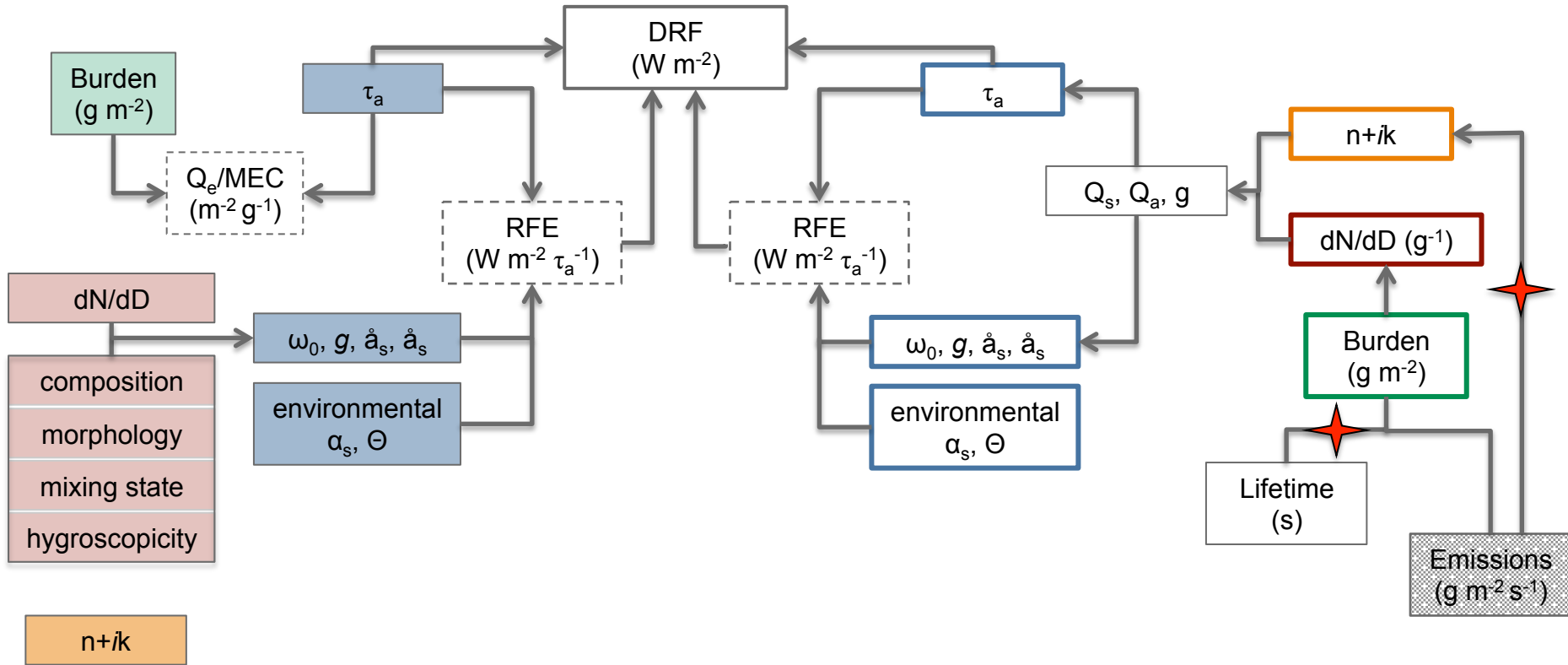
Pathways to Calculating Aerosol Direct Radiative Forcing



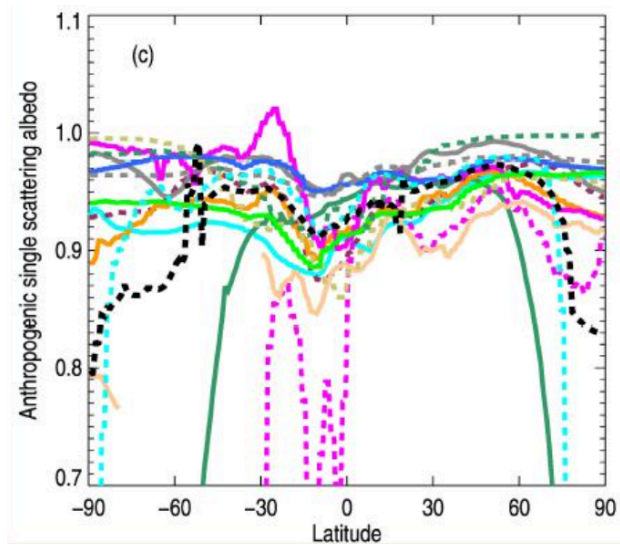
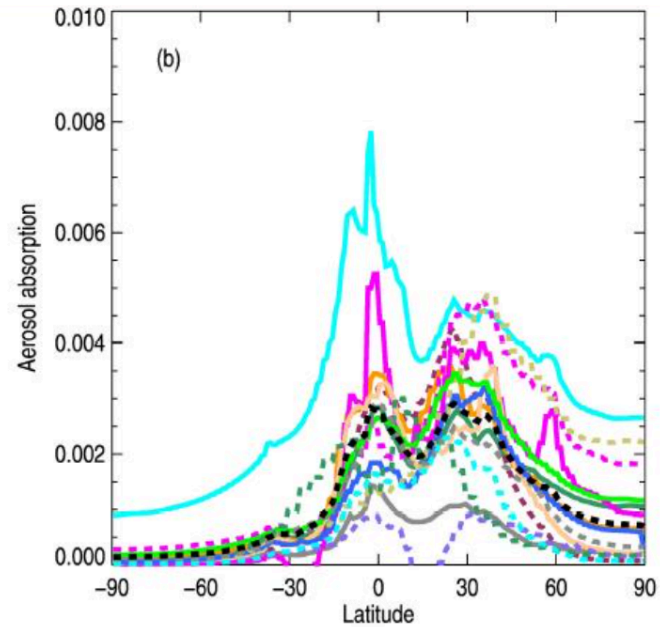
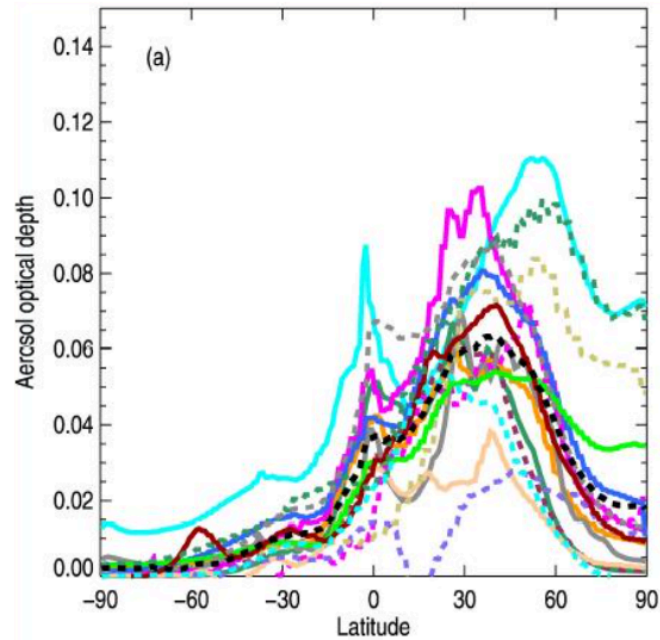
Pathways to Aerosol Direct Radiative Forcing: Observed and Modeled

Observed

Modeled



AeroCom Diversity in optical properties



What is an aerosol?

A liquid or solid particle suspended in a gas

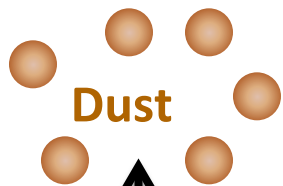
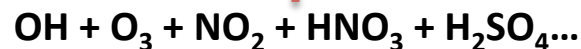
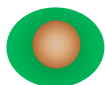
Originate from **natural** and **anthropogenic** sources



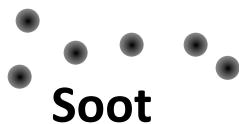
courtesy Jessie Creamean

Aerosol Formation Pathways

Transformation



Dust



Soot



Primary Formation

Directly emitted

Secondary Formation

Gaseous emissions

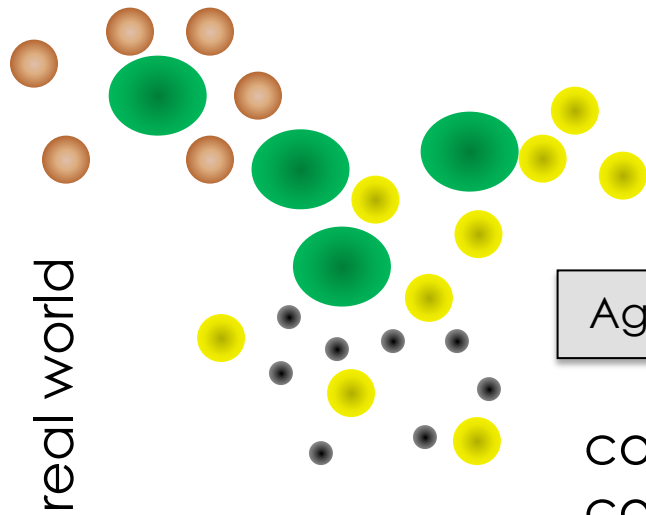
Formed/transformed in the atmosphere

courtesy Jessie Creamean

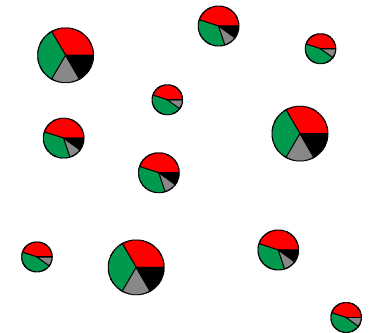
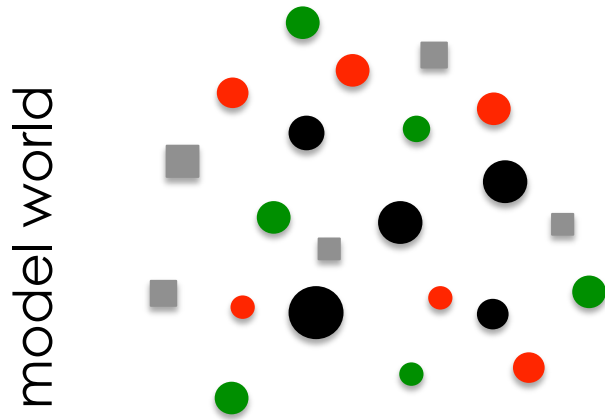
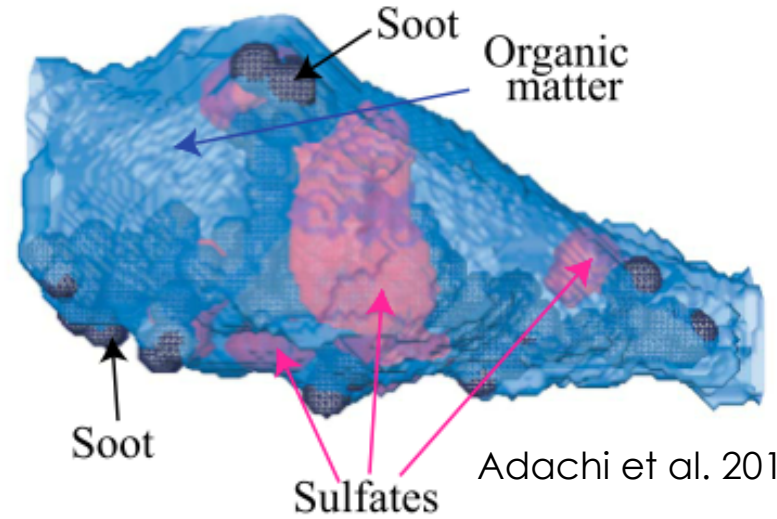
Aerosol Mixing State

External Mixture

Internal Mixture



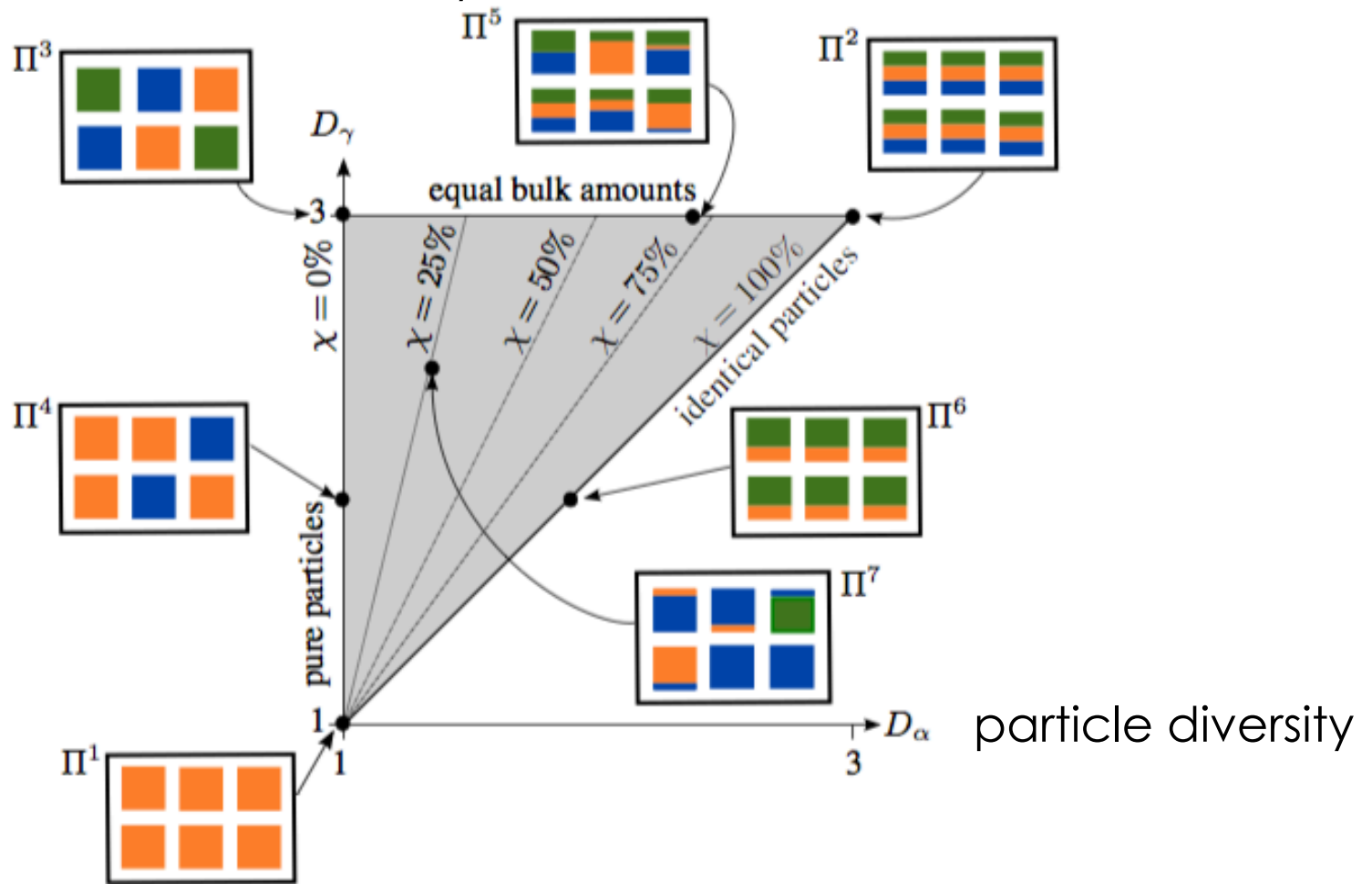
condensation
coagulation
chemical transformation

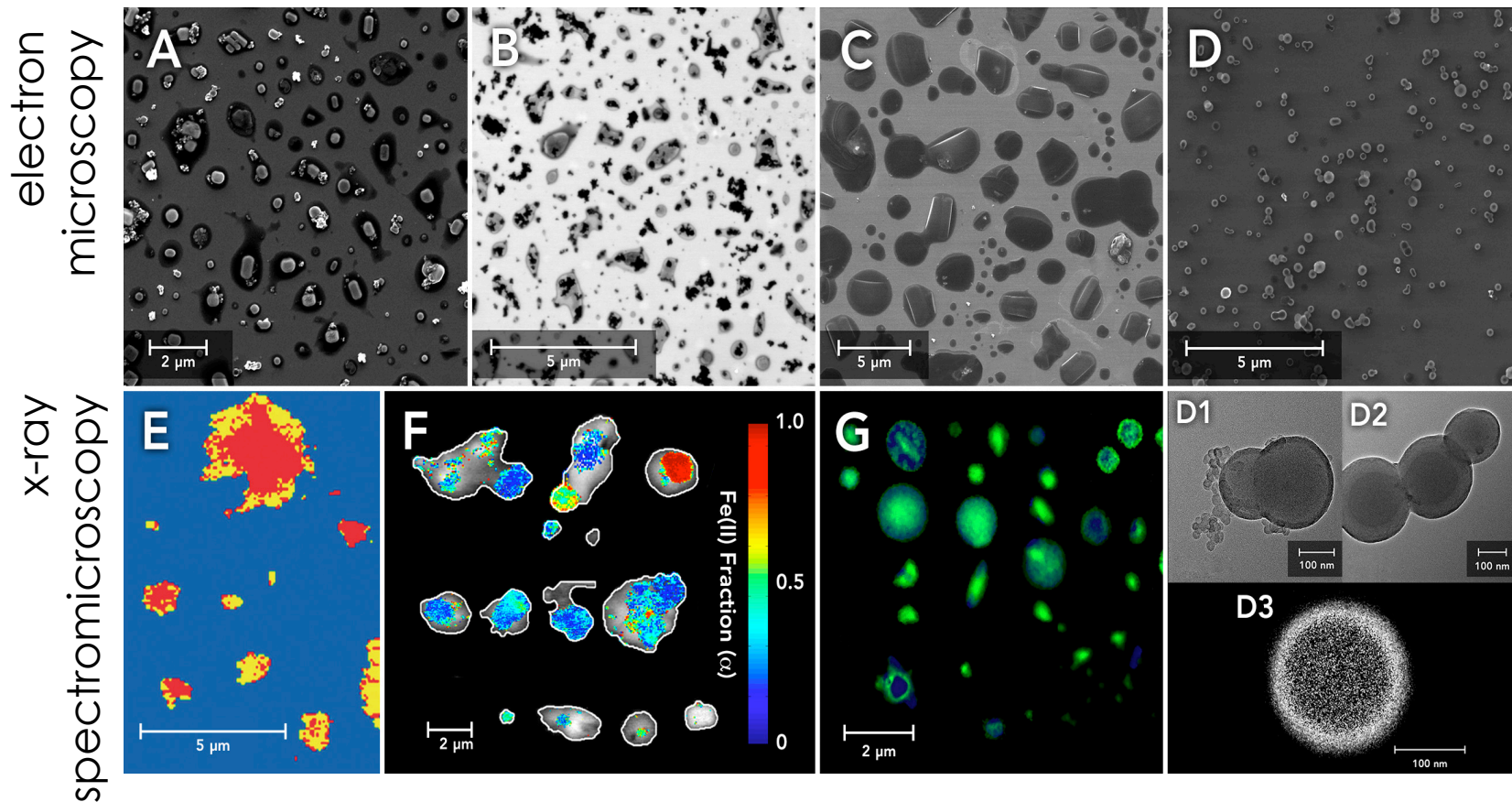


'parameterization'

the Mixing State Index χ

bulk diversity



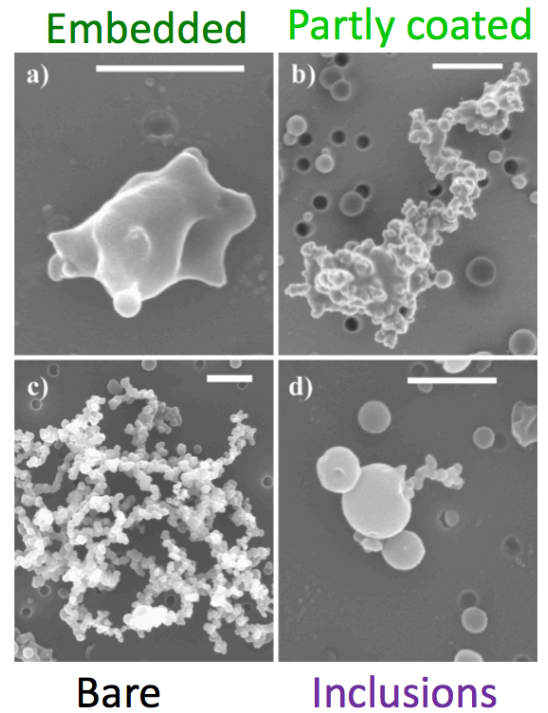
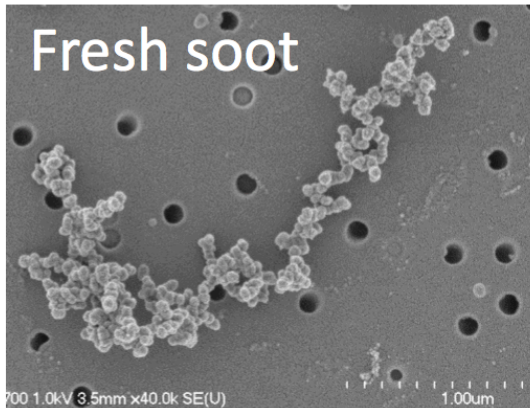


E - Marine particles collected at Pt. Reyes National Shore Park, California. Red and yellow colors indicate the internal distribution of two different oxidation states of sulfur (S(IV) and S(VI)) inside individual particles (Liu et al. 2011b)

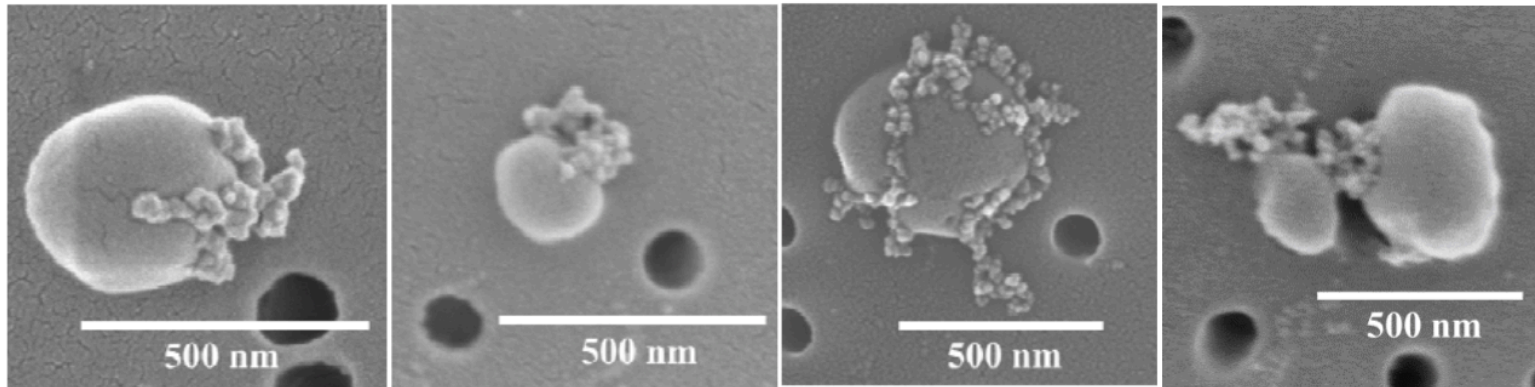
F - Atmospheric dust particles collected on Okinawa Island, Japan. Color scale indicates fraction of Fe(II) present in individual particles (Moffet et al. 2012)

G - Marine particles (blue) internally mixed with anthropogenic organic material (green) collected in vicinity of Sacramento, California (Laskin et al. 2012b)

Aerosol Morphology



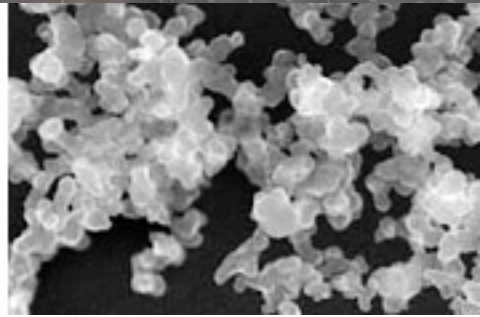
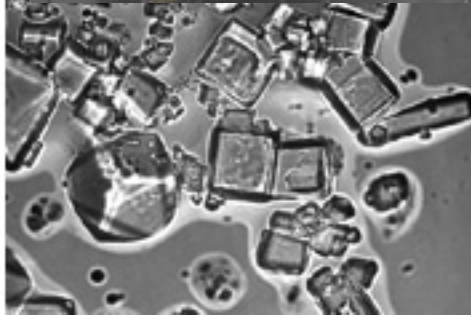
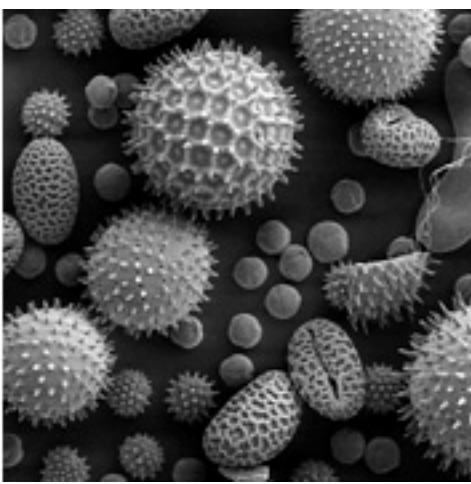
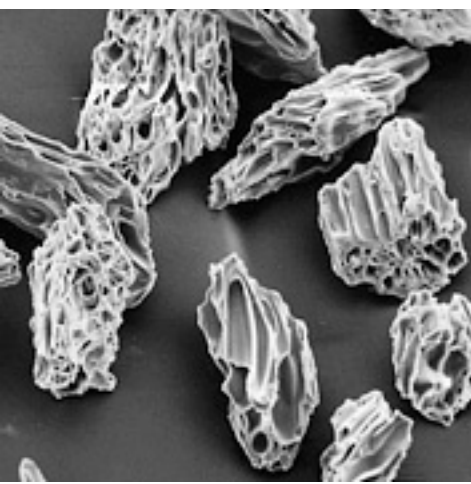
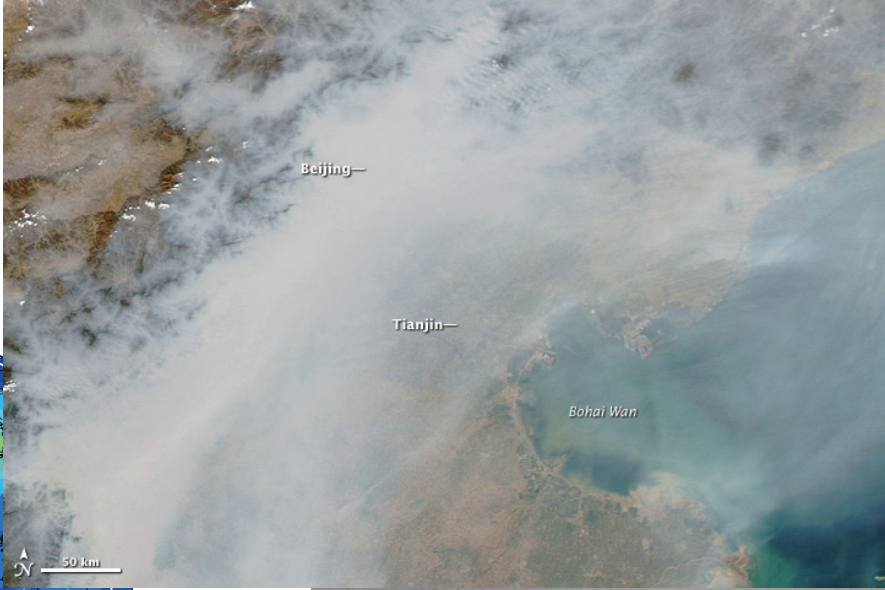
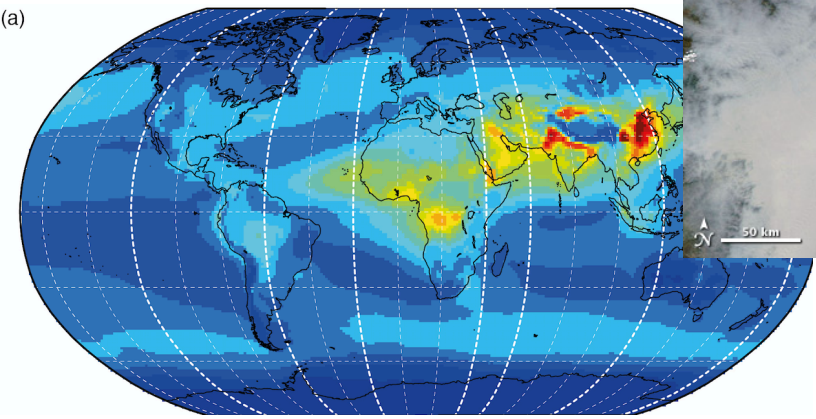
[China et al., 2013]



Soot morphology and mixing: Sacramento 2010

courtesy Claudio Mazzoleni

(a)



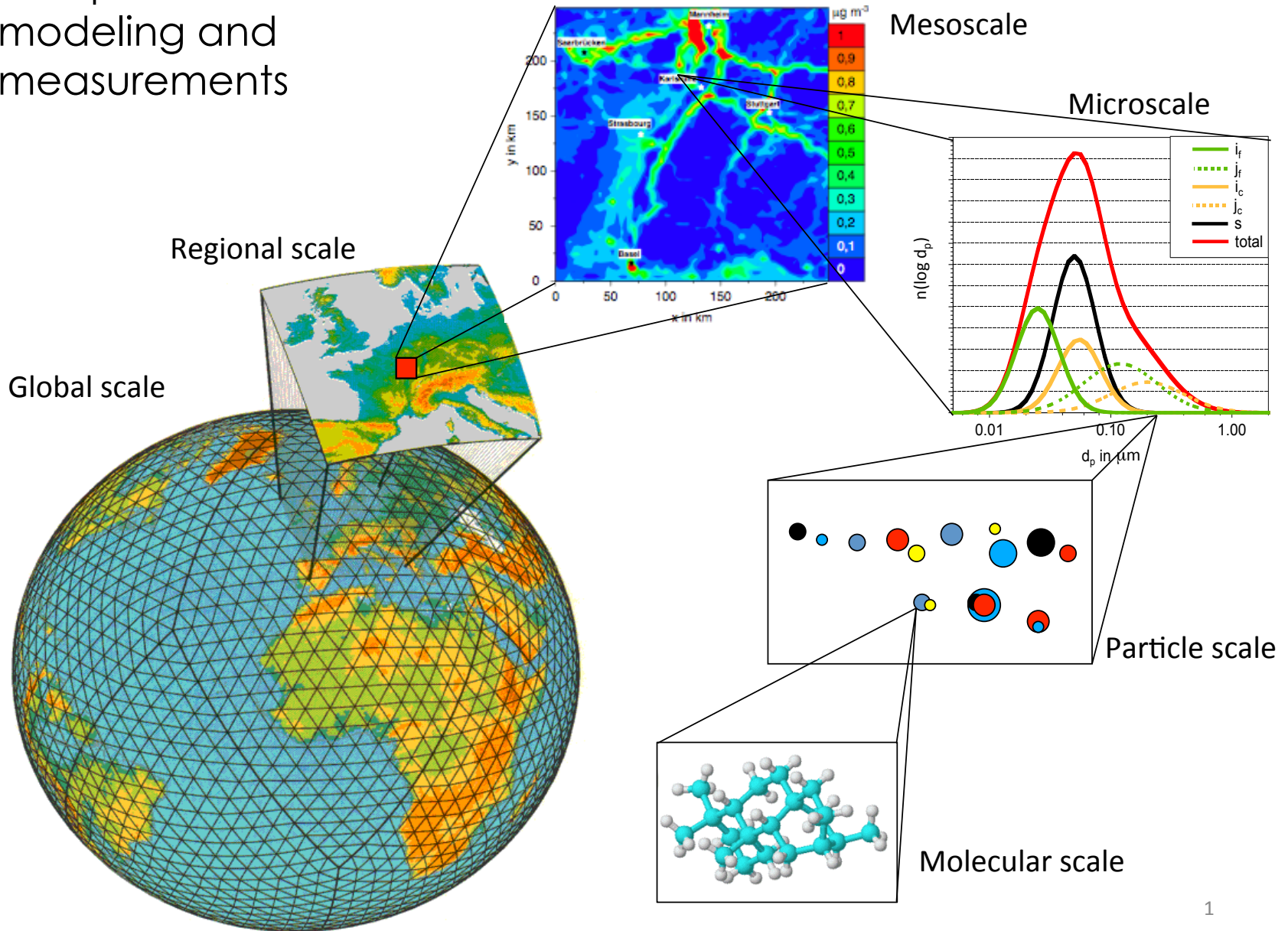
volcanic ash

pollen

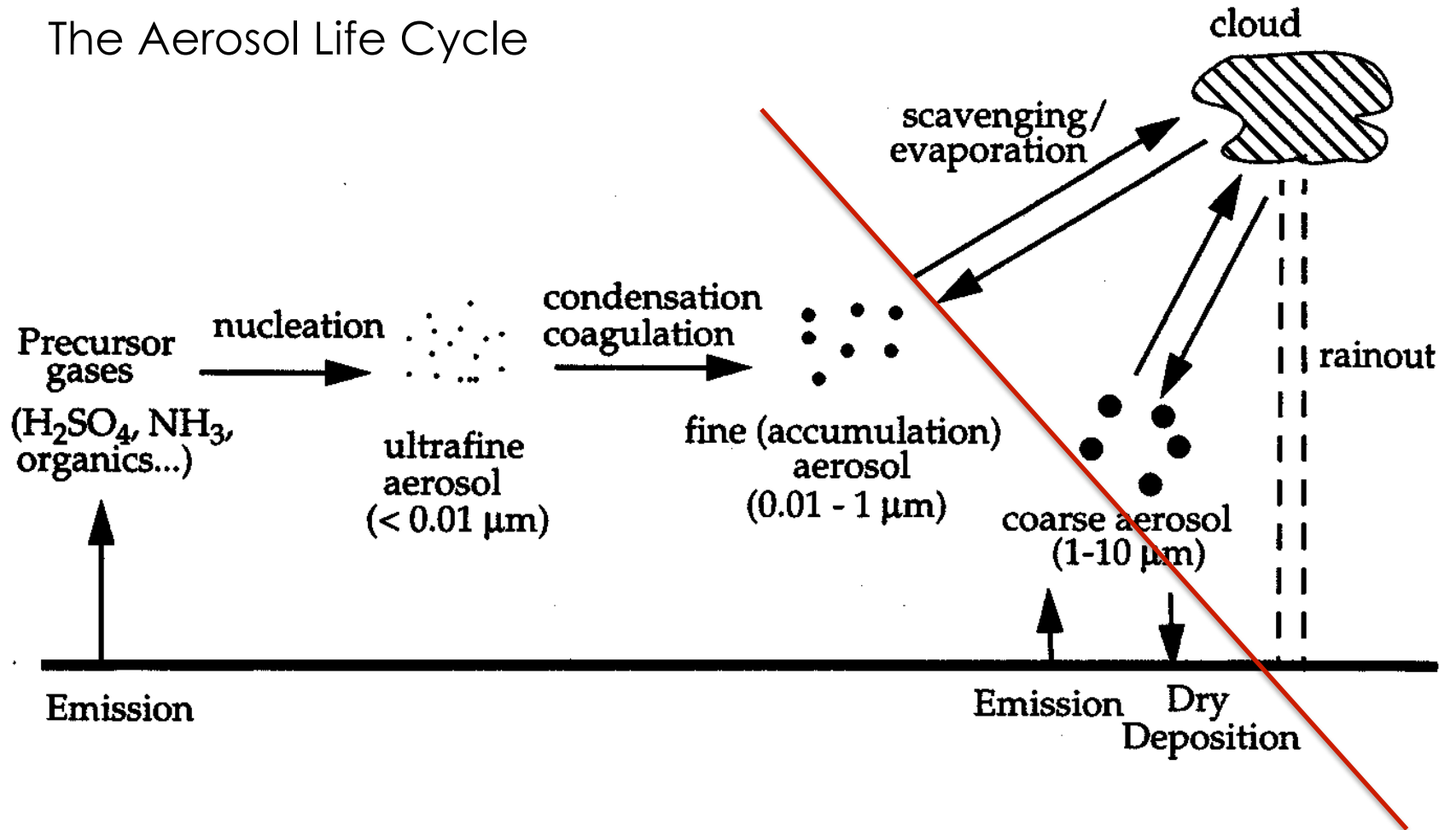
sea salt

soot

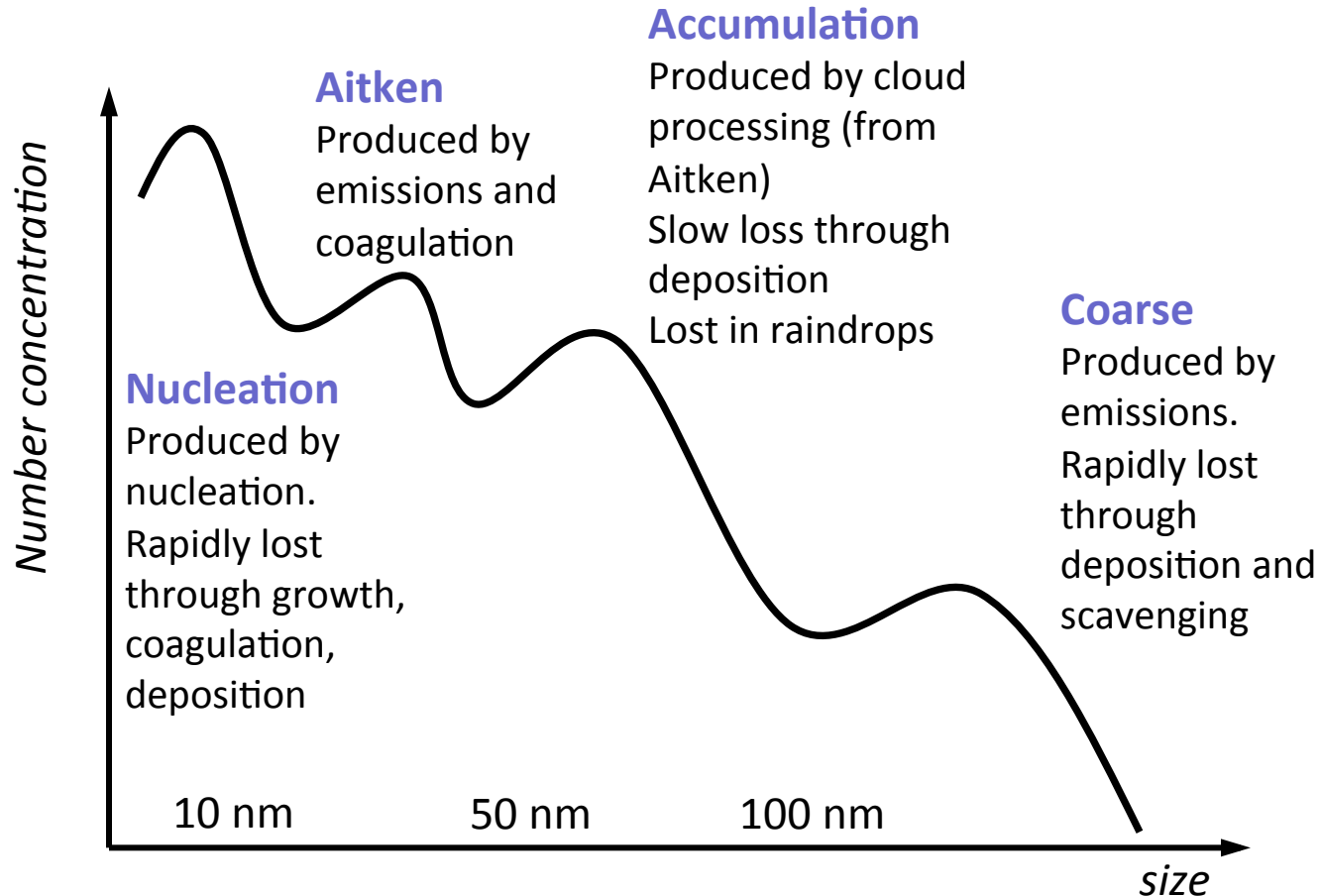
Multiple scales for modeling and measurements



The Aerosol Life Cycle



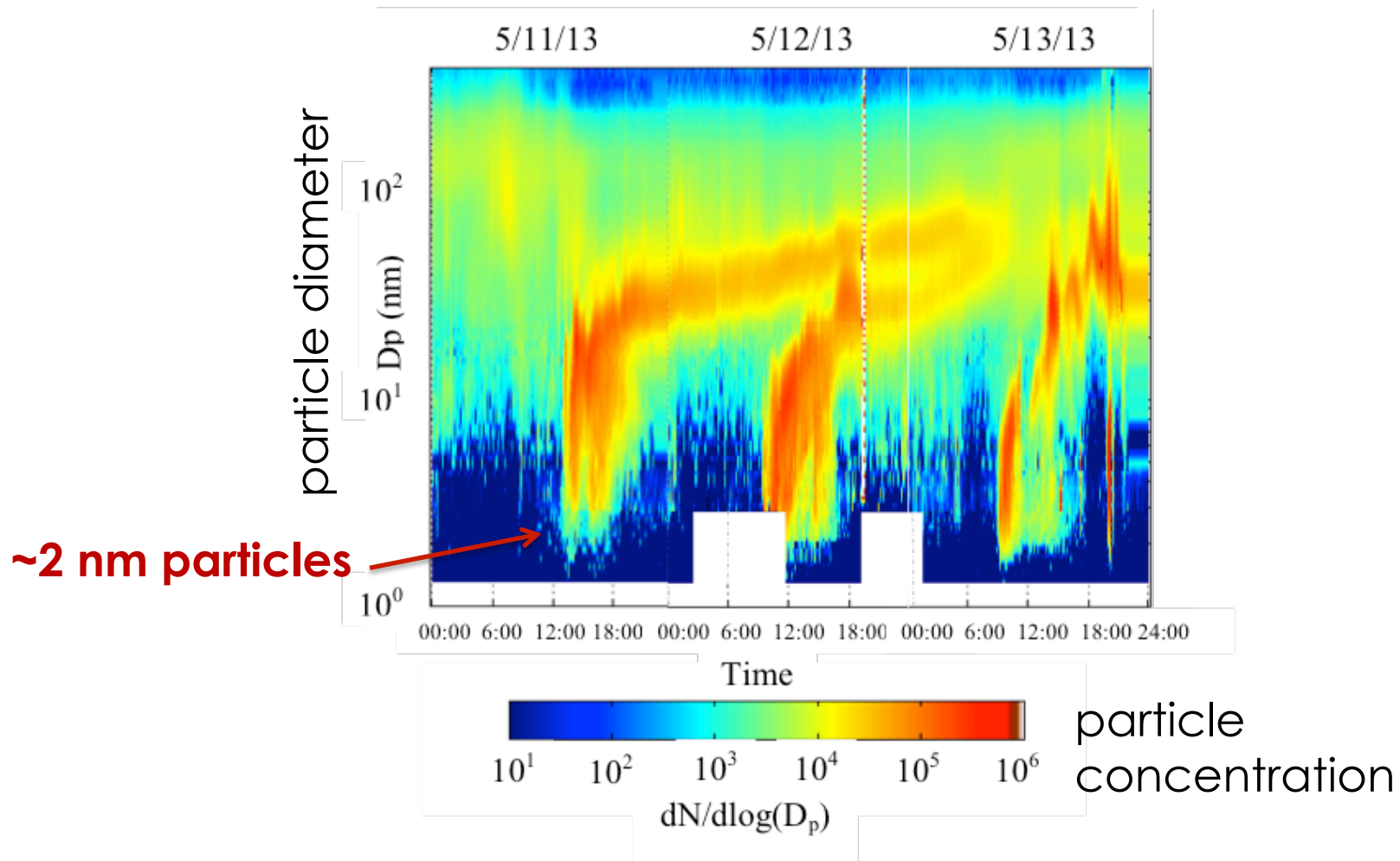
Aerosol size distributions: function of emissions, formation, and processing



Size is the most fundamental parameter for determining aerosol optical properties and radiative effects.

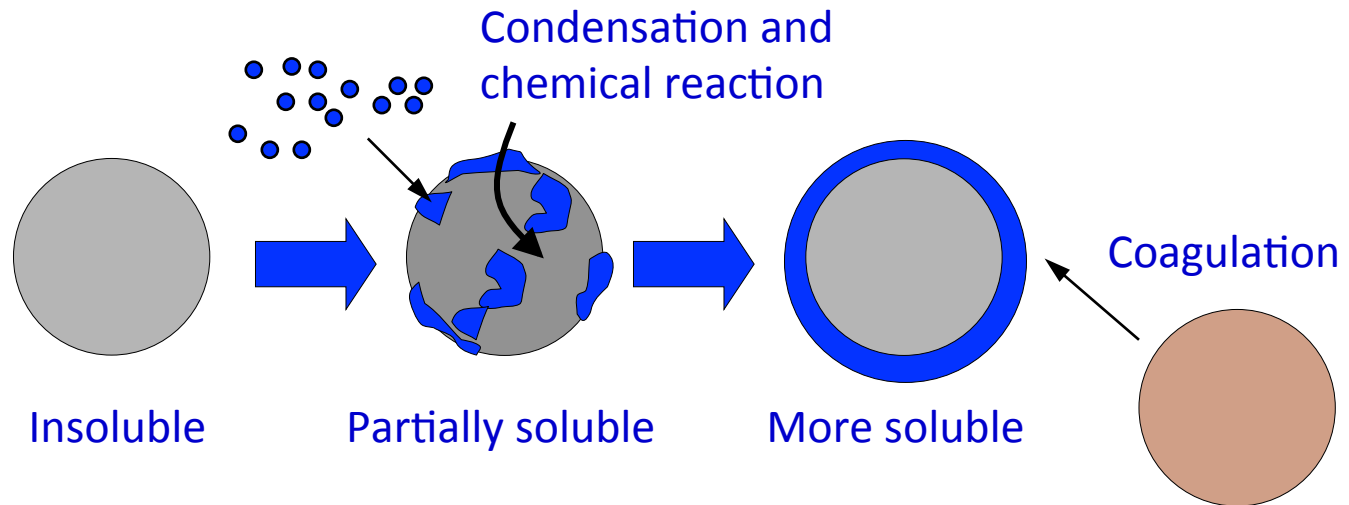
Aerosol Nucleation

New Particle Formations events at SGP



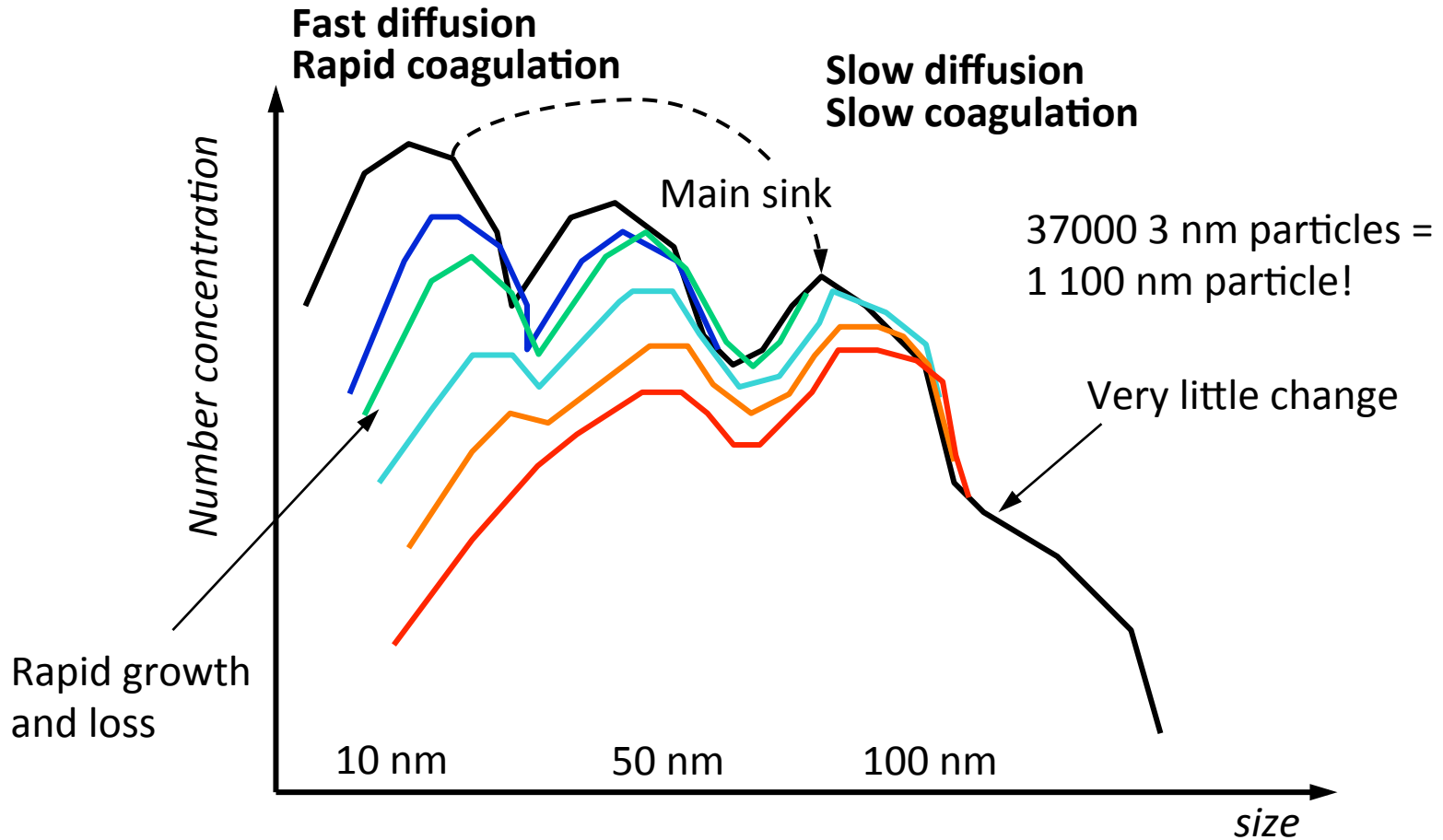
courtesy Peter McMurry and Jim Smith

Aging Processes

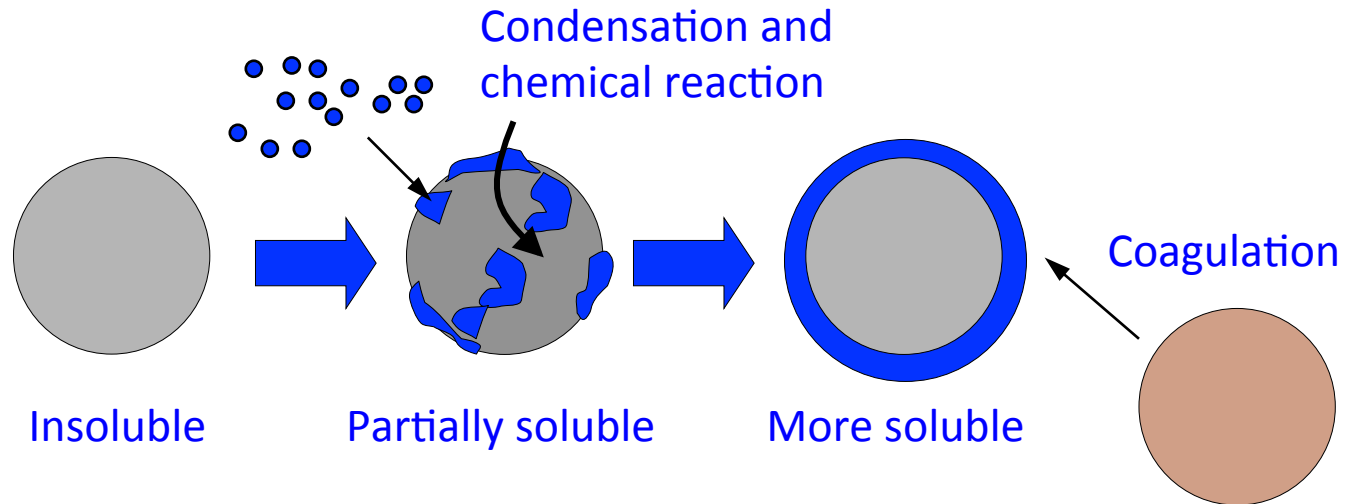


- some primary particles are initially insoluble
- can become partly soluble through
 - coagulation
 - chemical transformation
 - condensation
- results in
 - varied optical properties
 - greater CCN activity

Growth and coagulation



Aging Processes



Rate of ageing: time to form 1 monolayer ($\sim 10^{19}$ molec m^{-2}) of H_2SO_4 on a 100 nm particle

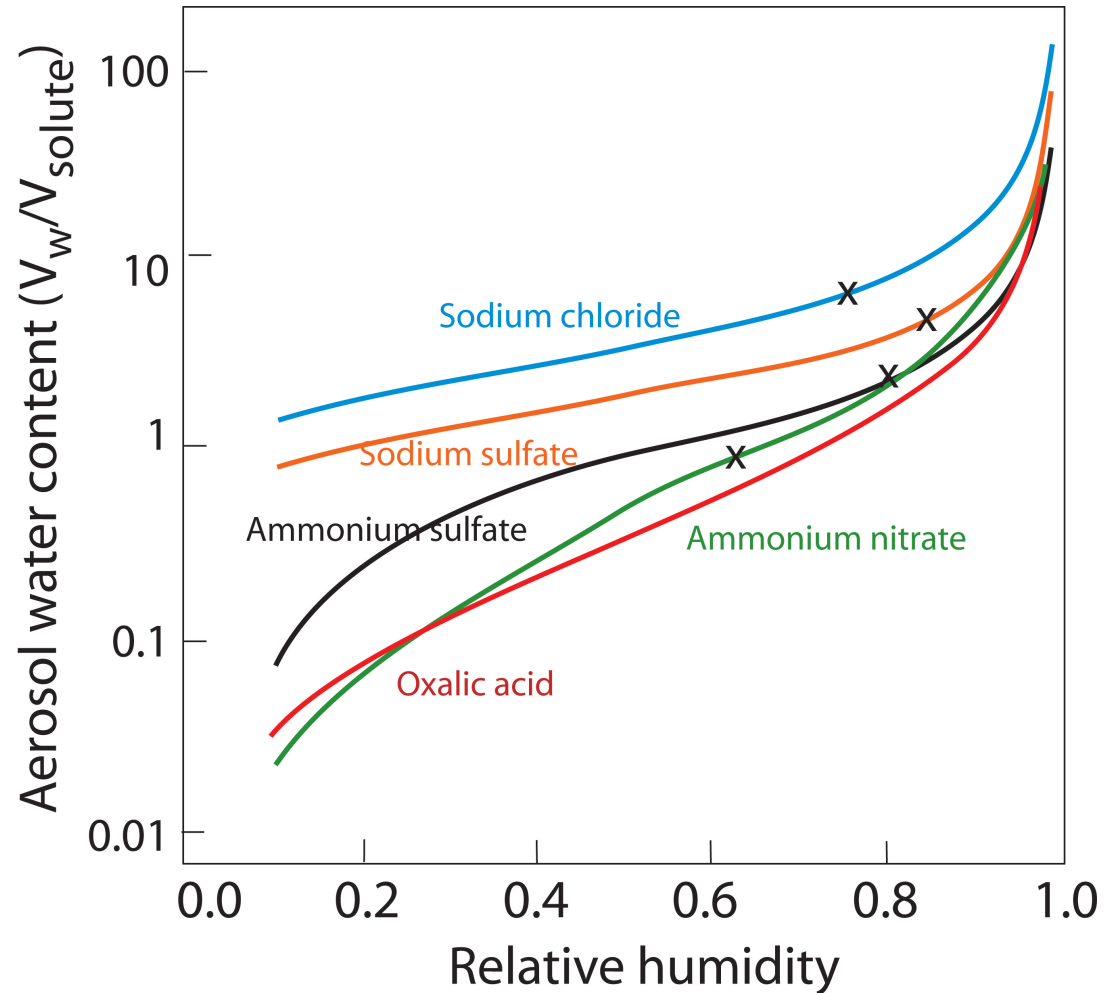
- **$\sim 10^4$ s (3 h)** in polluted conditions ($\text{H}_2\text{SO}_4 = 10^7 \text{ cm}^{-3}$)
- **$\sim 10^5 - 10^6$ s (1-10 days)** in clean conditions
- Factor >10 longer for coarse particles

these processes coupled with meteorology determine the spatial distribution of aerosol and their radiative properties

Hygroscopicity

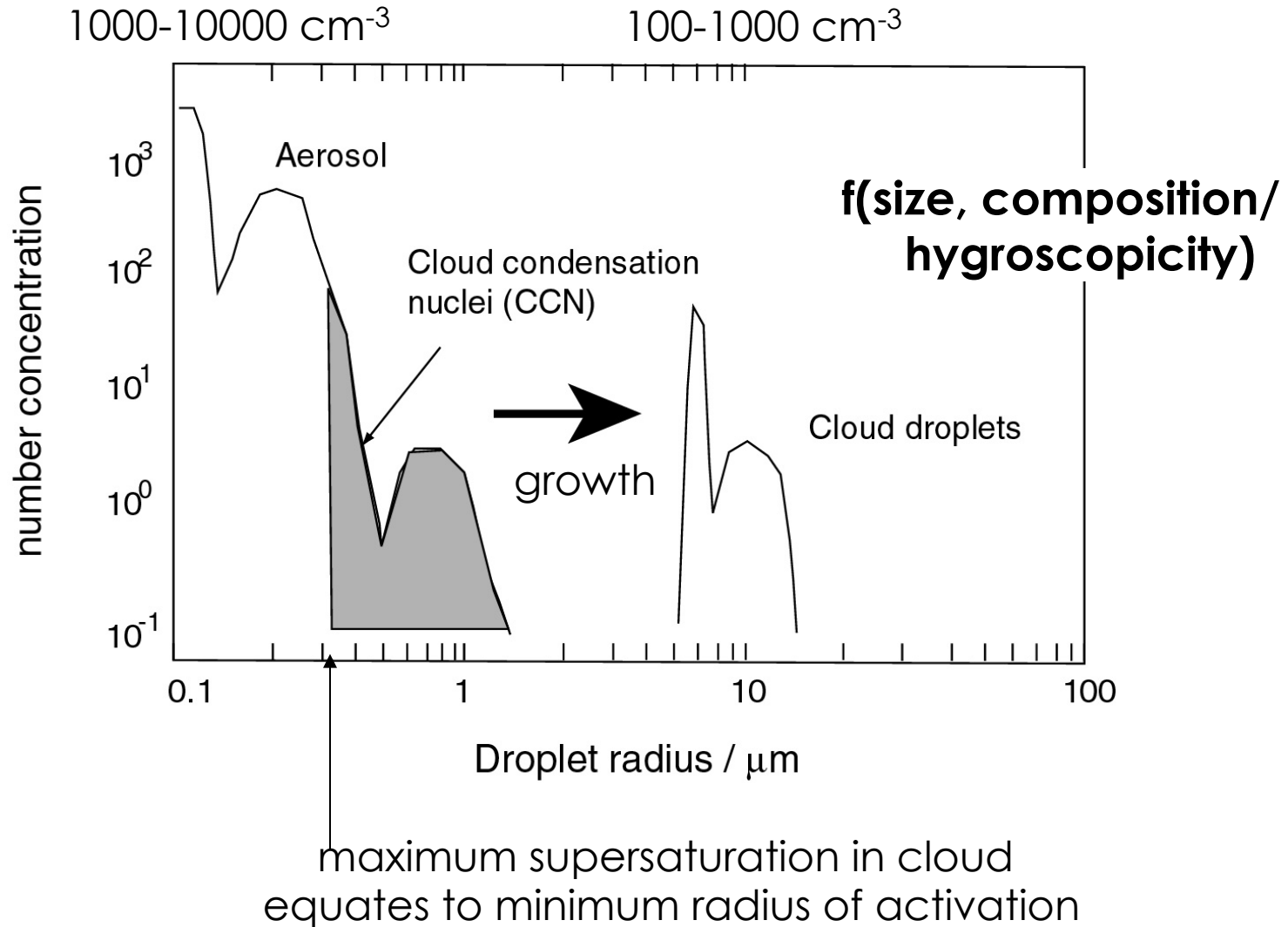
measure of water uptake by soluble particles

- Reversible process (deliquescence, efflorescence)
- $f(\text{RH, composition})$
- Timescale for water equilibration \sim seconds



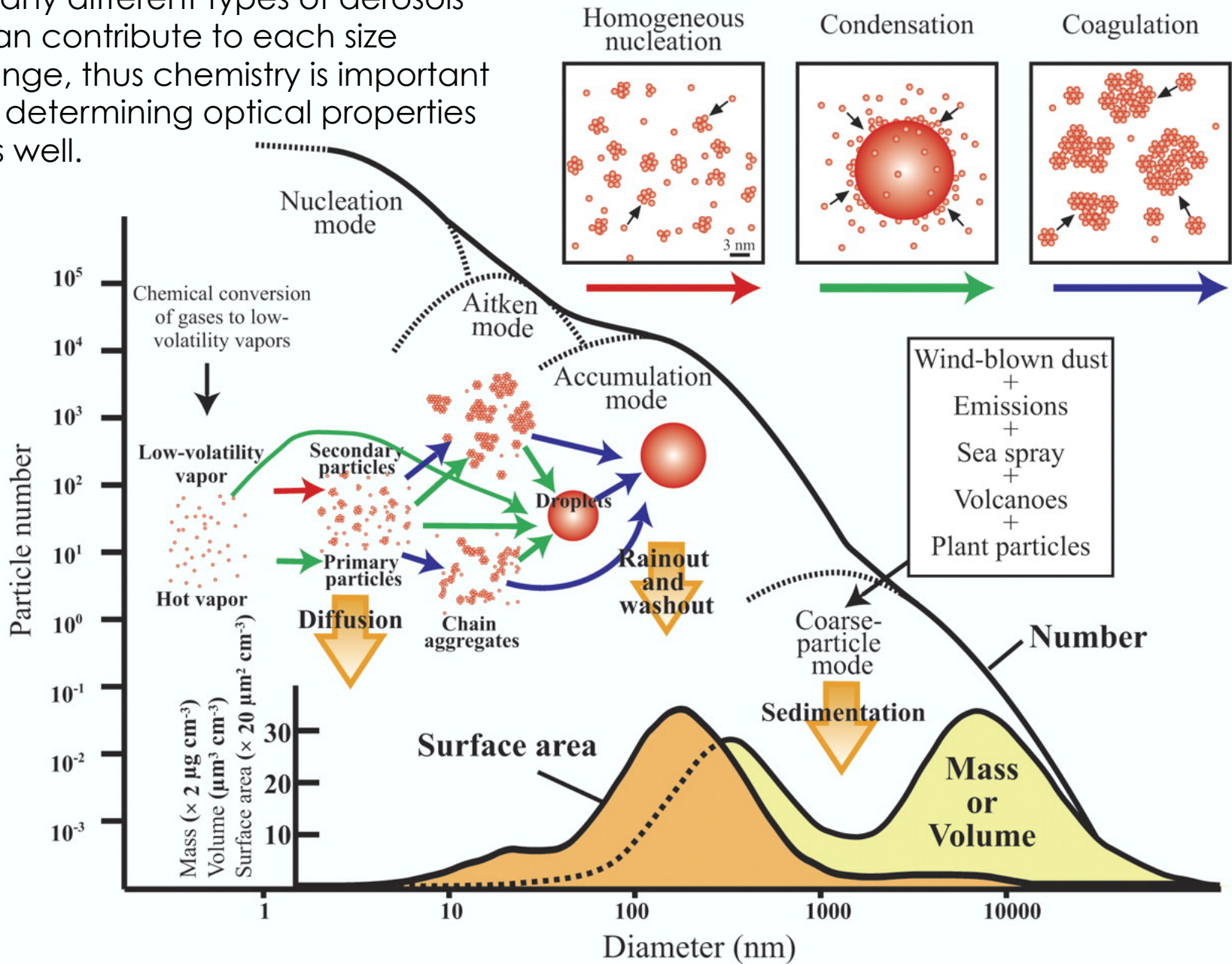
Petters and Kreidenweis, 2007

Particle activation in clouds



Size is the most fundamental parameter for aerosol radiative effects.

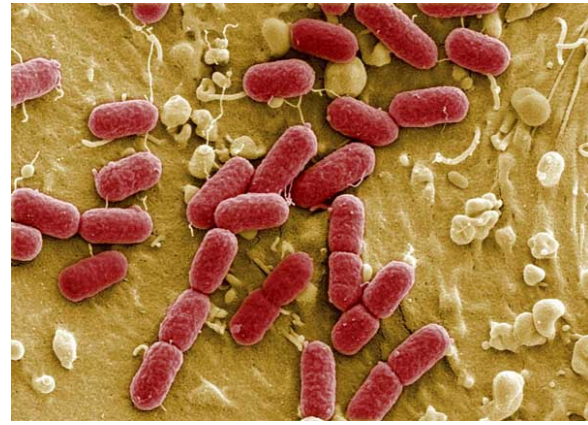
Many different types of aerosols can contribute to each size range, thus chemistry is important in determining optical properties as well.



Bioaerosol sources

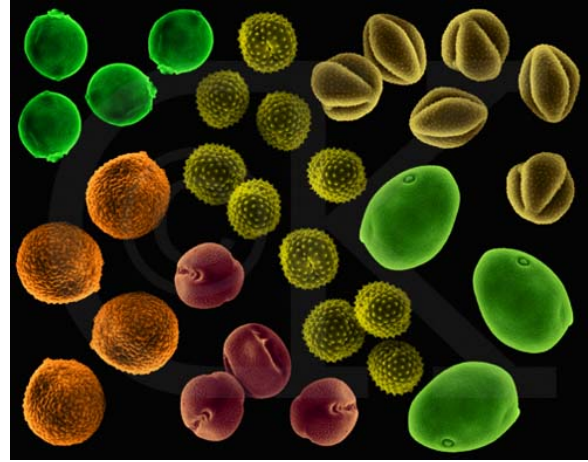
- “Bioaerosol” comprises particles from viruses (10’s of nm) to pollen and leaf fragments (100’s of um)
- Some are emitted actively and some passively

Bacteria
~1 um



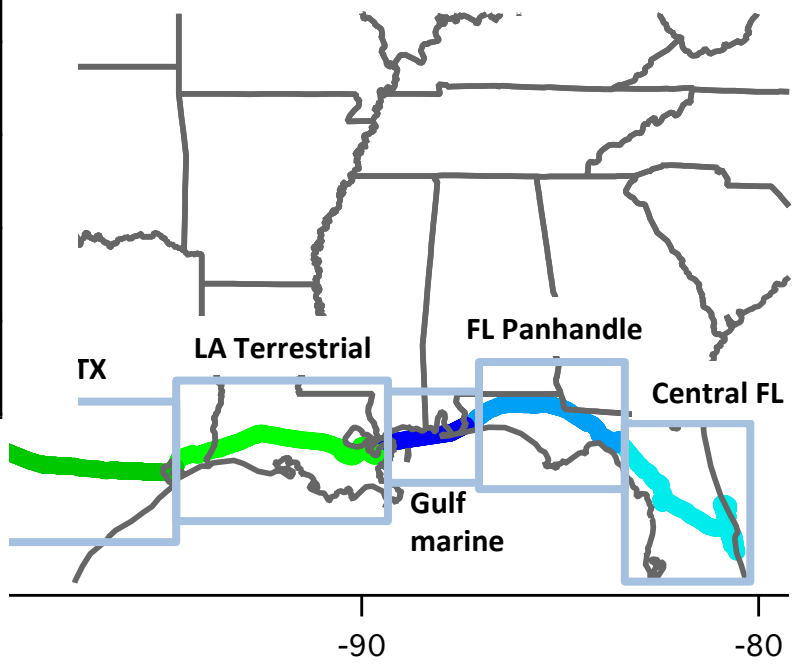
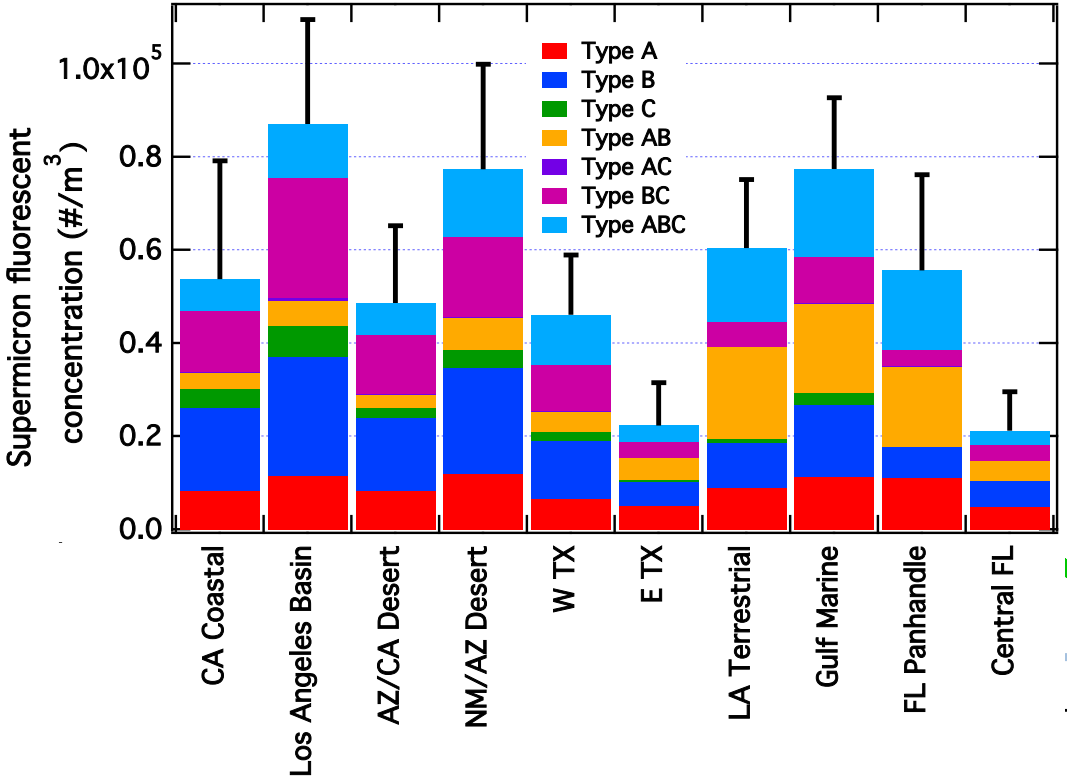
Fungal spores: 2-10 um

Pollen
> ~10 um

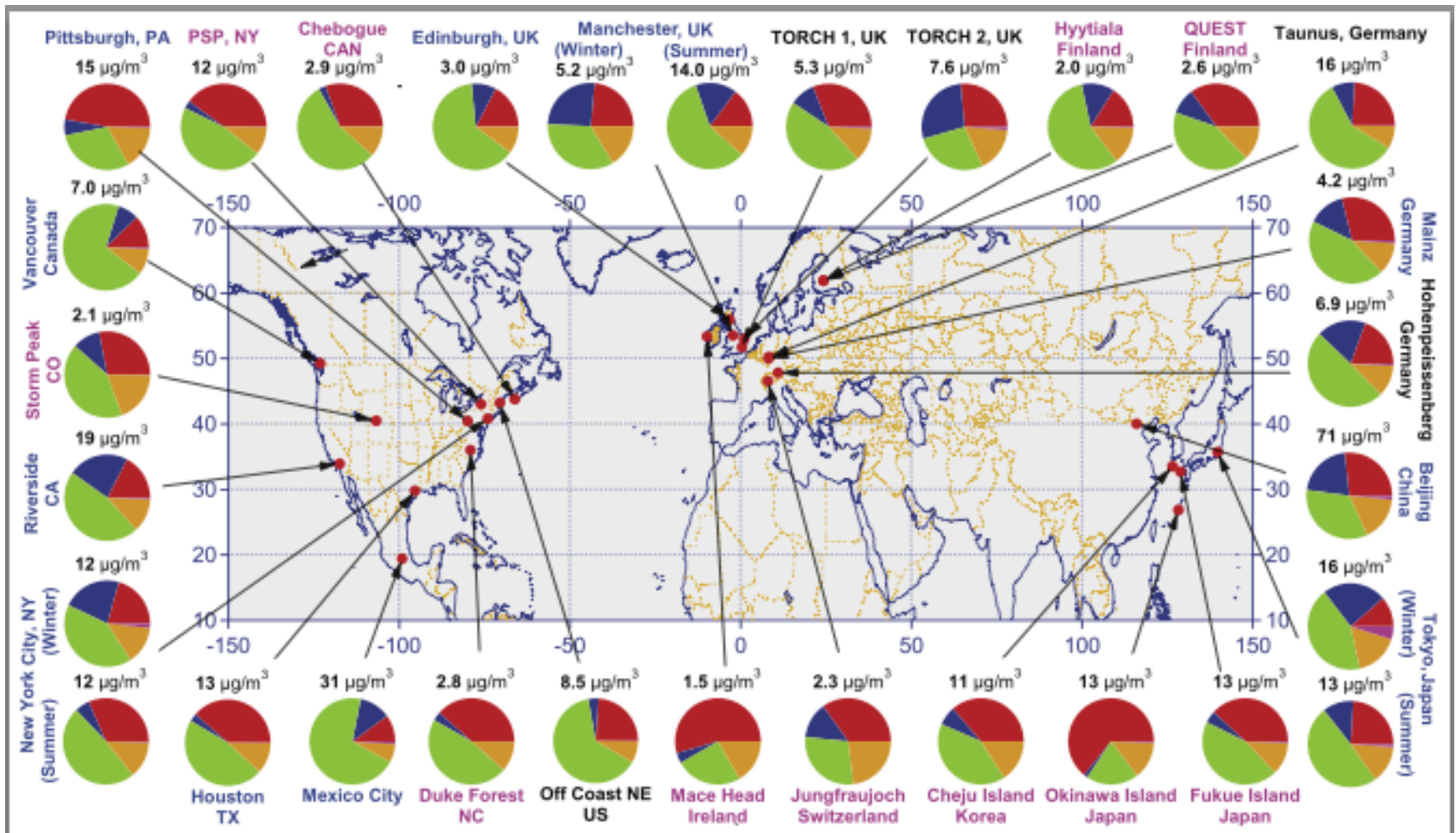


CloudLab

- ~1 month study from Sept – Oct 2013
- Most sampling at 300m, some excursions up to 1 km.
- In addition to the WIBS, the blimp was equipped with:
 - SP2 (Black carbon)

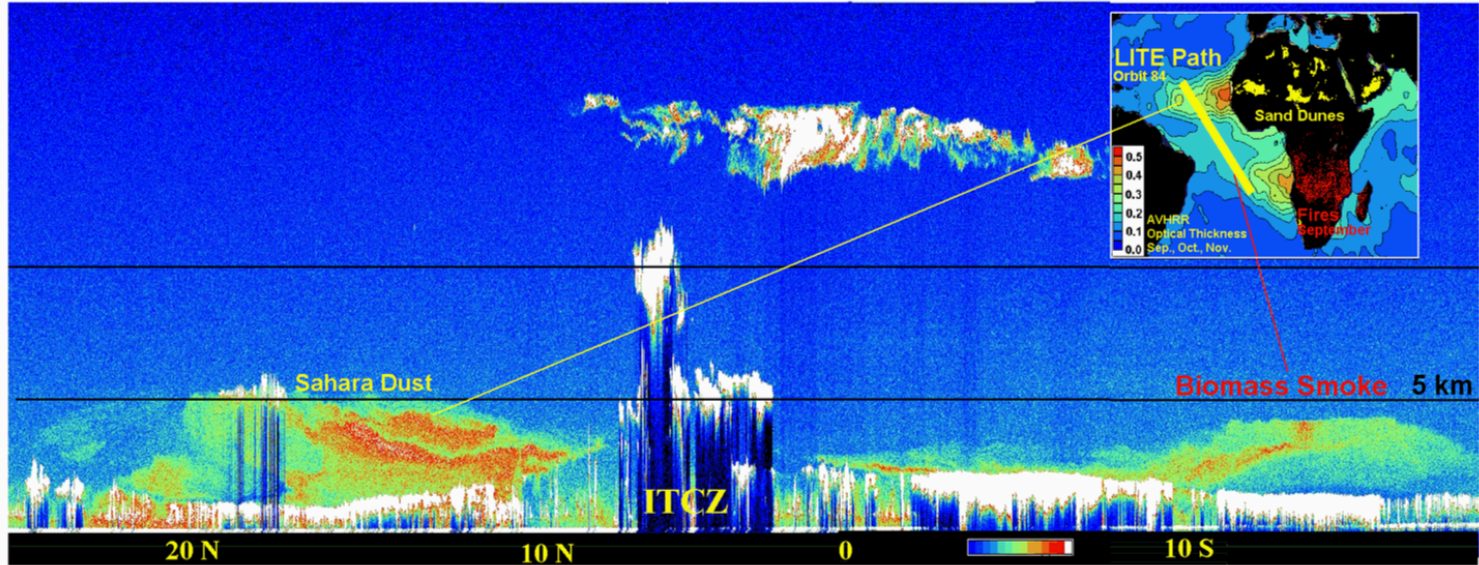


Global distribution of aerosol chemical composition

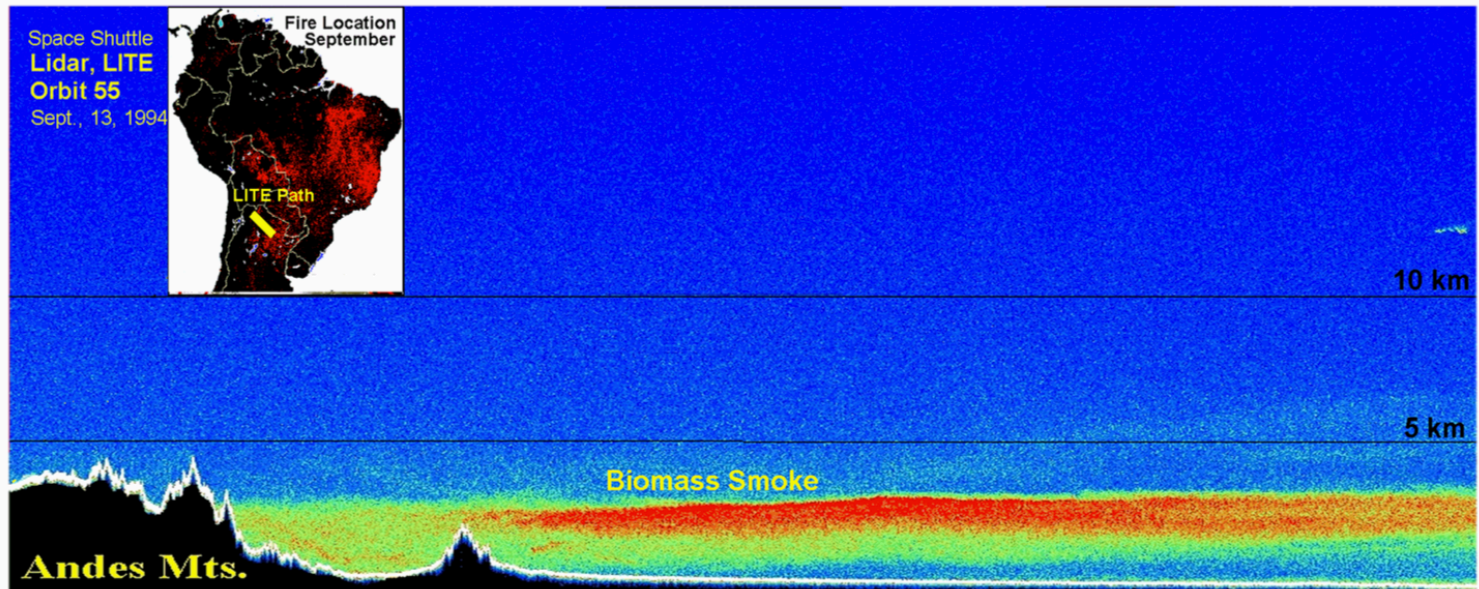


Zhang et al. 2007

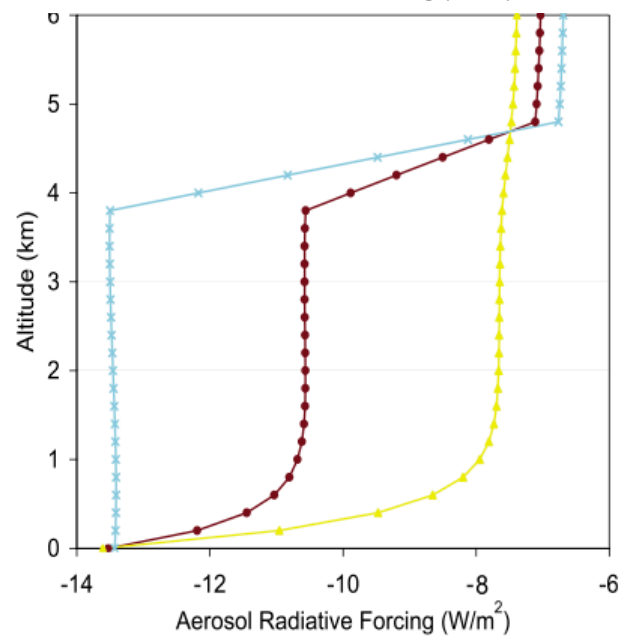
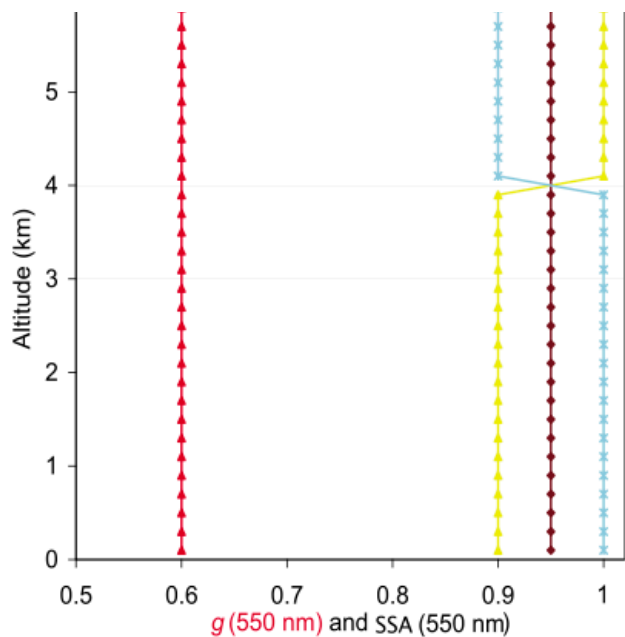
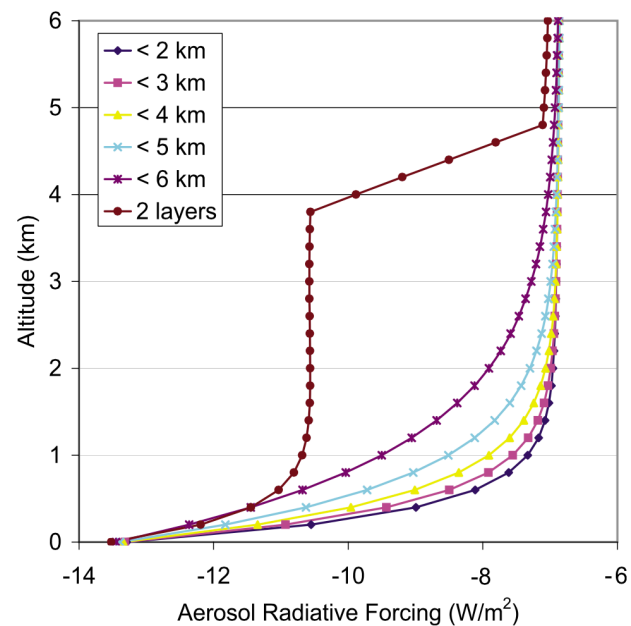
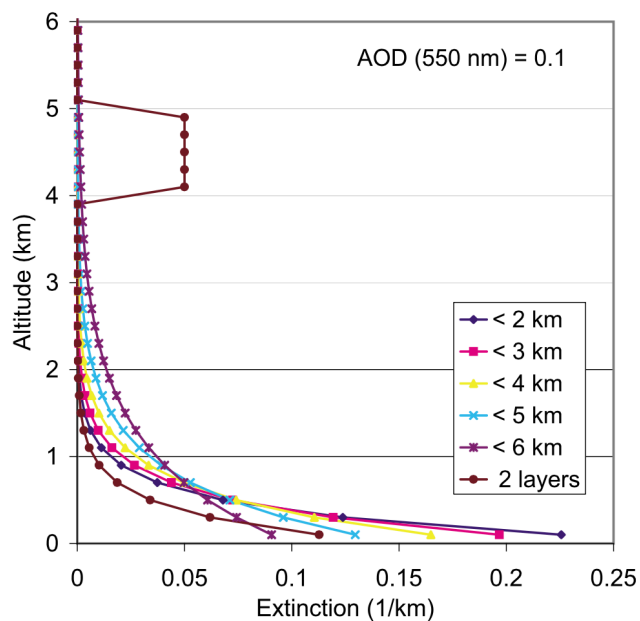
Sahara Dust and Biomass Smoke over the Atlantic



Biomass Smoke over South America



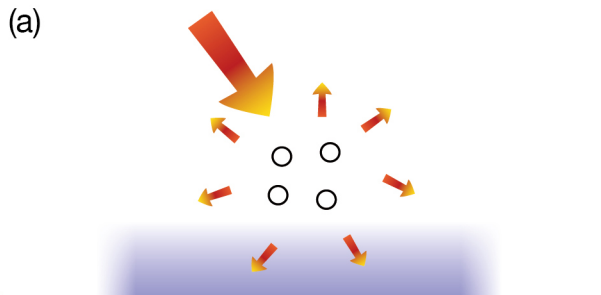
Vertical Distribution of Aerosol: Impacts on Radiative Forcing



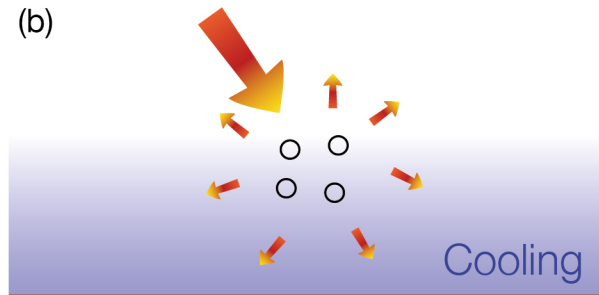
Aerosol-radiation interactions

Scattering aerosols

redistribution



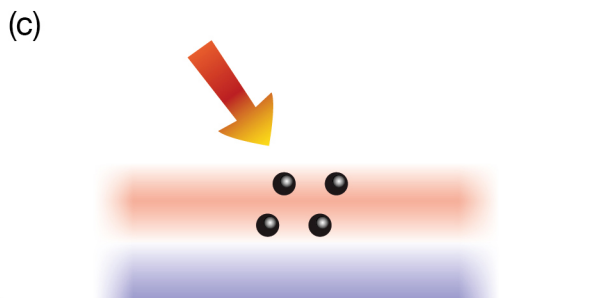
(a) Aerosols scatter solar radiation. Less solar radiation reaches the surface, which leads to a localised cooling.



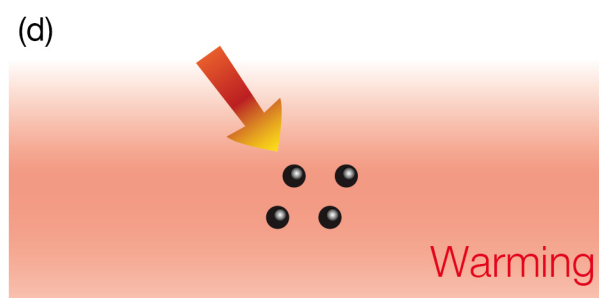
(b) The atmospheric circulation and mixing processes spread the cooling regionally and in the vertical.

Absorbing aerosols

extinction



(c) Aerosols absorb solar radiation. This heats the aerosol layer but the surface, which receives less solar radiation, can cool locally.



(d) At the larger scale there is a net warming of the surface and atmosphere because the atmospheric circulation and mixing processes redistribute the thermal energy.

Optical and Radiative Properties: Standard Long-Term Products

Measured quantities

σ_{sca} = scattering coefficient Mm^{-1}

σ_{abs} absorption coefficient Mm^{-1}

σ_{bsca} = backscattering coefficient Mm^{-1}

aerosol mass concentration (total/
speciated) $\mu\text{g m}^{-3}$

surface radiative fluxes W m^{-2}

Radiative Forcing

$$\text{RF} = (f_{\alpha} \downarrow - f_{\alpha} \uparrow) - (f_0 \downarrow - f_0 \uparrow) \text{ W m}^{-2}$$

$(f \downarrow - f \uparrow)$ = net irradiance (upwelling minus
downwelling flux)

f_{α} = flux with aerosol

f_0 = flux without aerosol

$$\text{RFE} = \text{RF} / \tau_{\alpha} \text{ W m}^{-2}$$

$$\tau_{\alpha} \sim Q_e$$

Derived quantities

extinction efficiency:

$$Q_e = \text{total mass} / (\sigma_{\text{sca}} + \sigma_{\text{abs}}) \text{ m}^2 \text{ g}^{-1}$$

single scattering albedo:

$$\omega_0 = \sigma_{\text{sca}} / (\sigma_{\text{sca}} + \sigma_{\text{abs}})$$

asymmetry parameter:

$$g = 1.011 - 1.036 \beta + 2.0059 \beta^2,$$

$$\beta = 0.0817 + 1.8495 \sigma_{\text{bsca}} - 2.9682 \sigma_{\text{bsca}}^2$$

scattering Ångström exponent:

$$\alpha_{\text{sca}} = \log(\sigma_{\text{sca} 450 \text{ nm}} / \sigma_{\text{sca} 700 \text{ nm}}) / \log(450/700)$$

absorption Ångström exponent:

$$\alpha_{\text{abs}} = \log(\sigma_{\text{abs} 450 \text{ nm}} / \sigma_{\text{abs} 700 \text{ nm}}) / \log(450/700)$$

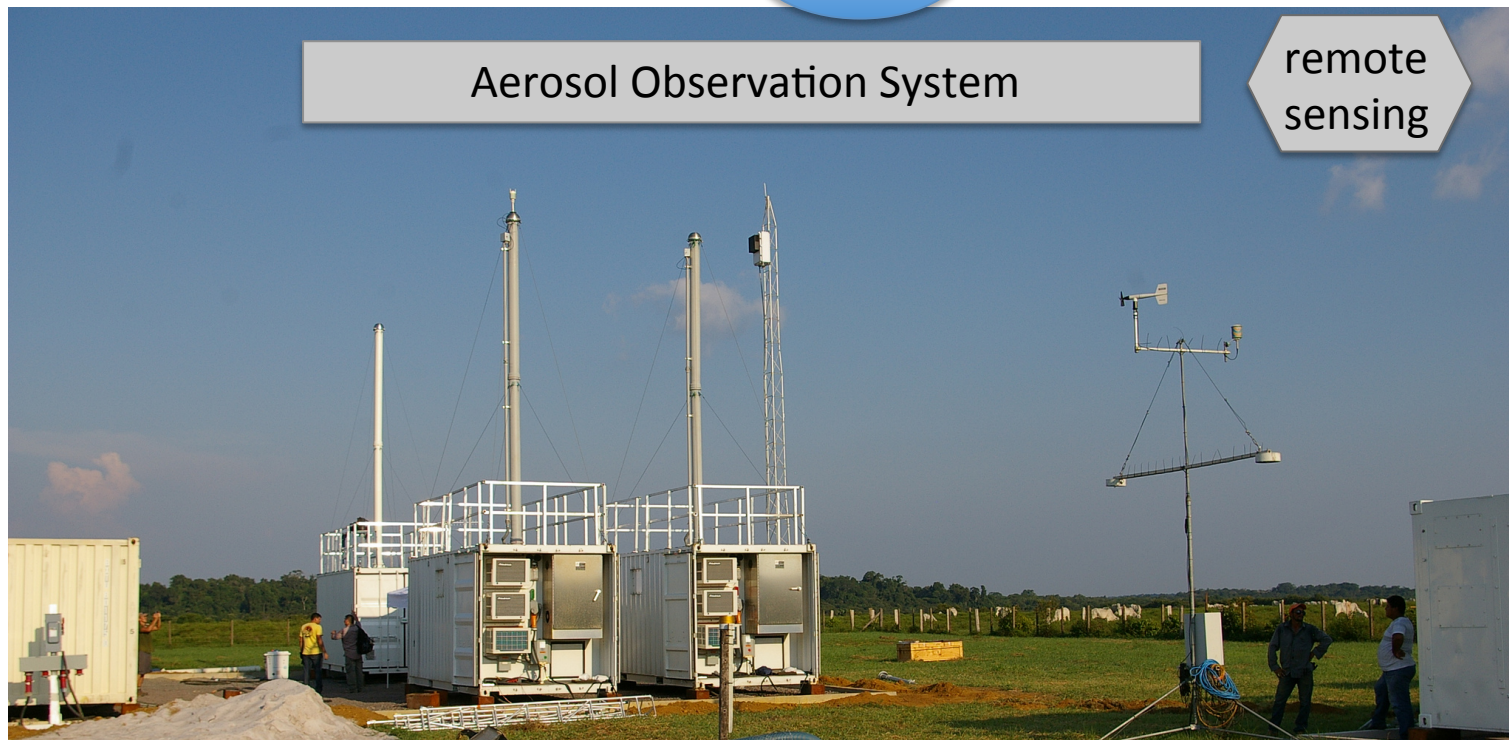
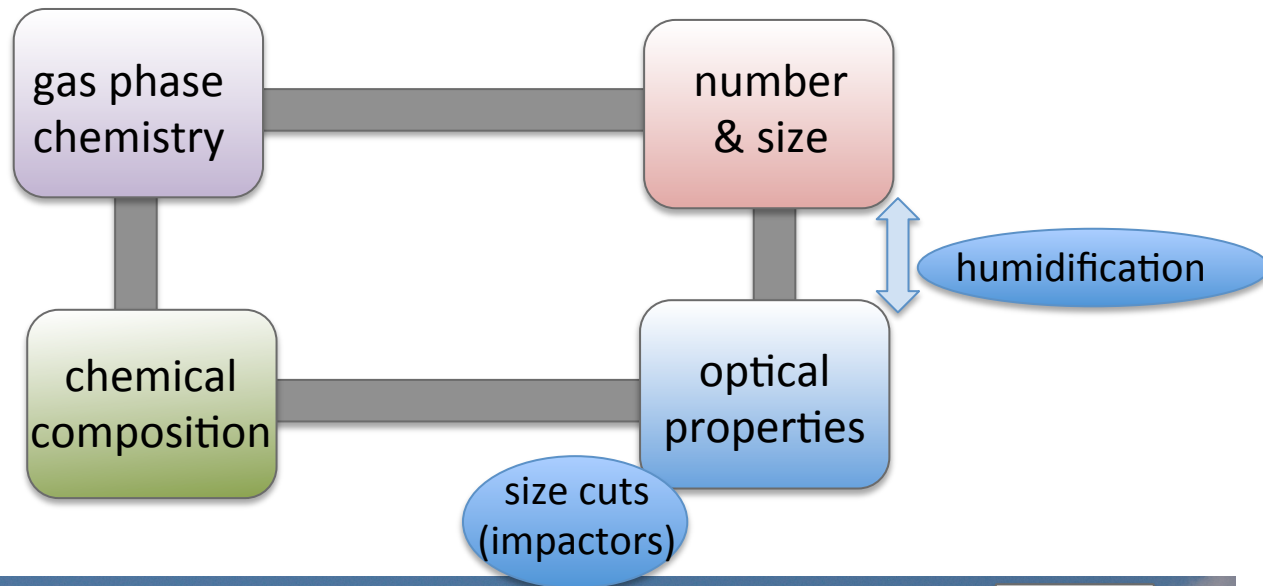
humidification factor:

$$\text{fRH} = \sigma_{\text{sca} 85\% \text{RH}} / \sigma_{\text{sca} 40\% \text{RH}}$$

sub-micron scattering fraction:

$$R_{\text{sca}} = \sigma_{\text{sca} 1 \mu\text{m}} / \sigma_{\text{sca} 10 \mu\text{m}}$$

ARM Aerosol Measurements



number & size

number conc

CPC 3772 (> 10 nm)

CPC 3776 (> 2.5 nm)

size distribution

50 nm – 1 μ m

UHSAS

size distribution

400 nm – 15 μ m

APS

CCN number conc.

spectrum with S

CCN-2

growth factor

HTDMA

optical properties

dry

light extinction

CAPS 3 λ

light scattering

nephelometer 3 λ

light absorption

PSAP (CLAP) 3 λ

humidified

light scattering

nephelometer +

humidograph 3 λ

aerosol chemical composition

non-refractory mass
concentration

(ToF-)ACSM

gas phase chemistry

CO

Los Gatos

CH₄, CO₂

Picarro

SO₂

ThermoScientific

NO_x/NO_y

NO_x Analyzer

O₃

Ozone Analyzer