

Global and Regional Aerosol Modeling

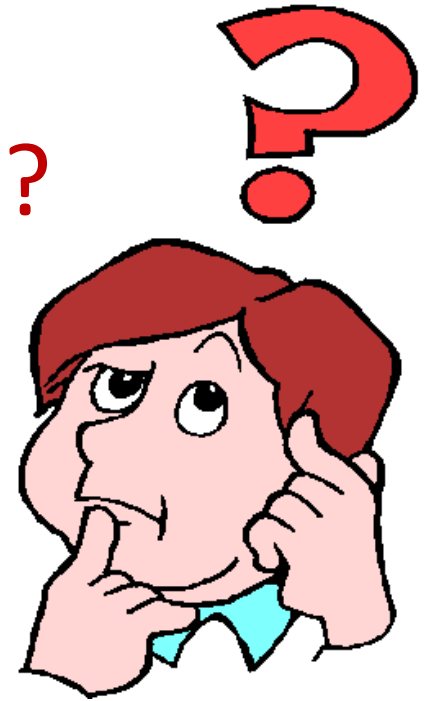
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- Utility of models
- Modeling concepts
- Aerosol representation in models
- Some results from aerosol simulations

All model are wrong!
Why do we need them?



All models are wrong!

Why do we need them?

- understand mechanisms, confirm understanding, generalize theory, analyze observations, guide experiments, predict changes,
- tools of synthesis of knowledge
- simplify the description of a complex system
- Account for feedback mechanisms.
- use a limited number of representative compounds or mixtures to describe the system's behavior
- Relatively simple parameterizations of processes can be used in global models of chemistry and climate.
- The accuracy of the models is evaluated by comparison to observations
- The different properties of atmospheric constituents need to be considered in the models: volatility, solubility, chemical reactivity, physical and optical properties

Surprises help learning and improving models.

Continuous dialogue

Integrated simplified view of the system

Heterogeneous/multiphase, k

γ

Modeling

$$-\frac{d[X]}{dt} = \frac{d[Y]}{dt} = k[X]$$

$$k = \omega \gamma A/4$$

$\gamma = f(V, T, \text{comp.}, \text{etc.})$
 $\gamma = f(H, D_1, k_1)$
Reconstruct γ from basic physicochemical parameters

Laboratory studies

Compute $A, V,$
composition, etc.
from basic data

Measure $A, V,$
composition,
phase, etc.

Field observation

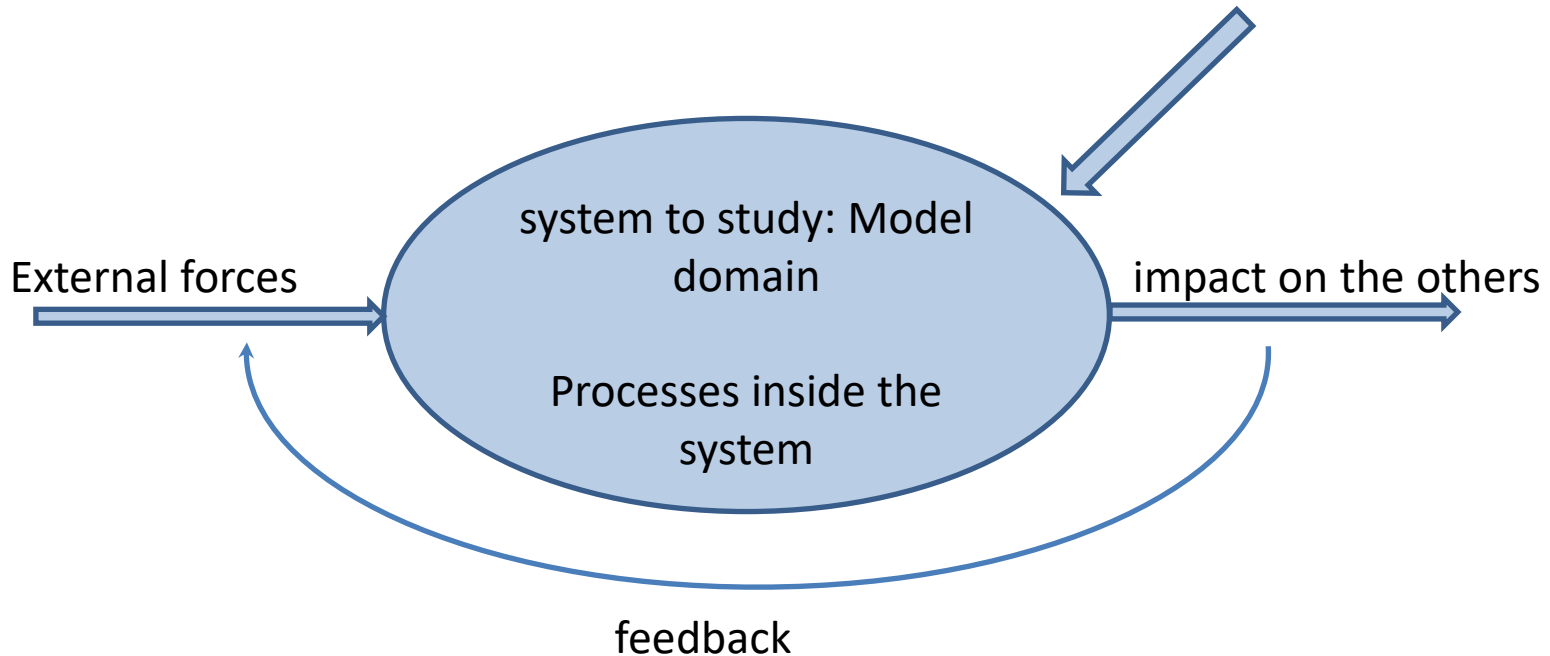
Ravishankara
Science,
1997

Controlled experiments for process understanding

Truth – benchmark for model results

To construct a model we need to:

Define the model domain and its boundaries



Main rule to follow → do not loose mass !

Mass balance (*also element balance*)

$$\int^{\infty} \frac{dc}{dt} = \left(\frac{\partial c}{\partial t} \right)_{transp} + \left(\frac{\partial c}{\partial t} \right)_{chem} + \left(\frac{\partial c}{\partial t} \right)_{emiss} + \left(\frac{\partial c}{\partial t} \right)_{dep} = 0$$

Energy balance!

Terminology

Technical

- Dimensions, grid resolution, geometry, sigma layers, nudging, sub-grid scale
 - Tracers, aerosol microphysics, time-step, parameter splitting

Scientific

- Chemistry-Transport Models (CTMs)
- General Circulation Models (GCMs)
- Coupled models –Earth System Models (ESMs)
 - Components (atmosphere, biosphere, lithosphere, cryosphere, etc)

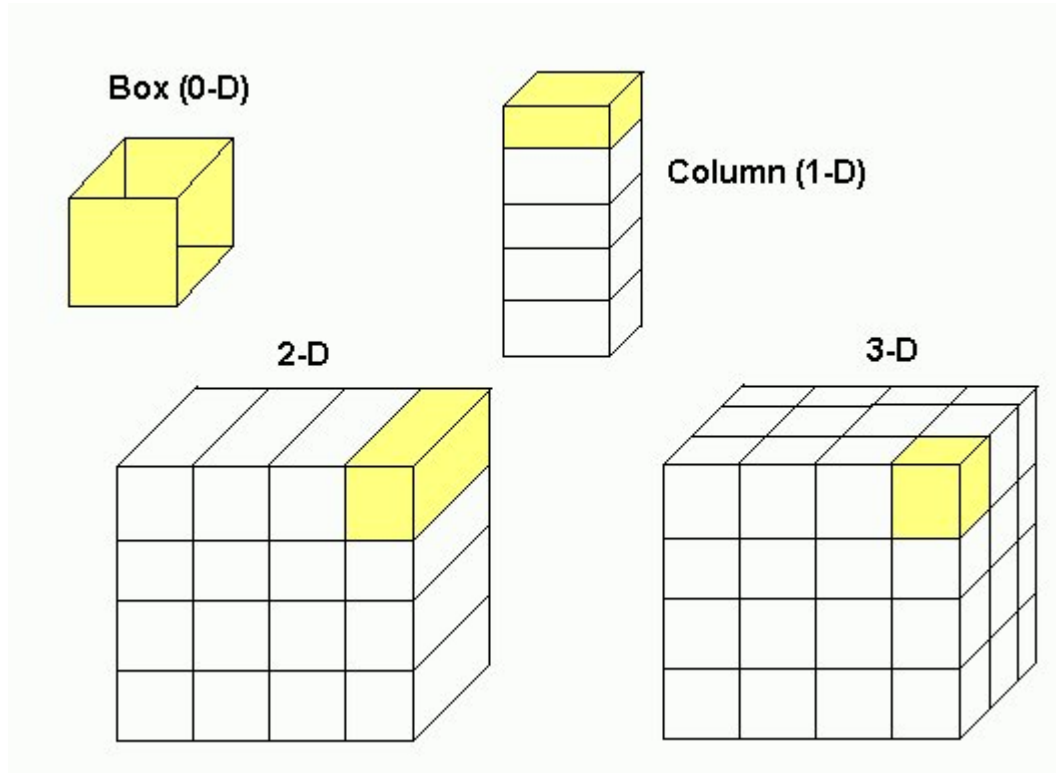
Processes

- emissions, transport, chemistry, radiation, deposition etc

Results

- diagnostic and prognostic

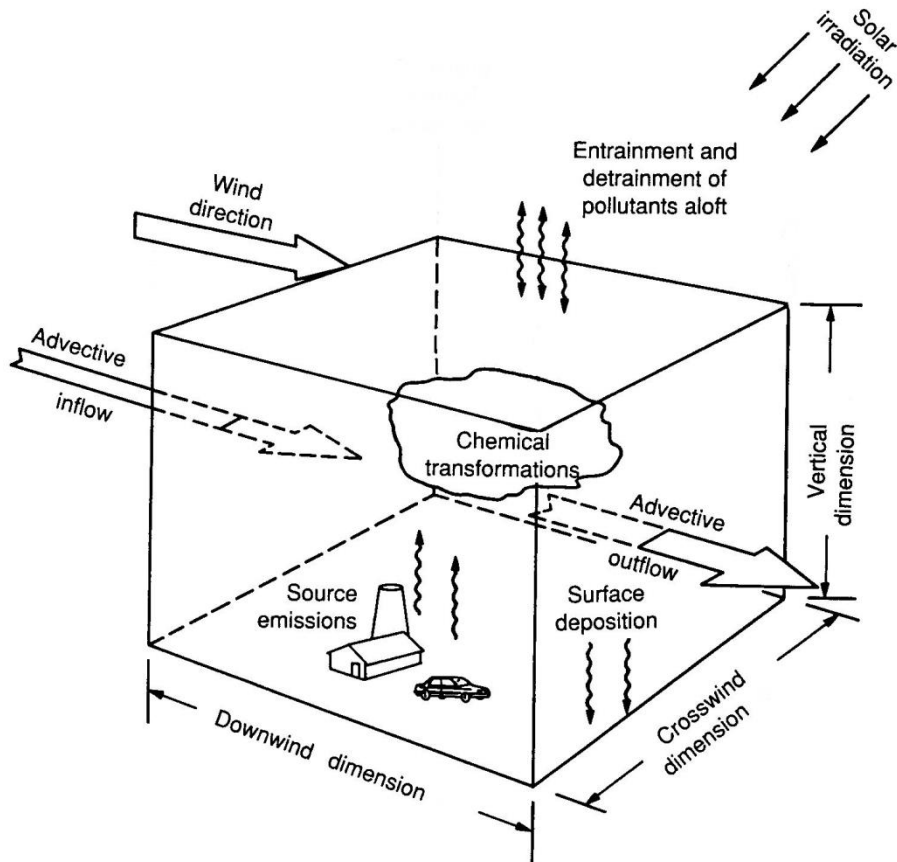
Model dimensions



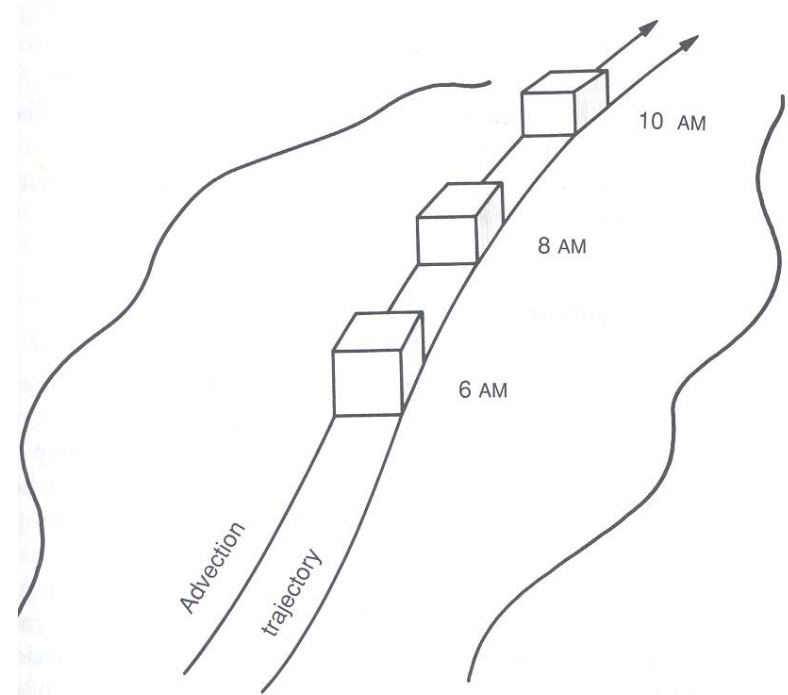
<http://irina.eas.gatech.edu/lectures/Lec29.htm>

Box (or 0D) models

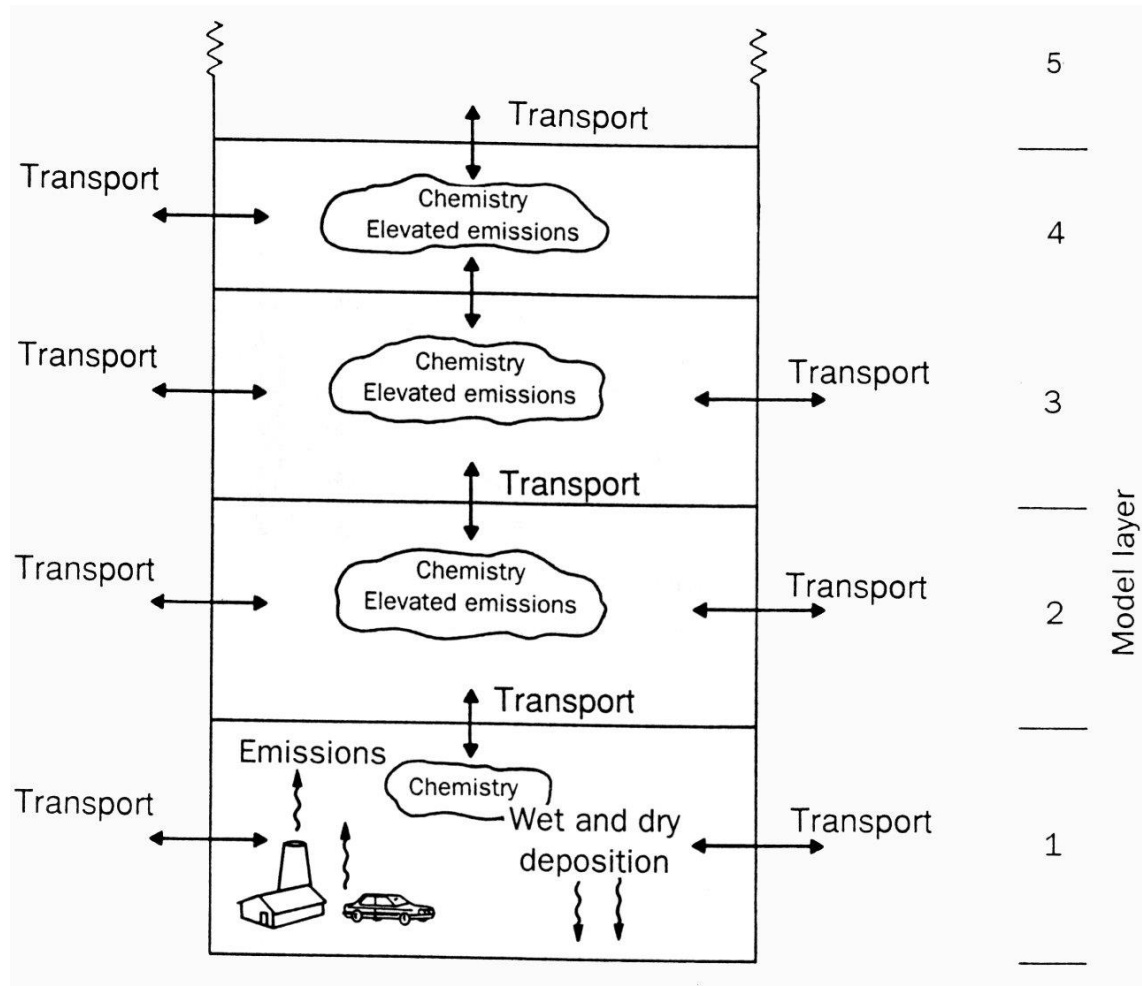
Eulerian



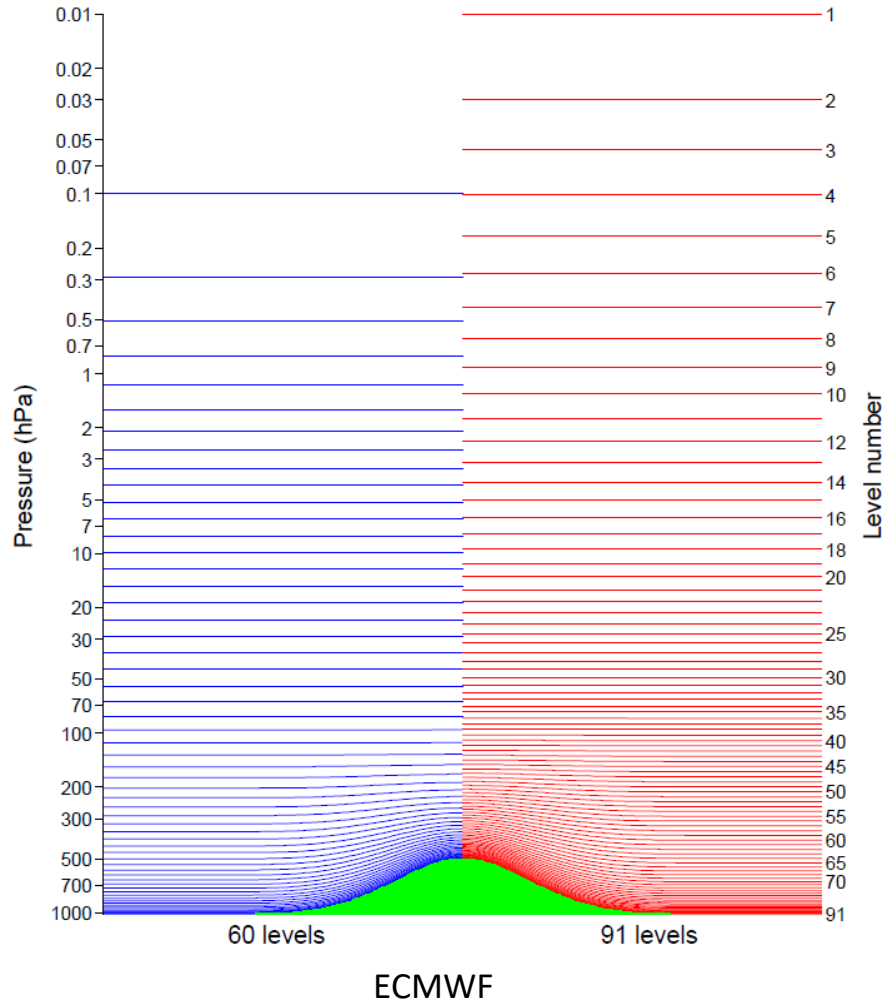
Lagrangian



Column (or 1D) models

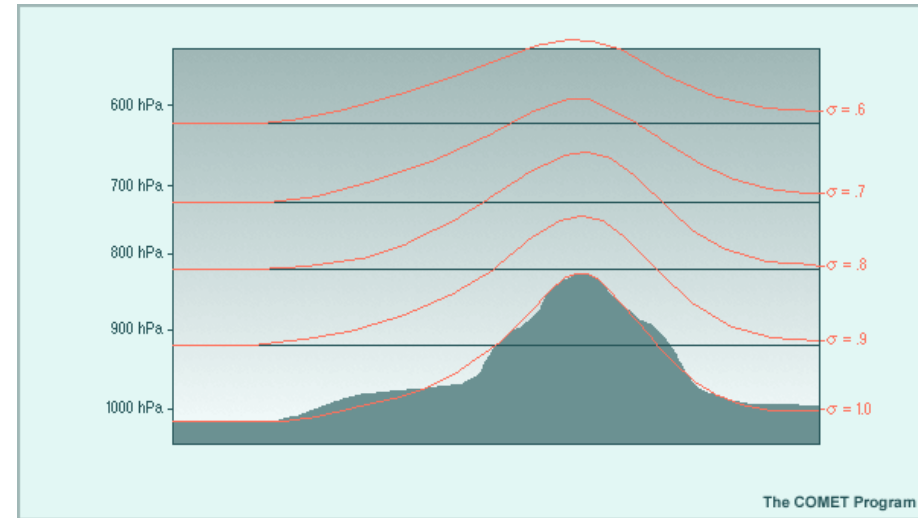


Vertical resolution – sigma levels



$$p_{k+1/2} = A_{k+1/2} + B_{k+1/2} p_{\text{surf}} \quad 0 \leq k \leq NLEV$$

$$p_k = \frac{1}{2} (p_{k-1/2} + p_{k+1/2}) \quad 1 \leq k \leq NLEV$$

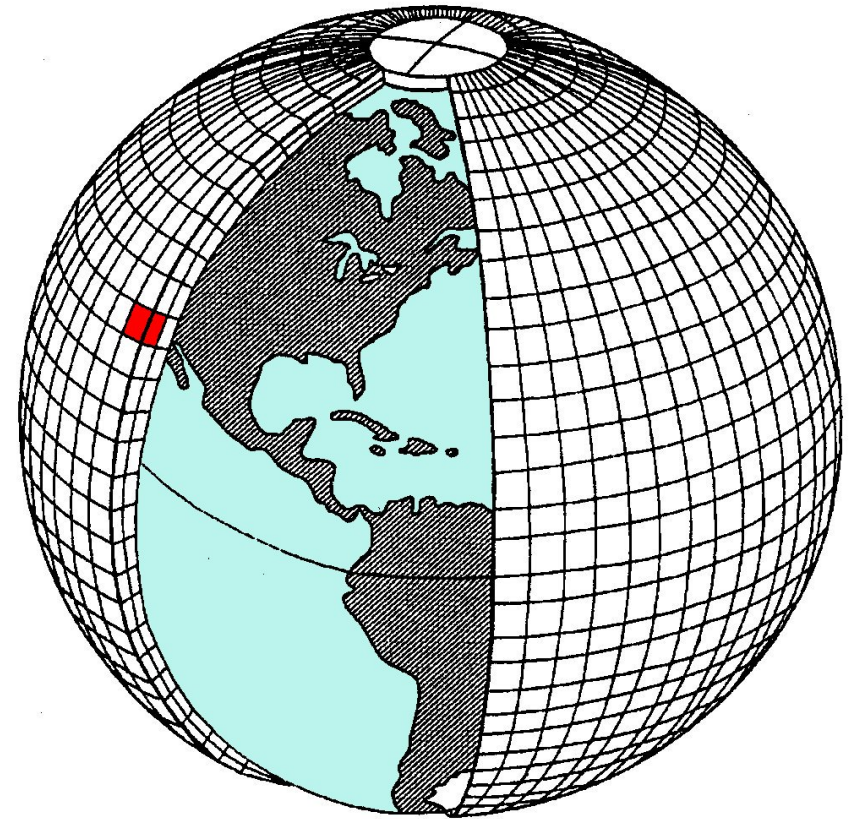
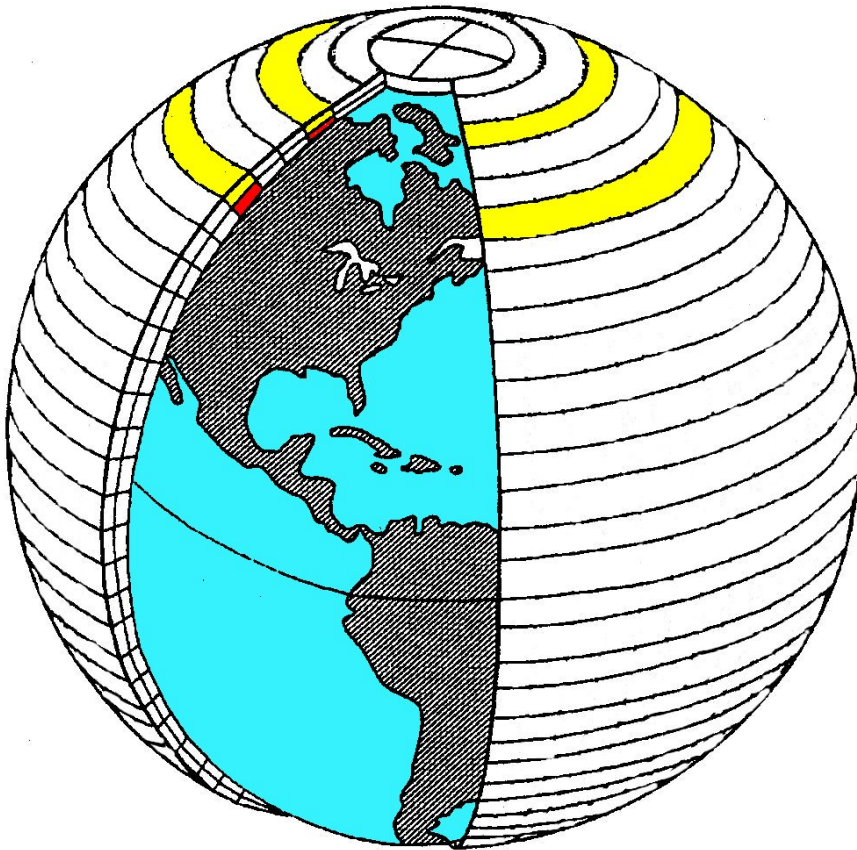


<http://atmo.tamu.edu/class/metr452/models/2001/sigma.png>

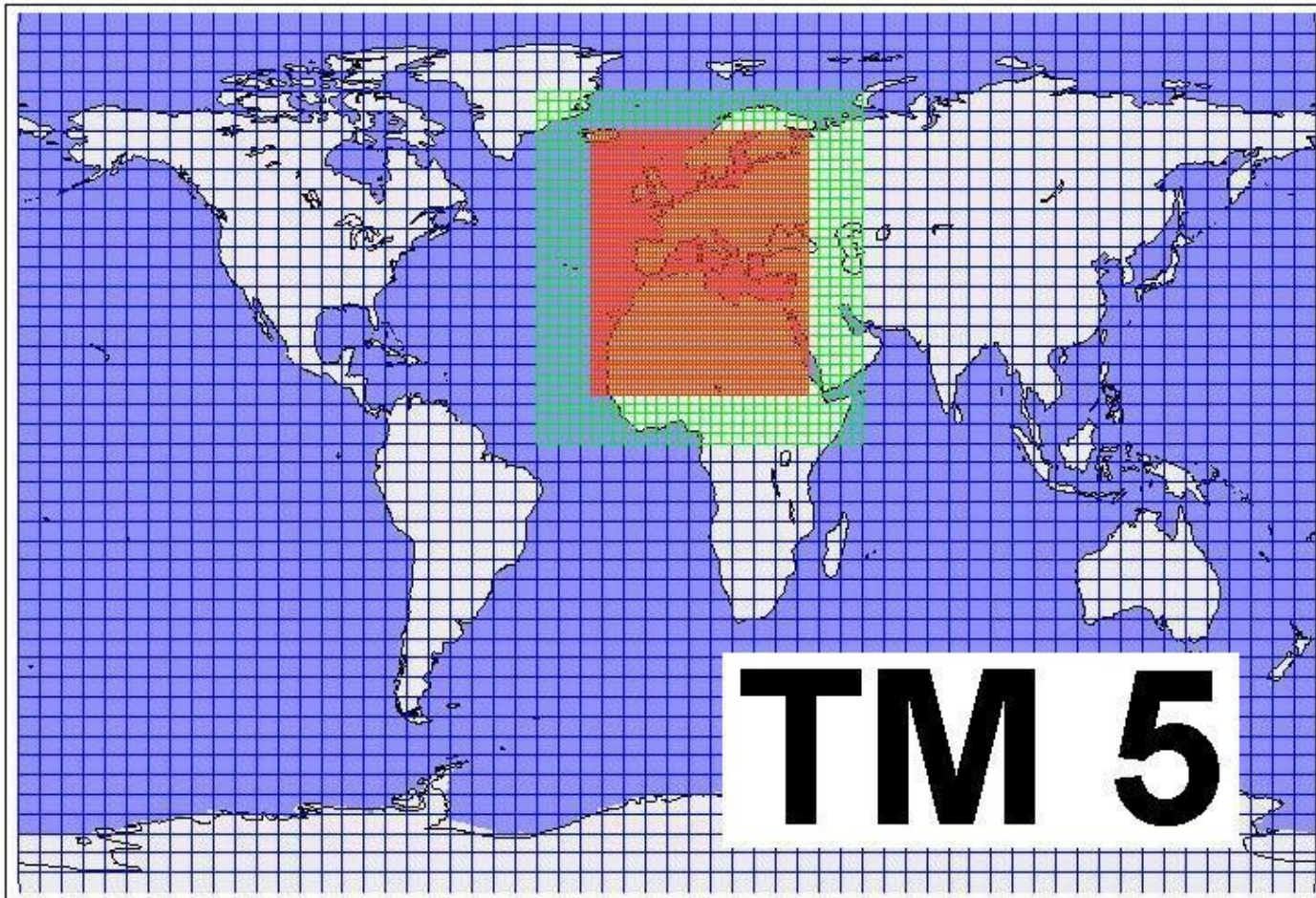
2D

Global models

3D



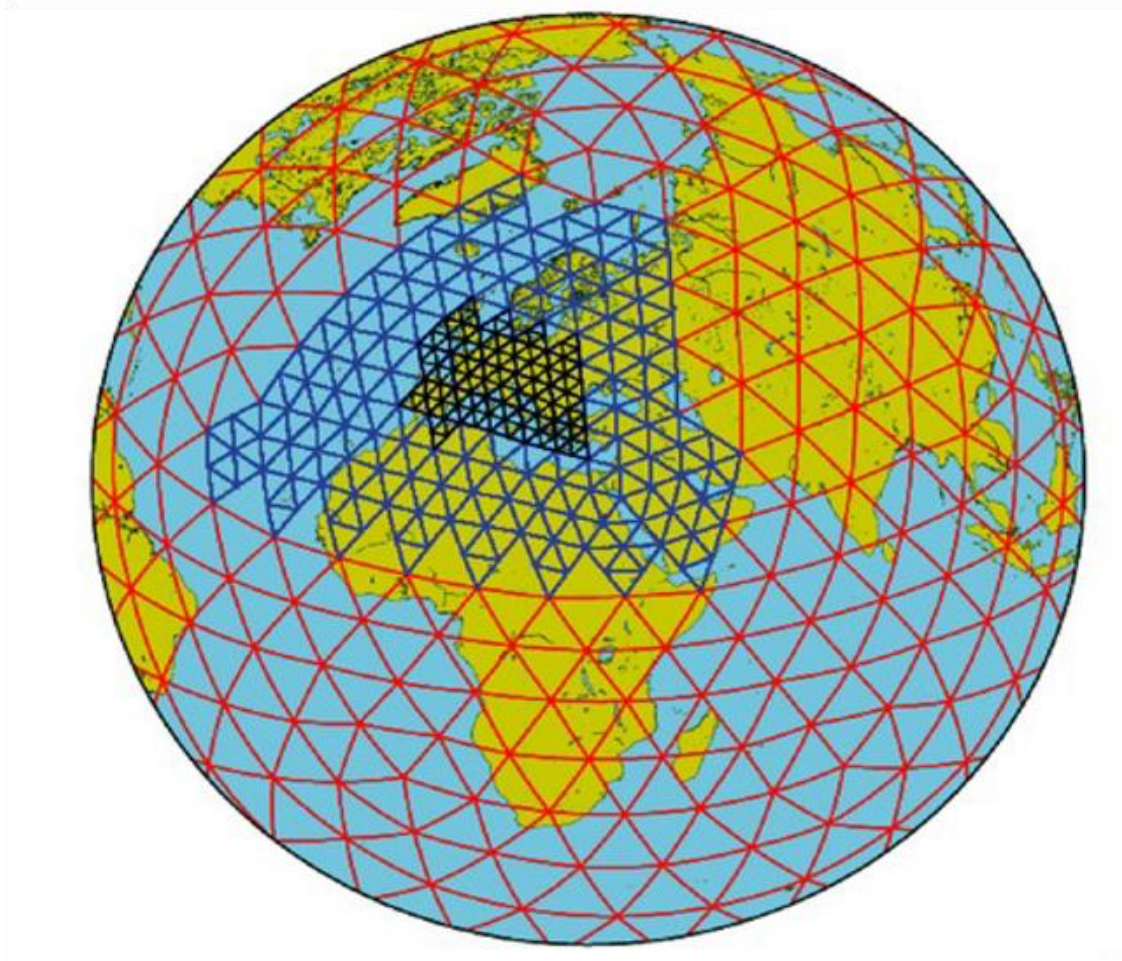
Nested global models



<http://www.phys.uu.nl/~tm5/>

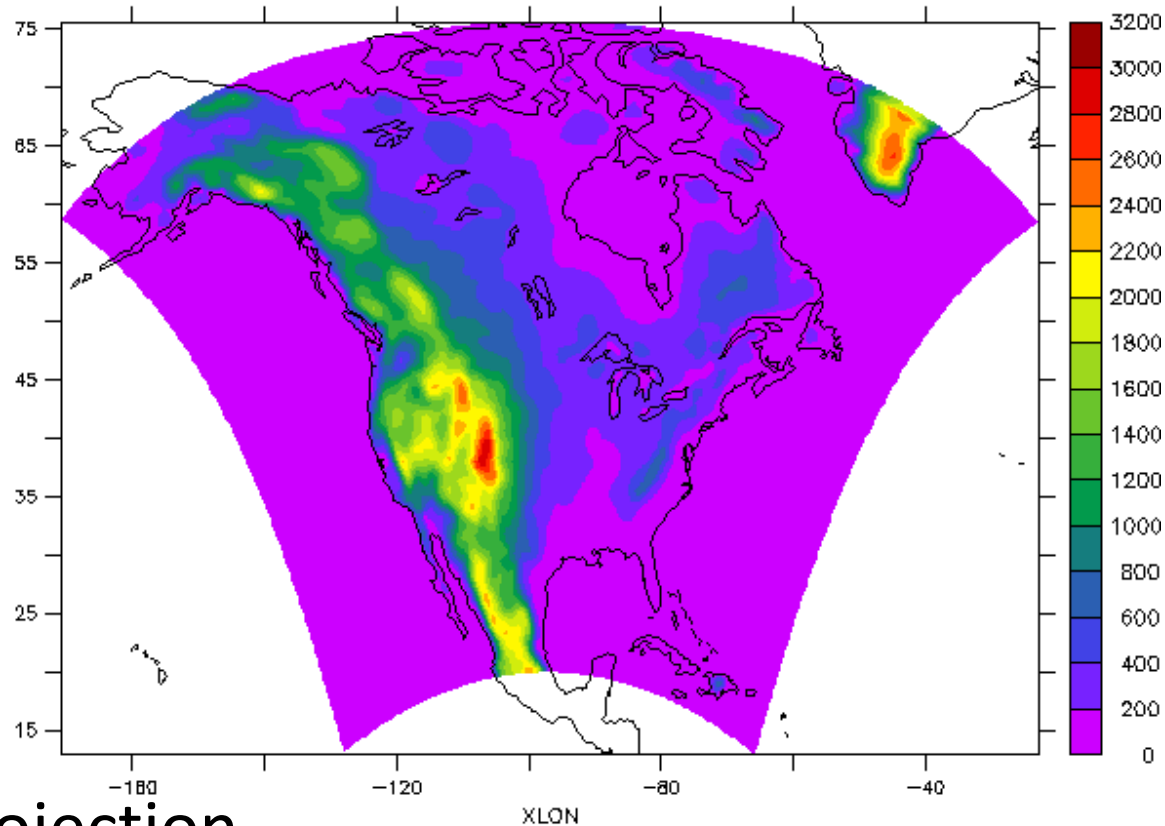
Emerging geometries

ICON (Icosahedral Nonhydrostatic) Model



https://www.meteo.physik.uni-muenchen.de/methoden/numerische_simulationen/index.html

Regional models

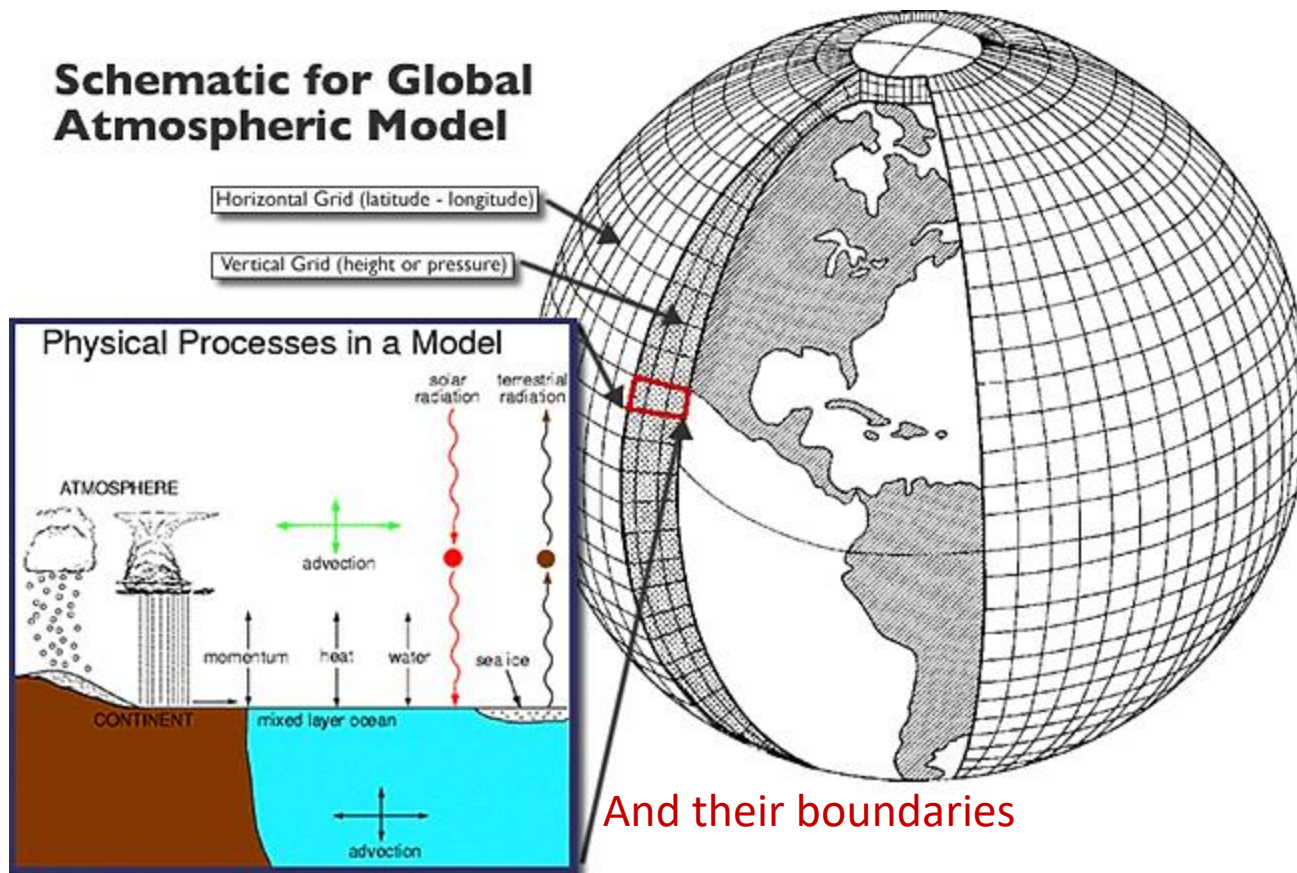


Lambert projection

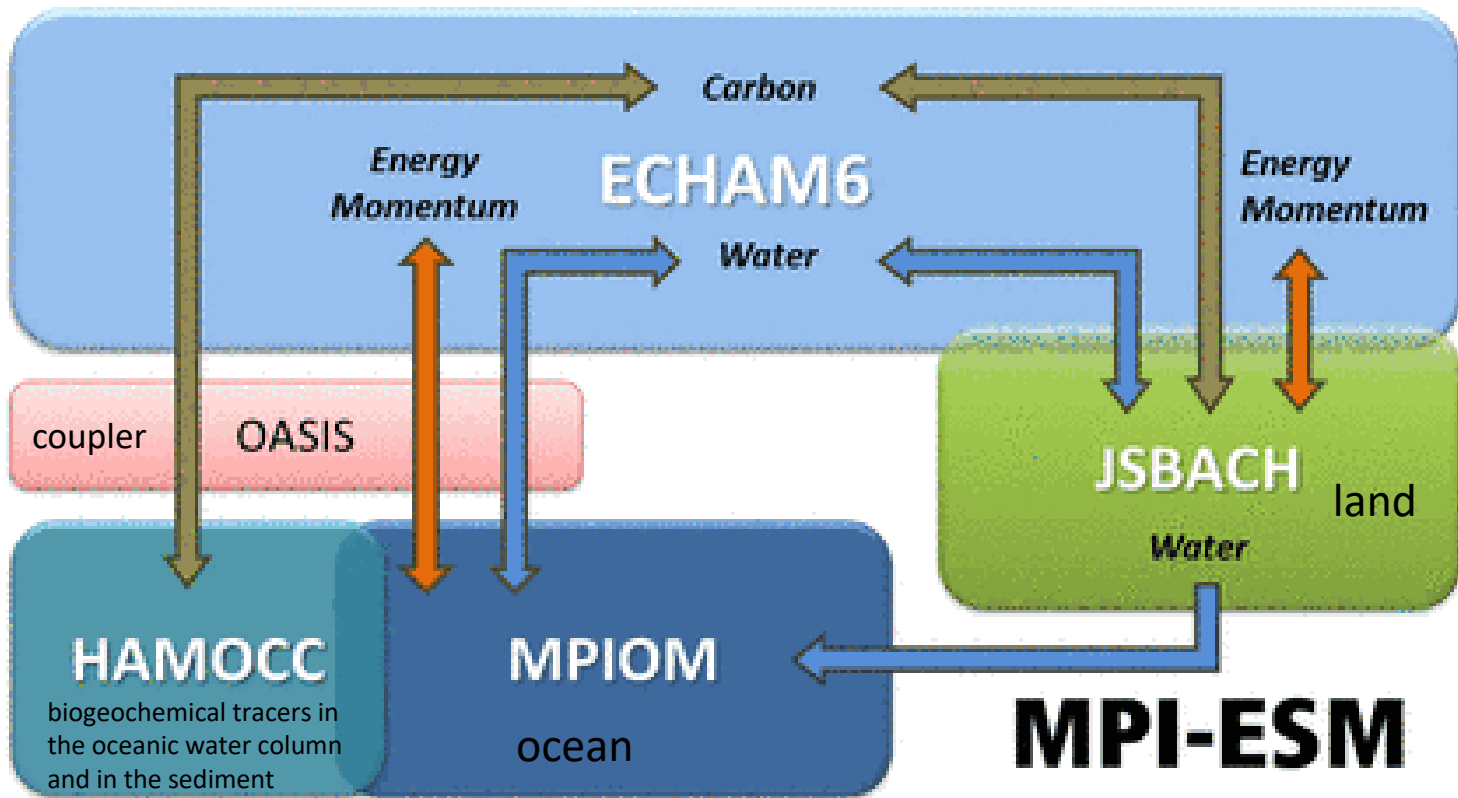
Grids in Degrees or km^{HT} → different projections

WRF – http://www.unidata.ucar.edu/newsletter/2006nov/Domain_narccap.gif

Global atmospheric models

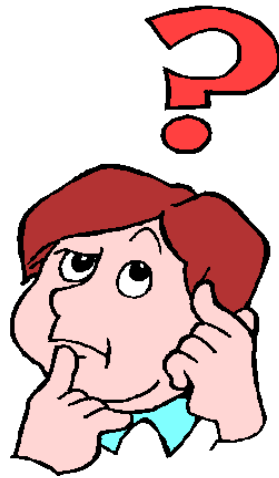


http://upload.wikimedia.org/wikipedia/commons/4/4a/Global_Atmospheric_Model.jpg

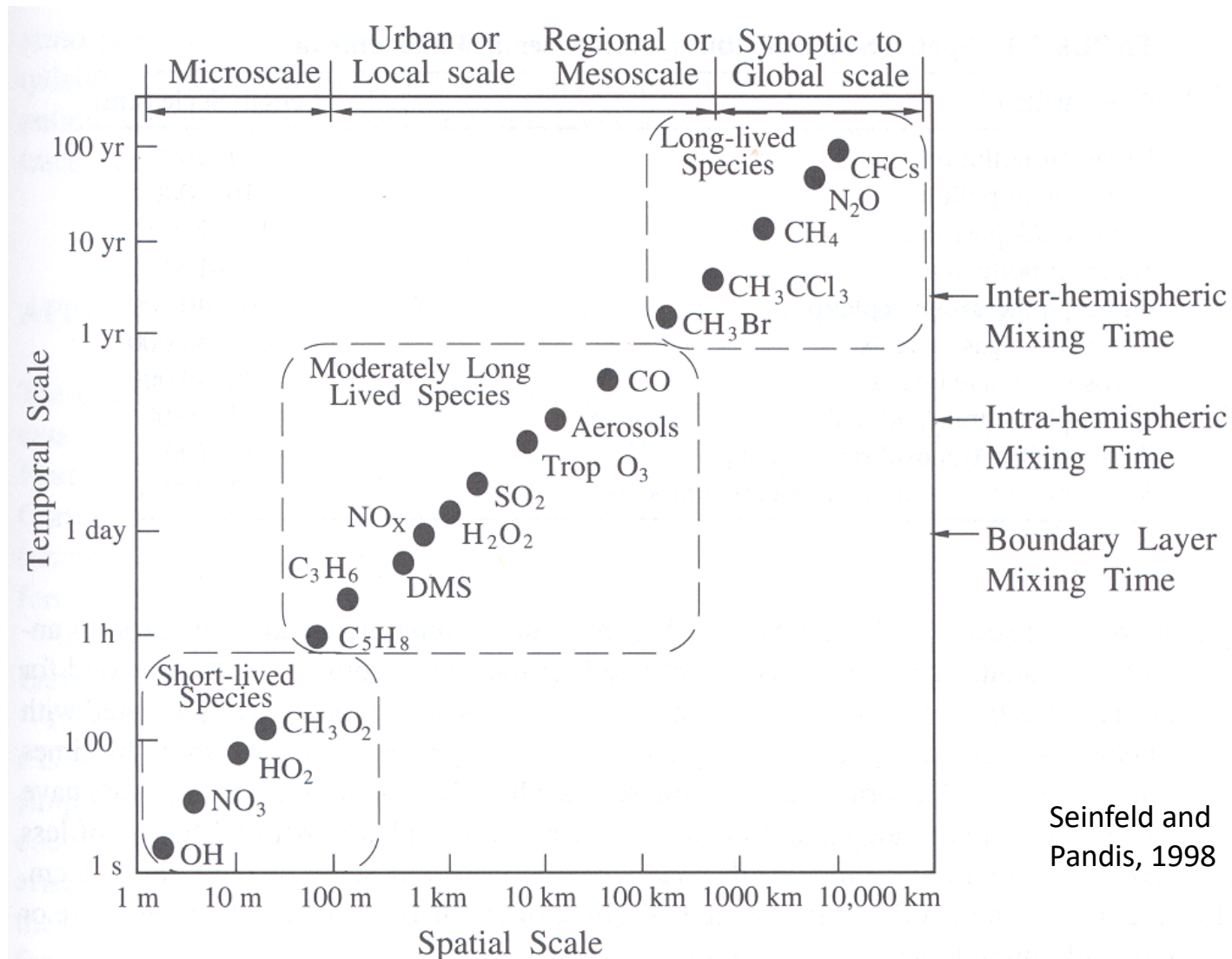


<http://www.mpimet.mpg.de/en/science/models/mpi-esm/>

Which is the best model to use?



Lifetime of studied pollutant and space-geographic extent of the model



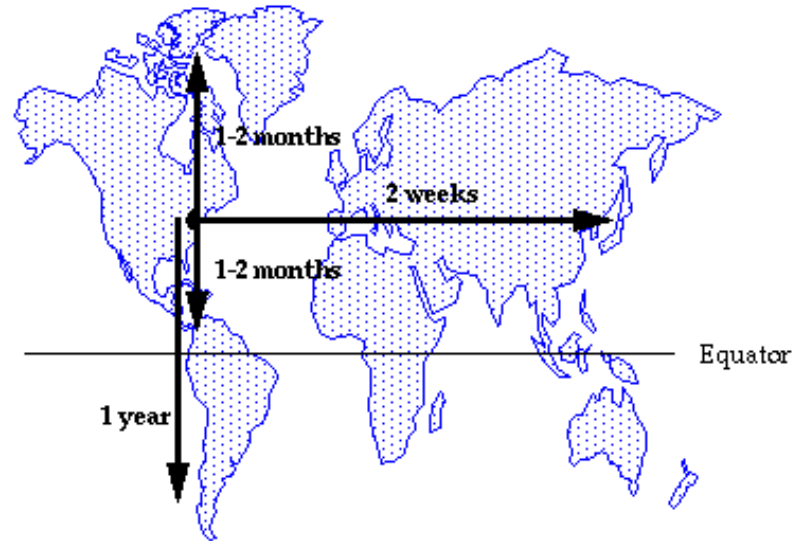
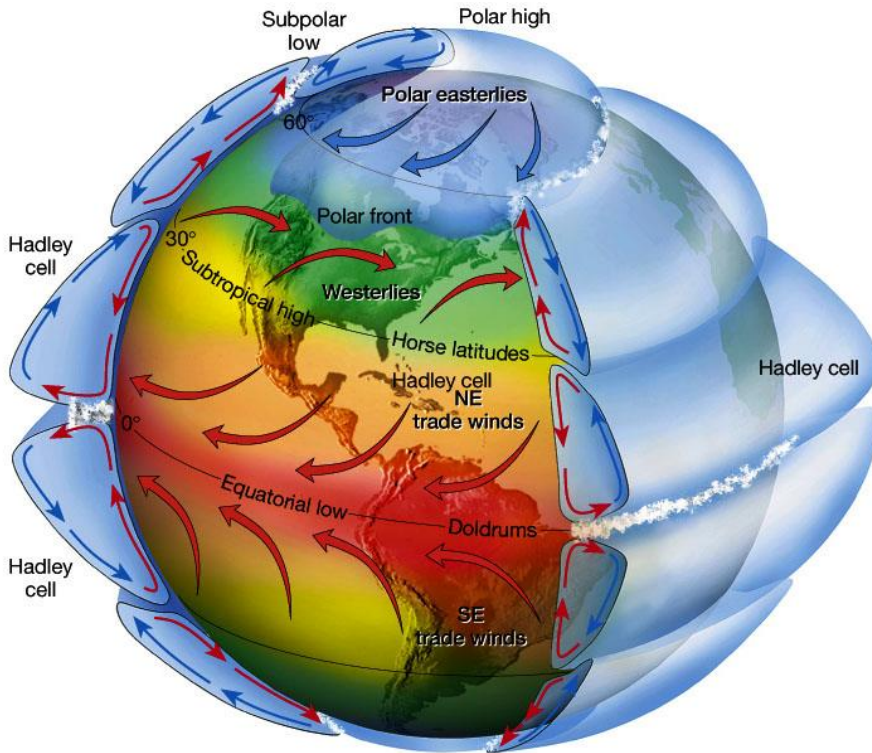
Seinfeld and Pandis, 1998

FIGURE 1.17 Spatial and temporal scales of variability for atmospheric constituents.

Atmospheric transport

Advection, deep and shallow convection

transport time scales for advection



http://www.ux1.eiu.edu/~cfjps/1400/FIG07_006.jpg

Jacob, 1999

Issues with mesoscale modeling

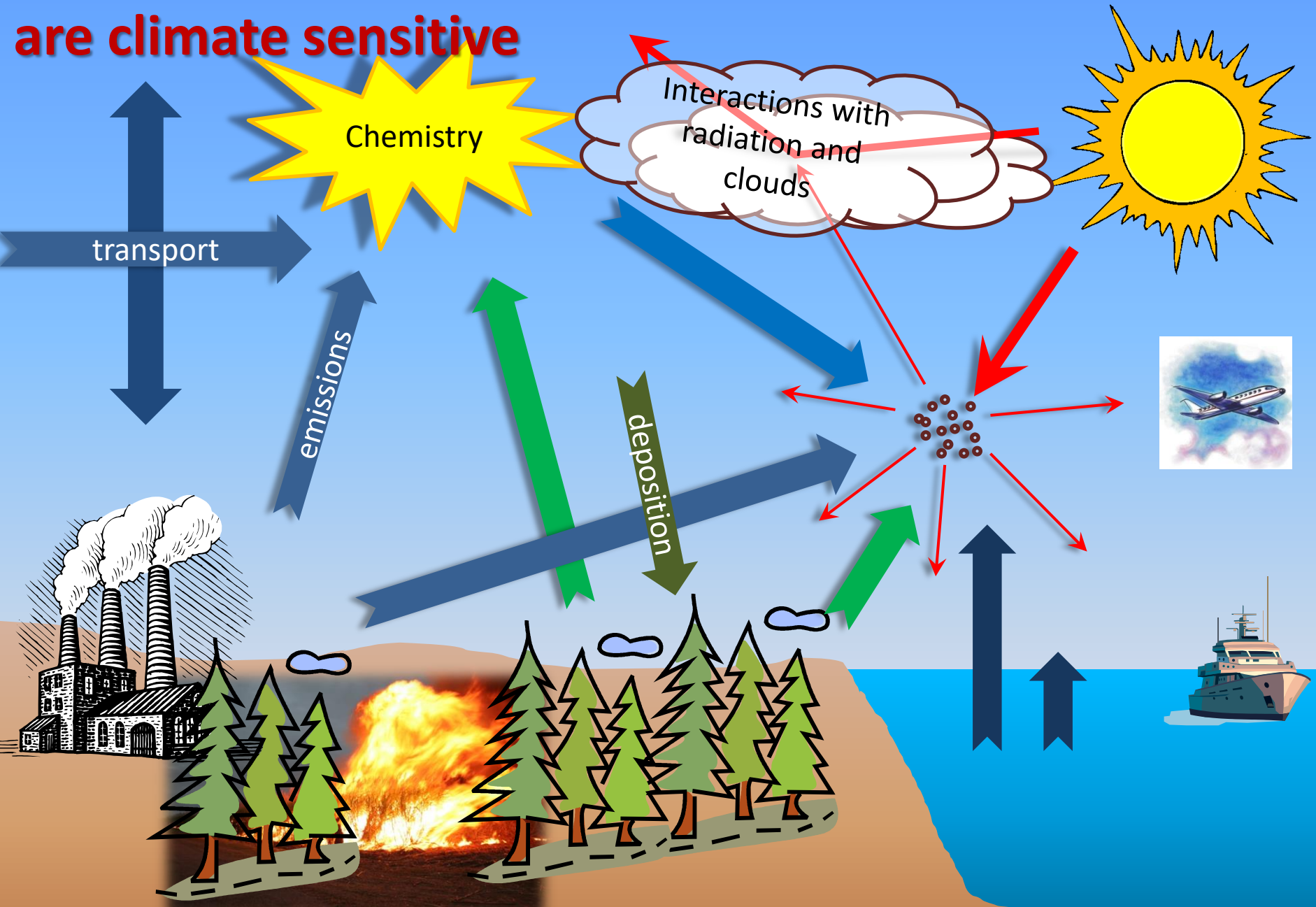


- Upper boundary conditions
- Lateral conditions
- Influx & outflux

(to represent sources or sinks that are not in the model domain)

- Time resolution

Processes affecting atmospheric composition – some are climate sensitive



AEROSOL SOURCES

- Emissions (primary aerosols)

	Over land	Over ocean
Anthropogenic*	Fossil Fuel (FF), Bio Fuel (BF), Biomass Burning (BB), dust (resuspension)	Shipping
Natural	BB, bioaerosols, dust, volcanoes	Sea-spray

- Chemistry (secondary aerosols, precursor emissions)

	Over land	Over ocean
Anthropogenic*	NO _x , SO ₂ , NH ₃ , Aromatics, Alkanes, etc. From FF, BF, BB, agricult.	NO _x , SO ₂ , NH ₃ , Aromatics, Alkanes, etc Shipping
Natural	Isoprene, Terpenes, OVOC From vegetation, soils	DMS, Isoprene, Terpenes OVOC

* Main activity sectors transport, energy, agriculture, industry

Table 2.1 Approximative emission fluxes from different types of primary aerosols and gaseous precursors of secondary aerosols. The climate importance of aerosols depends not only on the strength of their emissions, but also on their physical and chemical properties. Estimates are compiled from Penner et al. (2001), Dentener et al. (2006), Guenther et al. (1995), Jaenicke (2005), Burrows et al. (2009), Heald and Spracklen (2009). Tg = 10^{12} g = 1 million of tons. Gg = 10^9 g = 1 thousand of tons

AEROSOL SOURCES

Aerosol type	Emission flux (per year)
<i>Natural primary aerosols</i>	NATURAL
Desert dust	1000–3000 Tg
Sea spray	1000–6000 Tg
Biomass burning aerosols	20–35 Tg
Terrestrial primary biogenic aerosols	Order of 1000 Tg
Including bacteria	40–1800 Gg
Including spores	30 Tg
<i>Precursors of natural secondary aerosols</i>	
Dimethylsulphide (DMS)	20–40 Tg S
Volcanic SO ₂	6–20 Tg S
Terpenes , isoprene , ORVOC	40–1200 Tg

ANTHROPOGENIC

<i>Anthropogenic primary aerosols</i>	
Industrial dust	40–130 Tg
Biomass burning aerosols	50–90 Tg
Black carbon (from fossil fuel)	6–10 Tg
Organic carbon (from fossil fuel)	20–30 Tg
<i>Anthropogenic secondary aerosols</i>	
SO ₂	70–90 Tg S
Volatile organic compounds (VOCs)	100–560 Tg C
NH ₃	20–50 Tg N
NO _x	30–40 Tg N

Modified from Boucher, Atmospheric Aerosols, Springer, 2015

ISBN 978-94-017-9648-4

DOI 10.1007/978-94-017-9649-1

C carbon, S sulphur, N nitrogen

Which emissions can be calculated on line by a chemistry-climate model?



Interactive emissions

- Dust
- Sea-spray (sea-salt and organics)
- Fires
- Bioaerosols
- Biogenic VOC (MEGAN model)

databases

- Anthropogenic emissions
- Emissions from soils
- Volcanoes

1. Selection of emissions inventories
2. Time resolution typically very poor
3. Seldom updates

Aerosol emissions from arid soils

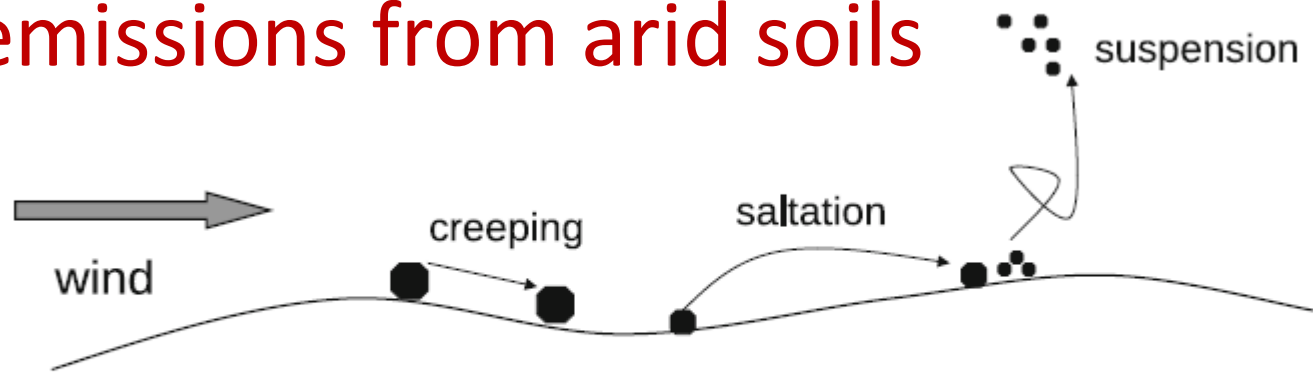


Fig. 4.2 Schematic description of the source mechanism for desert dust particles: creeping of the bigger particles which are rolling along the ground, saltation of the particles small enough to be lifted by the wind before falling, bouncing against and breaking up some of the soil aggregates into smaller particles, and suspension of the smallest particles (Boucher 2015)

Controlled by

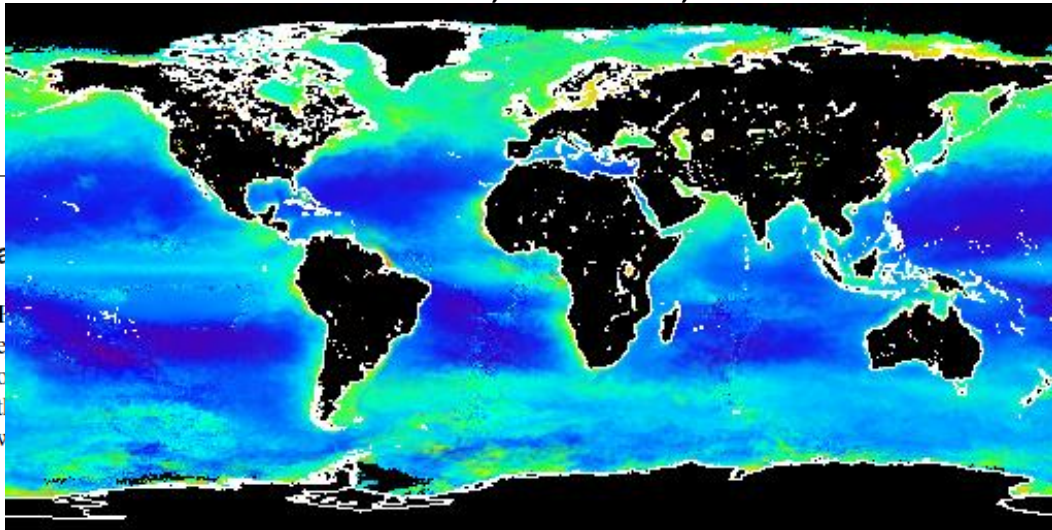
- quality of the soil (minerals, size of grains, soil moisture)
- wind speed (above friction velocity)

Examples in : Tegen et al, 2009 & Astitha et al. *Atmos. Chem. Phys.*, 12, 11057–11083, 2012



Sea-spray organic enrichment

Chl, SeaWiFS, 2000



O'Dowd et al., Nature, 2008

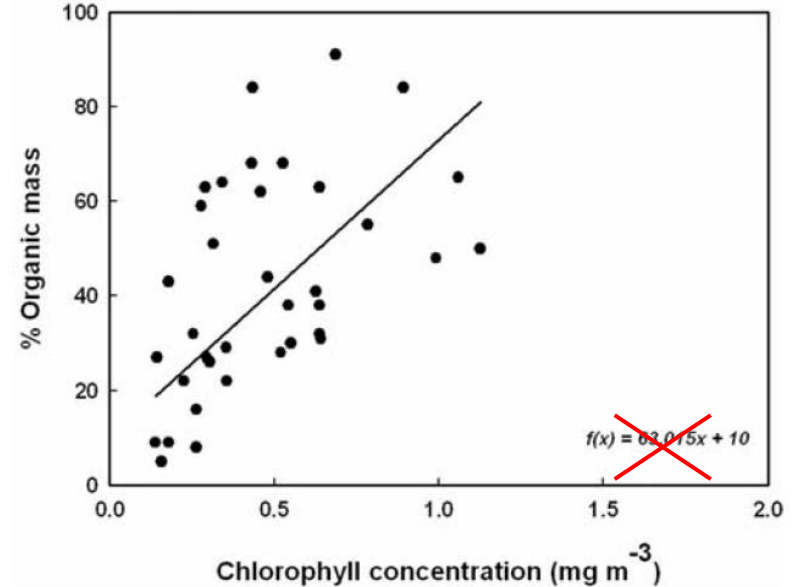
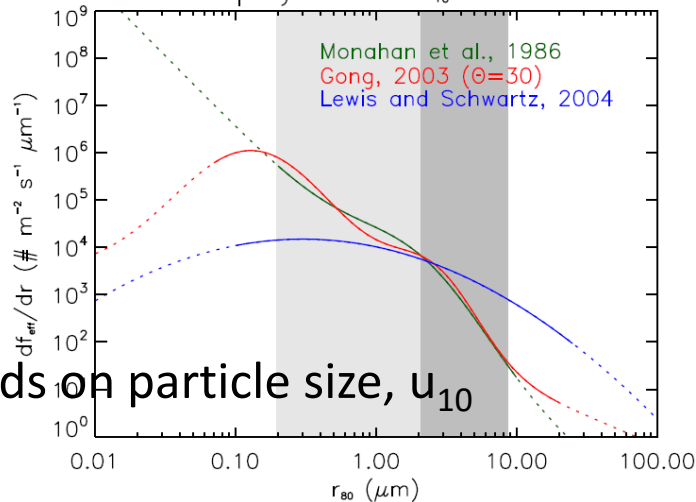


Figure 2. Correlation between fractional WIOC component of sea-spray as a function of grid-average chlorophyll-a concentration.

Sea spray flux at $U_{10}=10 \text{ m s}^{-1}$



depends on particle size, u_{10}

Vignati et al., AE, 2010

$$\% \text{ organic mass} = 43.5 \cdot \text{Chl} [\text{mg m}^{-3}] + 13.805,$$

$$\text{Chl} < 1.43 \mu\text{g m}^{-3}$$

Fire sources

$$E_i = A(x,t) \times B(x) \times \text{FB} \times \text{ef}_i \quad (1)$$

Where the emission of species i (E_i , mass of i emitted) is equal to the area burned at time t and location x [$A(x,t)$] multiplied by the biomass loading at location x [$B(x)$], the fraction of that biomass that is burned in the fire (FB), and the emission factor of species i (ef_i , mass of i emitted/mass of biomass burned). All biomass terms are on a dry weight basis.

FINN: Wiedinmyer et al., GMD, 2011

GFED: van der Werf et al ACP 2006, 2010

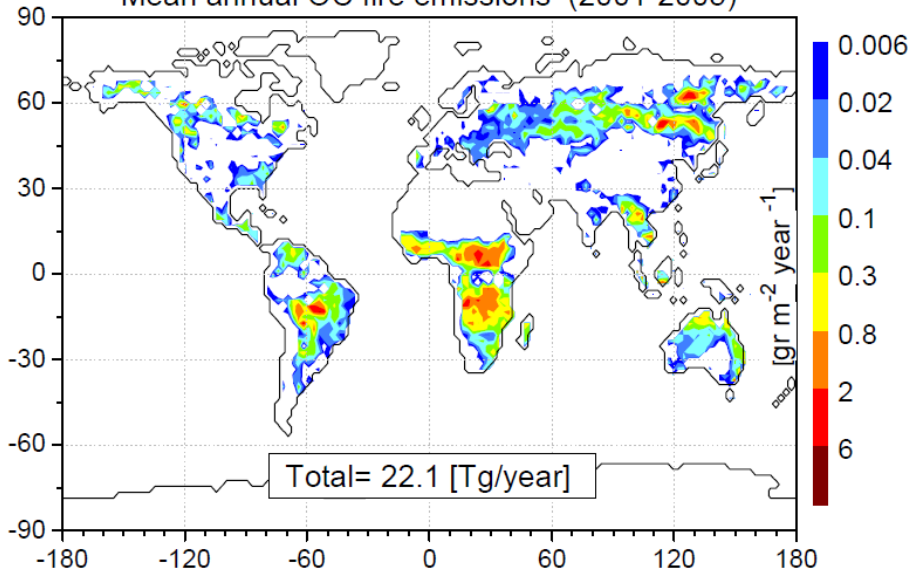
Online: Pechony and Shindell, *J Geophys Res* 2009

Flammability = f(RH, precipitation, vapor pressure deficit, vegetation density)

Fire counts = f(Flammability, Ignition by lightning + humans, fraction of non suppressed fires)

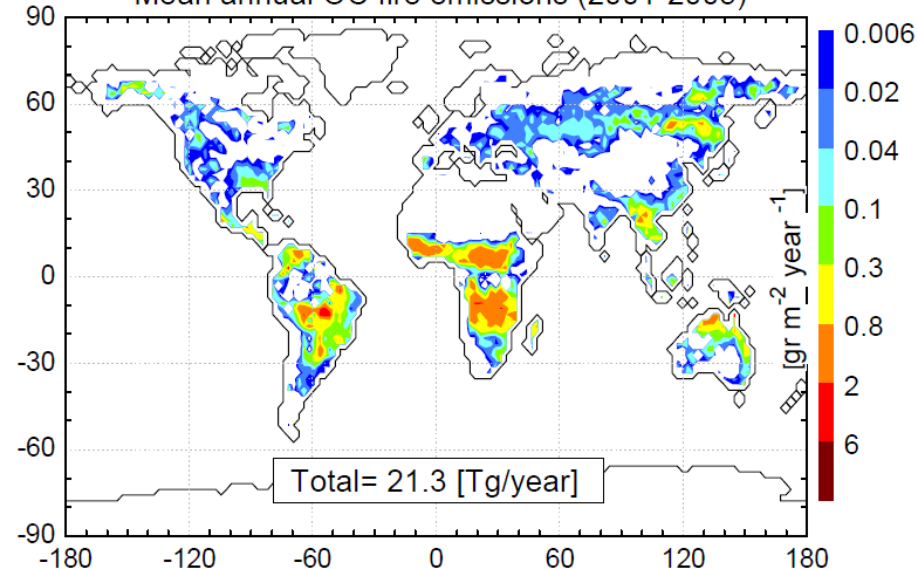
GFED

Mean annual OC fire emissions (2001-2005)

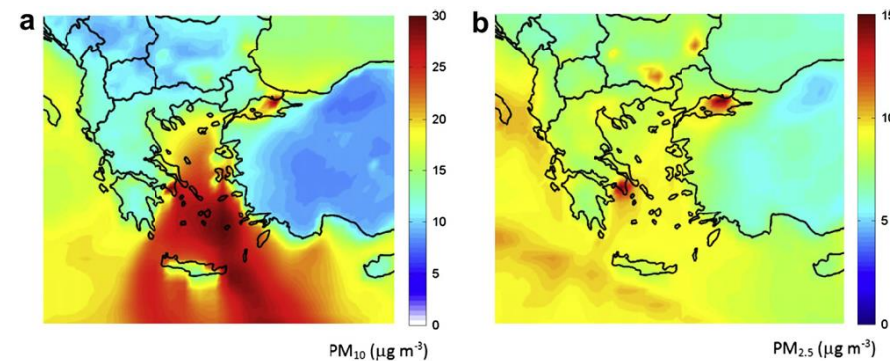
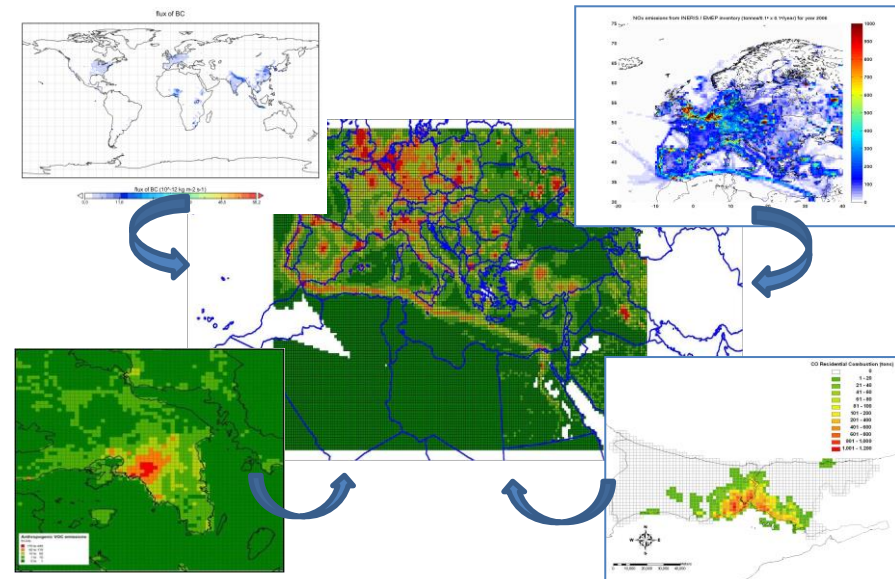
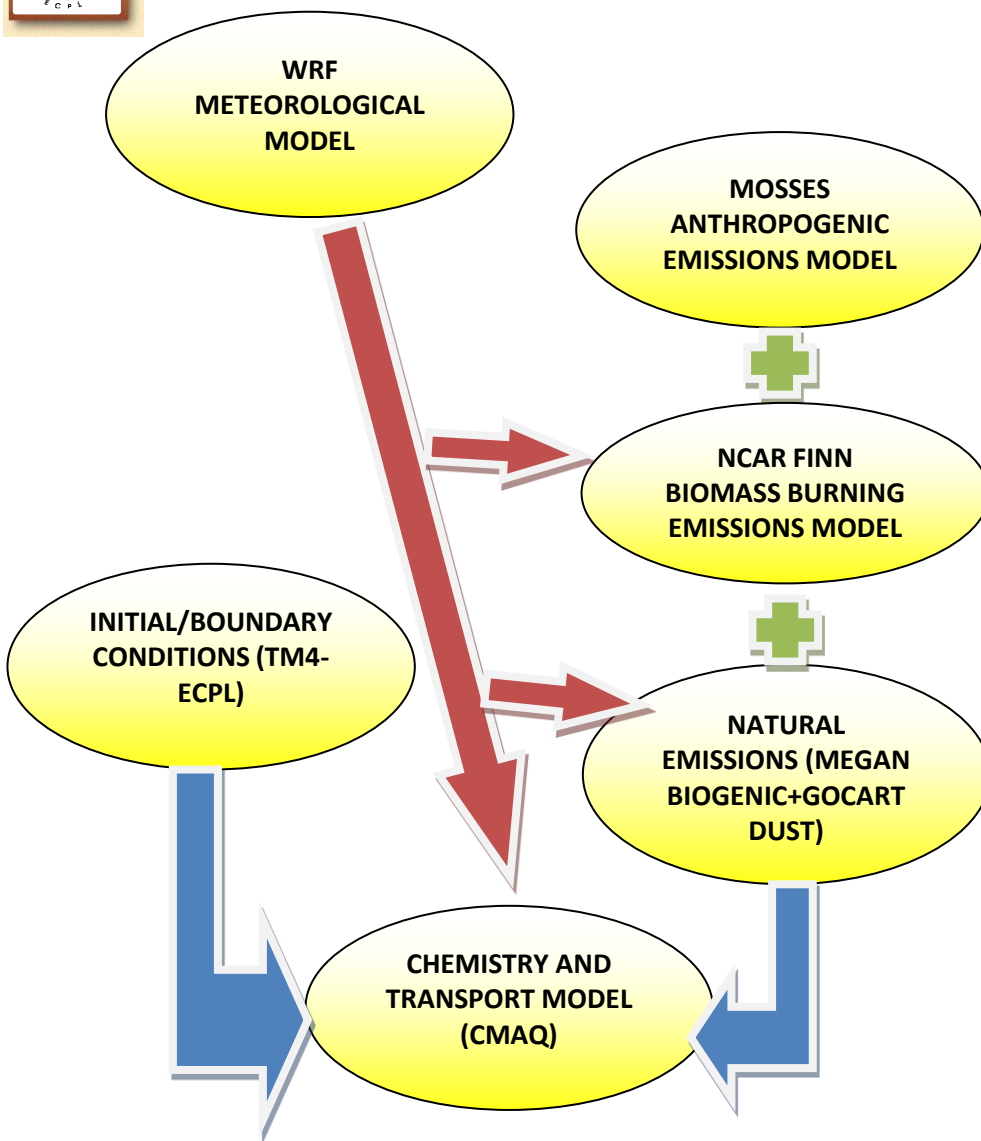


Reconstructed

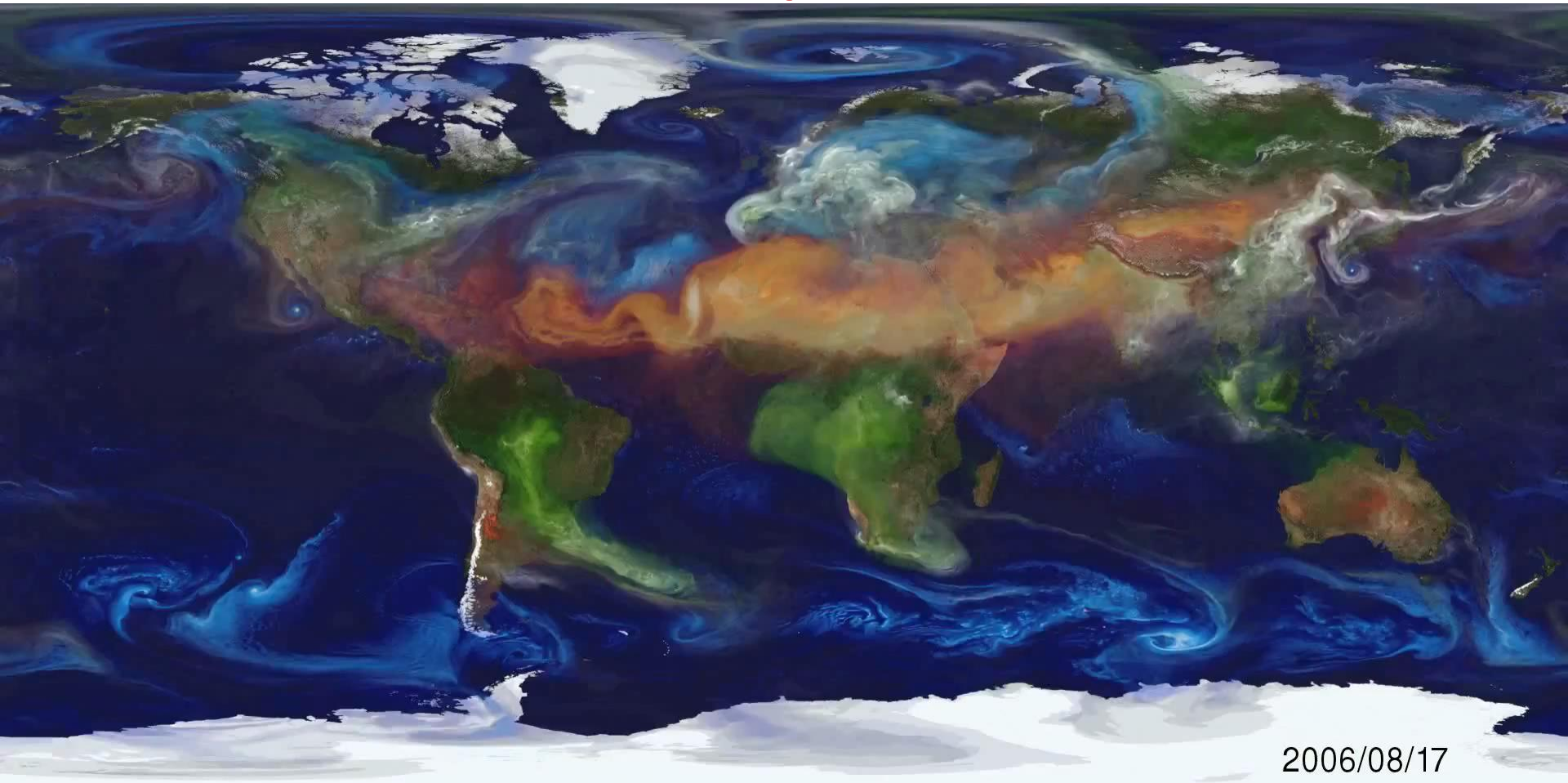
Mean annual OC fire emissions (2001-2005)



Methods – Model Configuration



Aerosols up to 10 km



2006/08/17

NASA GEOS 5

Desert dust

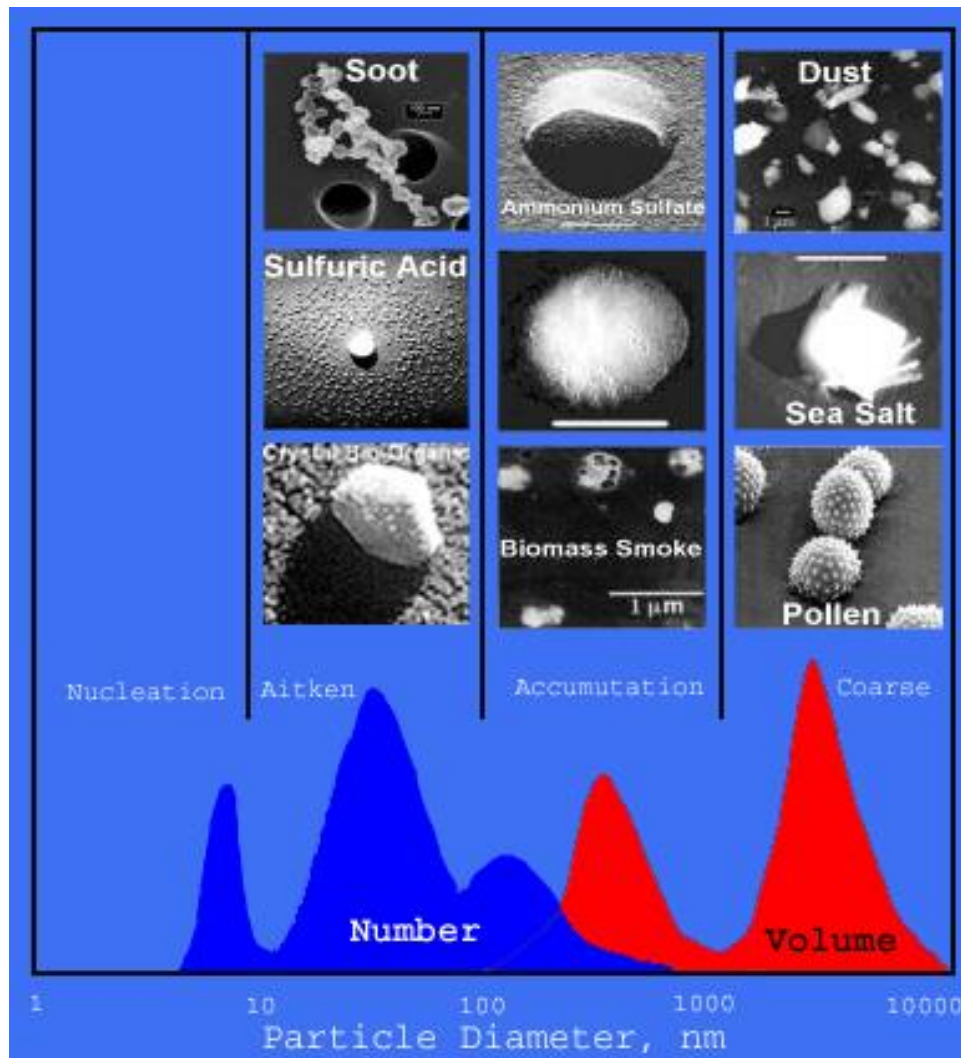
Sea-salt

Black and organic carbon

Sulfate aerosol

AEROSOL REPRESENTATION IN MODELS

Size distribution and shape



Models
assume all
aerosols are
spherical

From the IGAC first 10
year synthesis book,
edited by Brasseur et
al. (2002)

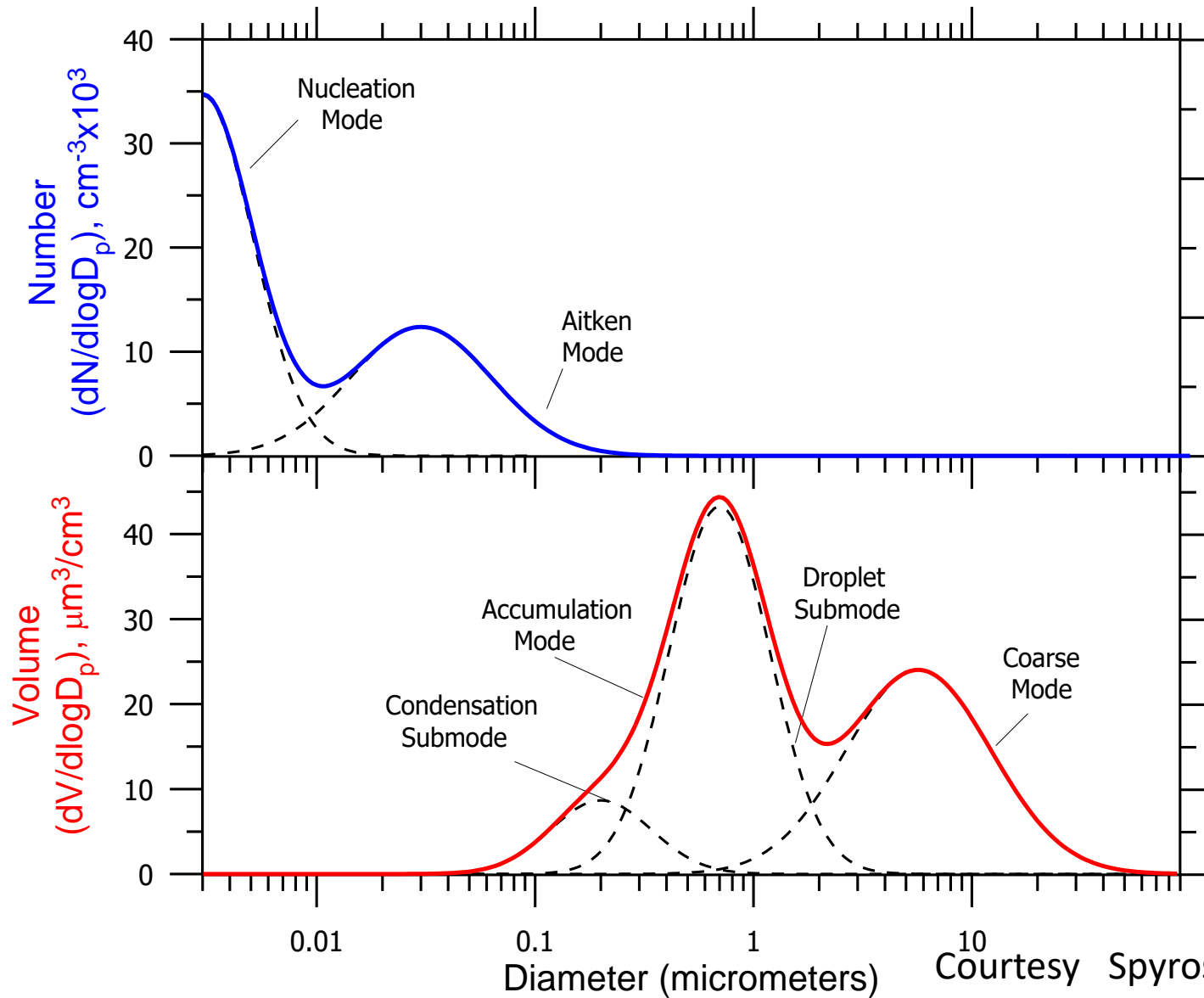


Do we need to resolve aerosol size ?



Is only one way to do this?

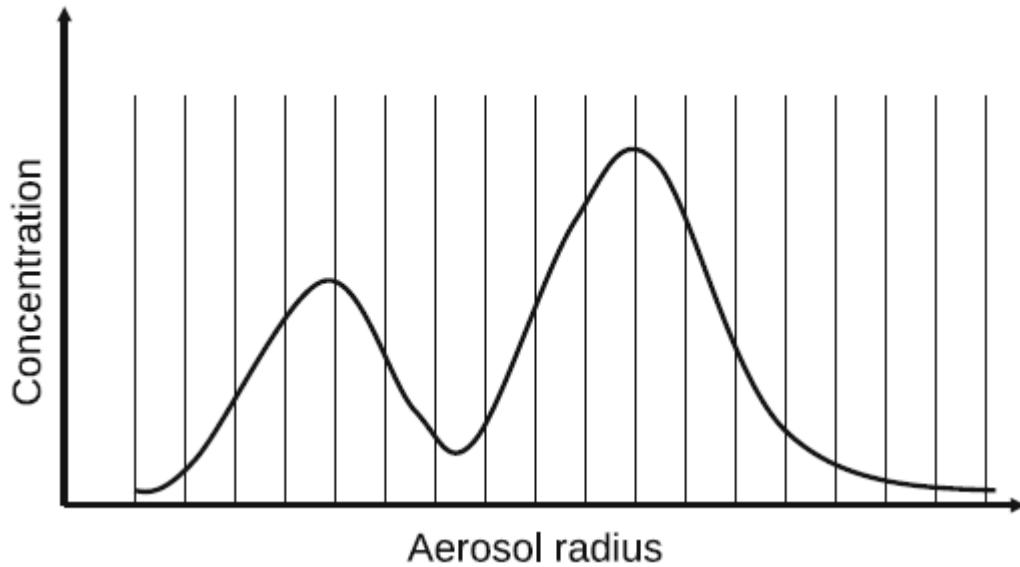
Atmospheric Aerosol Size Distribution



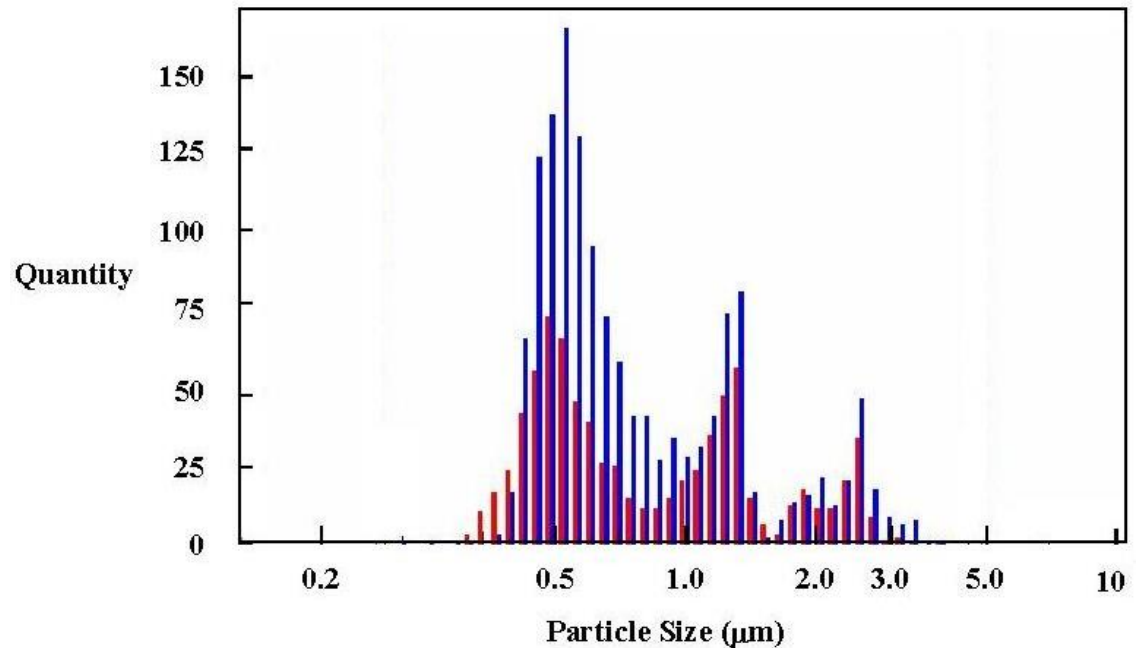
Size distribution –not resolved (bulk)

- Aerosols in models have a characteristic prescribed constant dry size, almost always lognormally distributed, with a constant width (sigma).
- Mixing is typically external, but can be internal as well, typically homogeneously mixed, but other mixing states are possible
- Aerosol total mass is calculated, out from which the aerosol number can be derived
- Very simple with very few tracers, but very fast

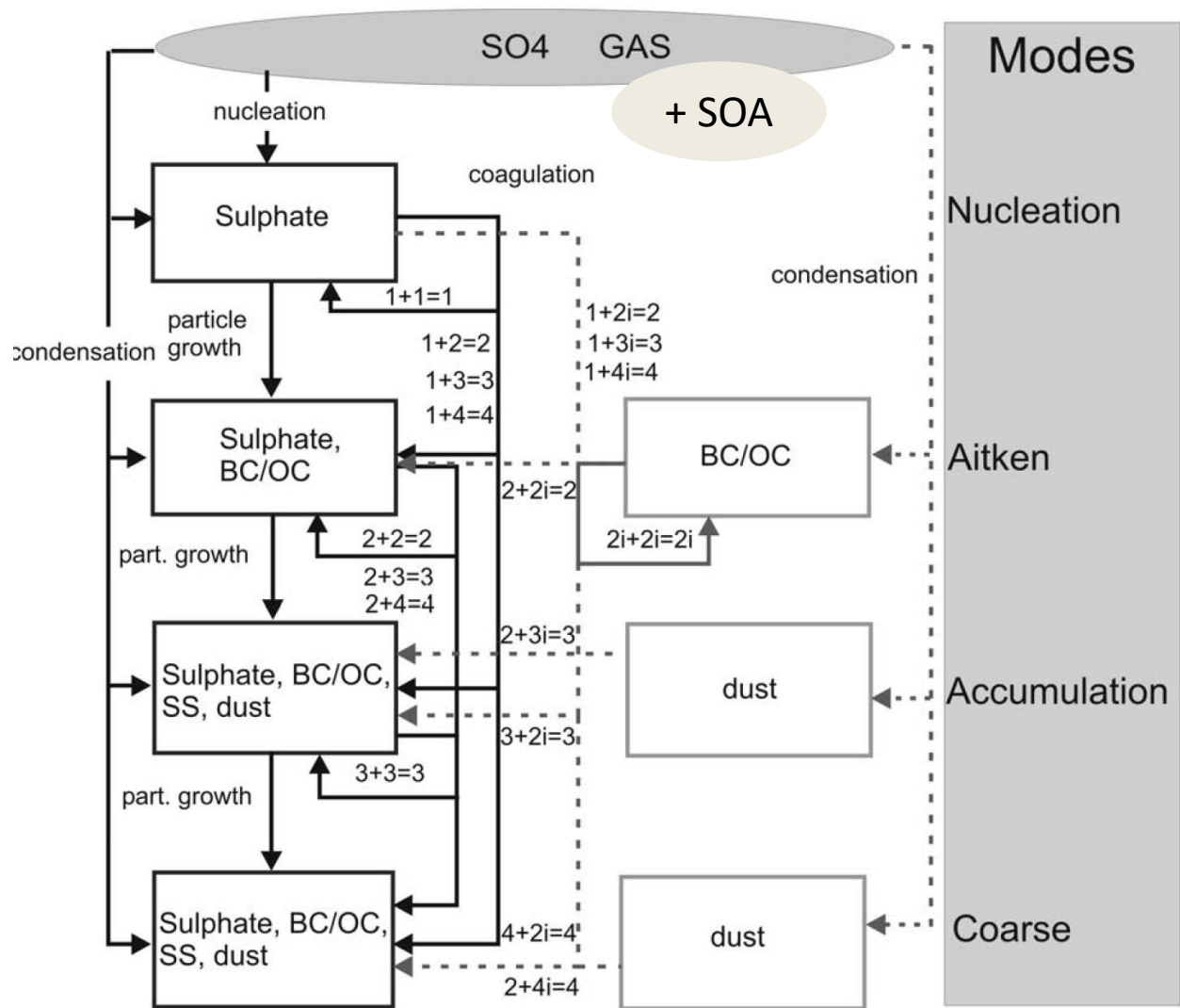
Size distribution modal vs sectional/bin approach



Boucher (text book) 2015



Modal approach & mixing of aerosols

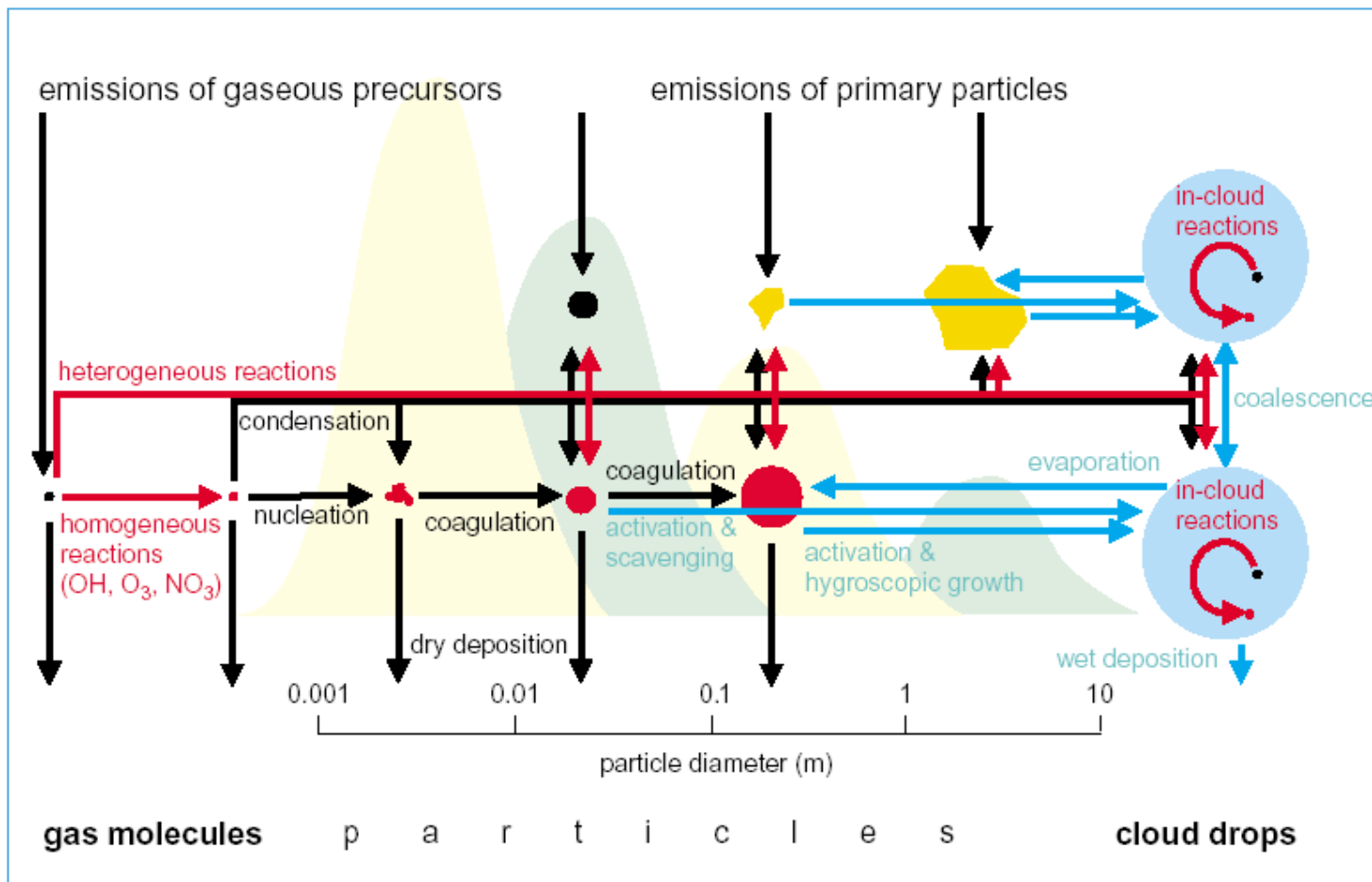


Aerosol
microphysics
model M7

soluble

In soluble

Processes affecting aerosols



Raes et al., 2000

Cluster formation can increase CCN number concentration by 2X – 9X:

Enhanced by organics

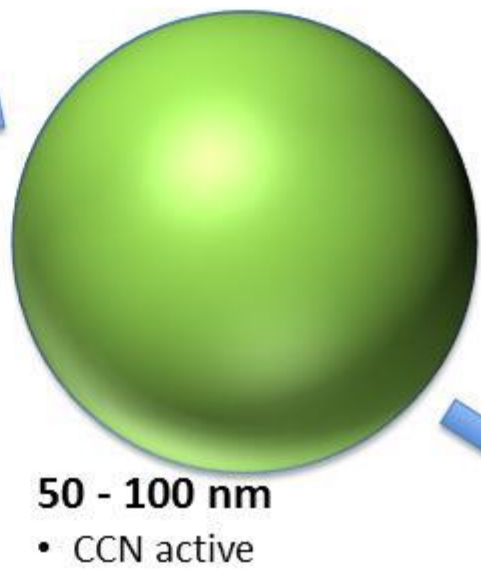
- < 1 nm**
- sulfuric acid
 - low volatility organics
 - ammonia/amines

- 1 – 1.5 nm**
- sulfuric acid
 - low volatility organics

Initial cluster growth determines cluster survival to larger sizes:
Dominated by organics



Nucleation & growth



Activation

> 1 μ m

Coagulation loss



Aerosol clouds interactions are one of the largest uncertainties in climate modeling

IPCC 2013

Nucleation mechanisms

1. Binary – sulfuric acid and water
2. Ternary- sulfuric acid – water – ammonia
3. Organics (HOM/ELVOC) contribute to the fast growth of the cluster. (Ehn et al., 2012)

$$JR = k_R [\text{H}_2\text{SO}_4]^2 0.5 [\text{BioOxOrg}] \text{ (Jöniken et al 2015)}$$

$$GR = 7.3 \times 10^{-8} [\text{H}_2\text{SO}_4] + 1.41 \times 10^{-7} [\text{HOM}] \text{ (Gordon et al., JGR, 2017)}$$

4. Ion induced nucleation by organics Kirkby et al., Nature 2016
5. Organics (amines, ELVOC) involved from the first step of nucleation

Nucleation of organics alone, have been parameterized as a sum of neutral (J_n) and ion-induced (J_{in}) components (Gordon et al., 2016):

$$J_{org} = J_n + J_{gcr} \tag{3}$$

$$J_n = a_1 [\text{HOM}]^{(a_2 + a_5)/[\text{HOM}]} \tag{4}$$

$$J_{in} = 2n_{\pm} a_3 [\text{HOM}]^{(a_4 + a_5)/[\text{HOM}]} \tag{5}$$

where the HOM concentration is given in units of 10^7 molecules per cm^3 , n_{\pm} is the ion concentration, and a_1 – a_5 are parameters fitted to the experimental data.

Currently in models:

(1), (2) where NH_3 levels are high, (3) consider organics for growth of clusters

Nucleation mechanisms

1. Binary – sulfuric acid and water
2. Ternary- sulfuric acid – water – ammonia
3. Organics (HOM/ELVOC) contribute to the fast growth of the cluster. (Ehn et al., 2012)

$$JR = k_R [H_2SO_4]^2 \cdot 0.$$

$$GR = 7.3 \times 10^{-8} [H_2SO_4] + 1.$$

4. Ion induced nucleation by
5. Organics (amines, ELVOC)

Nucleation of organics alone, have (J_{ion}) components (Gordon et al., 2017)

$$J_{org} = J_n + J_{gcr}$$

$$J_n = a_1 [HOM]^{(a_2 + a_5) / [HOM]}$$

$$J_{ion} = 2n \pm a_3 [HOM]^{(a_4 + a_5) / [HOM]}$$

where the HOM concentration is concentration, and a_1 – a_5 are parameters

Present day

Gordon et al., JGR 2017



■ org-ion

■ H₂SO₄-org-ion

■ H₂SO₄-org

■ H₂SO₄-NH₃

■ H₂SO₄-NH₃-ion

■ H₂SO₄-ion

Currently in models:

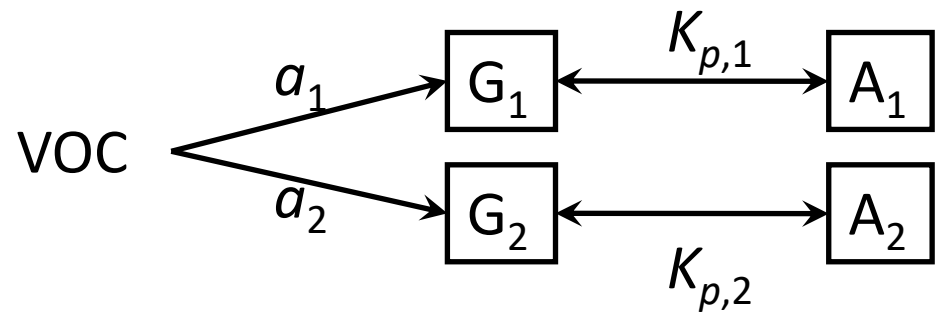
(1), (2) where NH₃ levels are high, (3) consider organics for growth of clusters

How SOA is parameterized in the models?

- Pseudo-emissions (~18% on terpenes emissions or terpenes oxidized amounts)

- 2-product model

- VOC reactivity
- SOA yield



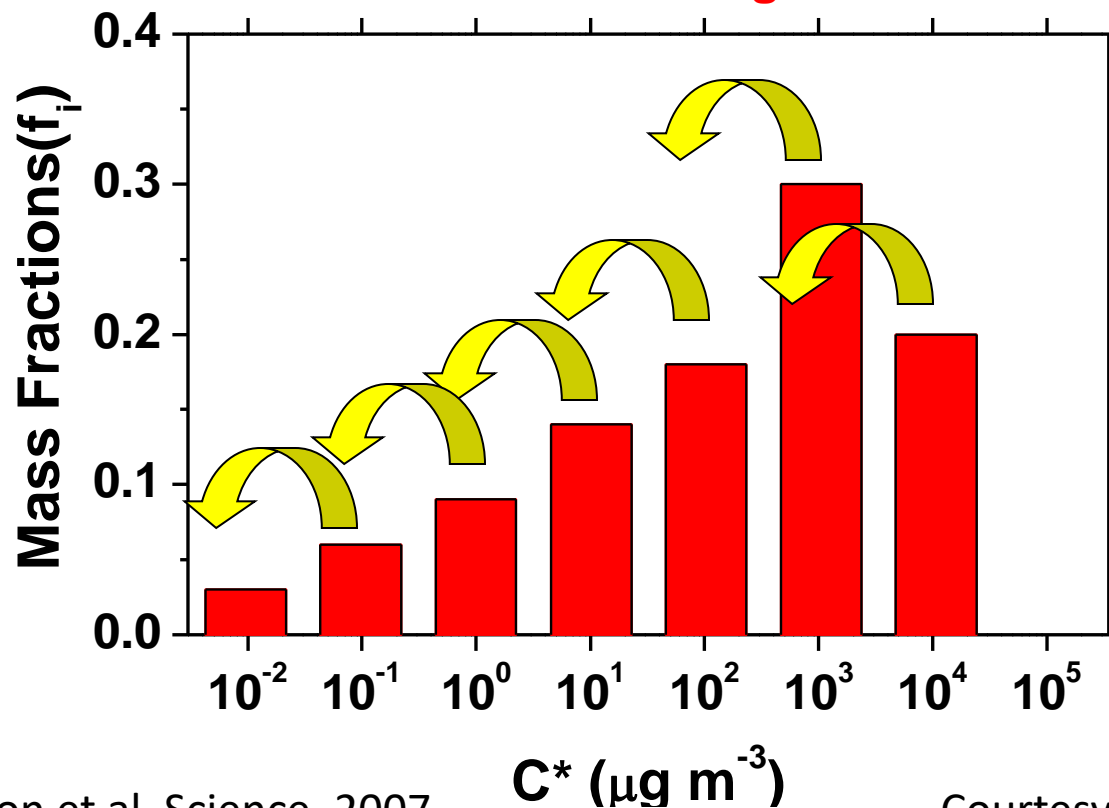
- Chemical dependence (e.g. NO_x levels) – a_i
- Physical dependence (e.g. temperature) – $K_{p,i}$

Odum et al., GRL, 2007;
Kanakidou et al., JGR, 2000

- Volatility bases set (VBS)

Organic Aerosol and its Chemical Aging

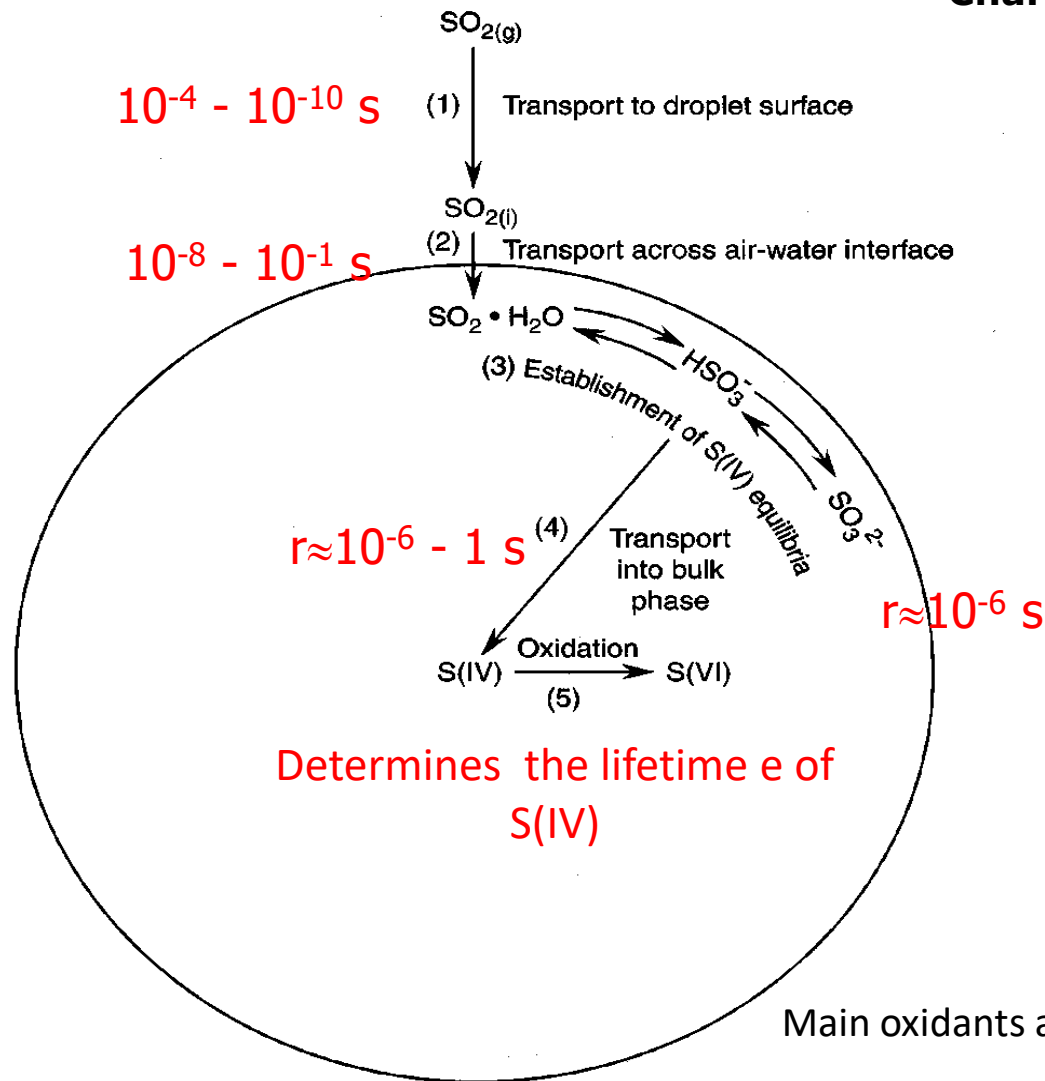
- Primary and secondary organics in the atmosphere also cover a wide range of volatilities
- Compounds react in the gas phase with OH producing material with lower volatility . Formation of very low volatility material ($10^{-5} \mu\text{g m}^{-3}$ from aging of semivolatile material assumed)



Oxidation in droplets/ aqueous phase chemistry

Characteristic times for the steps:

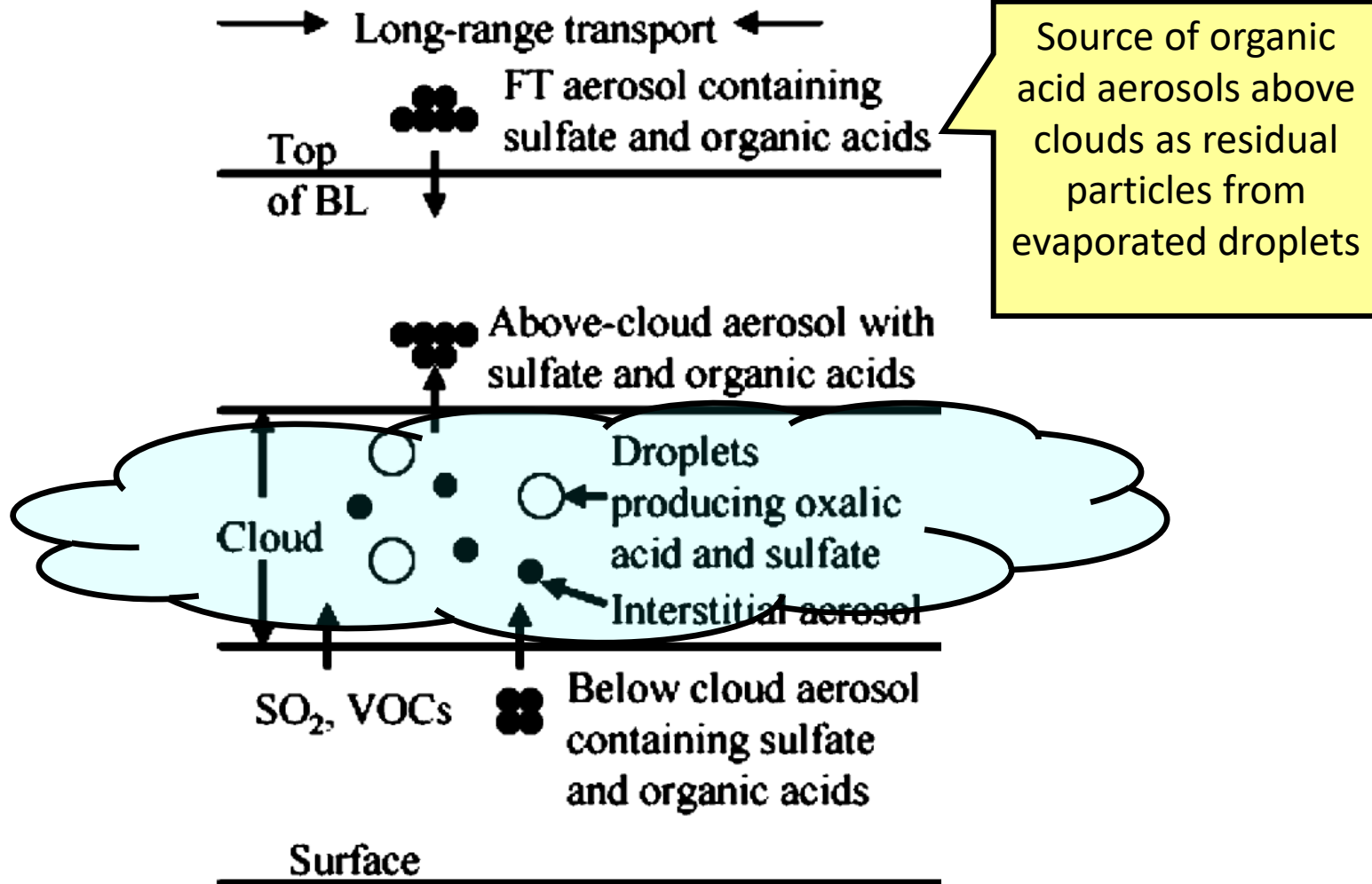
Typical products of interest inside the droplet
 SO_4^{2-} , organic acids



Main oxidants are O_2 , O_3 , H_2O_2 , $\bullet\text{OH}$, $\text{HO}_2\bullet$, NO_x

Importance of Fenton reactions $\text{Fe}^{2+}/\text{Fe}^{3+}$ & $\text{Cu}^+/\text{Cu}^{2+}$

Cloud processing



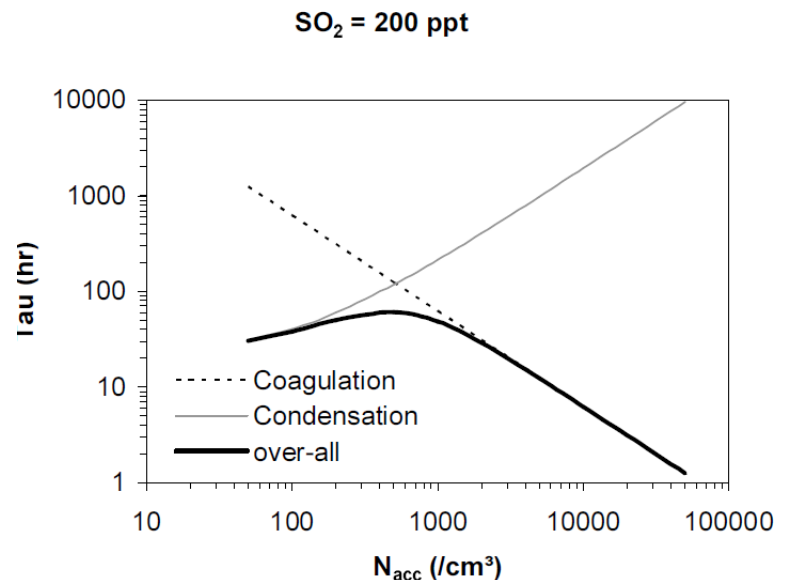
AEROSOL PROCESSING

Coagulation

- reduces the number of aerosols
- leads to larger sizes
- modifies size distribution
- does not remove mass from the atmosphere
- Important to consider when studying fine aerosol number concentrations.

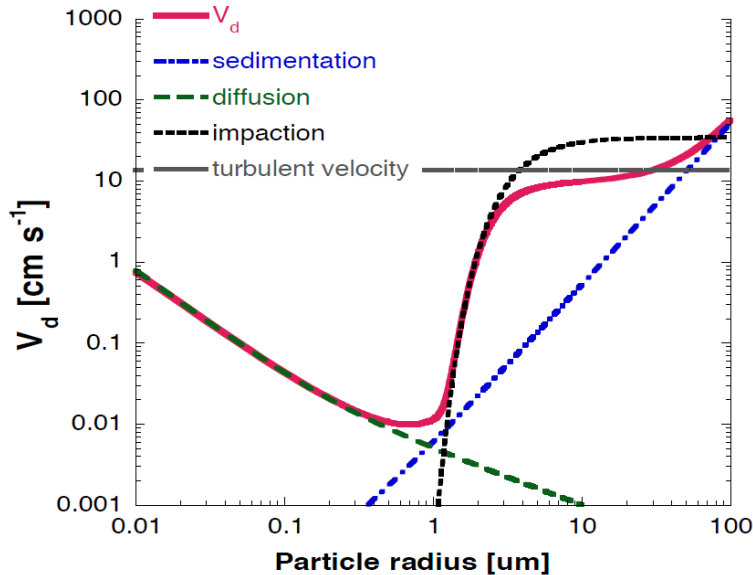
Importance of coagulation vs condensation for conversion of hydrophobic to hydrophilic particles. (turn overtime) for $\text{SO}_2=200$ ppt

Kanakidou et al., ACP 2005



Removal

Aerosol dry deposition and sedimentation



R_a = aerodynamic resistance

R_{cp} : quasi-laminar boundary layer resistance

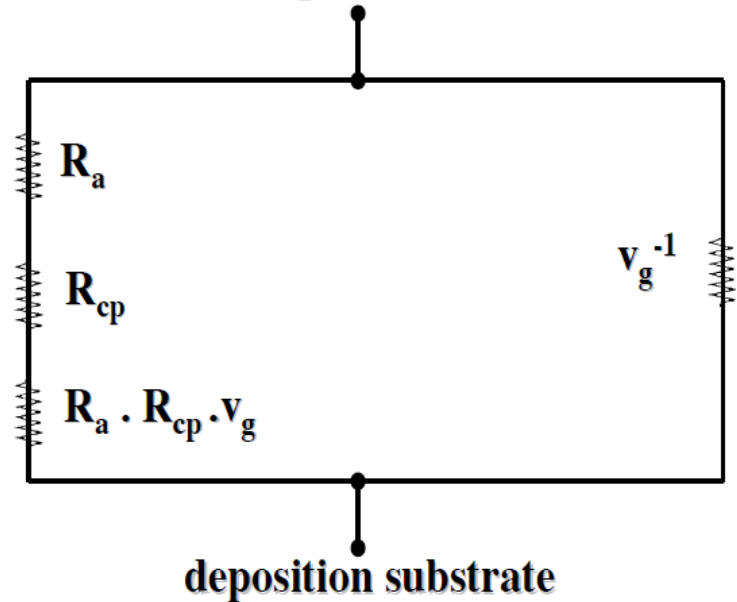
R_{surf} : surface resistance (all types of surface - soil, plant, buildings etc), becomes zero (if a particle reaches the ground, it has been deposited)

v_g : sedimentation velocity

V_d = deposition velocity

$$V_d = \frac{1}{\left(\frac{R_a}{a} + \frac{R_{cp}}{cp} + \frac{R_a R_{cp}}{a cp} \right) + v_g} + v_g$$

atmospheric source

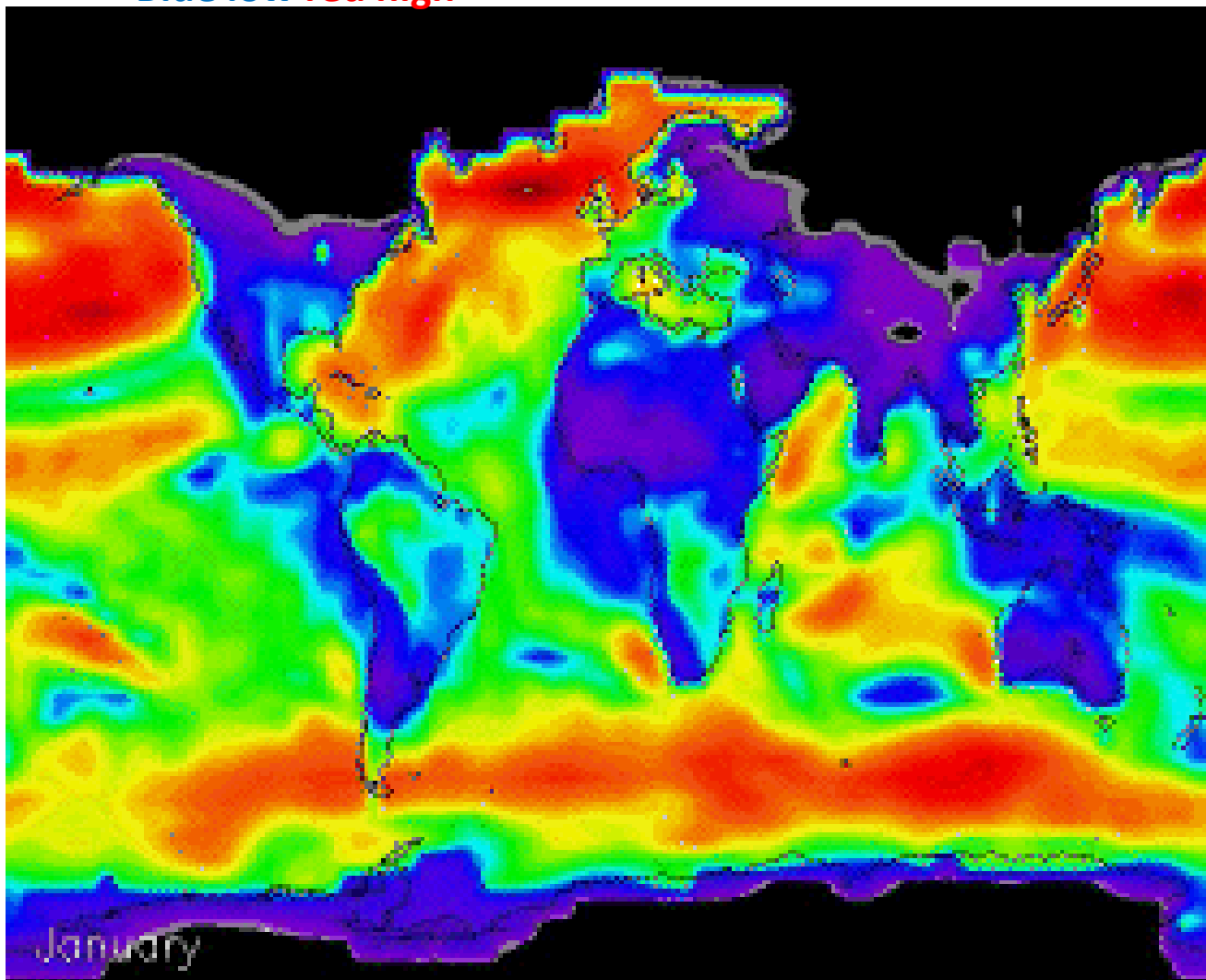


Monthly variation of SO₂ deposition as computed by ECHAM4

– Gazenveld Laurens

<http://www.atmosphere.mpg.de/enid/aaf662be6cd123f4c54c4d90d24b1373,0/hr.html>

Blue low red high



Annual cycle in global SO₂ dry deposition velocity constructed by using 2-week average global distributions.

Over the oceans V_{dSO_2} reflects the **turbulence intensity** → large removal rates over the **stormtracks** over the higher latitudes in the SH throughout the year and in the NH winter over the Atlantic & Pacific.

Over land **near-zero values (black areas)**, due to a large surface resistance for surface temperatures < -10 °C, reflecting the **snow/ice** cover.

Relative large values **over Europe in winter** due to the presence of a large fraction of **wetted surfaces** due to rainfall interception and dewfall.

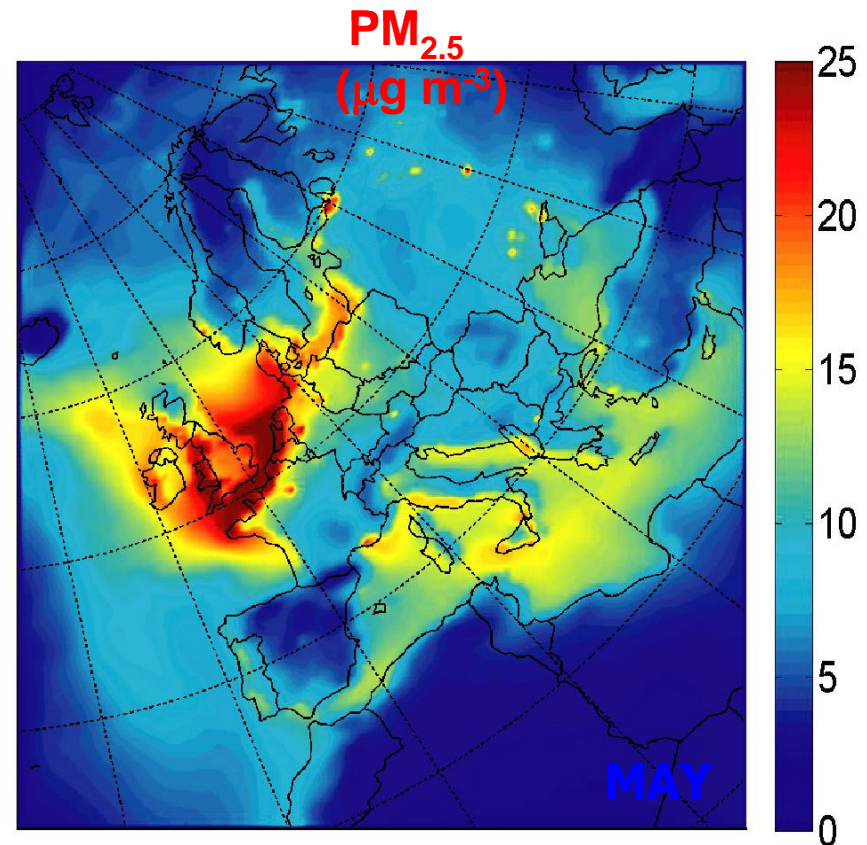
A dry deposition parameterization for sulfur oxides in a chemistry and general circulation model, Ganzeveld, Lelieveld, and Roelofs, J. Geophys. Res., 103. 1998, 5679-5694

Wet deposition

- In-cloud removal
 - By dissolution followed by precipitation
- Below-cloud removal
 - By falling droplets
- Function of size, both aerosol's and cloud droplet's
- **Not all wet-removed aerosols reach the ground**, due to rain-droplet evaporation, a process serving as a lower level source of cloud processed aerosols

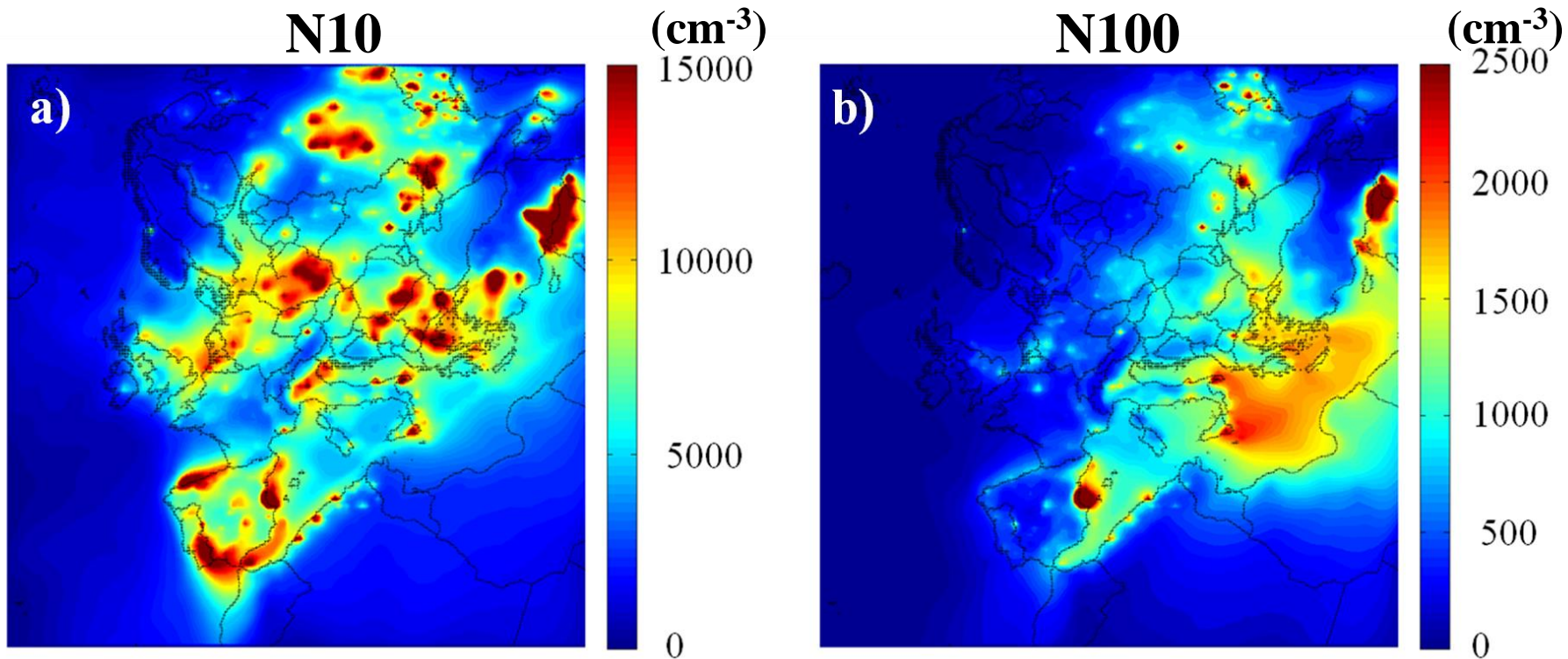
PMCAMx-UF

- Ultrafine PM-focused version of PMCAMx.
- Simulates both aerosol number and composition using the TOMAS algorithm
 - 43 size sections (1 nm-10 μm)
 - 13 aerosol species
 - H_2SO_4 in pseudo-steady state
- Grid nesting (down to a 3x3 km)
 - 3492x3240 km region
 - 36x36 km grid
 - 14 levels up to 6 km
- Carbon Bond-IV gas-phase chemistry
 - 34 gas species
- WRF Meteorology



Courtesy Spyros Pandis

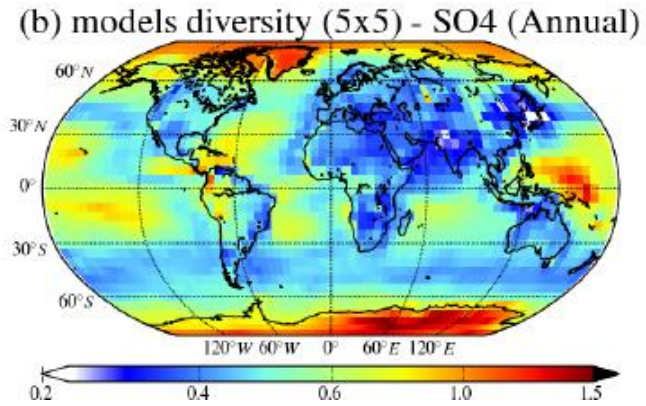
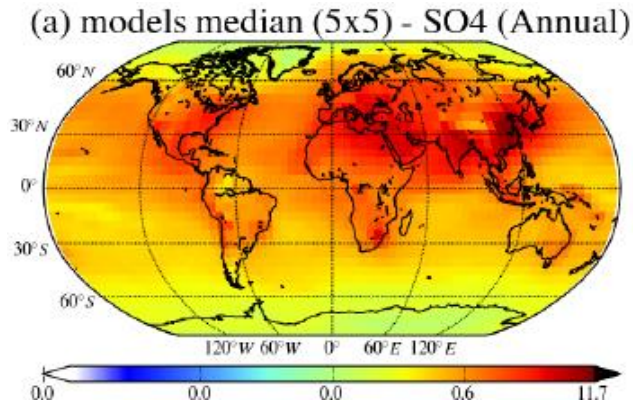
Particle number concentration fields (June 2012)



Video that shows how NPF are formed
in Europe in July 2012

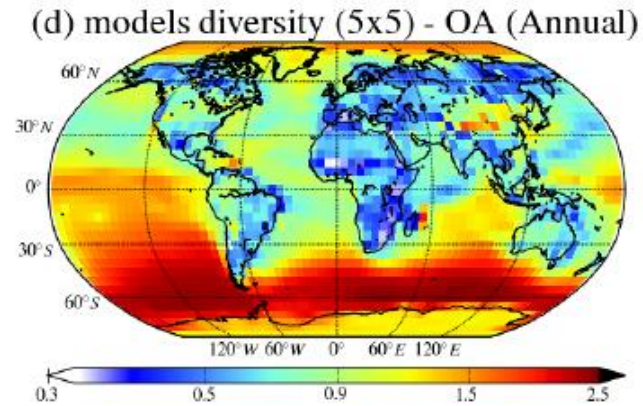
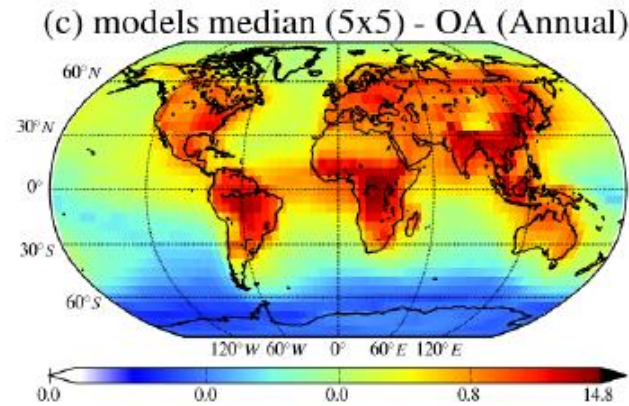
PM₁ multi-model median chemical composition- surface

median
sulfate

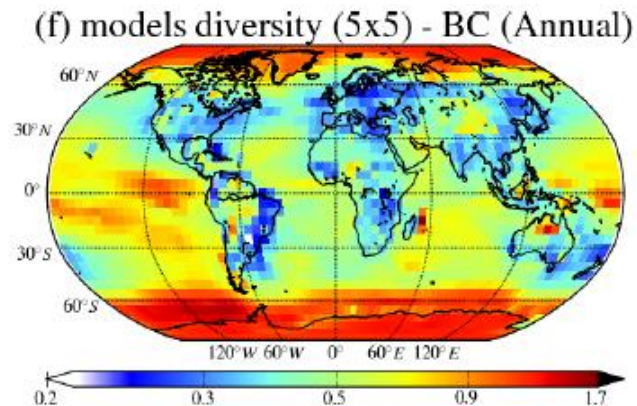
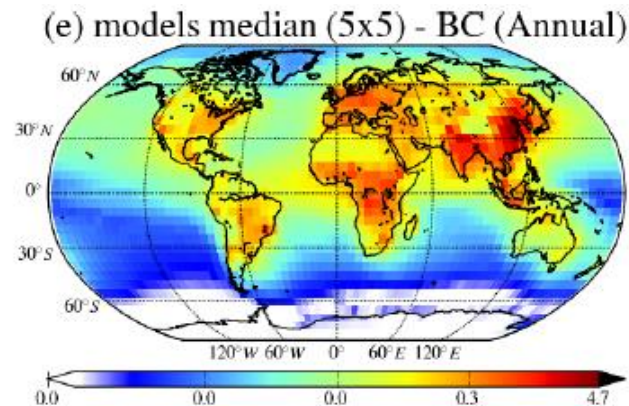


diversity

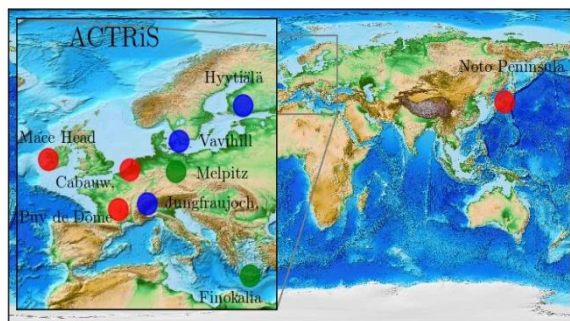
Organic
aerosol



Black
carbon



**BACCHUS model
intercomparison
15 models vs
AMS- organics**

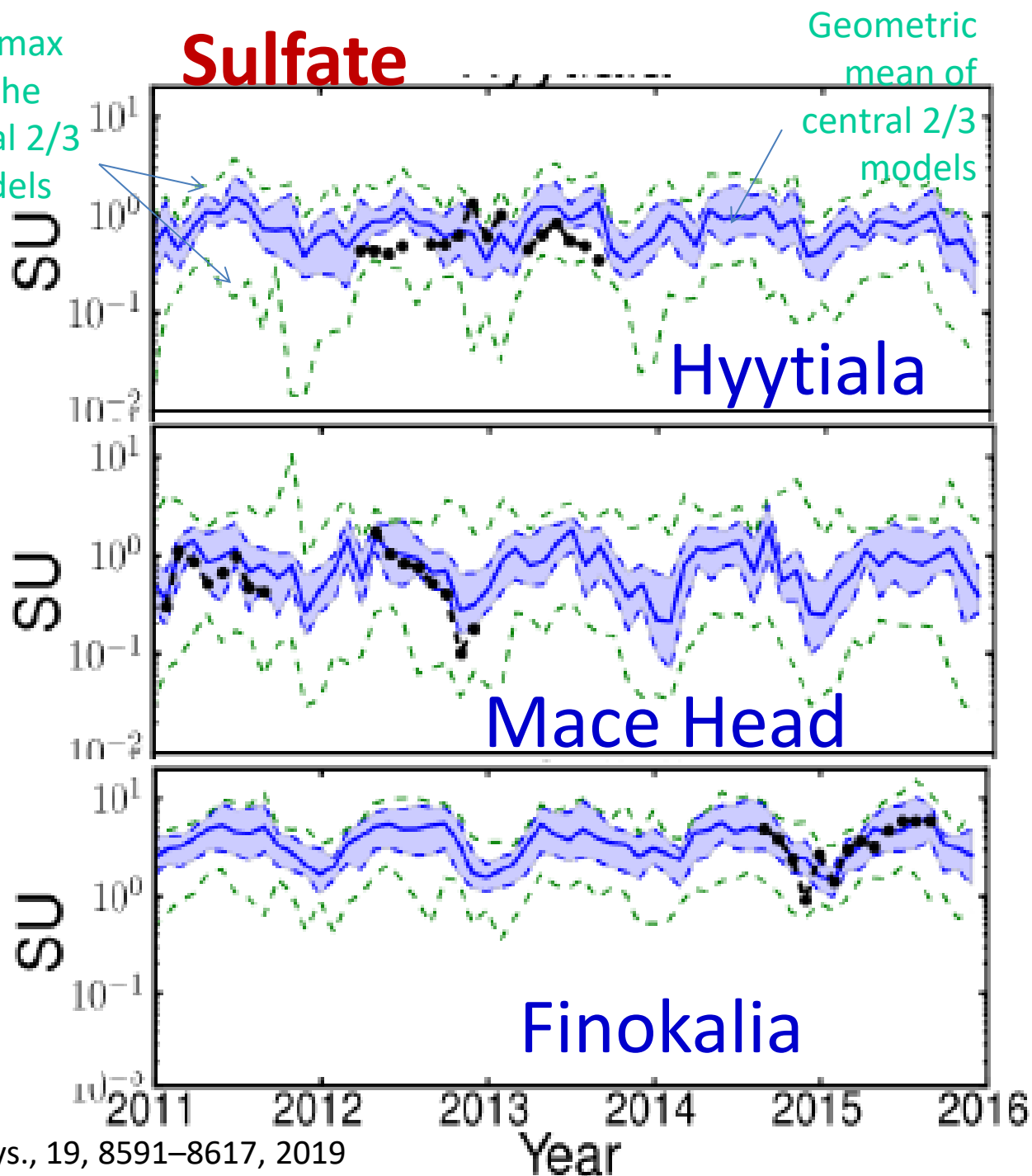


Data (black dots) from
ACTRIS - Schmale et al.
Scientific Reports, 2017

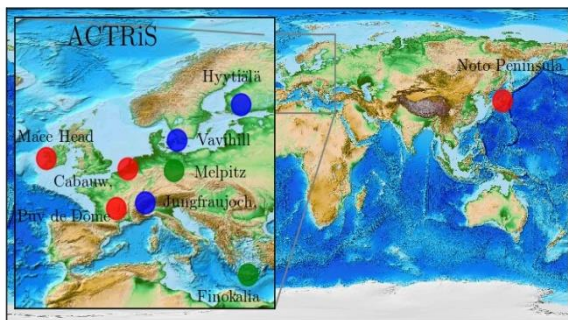
MMM – blue line
min, max model green
dashed

Min max
of the
central 2/3
models

Sulfate



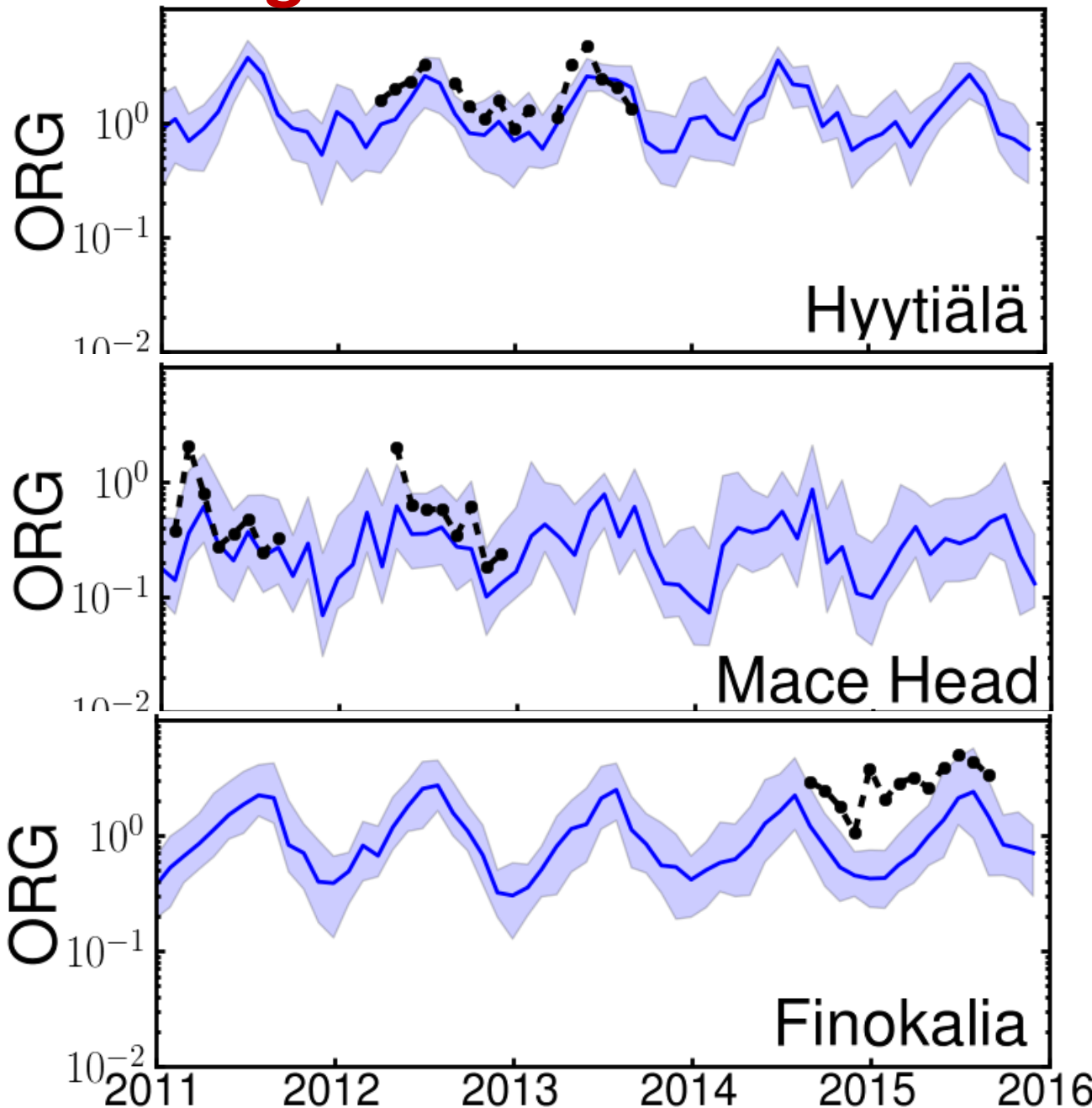
BACCHUS model intercomparison 15 models vs AMS- organics



Data from ACTRIS -
Schmale et al. Scientific
Reports, 2017

Fanourgakis et al., Atmos.
Chem. Phys., 19, 8591–8617,
2019

Organics



AEROSOL WATER

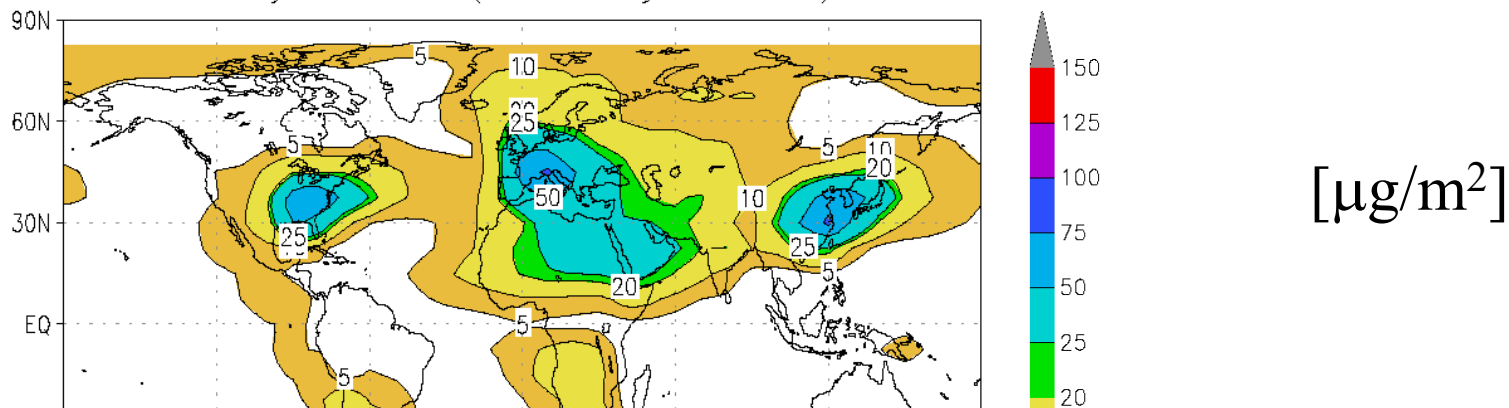
sum of water associated with the **inorganic and organic** components of the aerosols –
in most models only the inorganic aerosol is taken into account for this calculation

AEROSOL ACIDITY

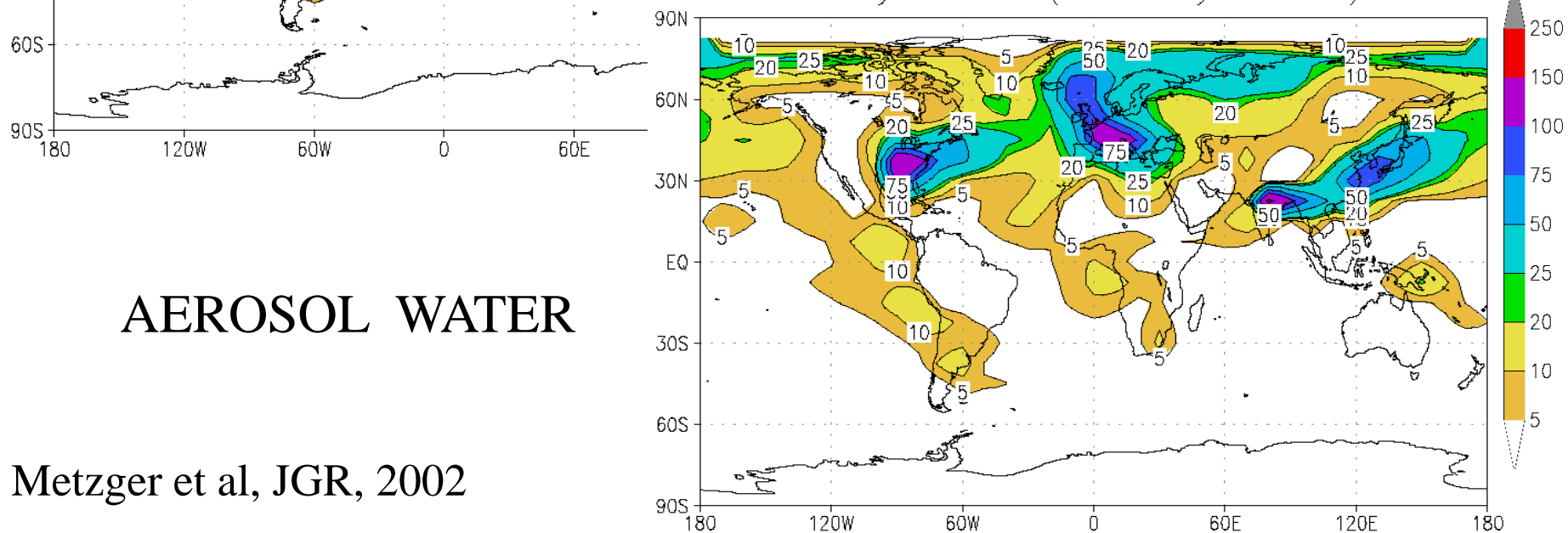
Important for aerosol reactions, for NO_3^- partitioning to aerosol phase, for nutrients solubilisation

TOTAL DRY AEROSOL MASS (SO₄, NO₃, NH₄)

July 1997 (monthly mean)



July 1997 (monthly mean)

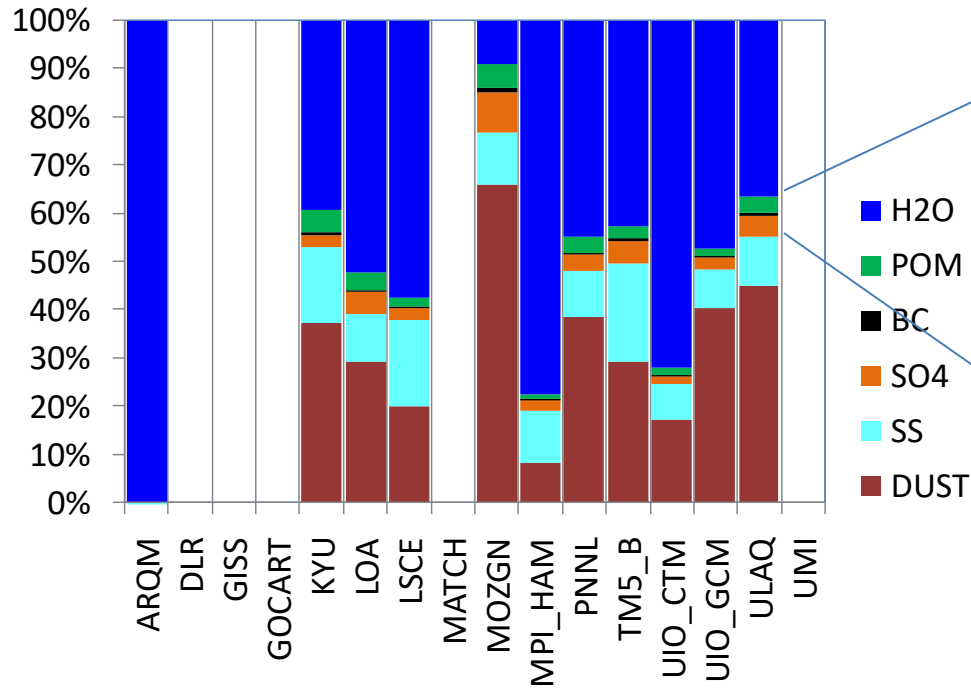


AEROSOL WATER

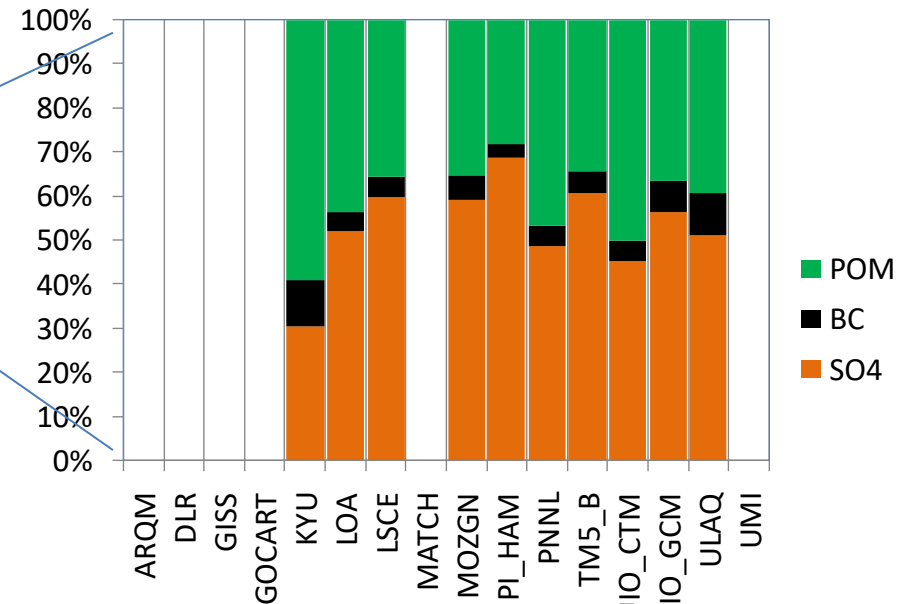
Metzger et al, JGR, 2002

Aerosol composition

Fraction of global aerosol composition



Fraction of global fine dry aerosol composition



Modified from Textor et al., 2006

AEROSOL HYGROSCOPICITY

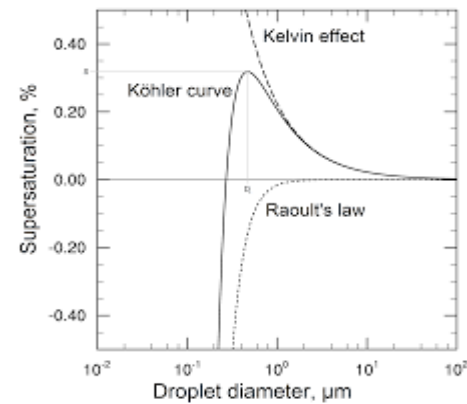
(K)

$$\kappa = \sum_i \varepsilon_i \kappa_i$$

$$\frac{1}{a_w} = 1 + \kappa \frac{V_s}{V_w},$$

$$S = a_w \exp\left(\frac{4\sigma_{s/a} M_w}{RT \rho_w D}\right)$$

$$S(D) = \frac{D^3 - D_d^3}{D^3 - D_d^3(1 - \kappa)} \exp\left(\frac{4\sigma_{s/a} M_w}{RT \rho_w D}\right)$$



a_w activity of water in solution

V_s dry volume

V_w volume of water

S saturation ratio

$\sigma_{s/a}$ surface tension

M_w, ρ_w molecular weight and density of water

D particle diameter

D_d dry particle diameter

M. D. Petters and S. M. Kreidenweis

Atmos. Chem. Phys., 7, 1961–1971, 2007

$$d_c = \left(\frac{4A^3}{27\kappa s_c^2} \right)^{1/3}$$

s_c supersaturation
 d_c critical diameter

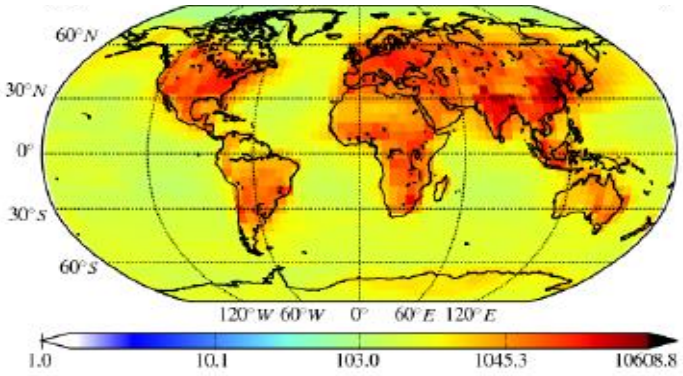
above which the particle acts as CCN

CCN at a specific supersaturation : number of particles of diameter $\geq d_c$

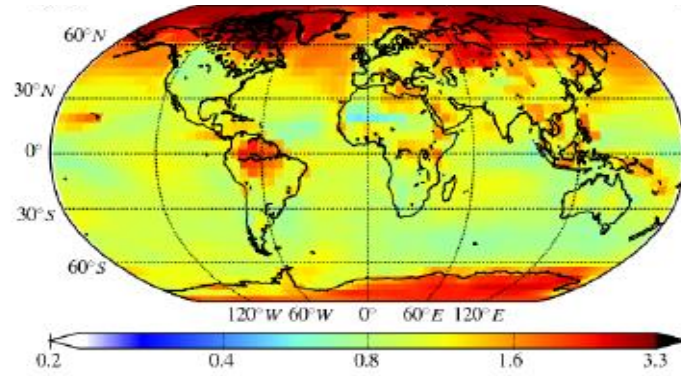
$$\text{CCN}(d_c) = \int_{d_c}^{\infty} n(d_p) dd_p$$

multi-model results at surface

Annual Multi-model median

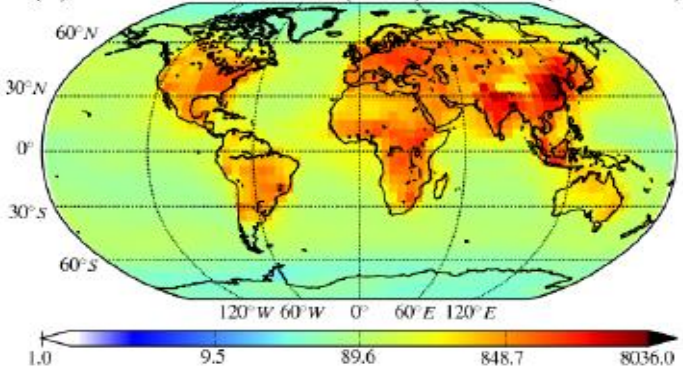


Model diversity (stdev/mean)

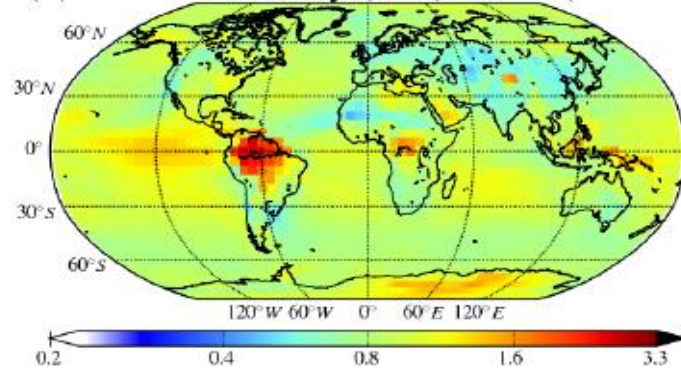


N3

(c) models median (5x5) - N50 (Annual)

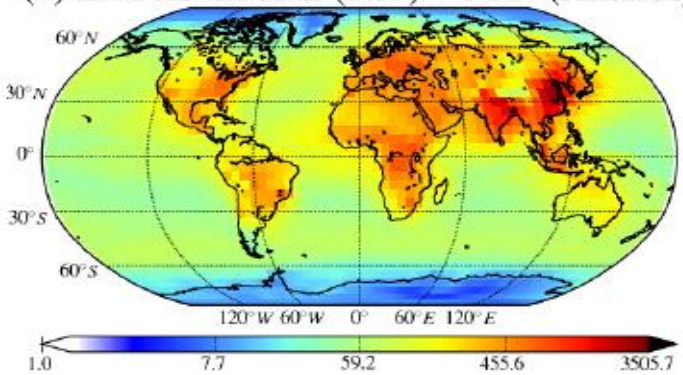


(d) models diversity (5x5) - N50 (Annual)

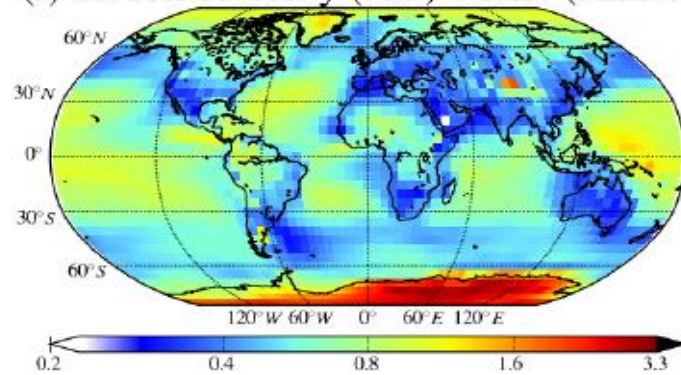


N50

(e) models median (5x5) - CCN (Annual)



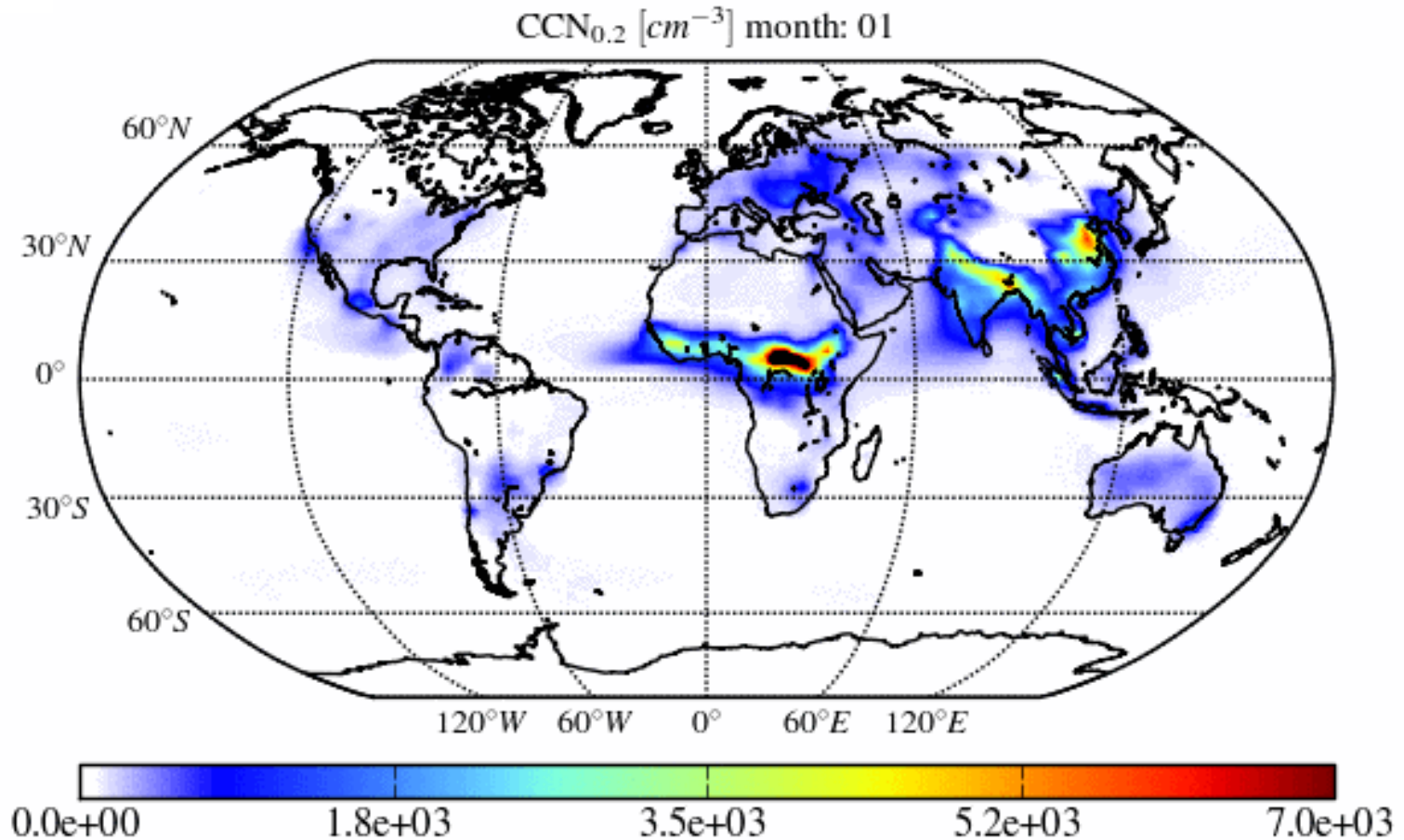
(f) models diversity (5x5) - CCN (Annual)



CCN

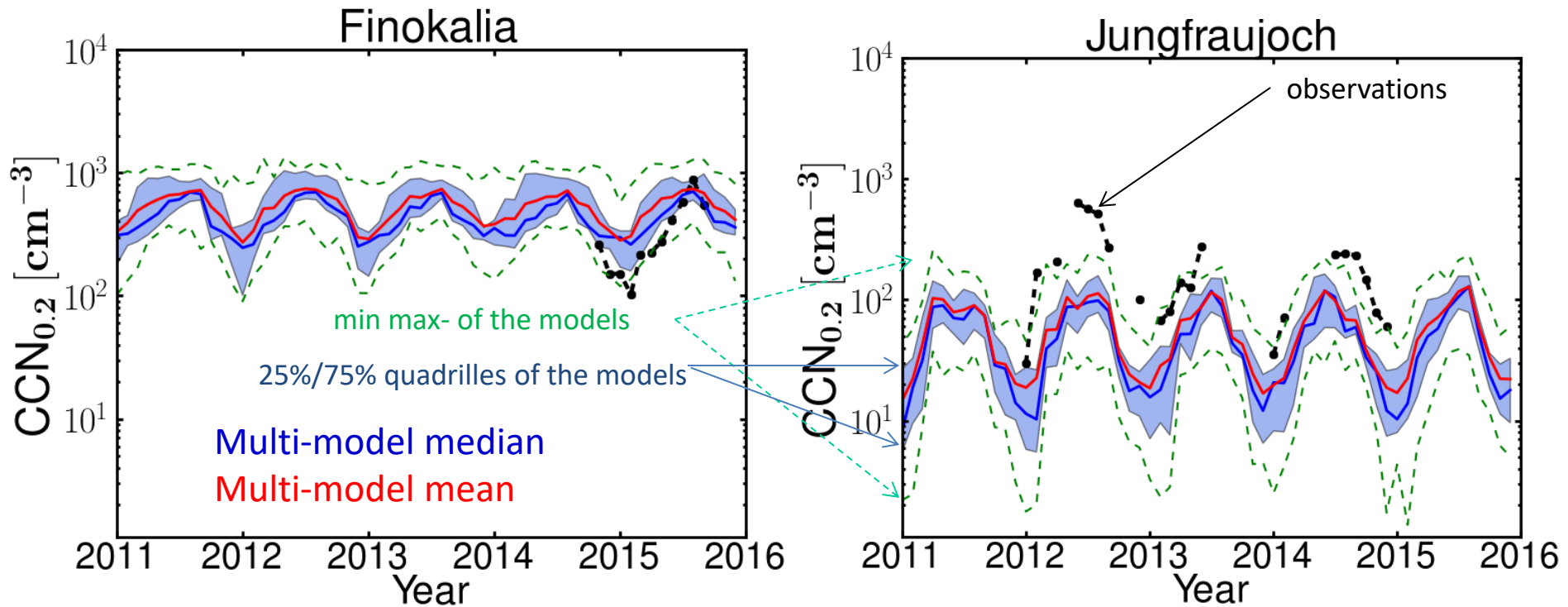


Seasonal variability of CCN at 0.2% ss



Near the surface

CCN at 0.2% supersaturation



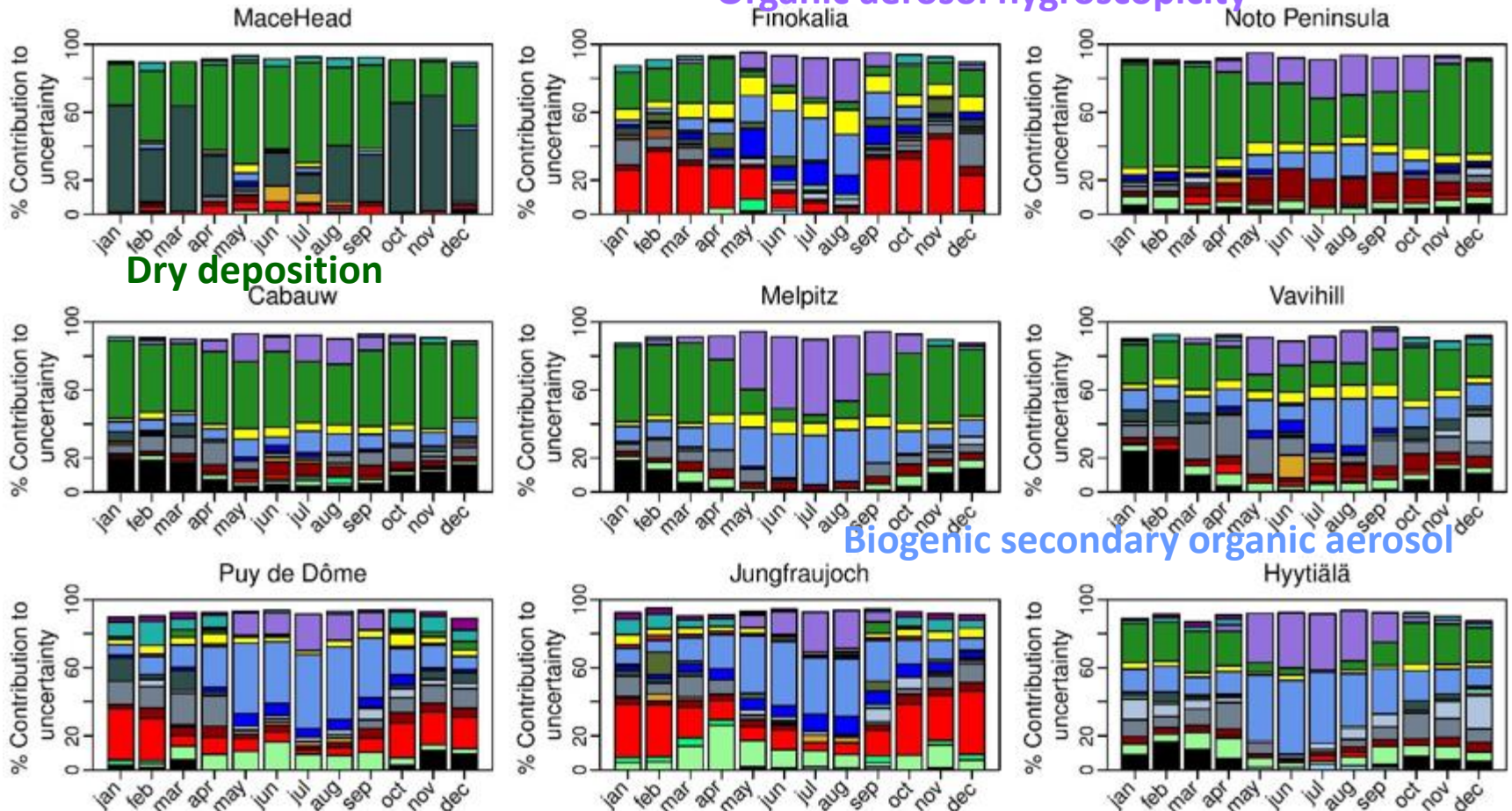
Overall NMB -37%

Major contributors to model uncertainty –

perturbed parameter ensemble

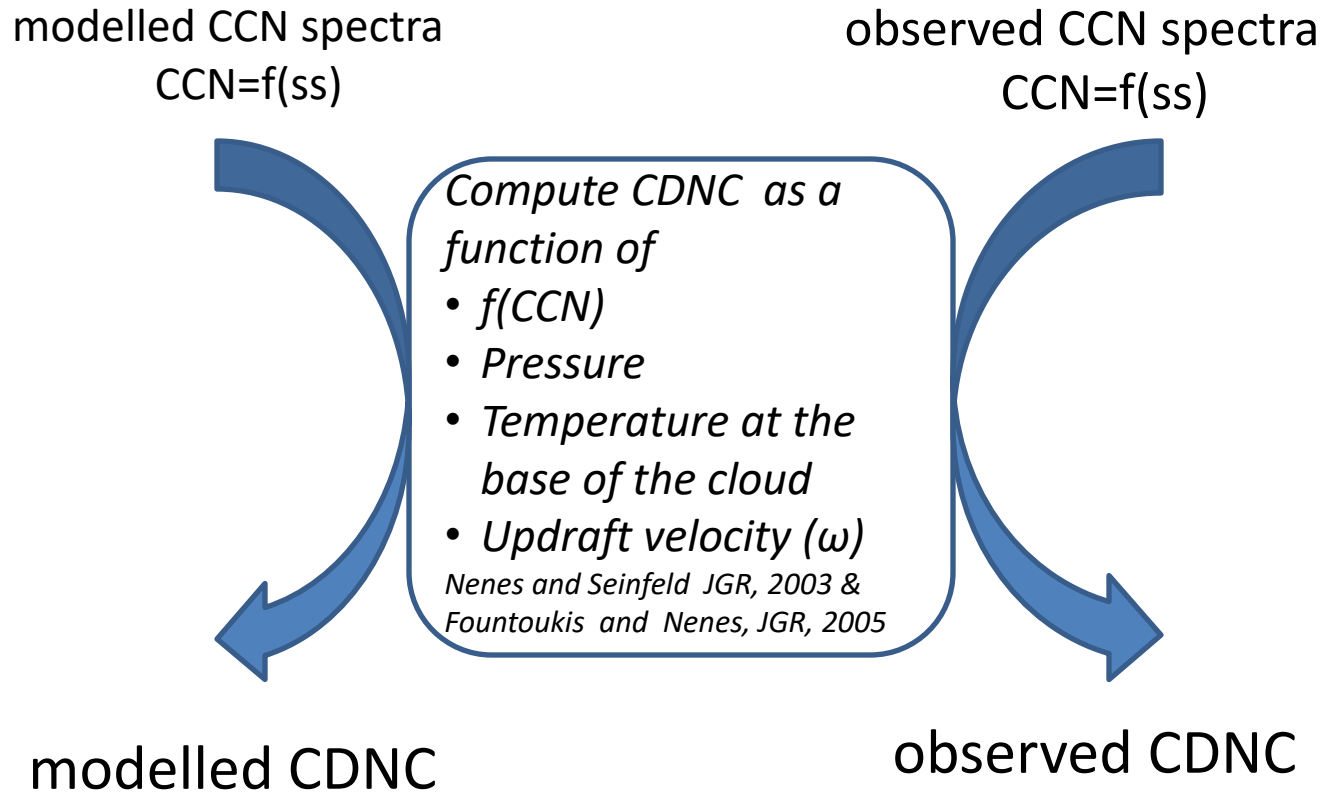
Fanourgakis et al.,
ACP, 2019

Organic aerosol hygroscopicity



HadGEM-UKCA
Yoshioka et al., in
prep.

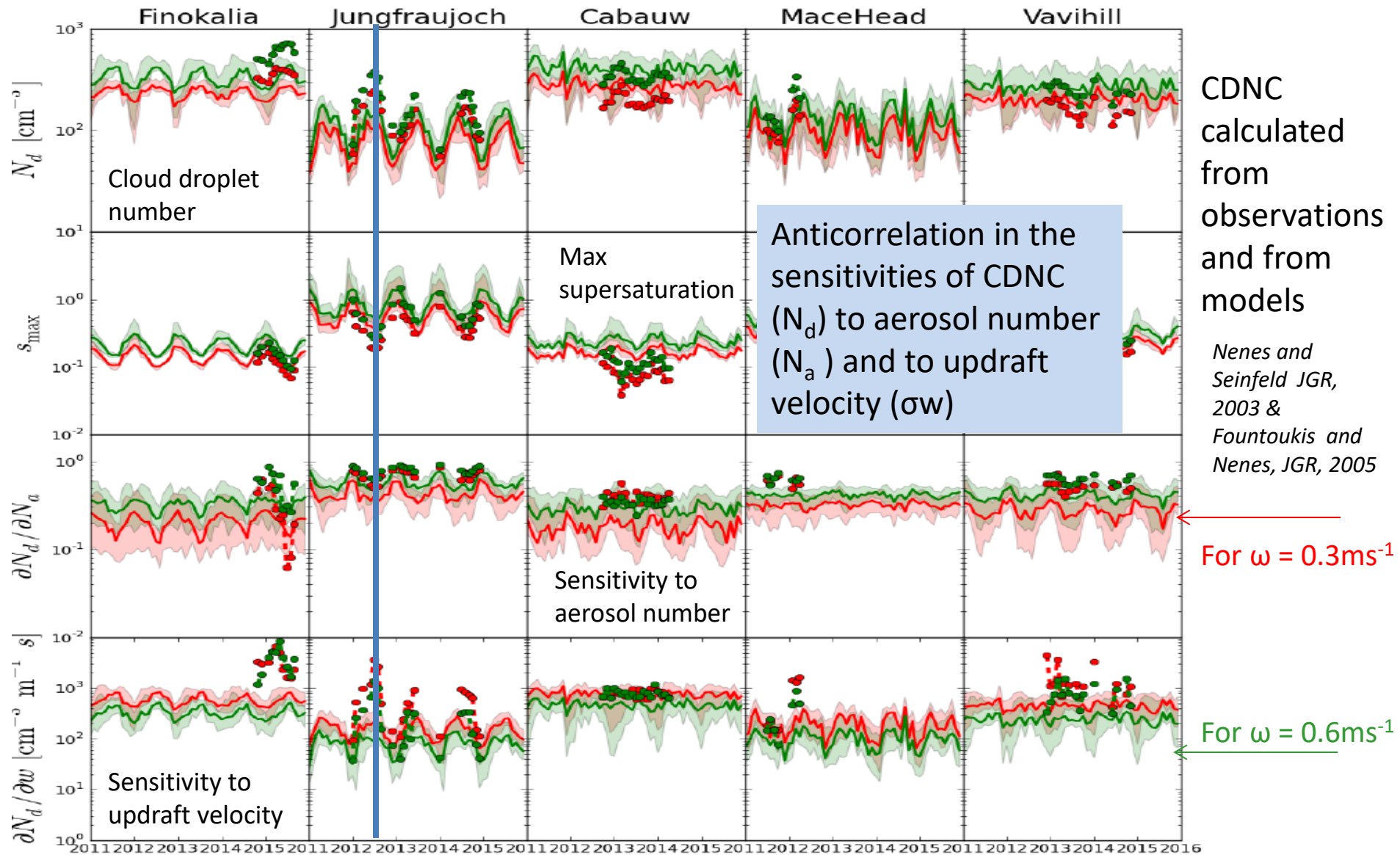
How the CCN uncertainty reflects in CDNC calculations?



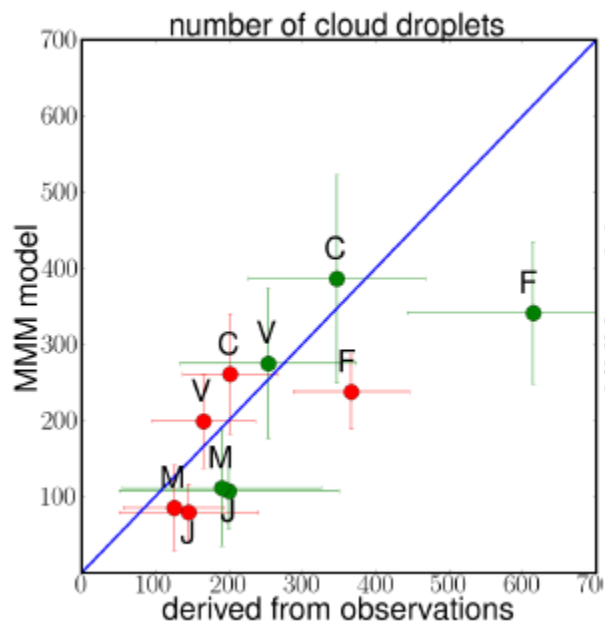
$\sigma_{\omega} = 0.3\text{ms}^{-1}$ typical for stratiform clouds

$\sigma_{\omega} = 0.6\text{ms}^{-1}$ typical for cumulus clouds

Cloud droplet number and its sensitivity to aerosol number and updraft velocity

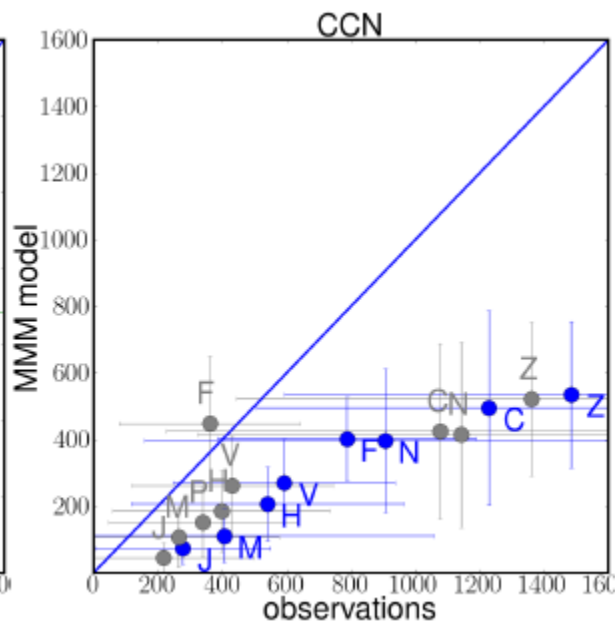


Cloud droplet number and its sensitivity to aerosol number and updraft velocity



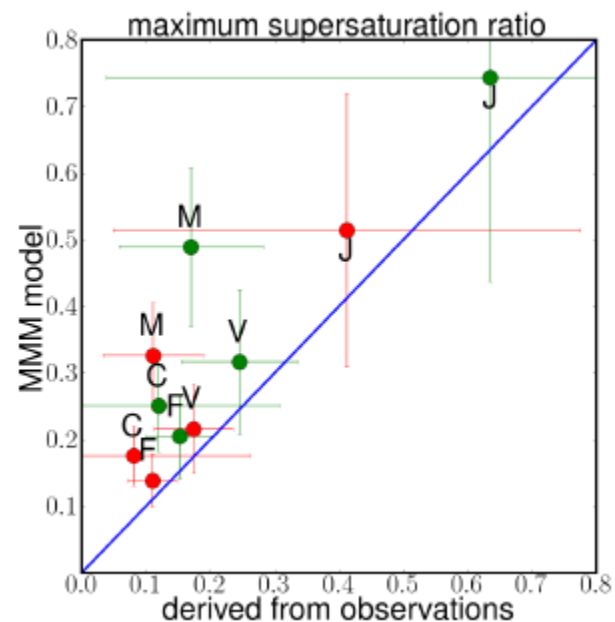
$\sigma_w 0.6 \text{ m s}^{-1}$

$\sigma_w 0.3 \text{ m s}^{-1}$



CCN at supersaturation 0.2%

CCN at max measured supersaturation (0.8%-1.0%)



$\sigma_w 0.6 \text{ m s}^{-1}$

$\sigma_w 0.3 \text{ m s}^{-1}$

The number of CCN at a prescribed supersaturation cannot be used as indicator of CDNC, as supersaturation is dynamically determined and can vary considerably for a given site



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