Aerosol Representation in GCMs

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Thanks to

M. Wang, S. J. Ghan, R. C. Easter, P. Rasch, J. Fast (PNNL)

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DOE ASR Aerosol Lifecycle WG
October 11, 2010

Pacific Northwest NATIONAL LABORATORY

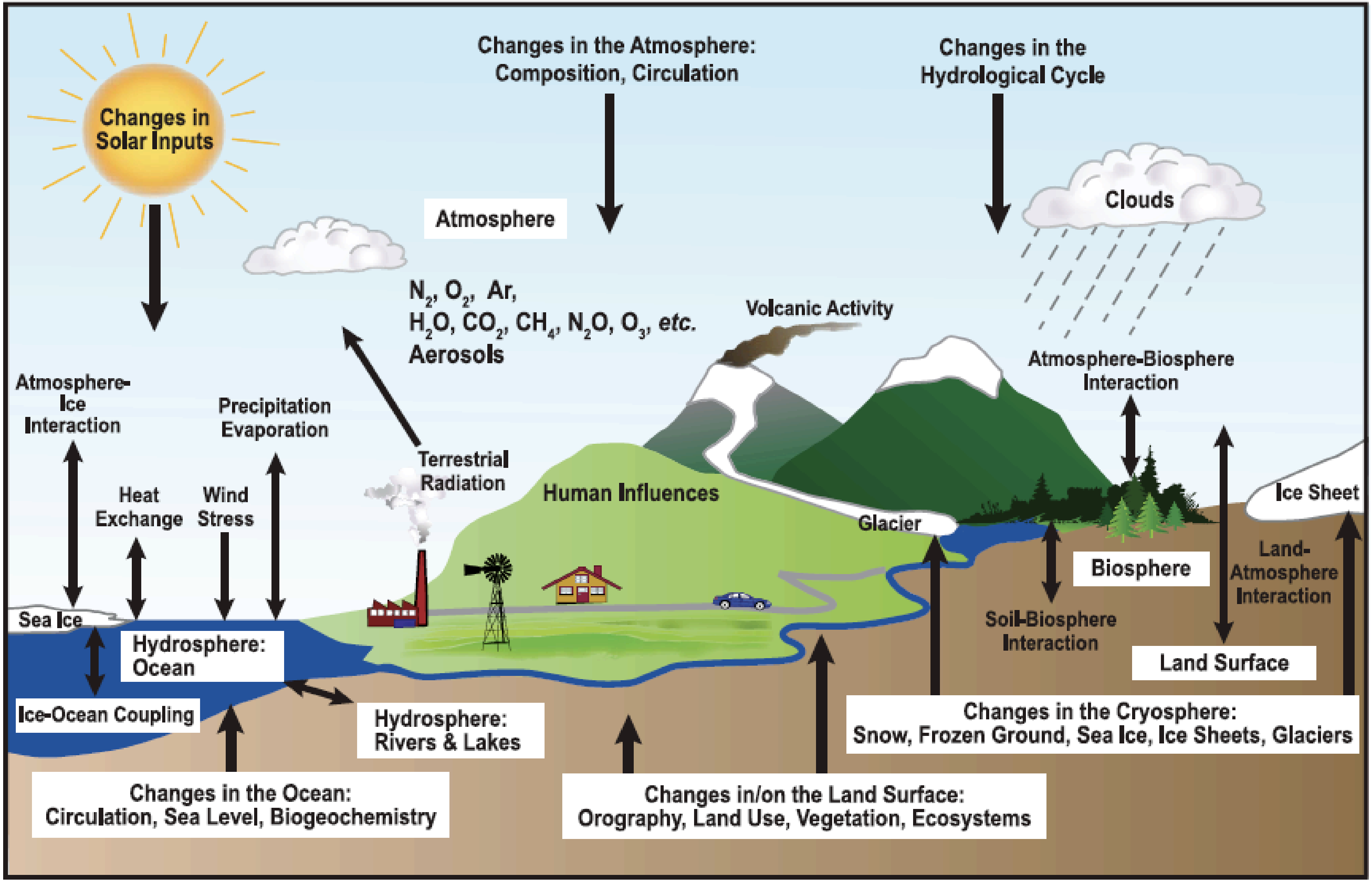
Questions :

1) How are aerosol properties and processes represented in current GCMs (including CAM, GISS, etc)? How do the models compare to each other?
2) What are the major assumptions/simplifications in the representations? What are the weaknesses in current representations?
3) Where are the trouble spots? Which types of aerosol, or which regions in which aerosols are not represented well, and/or simulated aerosols do not agree with existing measurements?
4) Following (2) and (3), how can current representations in GCMs be improved by process studies? What aerosol properties and/or processes need to be better understood and parameterized?

Outline

- Aerosol Representations in GCMs (CAM, GISS, ECHAM)
  - Size representation
  - Processes (sources & sinks)
  - Properties (physical, chemical & optical)
- Uncertainties in Aerosol Processes and Properties in GCMs
  - Primary emissions
  - Secondary aerosol formation (aerosol nucleation & SOA)
  - Water uptake
  - Wet removal
- How Can Aerosol Representation in GCMs be Improved (with the Help of ASR Process Studies)?

Components of the Climate System in GCMs

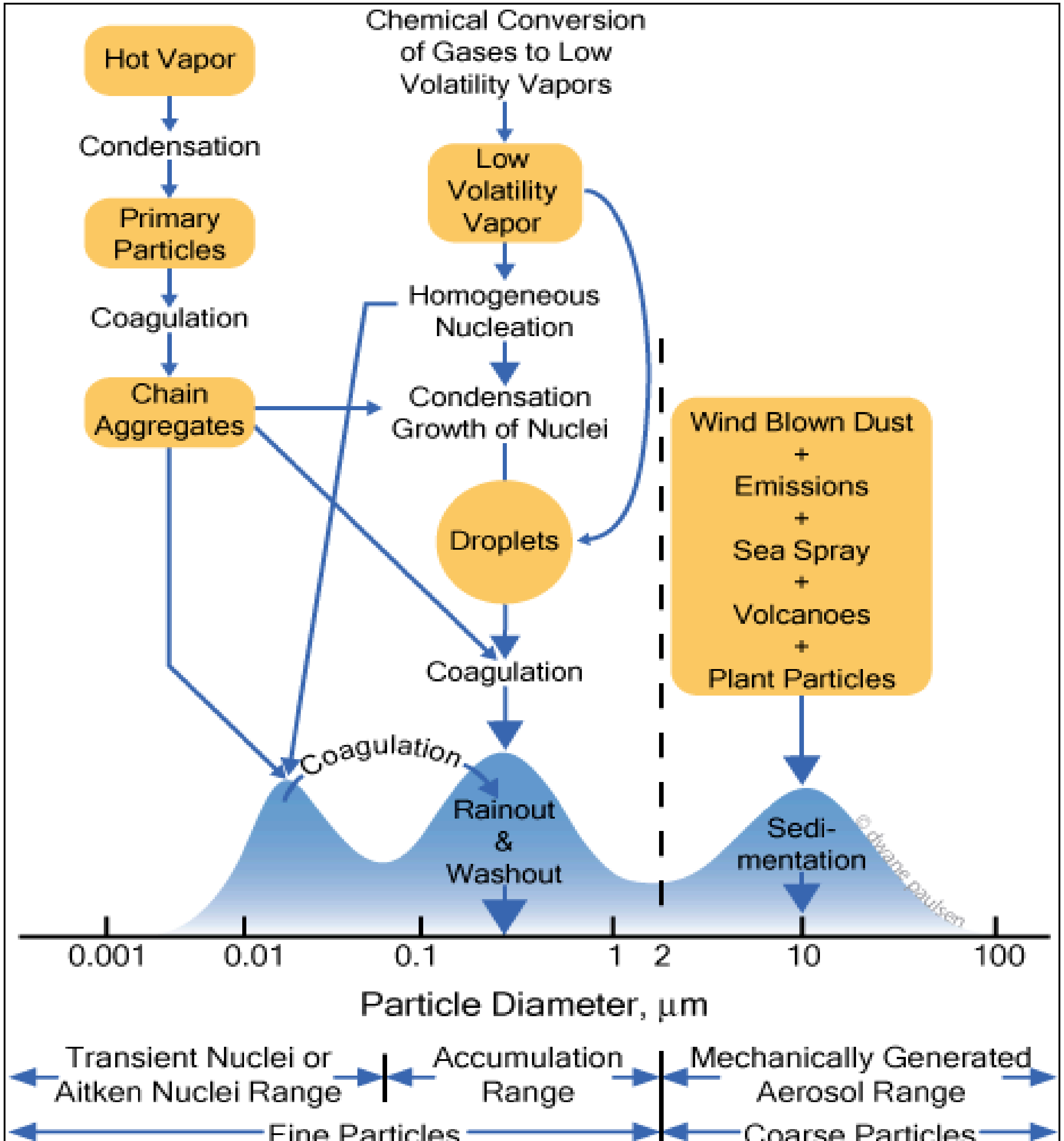


IPCC, 2007

Outline

- Aerosol Representations in GCMs (CAM, GISS, ECHAM)
  - Size representation
  - Processes (sources and sinks)
  - Properties (physical, chemical, and optical)

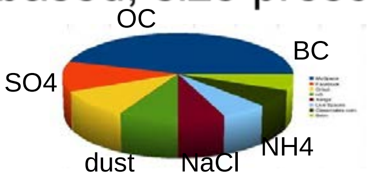
Aerosol Size and Composition in the Atmosphere

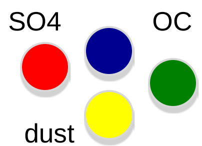


Aerosol Representation in GCMs

- **Bulk**

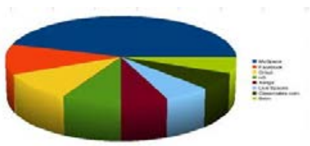
  Mass based, size prescribed, external mixture assumed, no aerosol microphysics

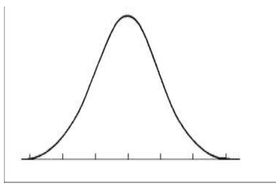


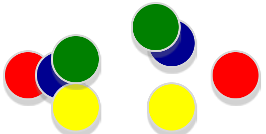


- **Moment-based (modal, 2-moment quadrature method of moments)**

  Assumed functional form of size distributions (log-normal), predict evolution of size distribution by predicting mass (3rd moment) and number (0 moment) mixing ratio in each mode, assumed standard deviation of log-normal, internal mixture within modes and external mixture between modes, aerosol microphysics

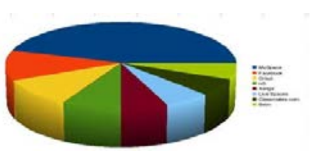


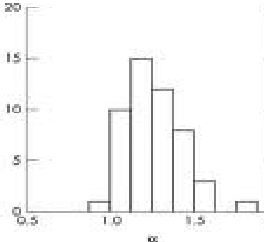


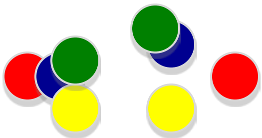


- **Sectional (bin) method**

  Split size distribution into bins, predict evolution of size distribution by predicting mass and number mixing ratio in each bins, aerosol microphysics







Bulk Aerosol Treatment in CAM3

sulfate	hydrophobic black carbon	sea salt 1	soil dust 1
ammonium	hydrophobic organic carbon	sea salt 2	soil dust 2
nitrate	hydrophilic black carbon	sea salt 3	soil dust 3
secondary organic carbon	hydrophilic organic carbon	sea salt 4	soil dust 4

7-Mode Modal Aerosol Model (MAM) in CAM5

Aitken
number
sulfate
ammmonium
secondary OM
sea salt

Accumulation
number
sulfate
ammonium
secondary OM
hydrophobic OM
BC
sea salt

Fine Soil Dust
number
soil dust
sulfate
ammonium

Fine Sea Salt
number
sea salt
sulfate
ammonium

coagulation
condensation

Primary Carbon
number
hydrophobic OM
BC

Coarse Soil Dust
number
soil dust
sulfate
ammonium

Coarse Sea Salt
number
sea salt
sulfate
ammonium

All modes log-normal with prescribed width.

Total transported aerosol tracers: 31

Cloud-borne aerosol and aerosol water predicted but not transported.

Computer time is ~100% higher than Bulk

Simplified 3-mode version of MAM in CAM5

Assume primary carbon is internally mixed with secondary aerosol.

Sources of dust and seasalt are geographically separate

Assume ammonium neutralizes sulfate.

Aitken
number
sulfate
secondary OM
sea salt

Accumulation
number
sulfate
secondary OM
primary OM
black carbon
soil dust
sea salt

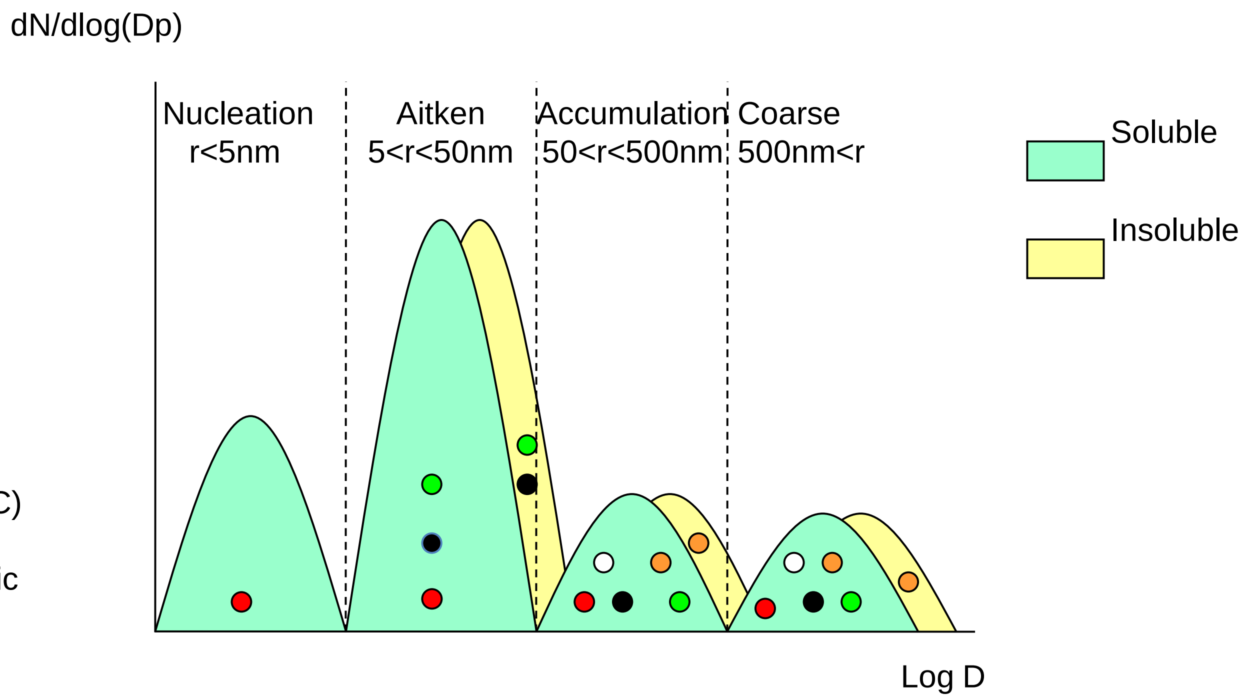
Coarse
number
soil dust
sea salt
sulfate

coagulation
condensation

Total transported aerosol tracers: 15

Computer time is 30% higher than Bulk

Modal Aerosol Module (ECHAM-HAM)



Sulphate (SU)

Black Carbon (BC)

Particulate Organic Matter (POM)

Sea Salt (SS)

Dust (DU)

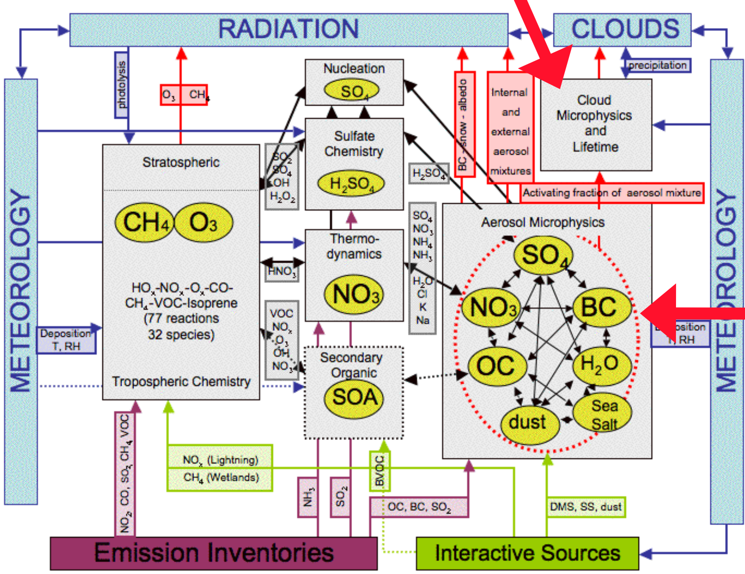
Predicted variables per mode:

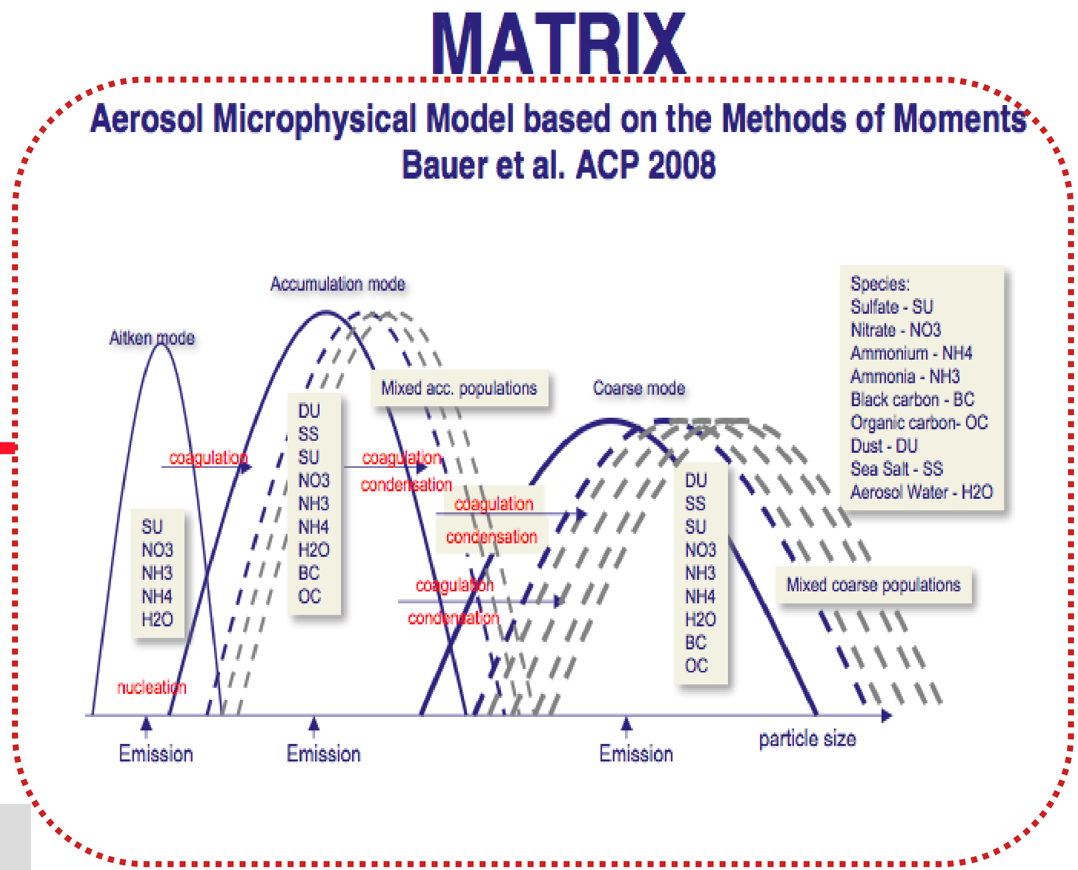
One number concentration and the mass mixing ratios of each chemical compound

Courtecy of Declan O'Donnell

GISS-MATRIX (QMOM)

Droplet activation:
Abdul Razzak and Ghan (1998, 2000)
Cloud droplet nucleation follows prognostic treatment of Morrison et al. 2005, 2008



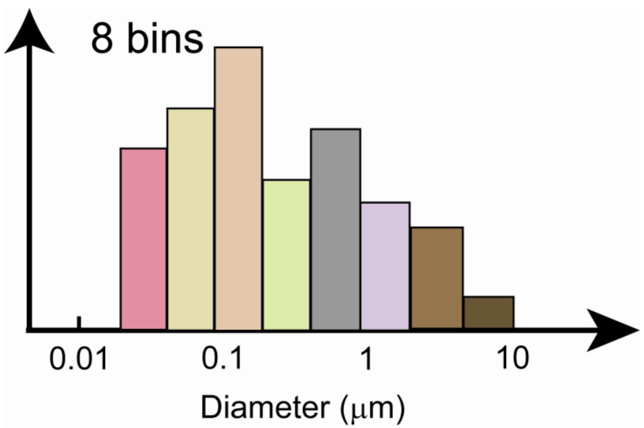


Aerosol Microphysics:

- Simulation of aerosol mass, mixing state and size distributions (1). Needed for:
- **Indirect effects**: Microphysical parameter. of aerosol - cloud activation (1,2)
- **Direct effects**: Radiation scheme coupled to aerosol shape and mixing state information (3)

(1) Bauer et al., Atmos. Chem. Phys., 2008
(2) Menon et al., Atmos. Chem. Phys, to be submitted
(3) Bauer et al., Atmos. Chem. Phys., 2010

Sectional Aerosol Treatment in CAM5



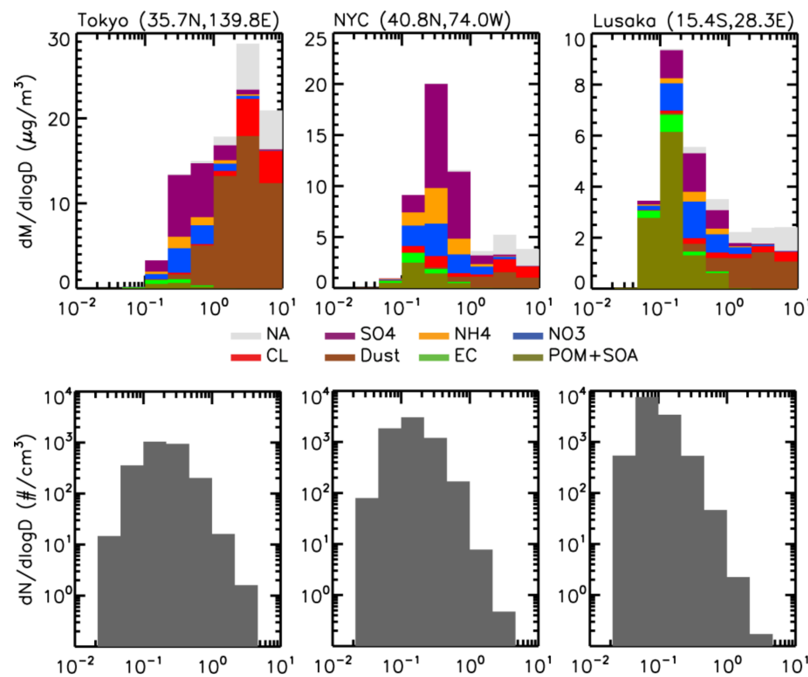
[Aerosol microphysics]

Based on MADRID 1 [Zhang et al., 2004]

- 27 aerosol species in each bin
  - Na, SO_4, NH_4, NO_3, Cl, Dust, H_2O, EC, POM
  - 14 biogenic SOAs, 4 anthropogenic SOAs
- 216 aerosol mass components
- 8 aerosol number components

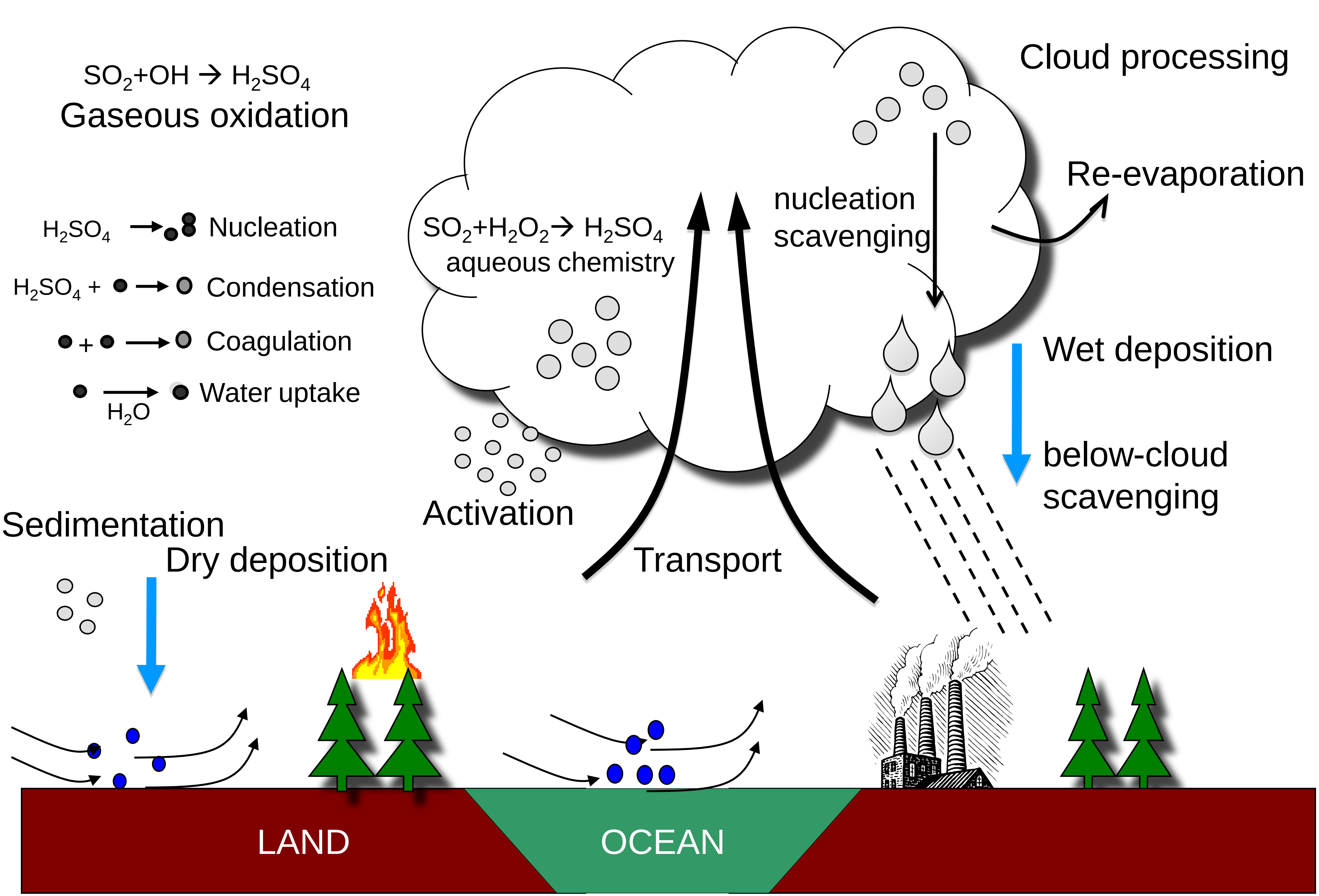
[Preliminary test using LLNL global chemistry/aerosol model, IMPACT]

Simulated aerosol mass (upper) and number (lower) distributions in regions of Tokyo, New York City, and Lusaka by IMPACT using 2004 GEOS4 meteorology.



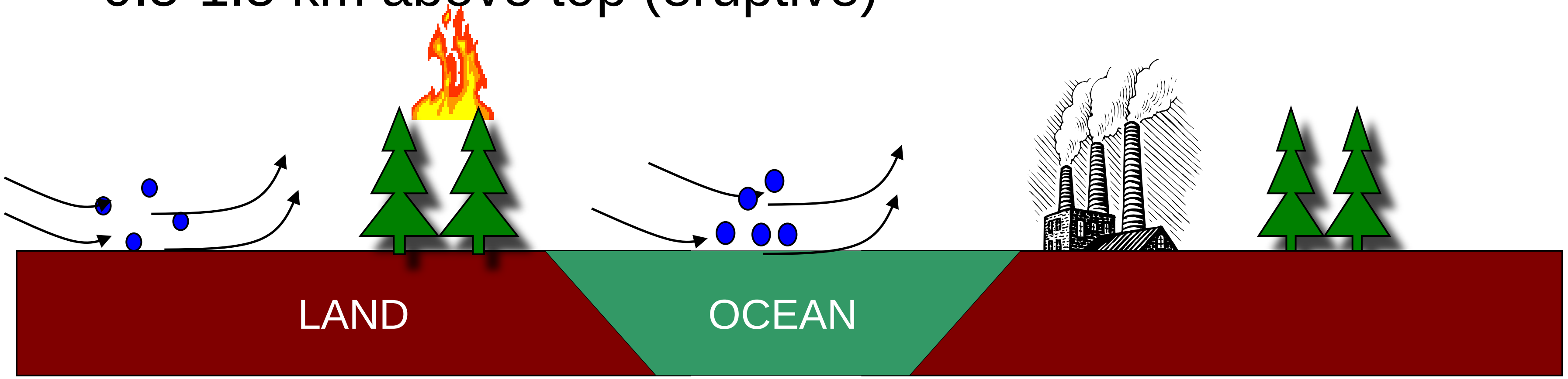
Cathy Chuang, Oct 2010

Global Aerosol Cycles



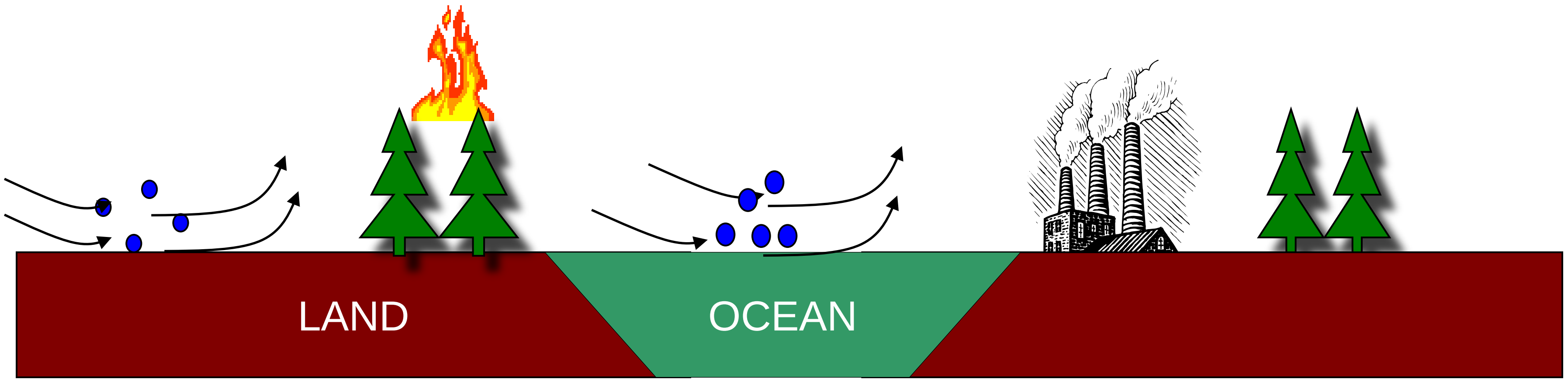
Aerosol Processes : Primary Emission

- Offline emission mass flux (for SO_2, POA, BC, DMS): prescribed from inventory
- Online emission mass flux (for dust, sea salt, ocean POA): f(u, r, soil moisture or ocean concentrations)
- Injection Heights:
  - Most emission fluxes applied at surface (lowest grid box), power plant SO_2 ~ 100-300 m;
  - Biomass burning applied an injection height profile;
  - Volcanic emission at 2/3-1/1 of volcano top (continuous) and 0.5-1.5 km above top (eruptive)

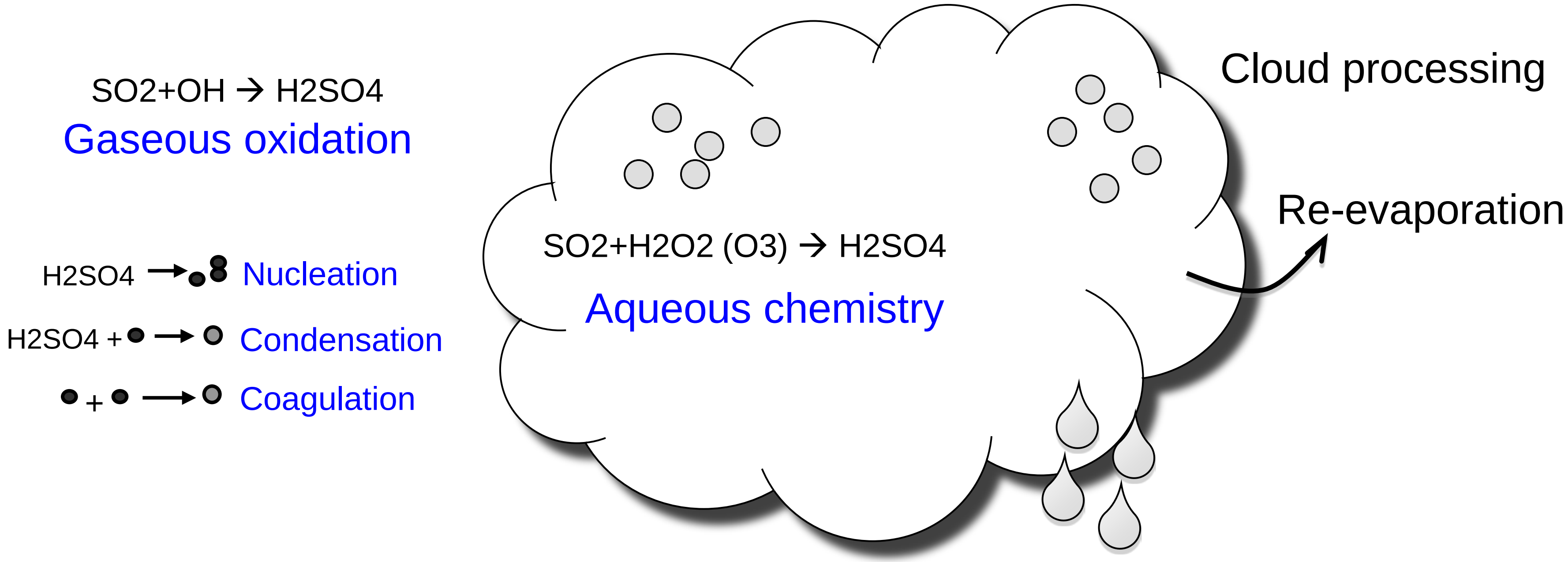


Aerosol Processes : Primary Emission

- Emission Number Flux:
  - Emission size distribution prescribed.
  - Fossil fuel having smaller emission size than biomass burning and biofuel.



Aerosol Processes (Secondary SO_4 Formation)



All models: include gas and aqueous phase SO_2 chemistry

Bulk models: assume instantaneous conversion of H_2SO_4 (g) to sulfate, no nucleation/condensation/coagulation

Modal (bin) models:

Nucleation of $H_2SO_4/NH_3/H_2O$: form new particles

Condensation of H_2SO_4/NH_3/SOA : thermo-dynamical transport, increase mass

Coagulation : reduce number

Aqueous chemistry: bulk chemistry depends on pH values, produces mass distributed to aerosol modes (bins) in proportional to number activated from modes (bins)

Aerosol Processes (SOA Formation)

Earlier Approaches:

SOA formed by assuming a fixed 15% SOA yield from the monoterpene emissions estimates of Guenther et al. (1995), with immediate non-volatile SOA production. Treat formed SOA as primary organics. ~15 Tg OC/yr.

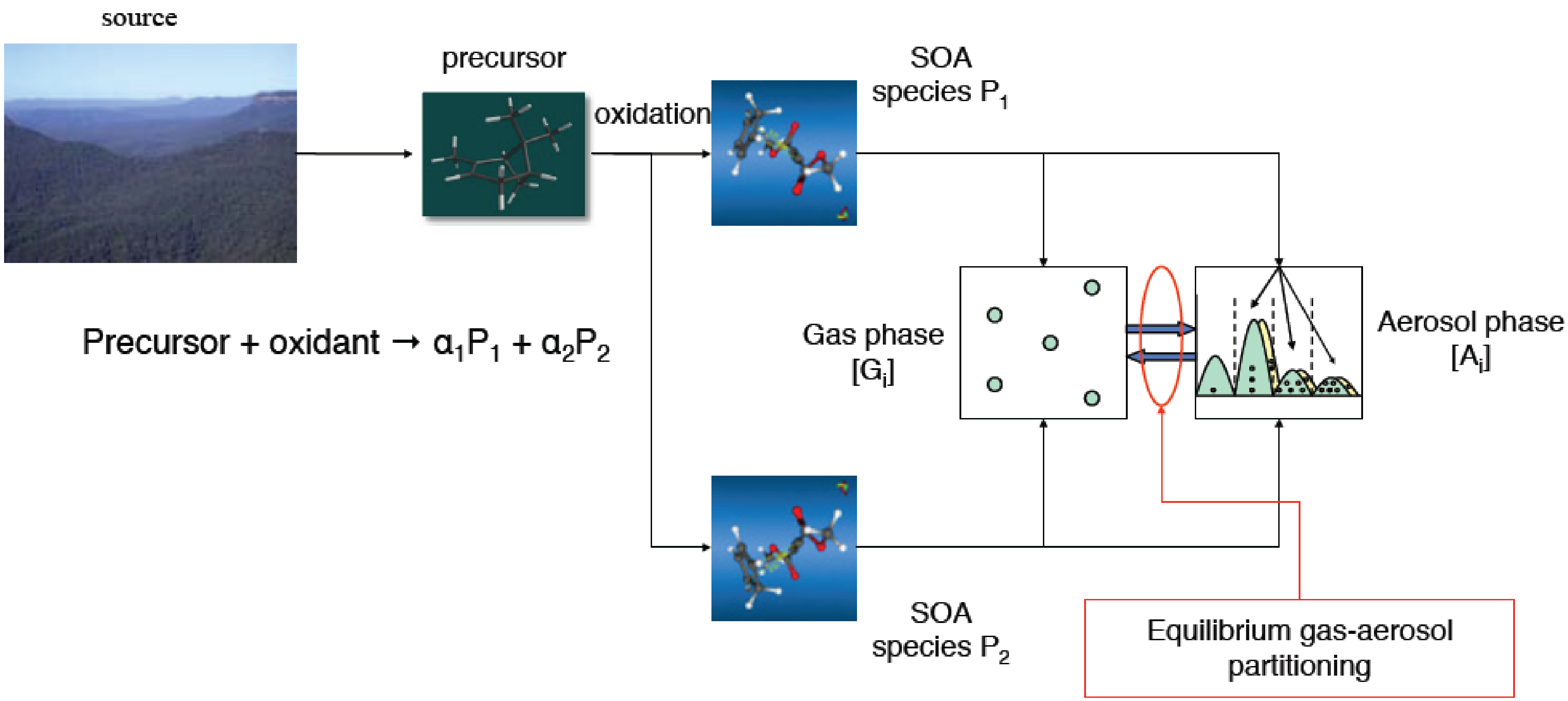
Newer Approaches:

Prognostic SOA scheme with explicit gas/aerosol partitioning

One step of more complexity : assumed fixed yields for biogenic and anthropogenic VOCs to form SOA (g). Treat SOA (g) as primary gas emission at surface. explicit gas/aerosol partitioning of SOA (g) -- CAM5.

Two steps of more complexity : primary VOCs emission and oxidation in atmosphere to form SOA (g). explicit gas/aerosol partitioning of SOA (g) – ECHAM & GISS.

SOA scheme in ECHAM-HAM2



Aerosol Processes (Nucleation)

CAM5: Ternary $H2SO_4$-NH_3-H_2O nucleation in MAM7 (Merikanto et al, 2007)
Binary H_2SO_4-H_2O nucleation in MAM3 (Vehkamaki et al. 2002);
Boundary layer nucleation: empirical 1st order nucleation rate
in H_2SO_4 (Sihtp et al. ,2006) with the rate coefficient of $1.0x10^{-6}$ s^{-1}

GISS: Ternary $H2SO_4$-NH_3-H_2O nucleation (Napari et al., 2002)
Binary H_2SO_4-H_2O nucleation (Jaecker-Voirol and Mirabel, 1989; Vehkamaki et al. 2002)

ECHAM:

Old: Binary H_2SO_4-H_2O nucleation (Vehkamaki et al. 2002);
New: Include charged nucleation induced by cosmic ray (Kazil et al., 2010)

Kerminen and Kulmala (2002) approach used to account for coagulation loss of new particles as they grow from critical cluster size (~1 nm) to Aitken mode size

Aerosol Processes (Aging)

Earlier Approaches:

Prescribed 1-2 days aging time from hydrophobic to hydrophilic for OC and BC – Bulk models

Instantaneous aging : assumed primary OC/BC mixing with other components instantly -- CAM5-MAM3, a good assumption for OC/BC away from sources. Underestimate OC/BC at remote regions due to wet scavenging

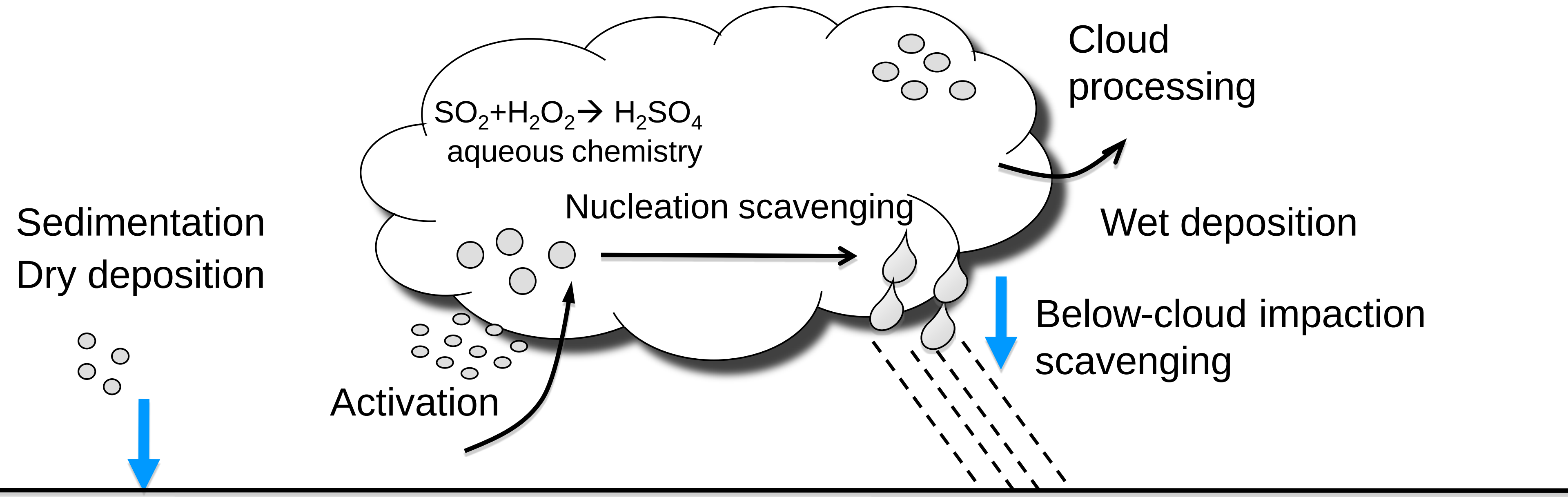
Newer Approaches:

Aging depending on coating of soluble materials : primary OC/BC aged to mixed mode depending on the surface coating of soluble materials (SO_4, NH_4, SOA, NO_3) – CAM5-MAM7, ECHAM & GISS

Aerosol Processes (Water Uptake)

CAM5:	Thermodynamical equilibrium based on K-Kohler theory. Volume mean K from each component for each mode. Hysteresis (averaging upper and lower curves between deliquesce and crystallization RH).
GISS:	Thermodynamical equilibrium based on EQSAM. E. Lewis formula for sea salt
ECHAM:	Old: ZSR method (Zdanovskii-Stokes-Robinson) New: K-Kohler theory

Aerosol Processes (Removal)



Dry Deposition : most models use the classical serial resistance approach.

$$F_d = C\rho_a v_d \qquad v_d = v_g + \frac{1}{r_a + r_s}$$

Wet Deposition : most models use conversion rate of cloud water to rain and precipitation rate, P_r/Q_c

Earlier models: prescribed soluble (activated) fraction depending on aerosol species (in-cloud nucleation scavenging); below-cloud scavenging coefficient (c_0) assumed

Improved models:

CAM5 : predicting aerosols in cloud water (through activation, aqueous chemistry, diffusion, and evaporation); size dependent of c_0

Caveat: very simple cloud microphysics in convective clouds

Aerosol Properties in GCMs

(CAM5, GISS, ECHAM)

- Mass and composition
  - interactive SO4, POA, SOA, BC, dust and sea salt,
  - ammonium, nitrate often not treated (CAM-MAM3, ECHAM)
- Size distribution
  - variable for each mode, or QMOM
- Mixing state
  - internal and external mixture
- Radiative properties and refractive index
  - parameterized in terms of bulk wet refractive index and wet mode radius or look-up tables
- Hygroscopicity
  - volume average of K from components in each mode

Outline

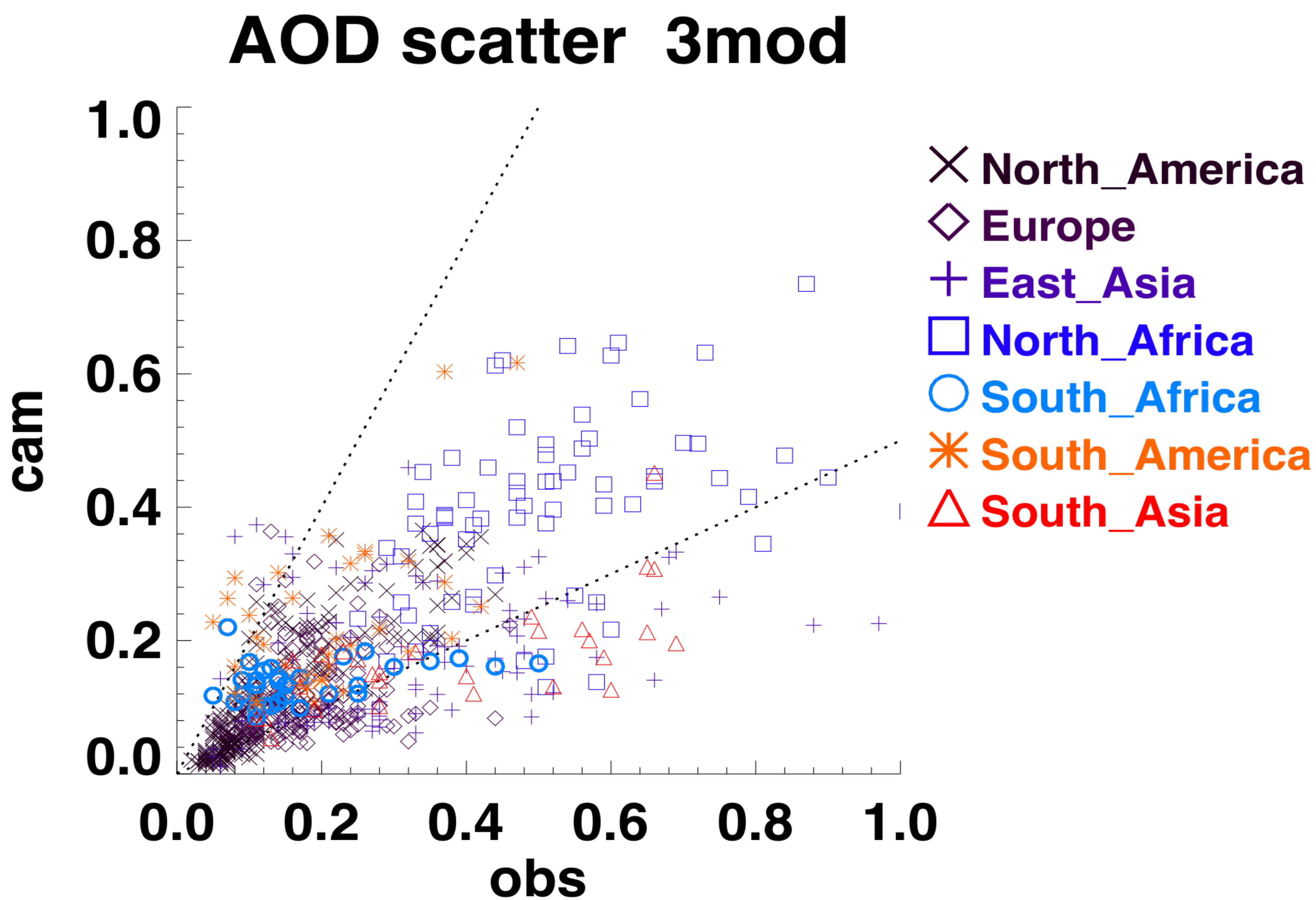
- Aerosol Representations in GCMs (CAM, GISS, ECHAM)
  - Size representation
  - Processes (sources, sinks)
  - Properties (physical, chemical, optical)
- Uncertainties in Aerosol Processes in GCMs
  - Primary emissions
  - Secondary aerosol formation (nucleation & SOA)
  - Water uptake
  - Wet removal

Uncertainties in Aerosol Processes in GCMs (1)

- Primary emissions: mass flux, size distribution, injection height

  - Anthropogenic emissions in developing counties
  - Biomass burning emissions
  - Mineral dust and sea salt emissions
    - Dust: 1640 Tg/yr ± 50% (AEROCOM-A); 3200 Tg/yr (CAM5)
    - Sea salt: 6280 Tg/yr ± 200% (AEROCOM-A); 5000 Tg/yr (CAM5)

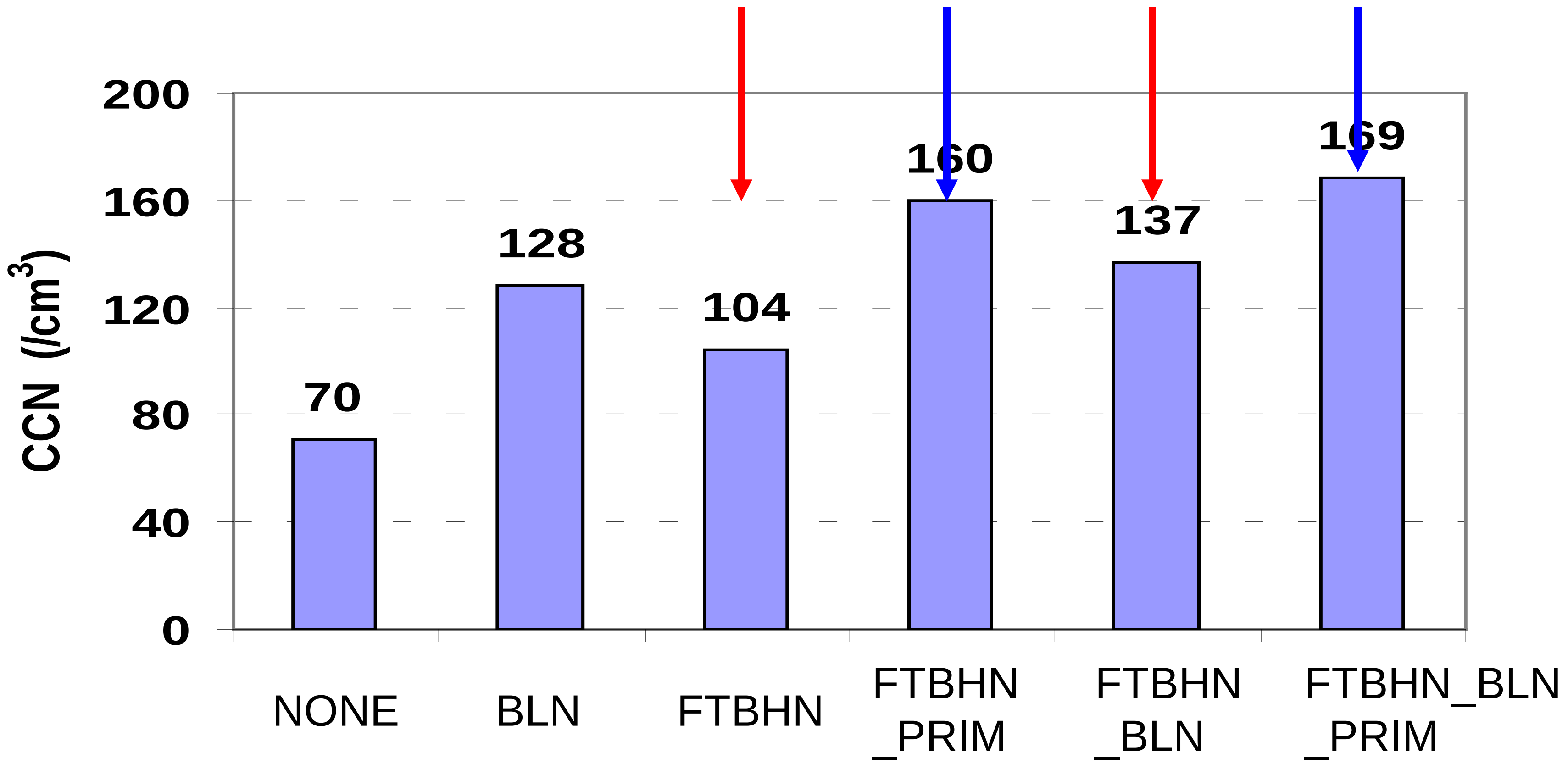
  - Primary organics from oceans


AOD scatter 3mod
cam
obs
0.0
0.2
0.4
0.6
0.8
1.0
North_America
Europe
East_Asia
North_Africa
South_Africa
South_America
South_Asia

Uncertainties in Aerosol Processes in GCMs (2)

- Secondary aerosol formation
  - Aerosol nucleation (in free troposphere and BL): how important to CCN in terms of climate effects?
  - SOA production and properties

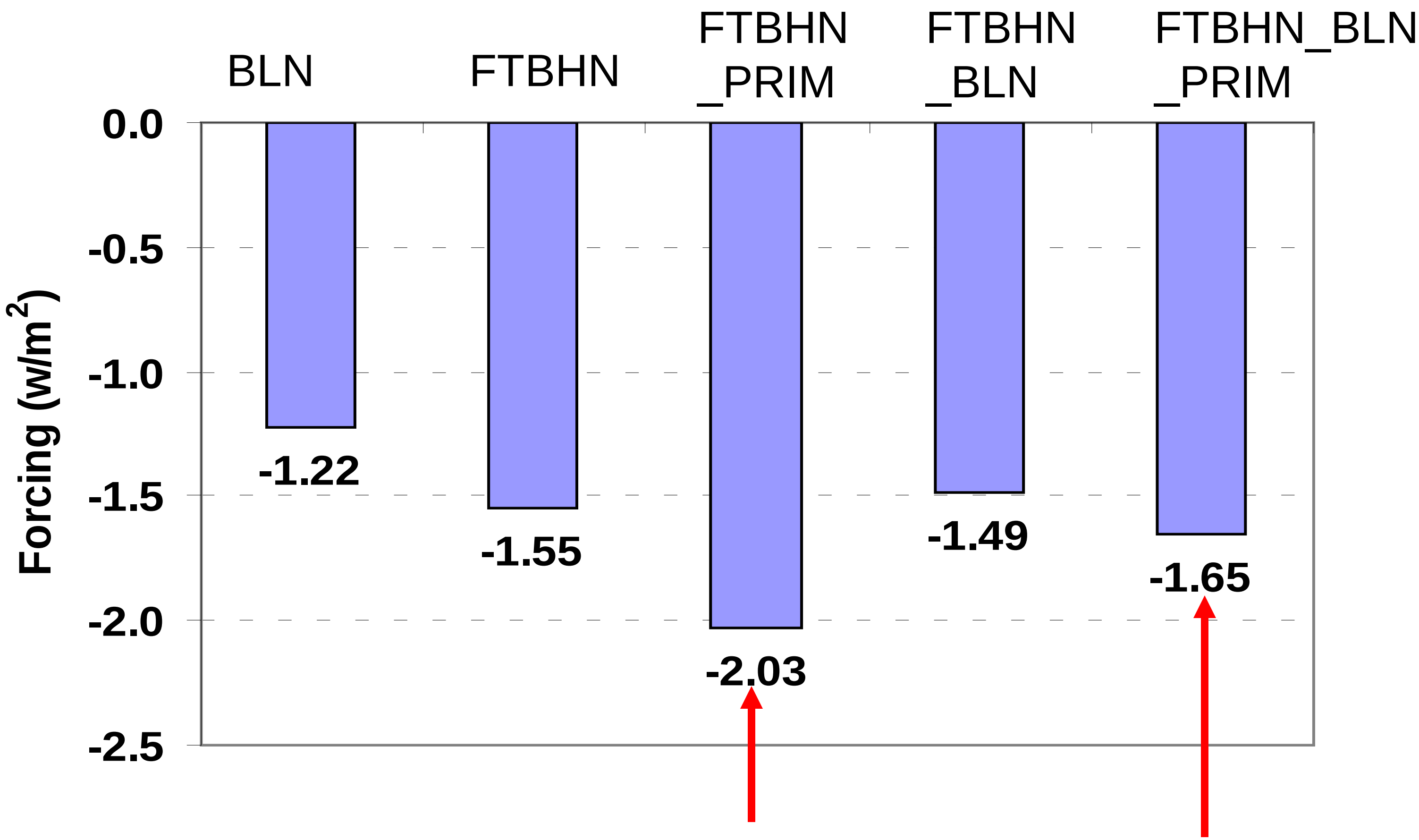
CCN CONCENTRATION IN THE BOUNDARY LAYER (930 hPa)



Case name	BLN	FTBHN	PRIM
NONE	NO	NO	NO
BLN	YES	NO	NO
FTBHN	NO	YES	NO
FTBHN_PRIM	NO	YES	YES
FTBHN_BLN	YES	YES	NO
FTBHN_BLN_PRIM	YES	YES	YES

Wang & Penner (2008)

AEROSOL FIRST INDIRECT FORCING



The forcing from various treatments of aerosol nucleation ranges from -1.22 to -2.03 W/m^2.

Wang & Penner (2008)

Effect of the new SOA scheme

Original

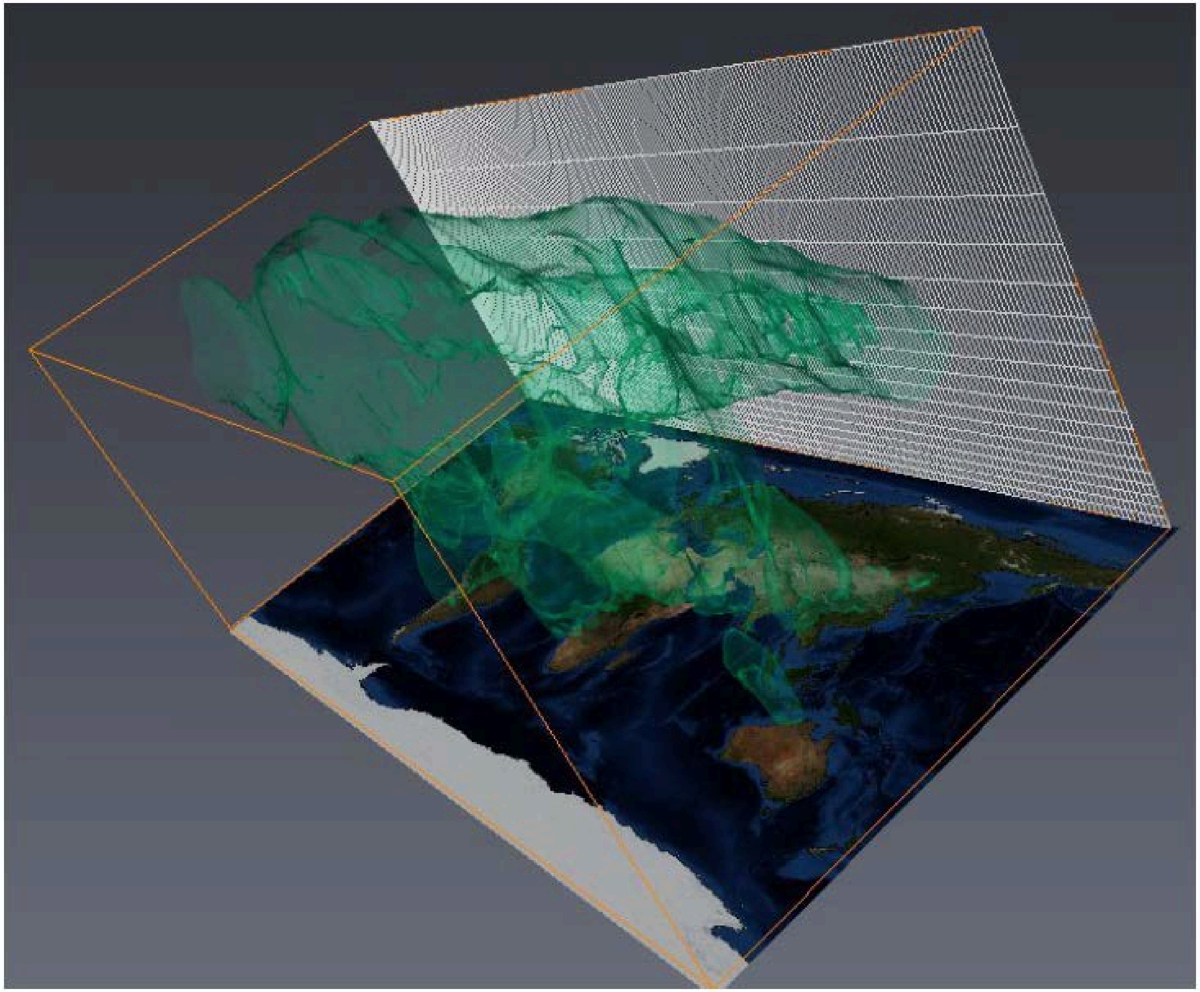
“organic carbon” refers to POA plus SOA formed by assuming a fixed 15% SOA yield from the monoterpene emissions estimates of Guenther et al. (1995), with immediate non-volatile SOA production.

New

Prognostic SOA scheme with explicit gas/liquid partitioning

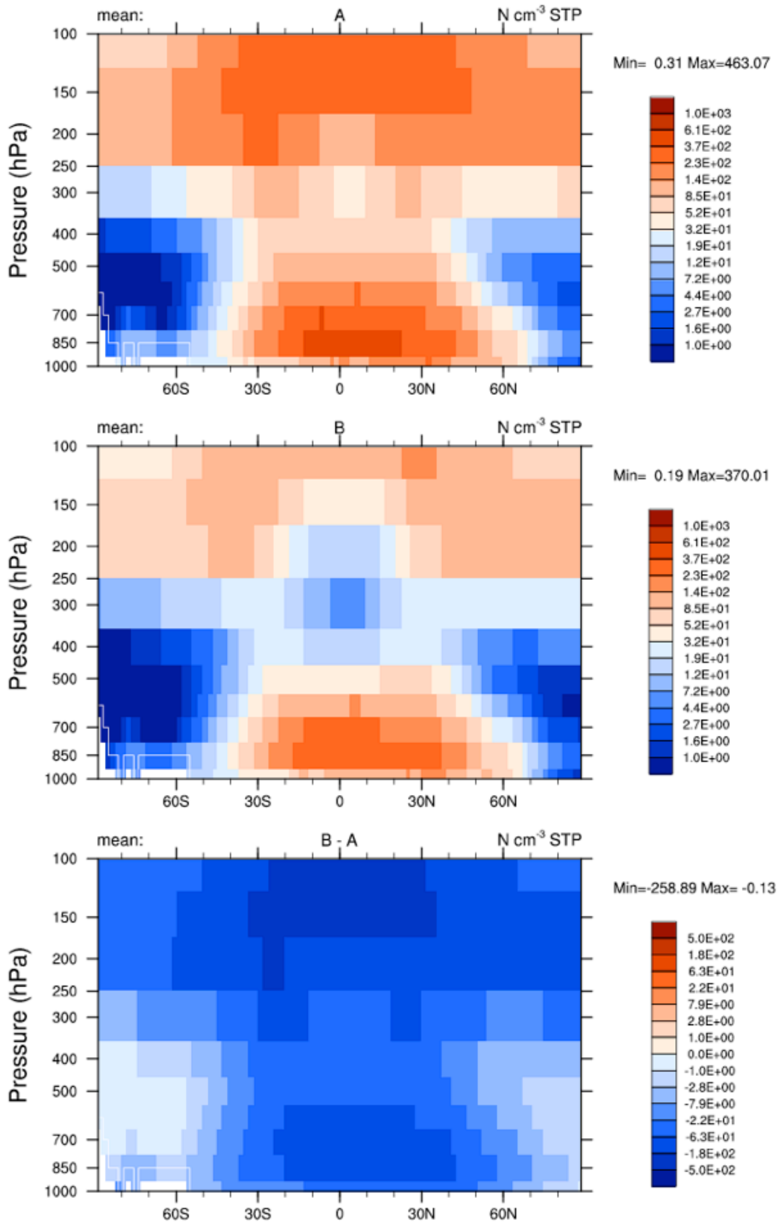
(from K. Zhang, ECHAM-HAM)

Annual mean vertical distribution of high-volatility isoprene SOA



O'Donnell (2010)

Aerosol number (soluble accumulation mode)



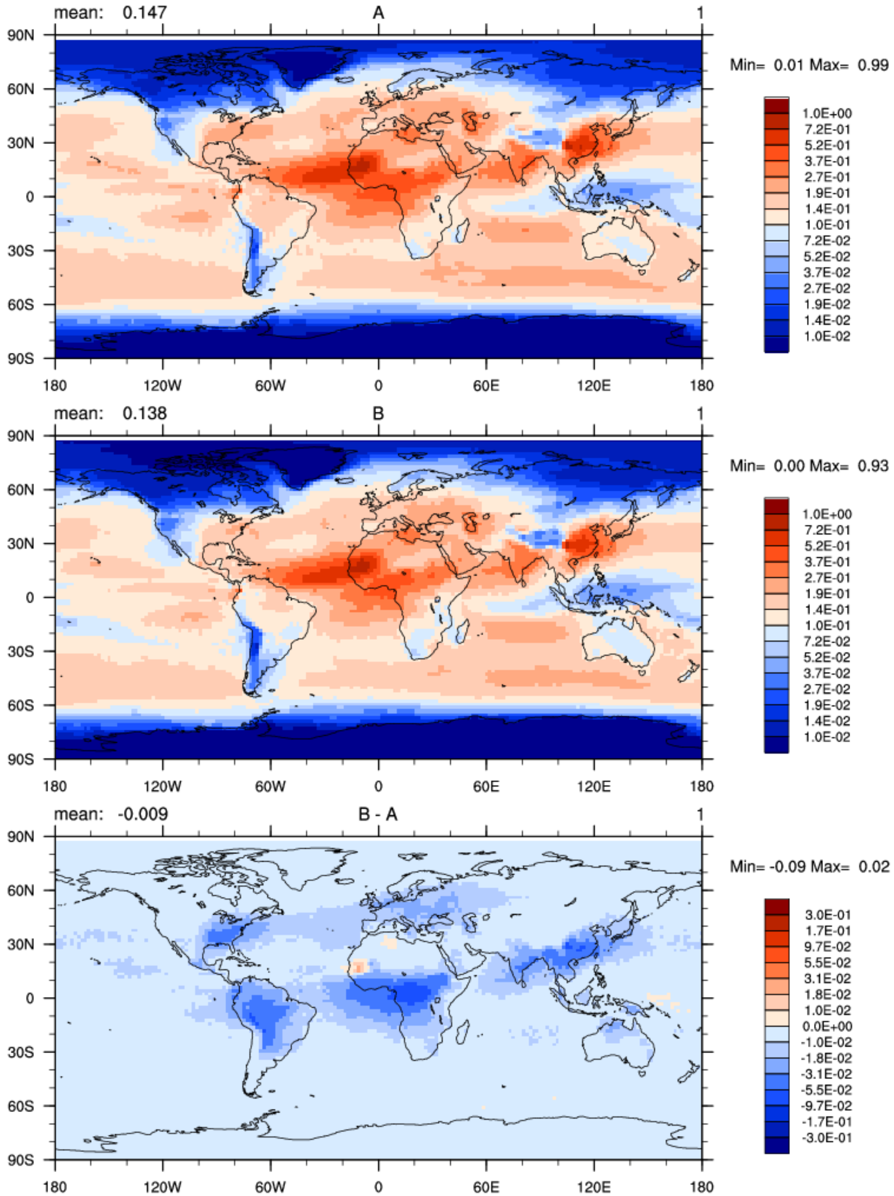
A: CTRL (with explicit SOA)

B: without explicit SOA

B - A

much more accumulation mode particles in the upper atmosphere

Aerosol optical depth (AOD)



A: CTRL (with explicit SOA)

B: without explicit SOA

B - A

without explicit treatment of SOA, global mean AOD decreases by ~7%

Uncertainties in Aerosol Processes in GCMs (3)

Effect of water uptake schemes

Original: ZSR based scheme

- take aerosol as a solution of mixed electrolytes
- sensitive to high RH

Jacobson et al. JGR-1996

New: Κ-Köhler theory based scheme

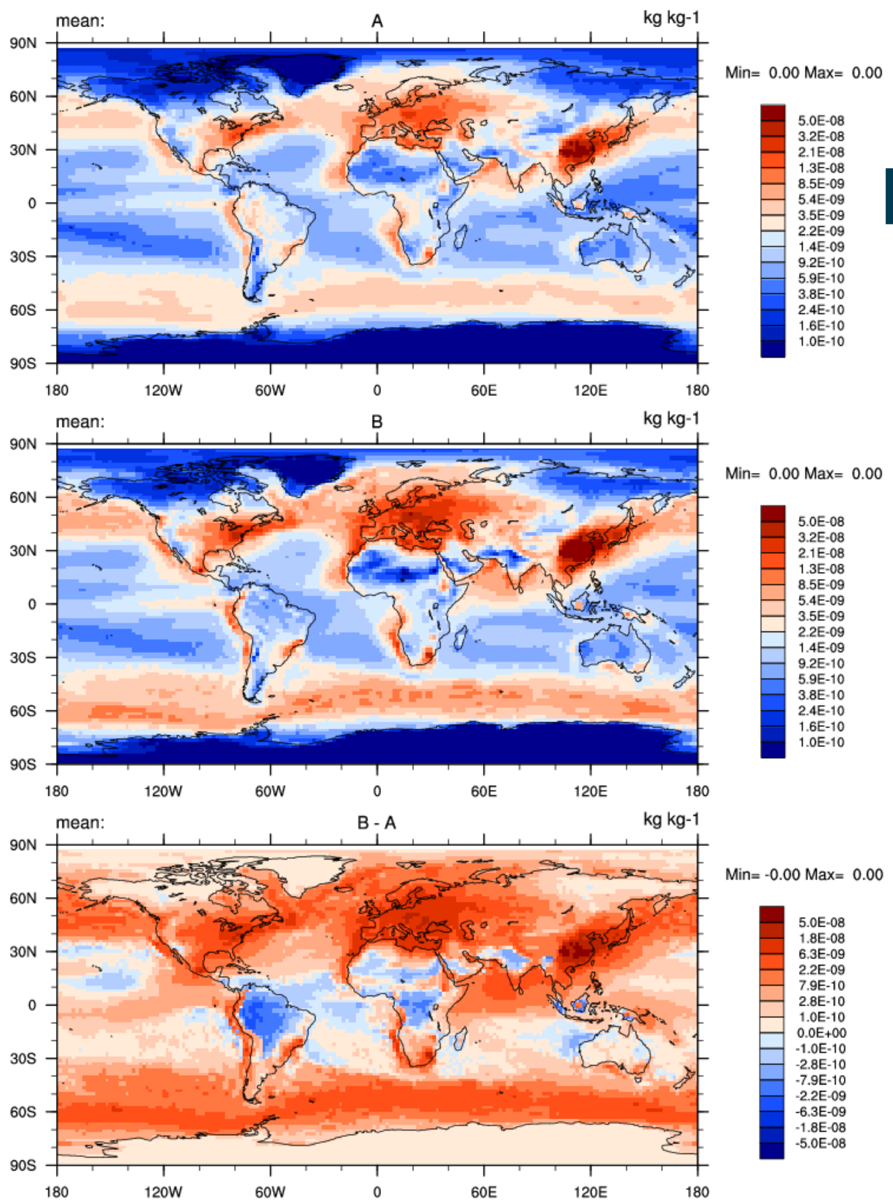
- can easily be applied for non-electrolytes (e.g. organic specie)
- a hygroscopicity parameter κ for each chemical component

Petters and Kreidenweis ACP-2007

Growth factor of an aerosol particle can be expressed as a function of temperature, relative humidity, aerosol dry diameter and Κ

K. Zhang (ECHAM-HAM)

Aerosol water content (soluble accumulation mode)



κ method (new)

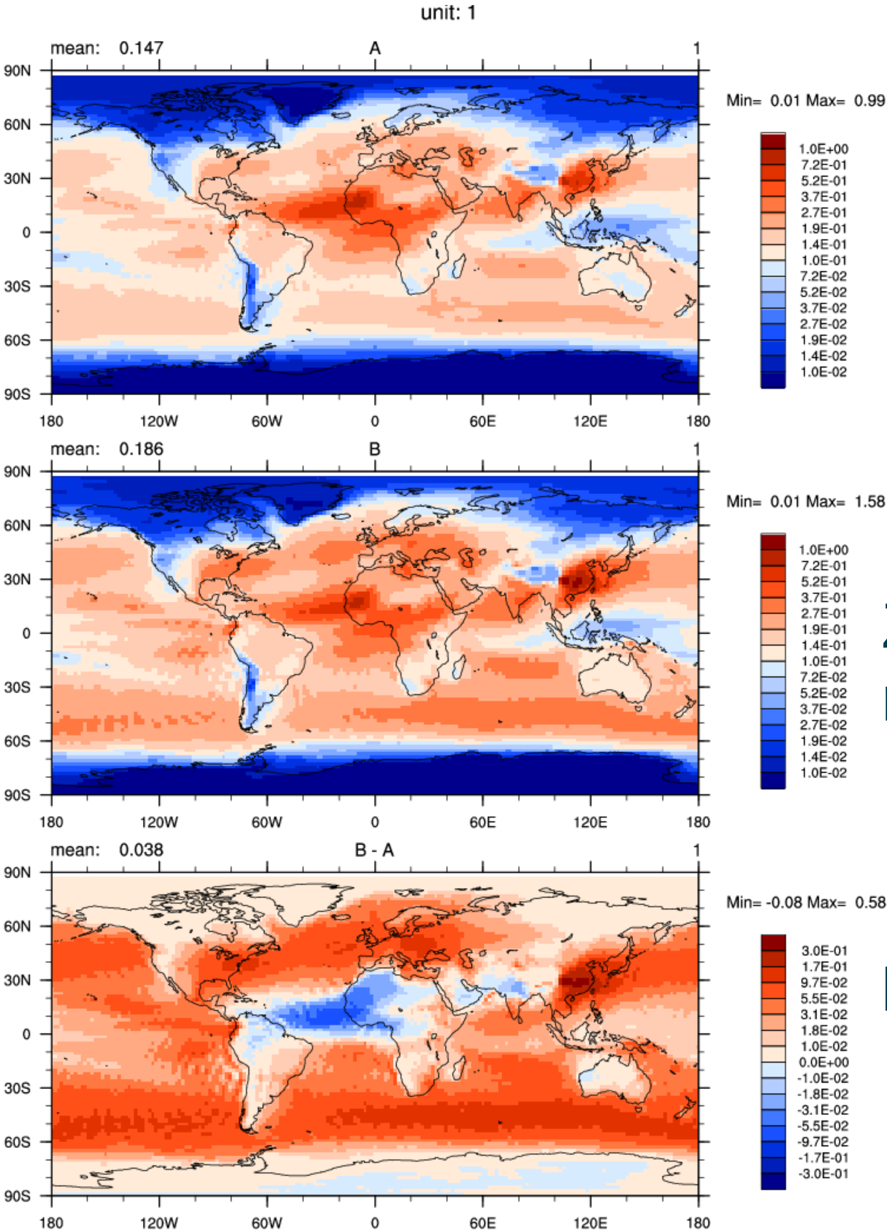
ZSR method

Difference (ZSR - κ)

The ZSR based method produces much larger aerosol water-uptake over both the ocean and the industrial area

K. Zhang (ECHAM-HAM)

Aerosol optical depth (AOD)



K method (new)

ZSR method

Difference (ZSR - K)

with the new scheme, global mean AOD decreases by 0.04 (~25%)

K. Zhang (ECHAM-HAM)

Uncertainties in Aerosol Processes in GCMs (4)

- Wet removal
  - Precipitation rate (conversion of cloud water to rain)
  - Sub-grid cloud and precipitation processes
  - Cloud microphysics in convective clouds

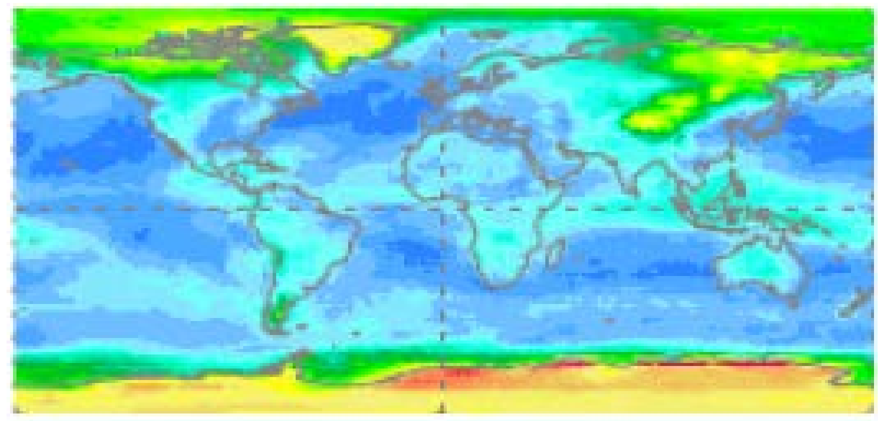
Aerosol Models Have Particular Trouble Simulating Aerosol Beyond the Polar Front

- Most relative uncertainty in simulated AOD/mass poles.
- Arctic aerosol sources primarily from midlatitudes.
- Uncertainty in transport treatment unlikely to cause x10-uncertainty.
- Large uncertainty could be from treatment of wet scavenging.

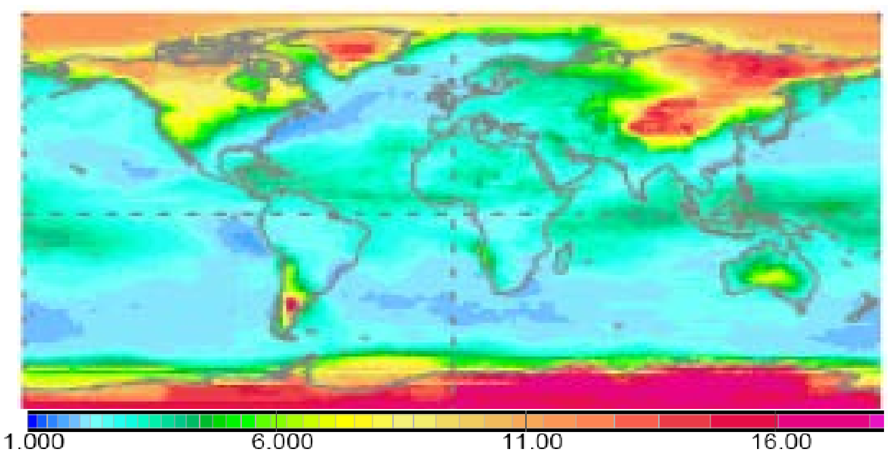
Major differences in poles

Max/Min of Central 2/3 of !6 Models

Aerosol Optical Depth

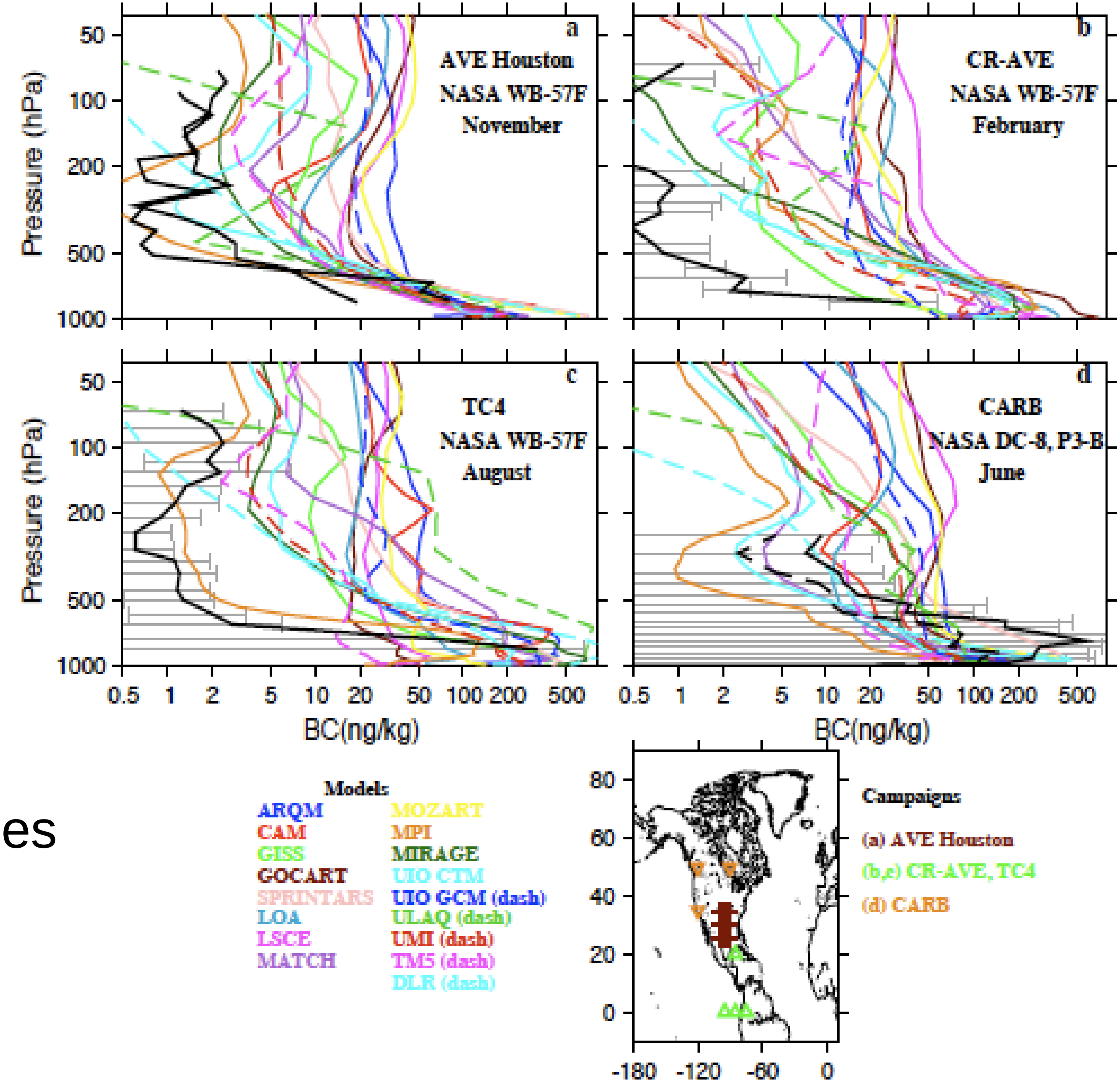


Aerosol Column Mass



Kinne et al., An AeroCom initial assessment. *Atmos. Chem. & Phys.*, 2006.

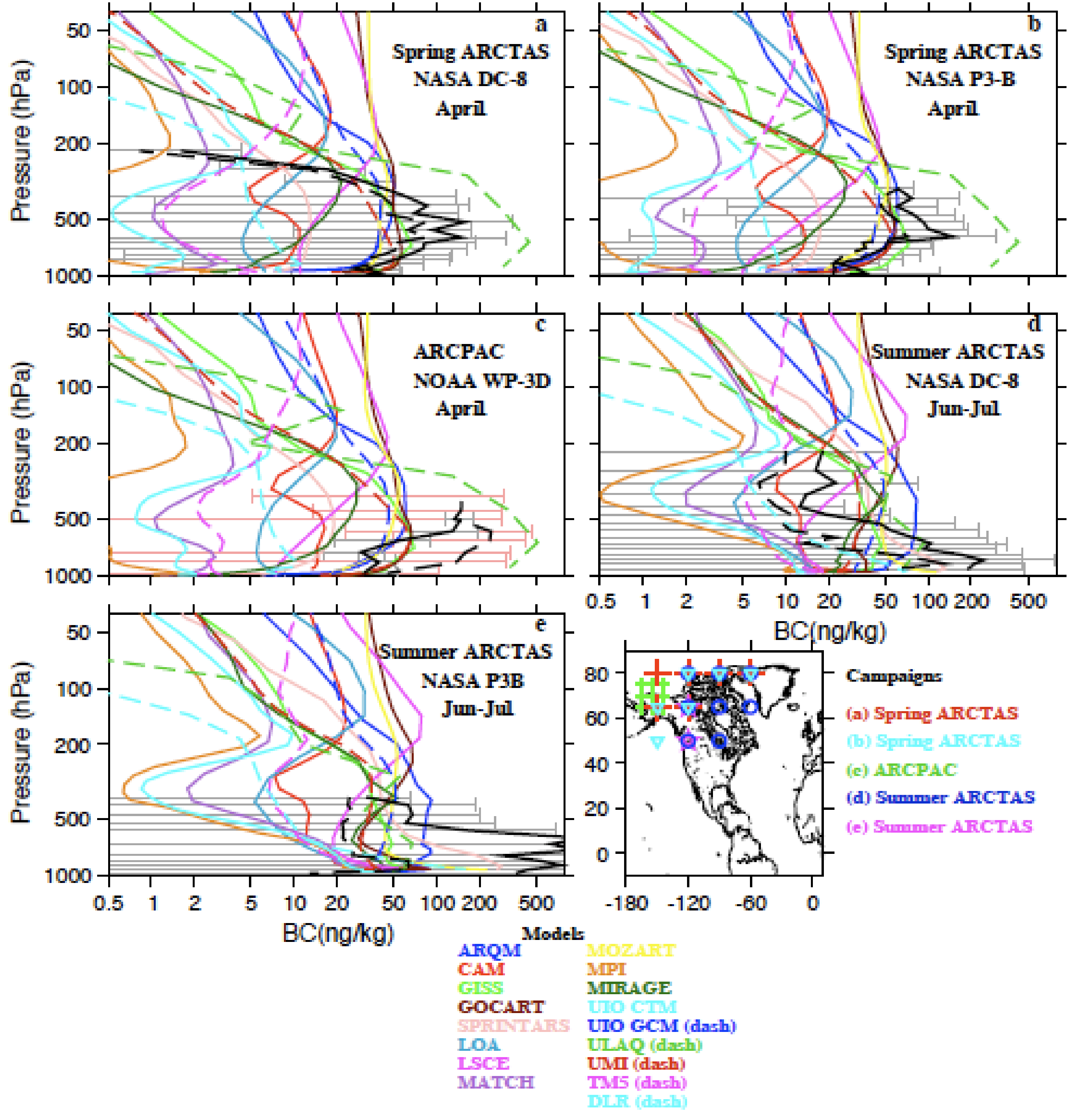
BC compared with SP2 (tropics and midlat.)



Major differences in free troposphere

Koch et al. (2009)

BC compared with SP2 (highlat.)



Koch et al. (2009)

Outline

- Aerosol Representations in GCMs (CAM, GISS, ECHAM)
- Uncertainties in Aerosol Processes and Properties in GCMs
- How Can Aerosol Representation be Improved in GCMs (with the Help of ASR Process Studies)?

How Can Aerosol Representation in GCMs be Improved (with the Help of ASR Process Studies)?

Processes :

- Improve primary emissions: size distribution and injection heights, organics from oceans
- Aerosol nucleation and growth (BL nucleation, role of organics)
- SOA production and evaporation
- Water uptake
- Wet scavenging (cloud and precipitation in GCMs, link to CAPI & CLWG)

Properties :

- Hygroscopicity of organics and mineral dust
- Mixing state (e.g., BC)
- Refractive index (dust, brown carbon)

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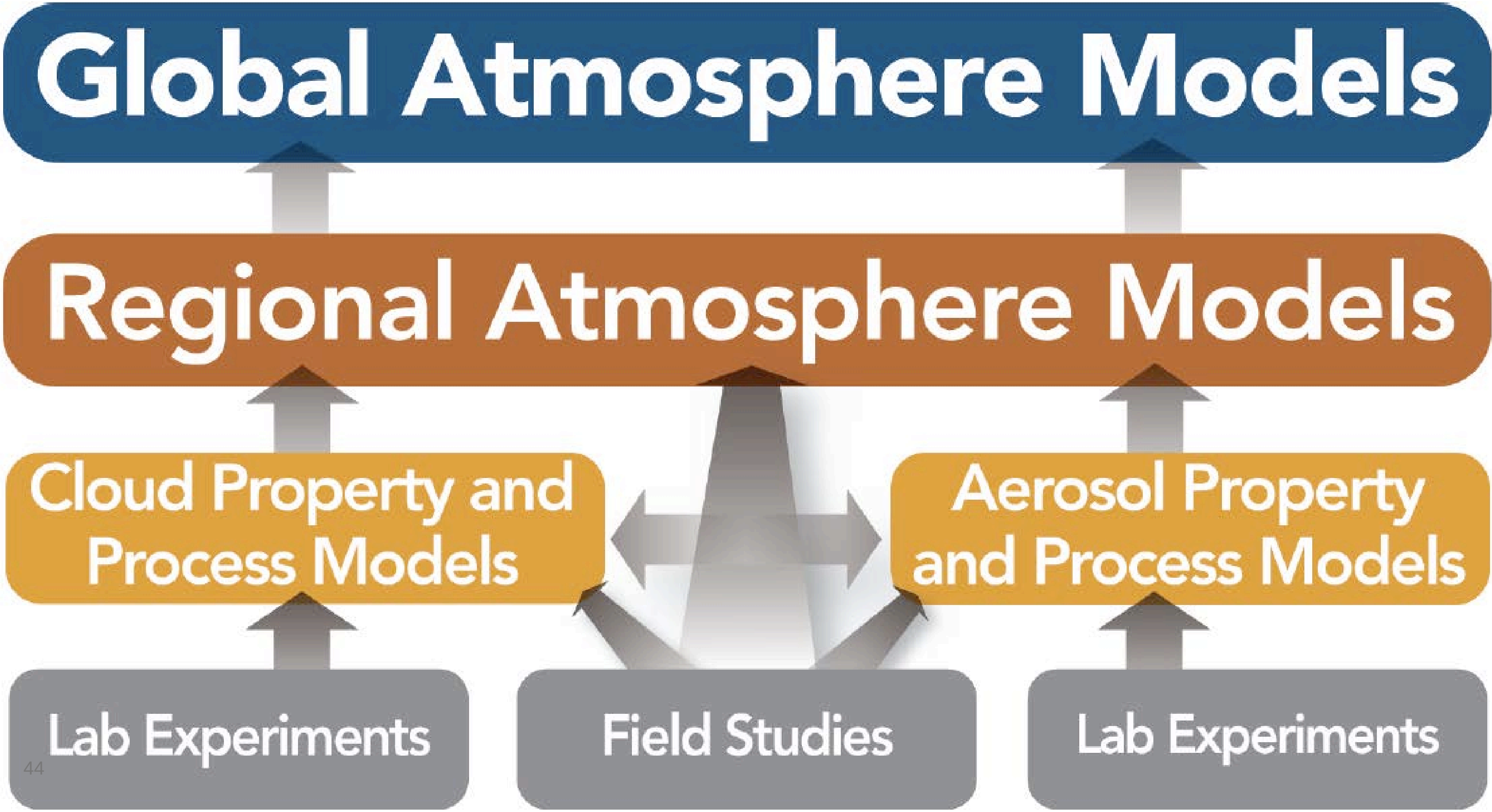
Road Map from Process Studies to GCMs
(Ghan and Schwartz, BAMS, 2007)



THANKS!

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