# Overview of Aerosols in GCMs

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# Liu Group at Univ. of Wyoming

# Questions :

1) What are main sources and sinks of the sulfate and black carbon lifecycles, respectively?
2) What roles do aerosols play in the Earth's climate system?

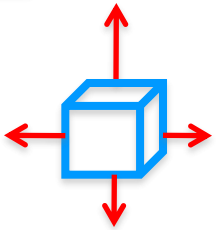
# Outline

- Aerosol Representations in GCMs
  - Processes (sources & sinks)
  - Properties (physical, chemical & optical)
- Uncertainties in Aerosol Processes and Properties in GCMs
  - Primary emissions
  - Secondary aerosol formation (aerosol nucleation & SOA)
  - Wet removal

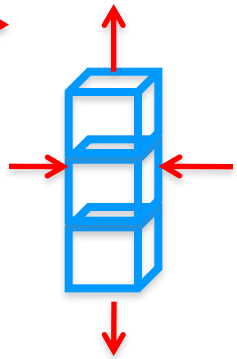
# Host Models

**Box Model**
0D, no transport, no external forcing

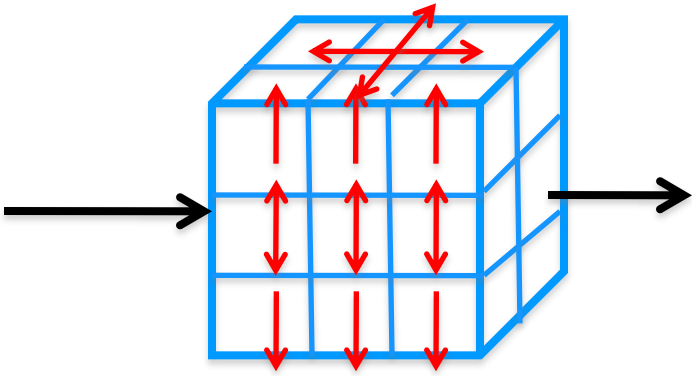
**Parcel Model**
0D, moved by prescribed external forcing



**Single Column Model (SCM)**
1D, vertical transport
External forcings (e.g., campaign)



**Chemical Transport Model (CTM)**
3D, regional or global
Met fields prescribed from GCMs or reanalysis,
no feedbacks of aerosol & chemistry on met fields

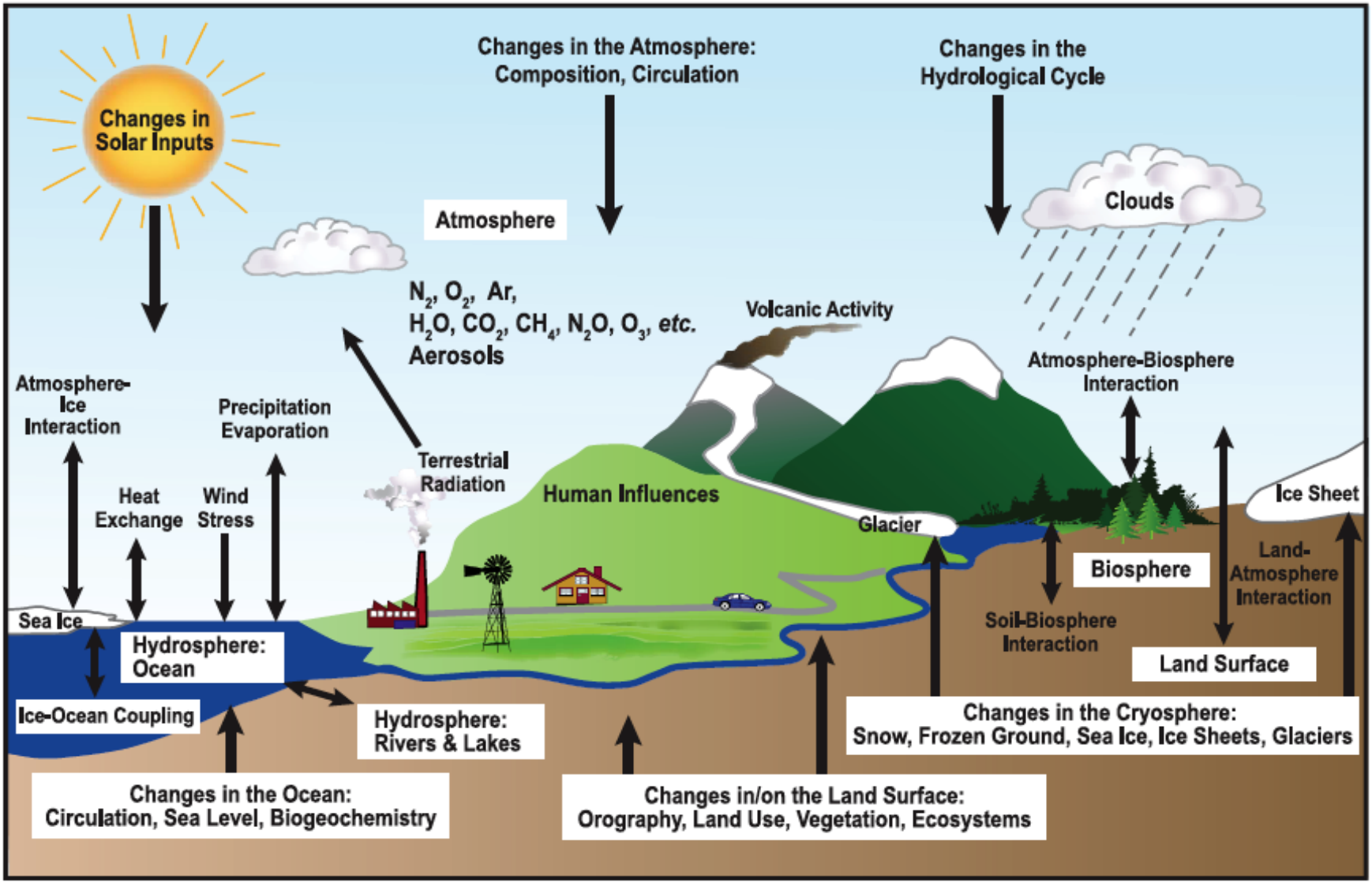


**Regional Circulation Model (e.g., WRF-CHEM)**
3D, regional
Met-fields predicted with boundary conditions from GCMs or reanalysis data

**Global Circulation Model (GCM)**
3D, global, met-fields predicted, nudged with reanalysis met-data,
online or offline aerosol

(From P. Stier)

# Components of the Climate System in GCMs




IPCC, 2007

# Outline

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

# What is an aerosol?

- An *aerosol* (particulate matter) is a suspension of fine solid particles or liquid droplets in air.
- Size: 1 nm to ~ 10 micrometer in diameter.
- Composition: sulfate, nitrate, ammonium, organic carbon, black carbon, dust, sea salt.

Los Angeles smog on 29 January 2004 Photo by Alan Clements

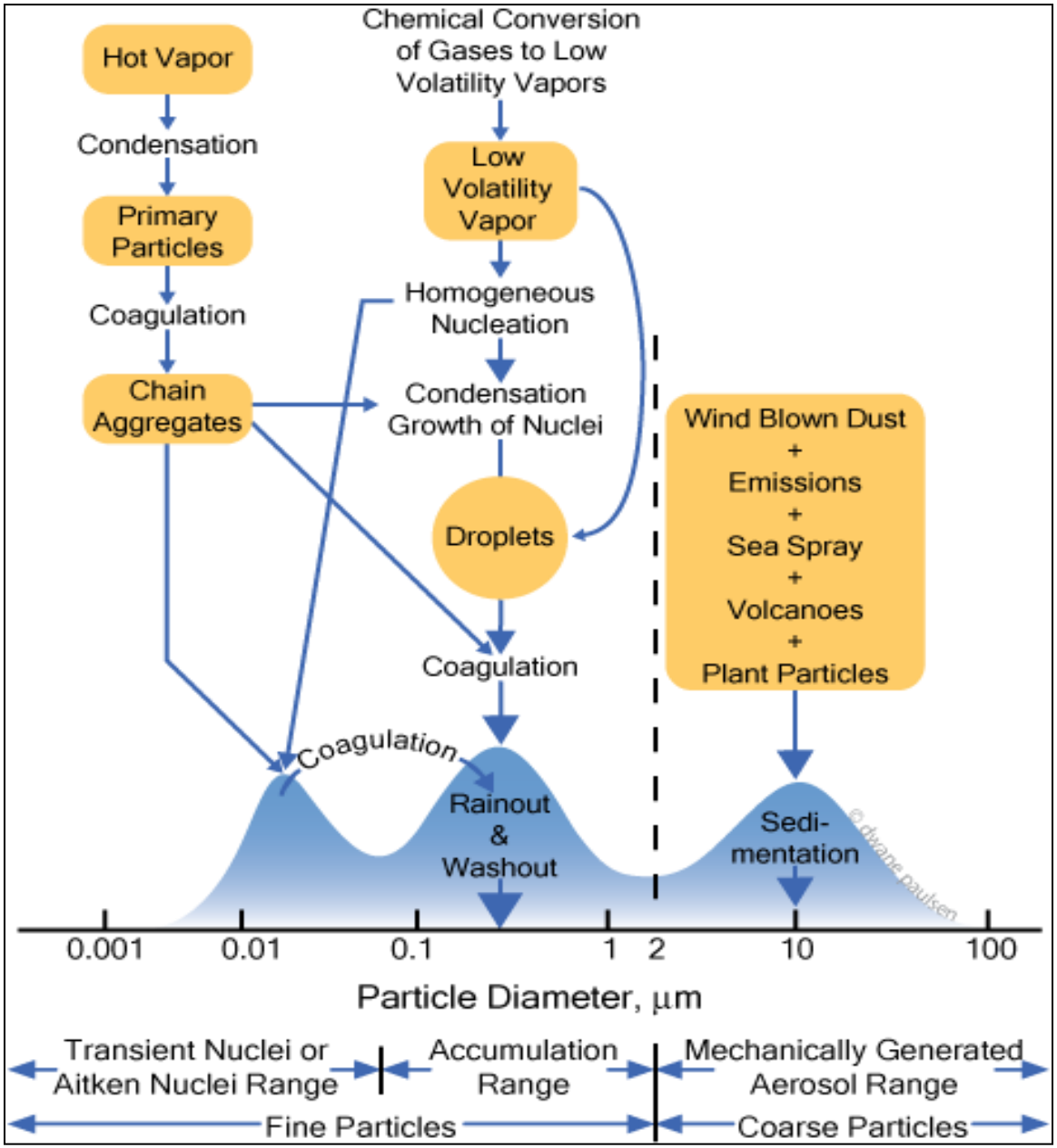
Beijing haze

# Where do aerosols come from?

Primary

Secondary

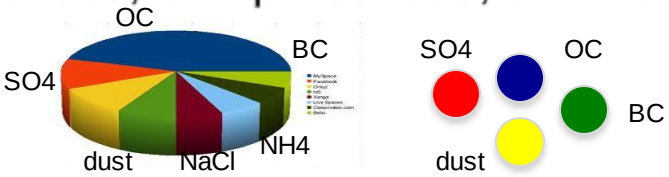
# Aerosol Size and Composition in the Atmosphere



# Aerosol Representation in GCMs

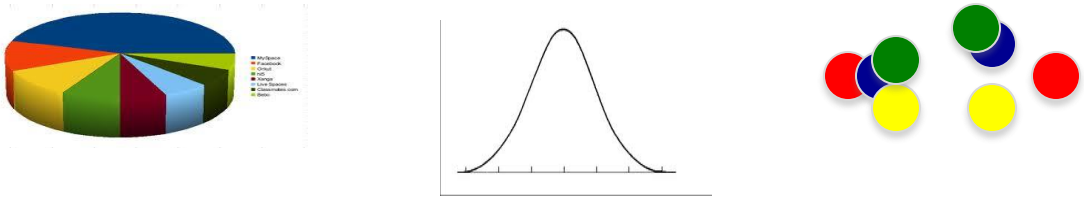
- **Bulk**

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



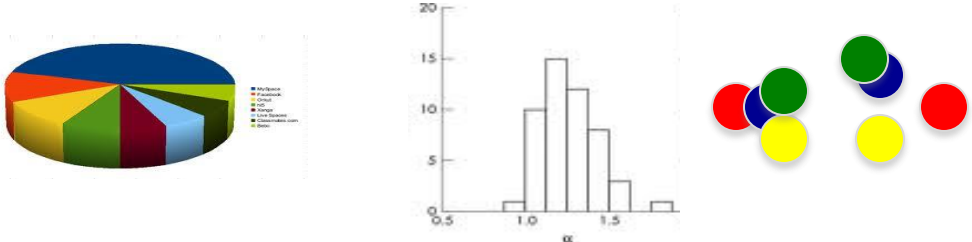

- **Moment-based (modal, 2-moment quadrature method of moments)**

  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**

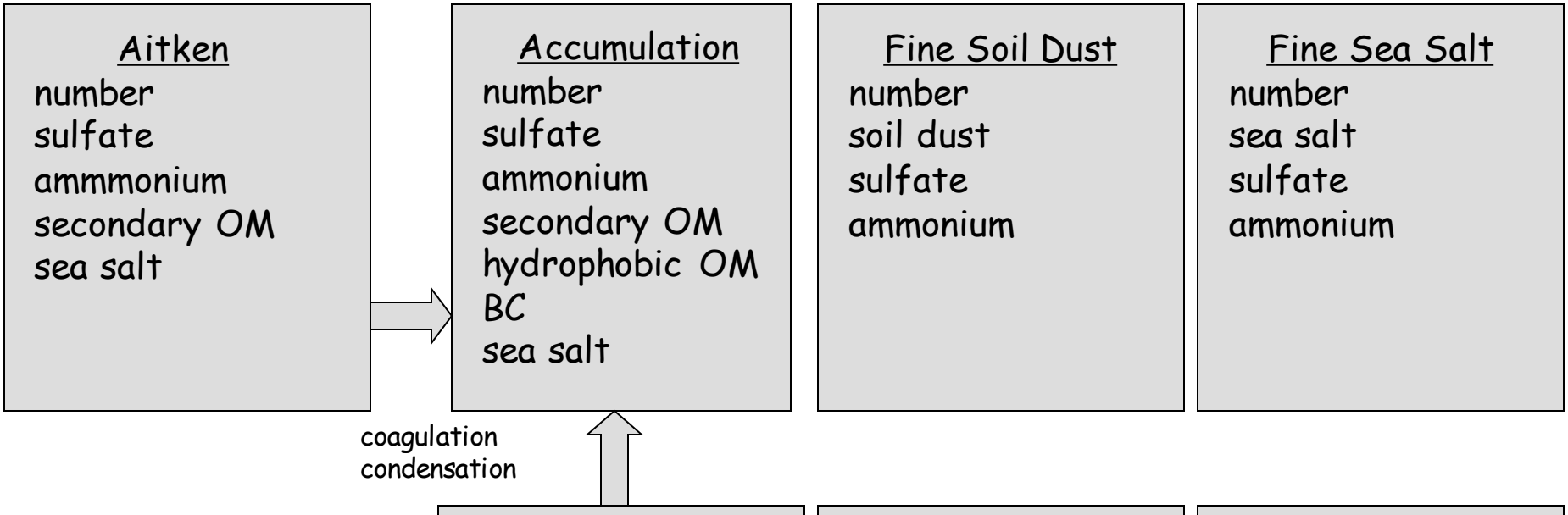
  Split size distribution into bins, predict evolution of size distribution by predicting mass and number mixing ratio in each bins, aerosol microphysics



# Bulk Aerosol Module (BAM) 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 Module (MAM) in CAM5




All modes log-normal with prescribed width.

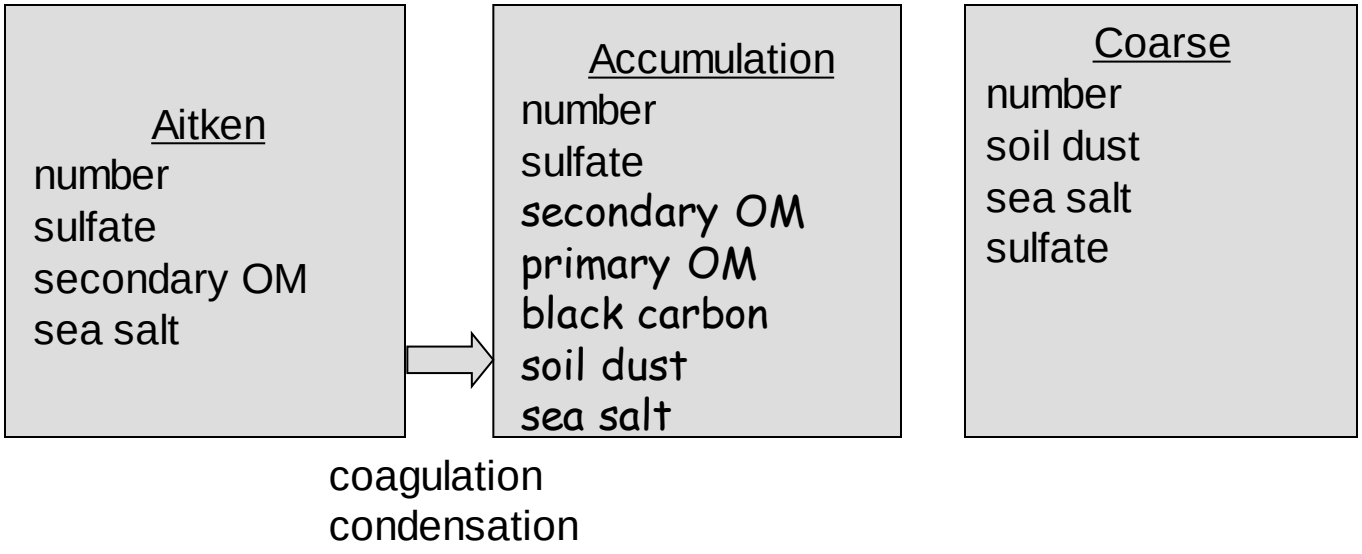
Total transported aerosol tracers: 31

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

**Computer time is ~100% higher than BAM**

# Simplified 3-mode version of MAM in CAM5

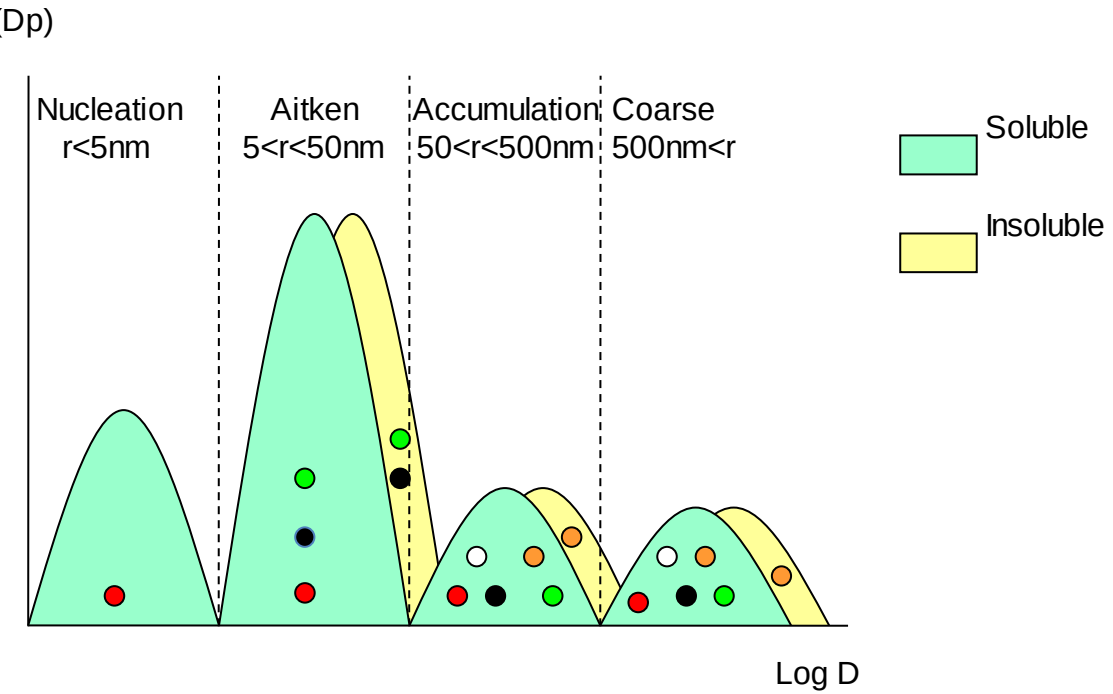
Assume primary carbon is internally mixed with secondary aerosol.
Sources of dust and seasalt are geographically separate
Assume ammonium neutralizes sulfate.




Total transported aerosol tracers: 15

**Computer time is 30% higher than BAM**

# M7 (ECHAM-HAM)



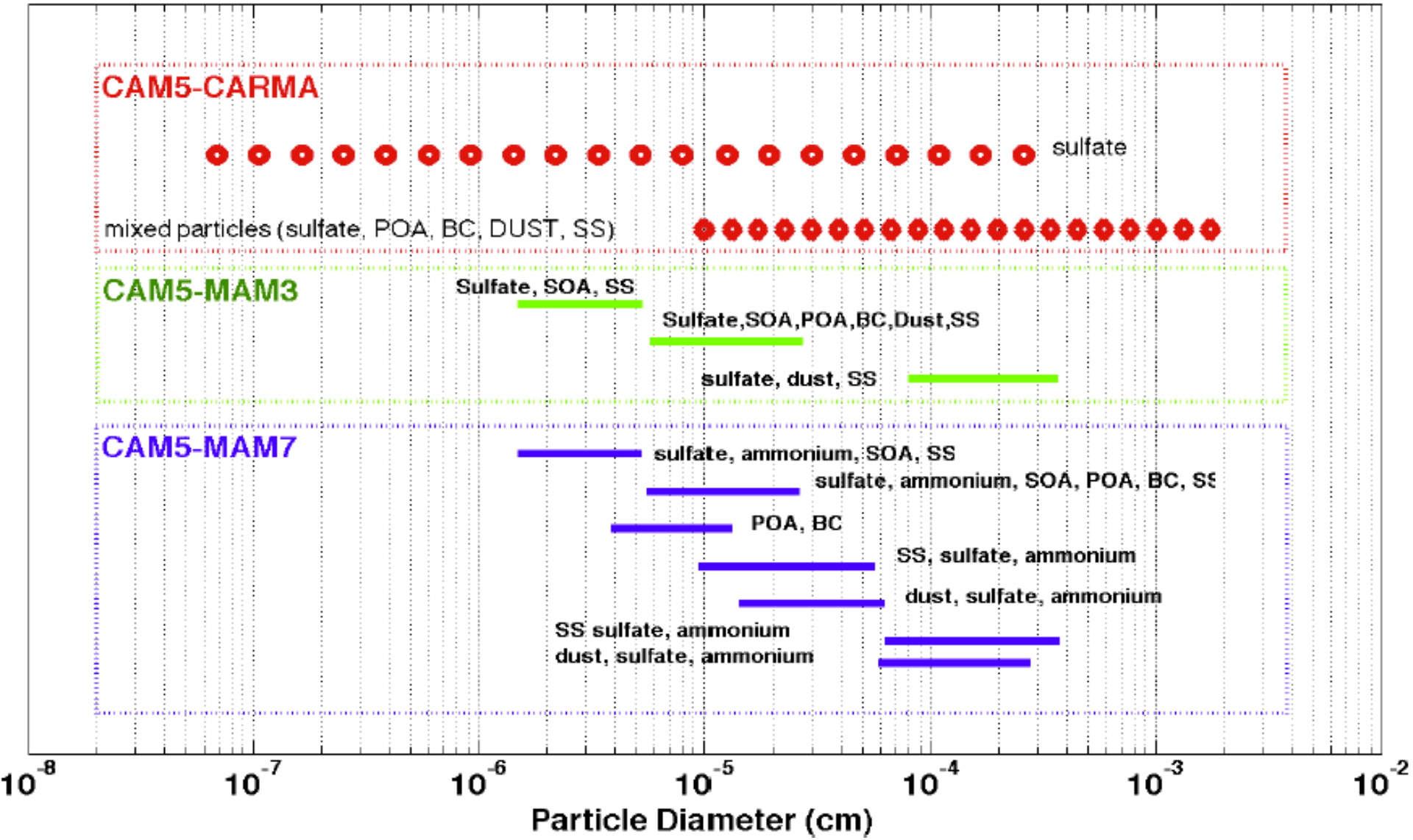

- Sulphate (SU)
- Black Carbon (BC)
- Particulate Organic Matter (POM)
- Sea Salt (SS)
- Dust (DU)

Predicted variables per mode:

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

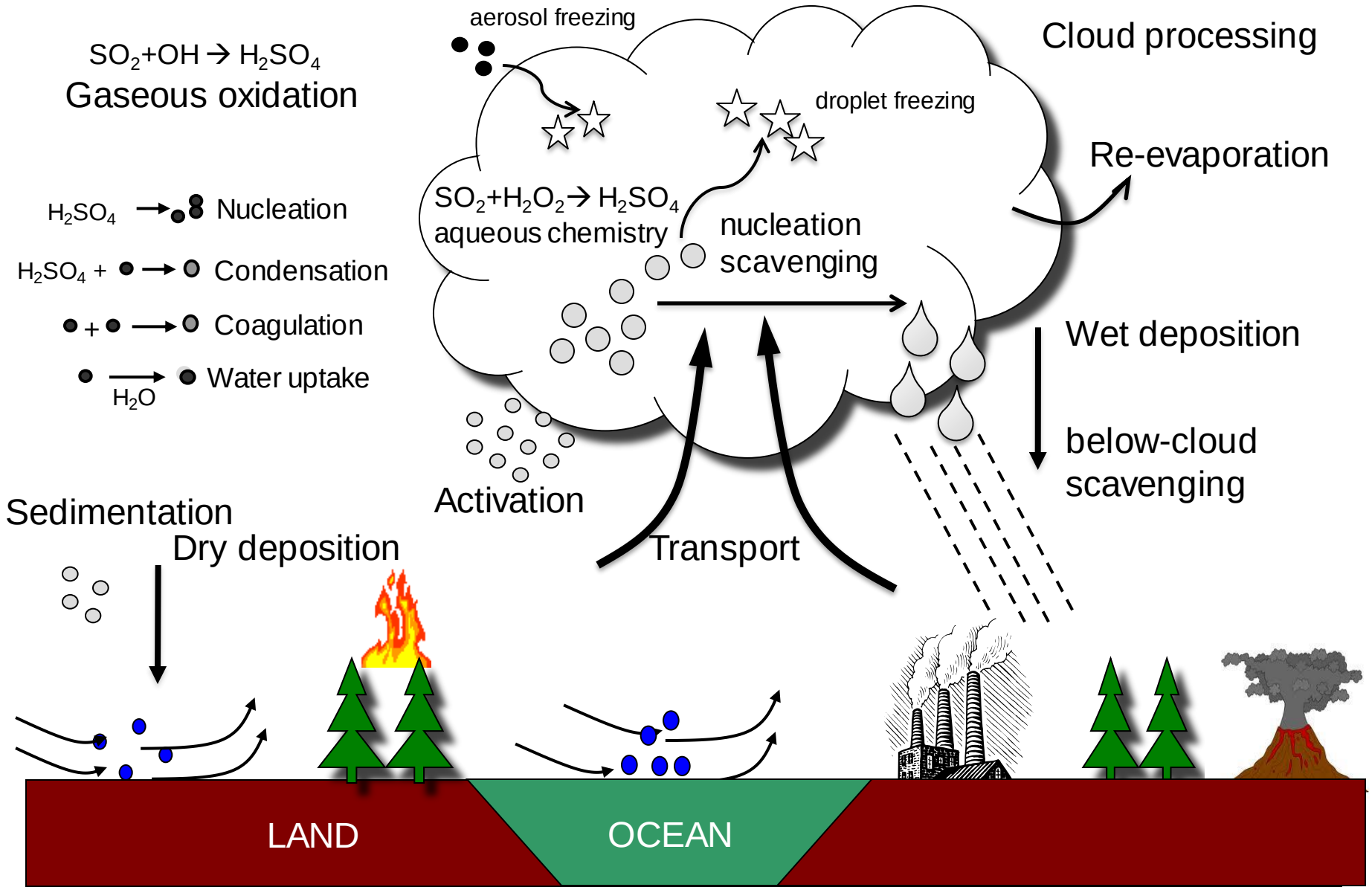
Courtecy of Declan O ‘Donnell

# Sectional Aerosol Treatment in CESM-CAM5



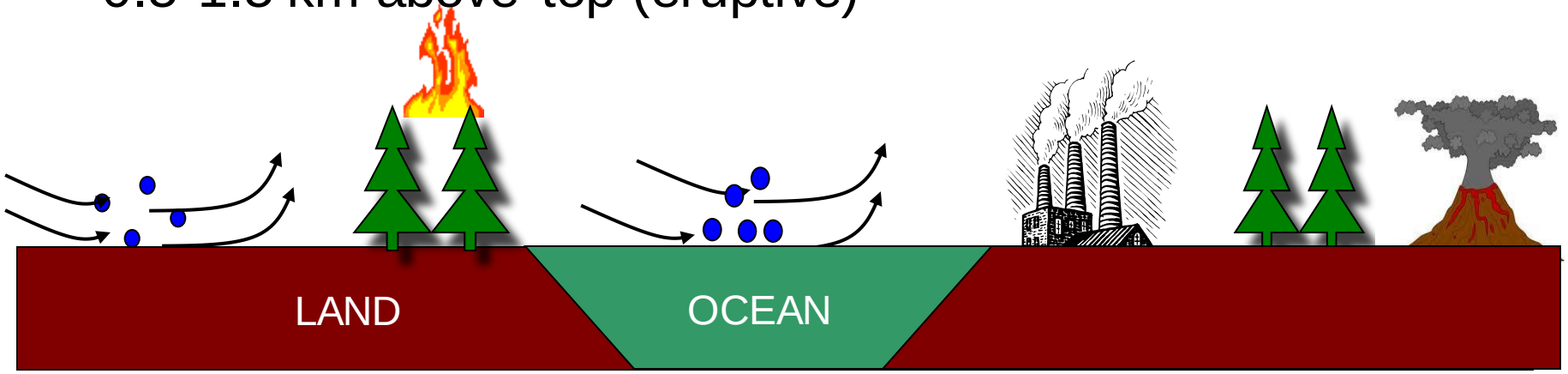

Pengfei Yu, 2015

# Global Aerosol Cycles

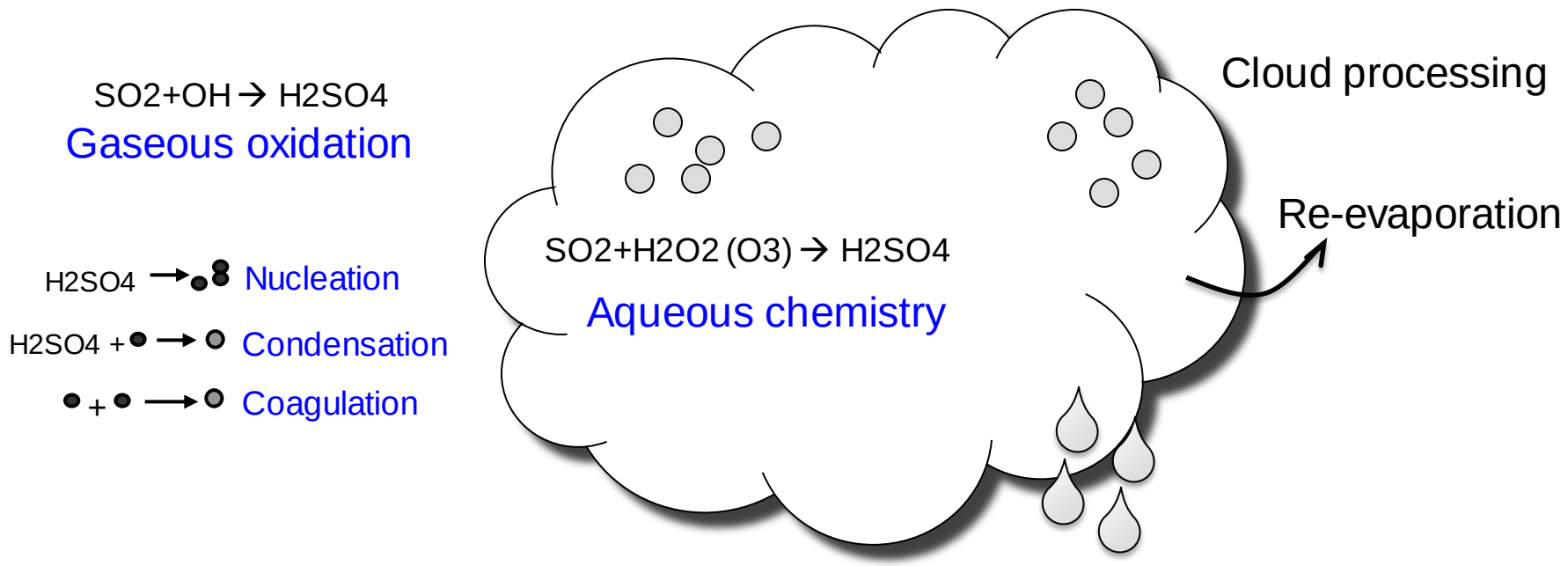


# Aerosol Processes : Primary Emission

- 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 (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(g) : 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:

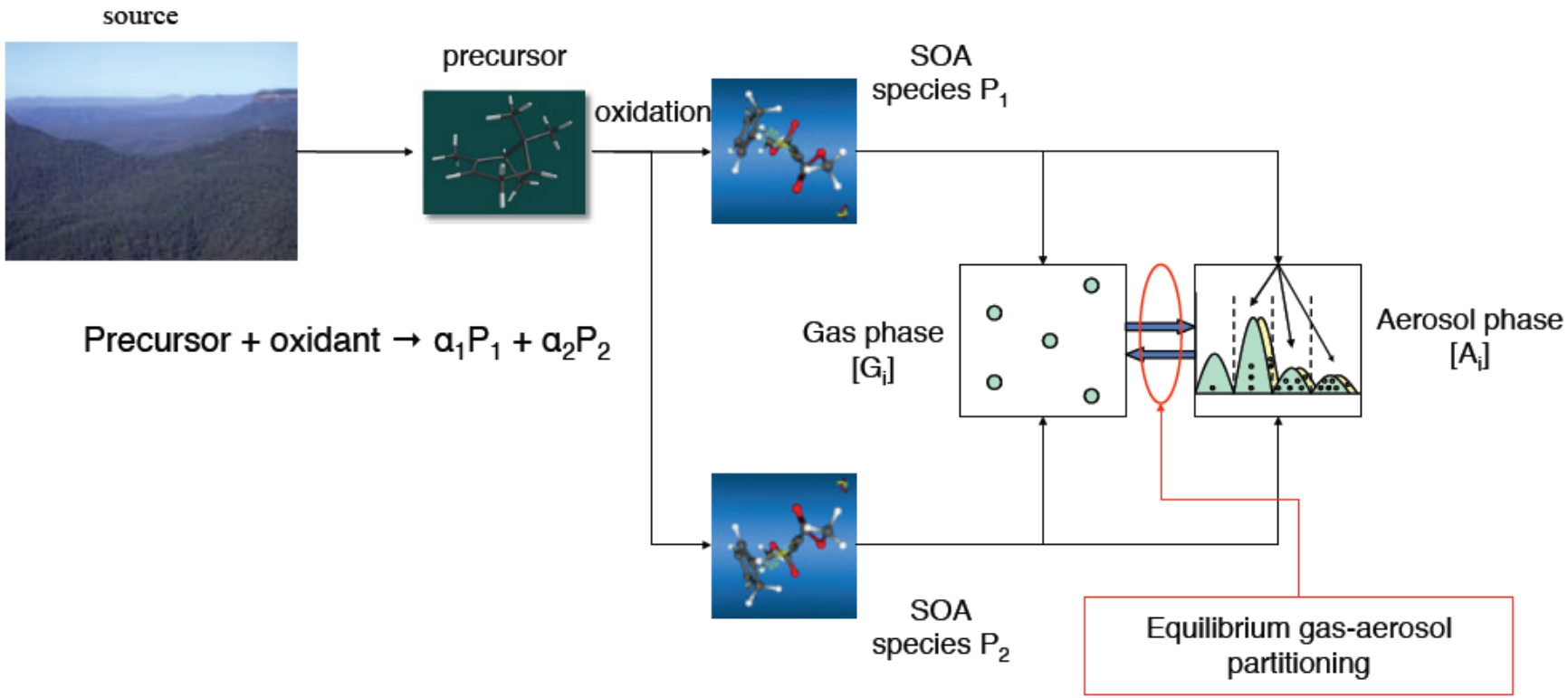
Prognostic SOA scheme with explicit gas/aerosol partitioning

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.

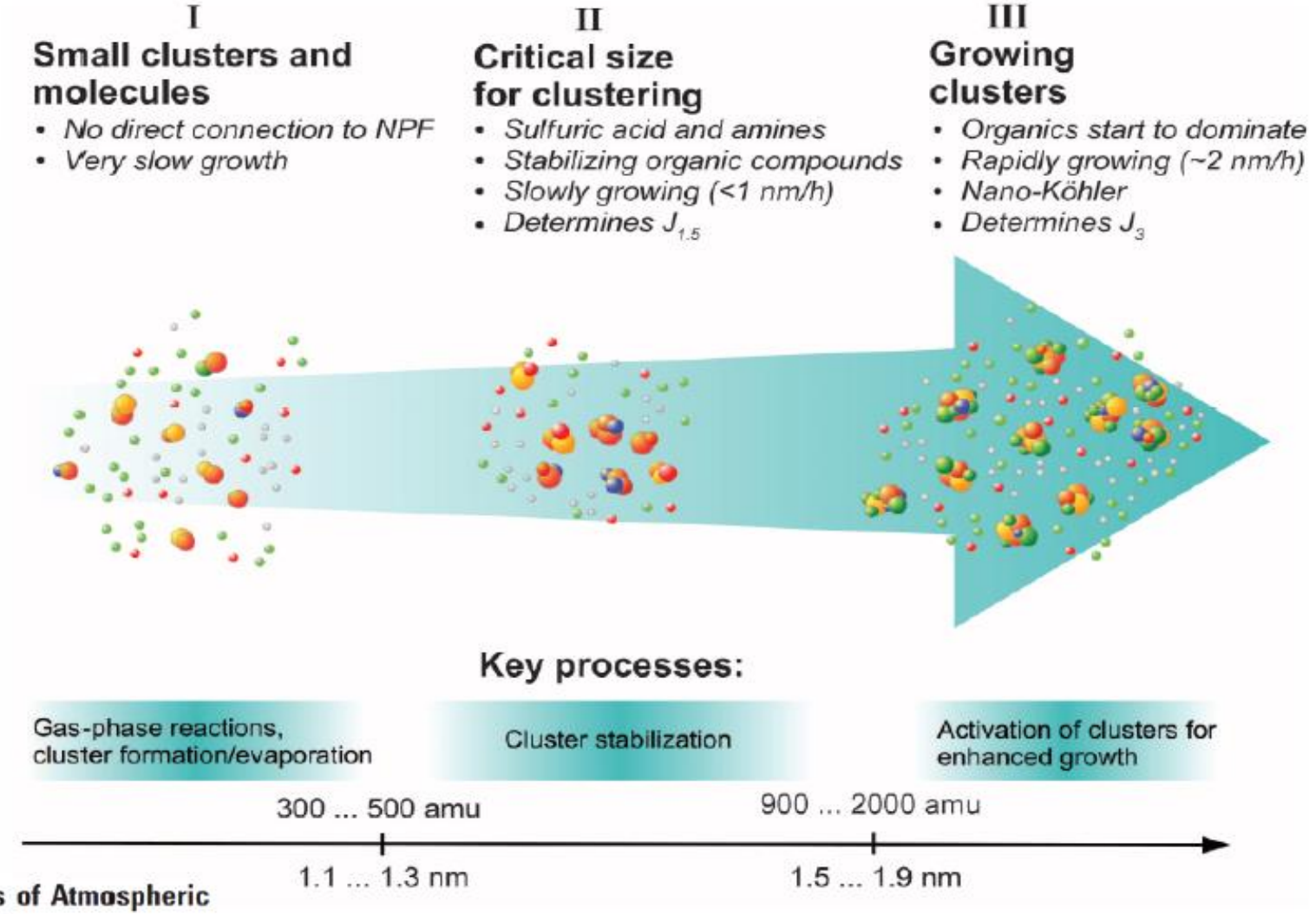
Two steps of more complexity : primary VOCs emission and oxidation in atmosphere to form SOA (g). explicit gas/aerosol partitioning of SOA (g) – ECHAM & GISS.

Multi-generational aging of organic vapors (VBS scheme) & treating SOA as non-volatile semi-solid (glassy) – CAM5

# SOA scheme in ECHAM-HAM2



# Aerosol Processes (Nucleation)

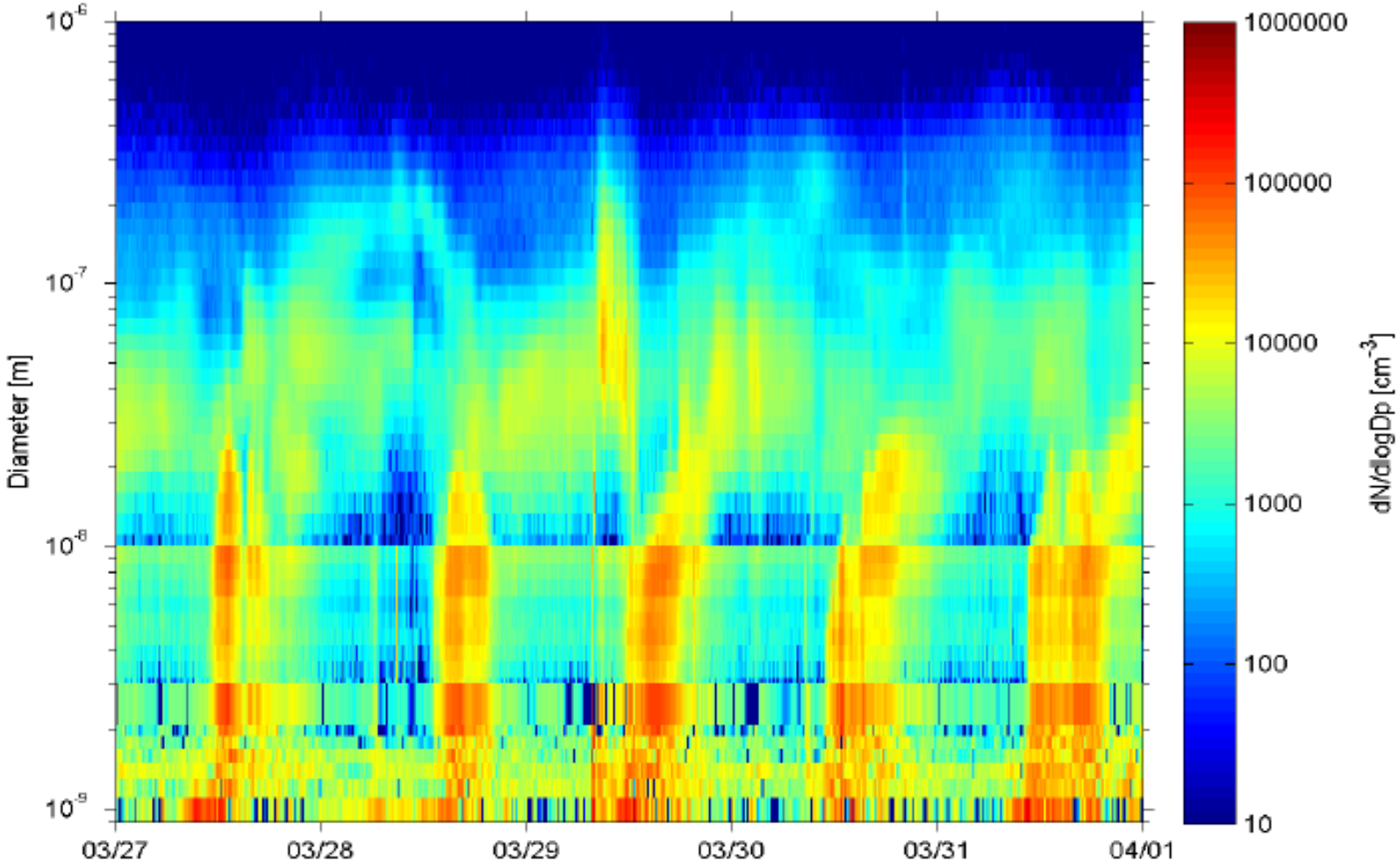



**irect Observations of Atmospheric erosol Nucleation**

ku Kulmala,[1*] Jenni Kontkanen,[1] Heikki Junninen,[1] Katrianne Lehtipalo,[1] a E. Manninen,[1] Tuomo Nieminen,[2,3†] Tuukka Petäjä,[1] Mikko Sipilä,[1] ried Schobesberger,[1] Pekka Rantala,[1] Alessandro Franchin,[1] Tuija Jokinen,[1] u Järvinen,[1] Mikko Äijälä,[1] Juha Kangasluoma,[1] Jani Hakala,[1] Pasi P. Aalto,[1] Paasonen,[1] Jyri Mikkilä,[1] Joonas Vanhanen,[2] Juha Aalto,[1] Hannele Hakola,[4] Makkonen,[4] Taina Ruuskanen,[1] Roy L. Mauldin III,[1,5] Jonathan Duplissy,[1] a Vehkamäki,[3] Jaana Bäck,[6] Aki Kortelainen,[7] Ilona Riipinen,[8] Theo Kurtén,[2,9] ay V. Johnston,[10] James N. Smith,[7,11] Mikael Ehn,[1,12] Thomas F. Mentel,[12] E. J. Lehtinen,[4,7] Ari Laaksonen,[4,7] Veli-Matti Kerminen,[1] Douglas R. Worsnop[1,4,7,13]

**Direct Observations of Atmospheric Aerosol Nucleation**
Markku Kulmala *et al.*
*Science* **339**, 943 (2013);
DOI: 10.1126/science.1227385

# Aerosol Processes (Nucleation)




Kulmala et al., 2013, Science, SOM

# Aerosol Processes (Aging)

## Earlier Approaches:

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

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 (SO4, NH4, SOA, NO3) – CAM5-MAM4/7, ECHAM & GISS

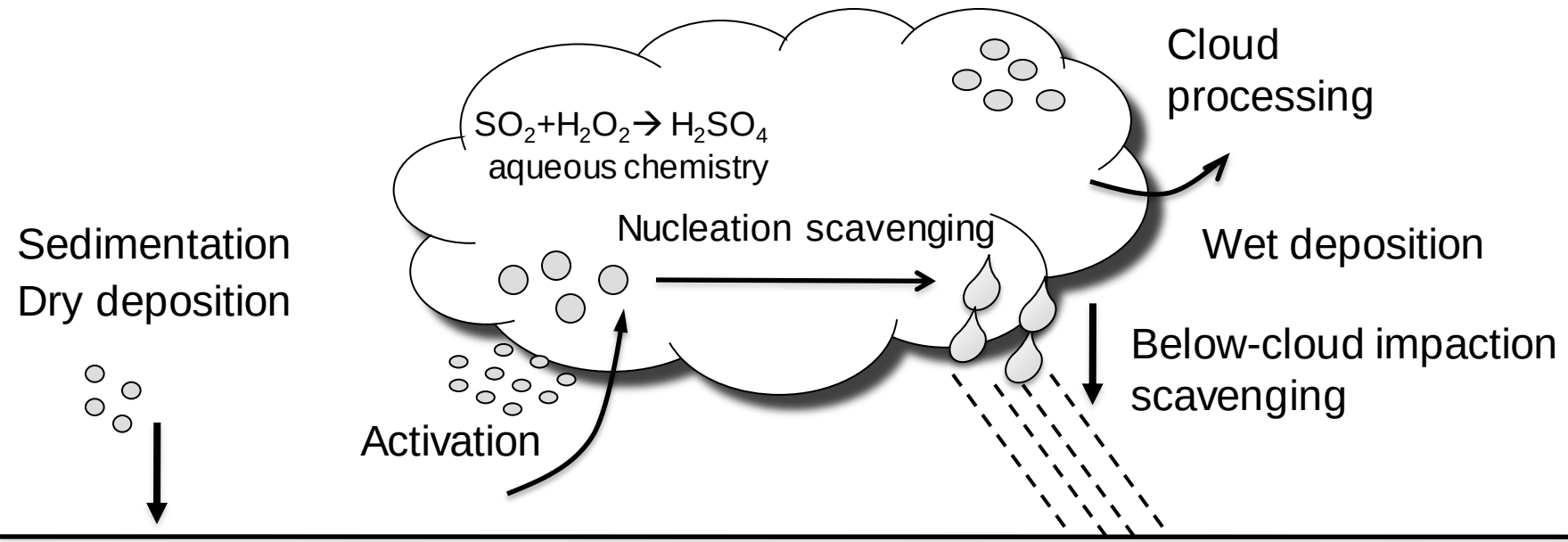
# Aerosol Processes (Water Uptake)

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.

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

Wet Deposition : most models calculate 1st order loss rate of cloud water with cloud water 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

# Sulfate budget

Table 3. Global annual budget for sulfate. The means and normalized standard deviations (in %) from available models participating in AeroCom (Textor et al., 2006) are listed. The values in parentheses are mean removal rates (in 1/day) and normalized standard deviations (in %) as budget terms are not given in Textor et al. (2006). For comparison removal rates (in 1/day) from MAM3 and MAM7 are listed in parentheses.

| | MAM3 | MAM7 | AeroCom |
|---|---|---|---|
| Sources | 44.30 | 45.71 | 59.67, 22 |
| Emission | 1.66 | 1.66 | |
| $SO_2$ aqueous-phase oxidation | 28.03 | 29.74 | |
| from $H_2O_2$ chemistry (%) | 53.9 | 48.1 | |
| $H_2SO_4$ aqueous-phase uptake | 0.59 | 0.51 | |
| $H_2SO_4$ nucleation | 0.030 | 0.030 | |
| $H_2SO_4$ condensation | 13.98 | 13.74 | |
| Sinks | 44.30 | 45.71 | |
| Dry deposition | 4.96 (0.03) | 5.51 (0.03) | (0.03, 55) |
| Wet deposition | 39.34 (0.23) | 40.20 (0.23) | (0.22, 22) |
| Burden | 0.46 | 0.47 | 0.66, 25 |
| In modes (%) | 2.8 (Aitken), 95.5 (accum.), 1.7 (coarse) | 2.9 (Aitken), 88.9 (accum.), 1.1 (fine sea salt), 5.9 (fine dust), 0.32 (coarse sea salt), 0.88 (coarse dust) | |
| Lifetime | 3.77 | 3.72 | 4.12, 18 |

Units are sources and sinks, Tg S $yr^{-1}$; burden, Tg S; lifetime, days.

# BC budget

Table 6. Global budgets for BC. The means and normalized standard deviations (in %) from available models participating in AeroCom (Textor et al., 2006) are listed. The values in parentheses are mean removal rates (in 1/day) and normalized standard deviations (in %) as budget terms are not given in Textor et al. (2006). For comparison removal rates (in 1/day) from MAM3 and MAM7 are listed in parentheses.

| | MAM3 | MAM7 | AeroCom |
|---|---|---|---|
| Sources | 7.76 | 7.76 | 11.9, 23 |
| Fossil and bio-fuel emission | 5.00 | 5.00 | |
| Biomass burning emission | 2.76 | 2.76 | |
| Sinks | 7.75 | 7.75 | |
| Dry deposition | 1.27 (0.04) | 1.41 (0.04) | (0.03, 55) |
| Wet deposition | 6.48 (0.20) | 6.34 (0.19) | (0.12, 31) |
| Burden | 0.088 | 0.093 | 0.24, 42 |
| In modes (%) | 100 (accum.) | 10.8 (primary carbon)<br>89.2 (accum.) | |
| Lifetime | 4.17 | 4.37 | 7.12, 33 |

Units are sources and sinks, Tg $yr^{-1}$; burden, Tg; lifetime, days.

# Aerosol Properties in GCMs

- Mass and composition
  - interactive SO4, POA, SOA, BC, dust and sea salt,
  - ammonium, nitrate often not treated (CAM, ECHAM)
- Size distribution
  - variable for each mode, bin
- Mixing state
  - internal and external mixture
- Radiative properties and refractive index
  - parameterized in terms of bulk refractive index and wet effective 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

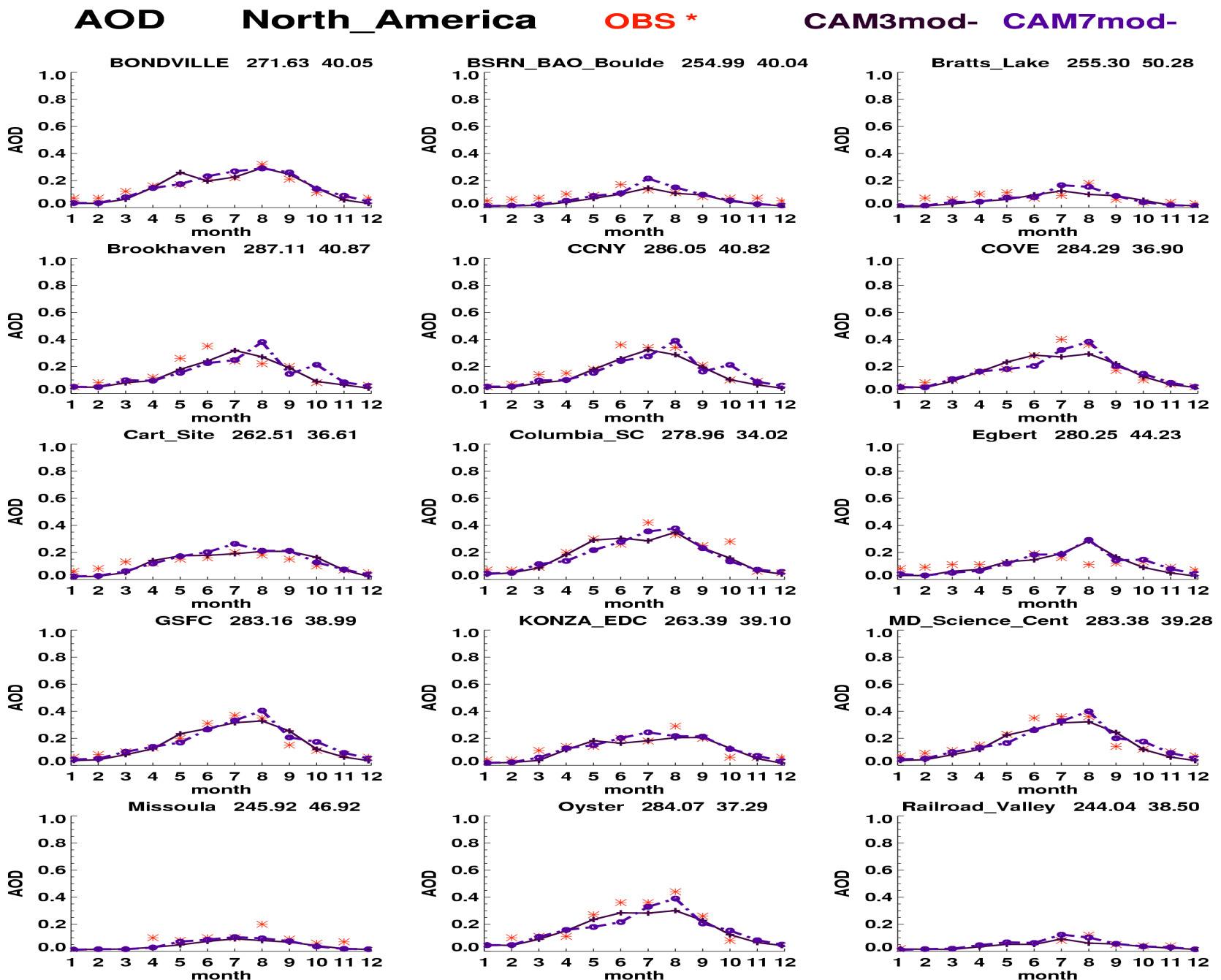
# Uncertainties in Aerosol Processes in GCMs

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

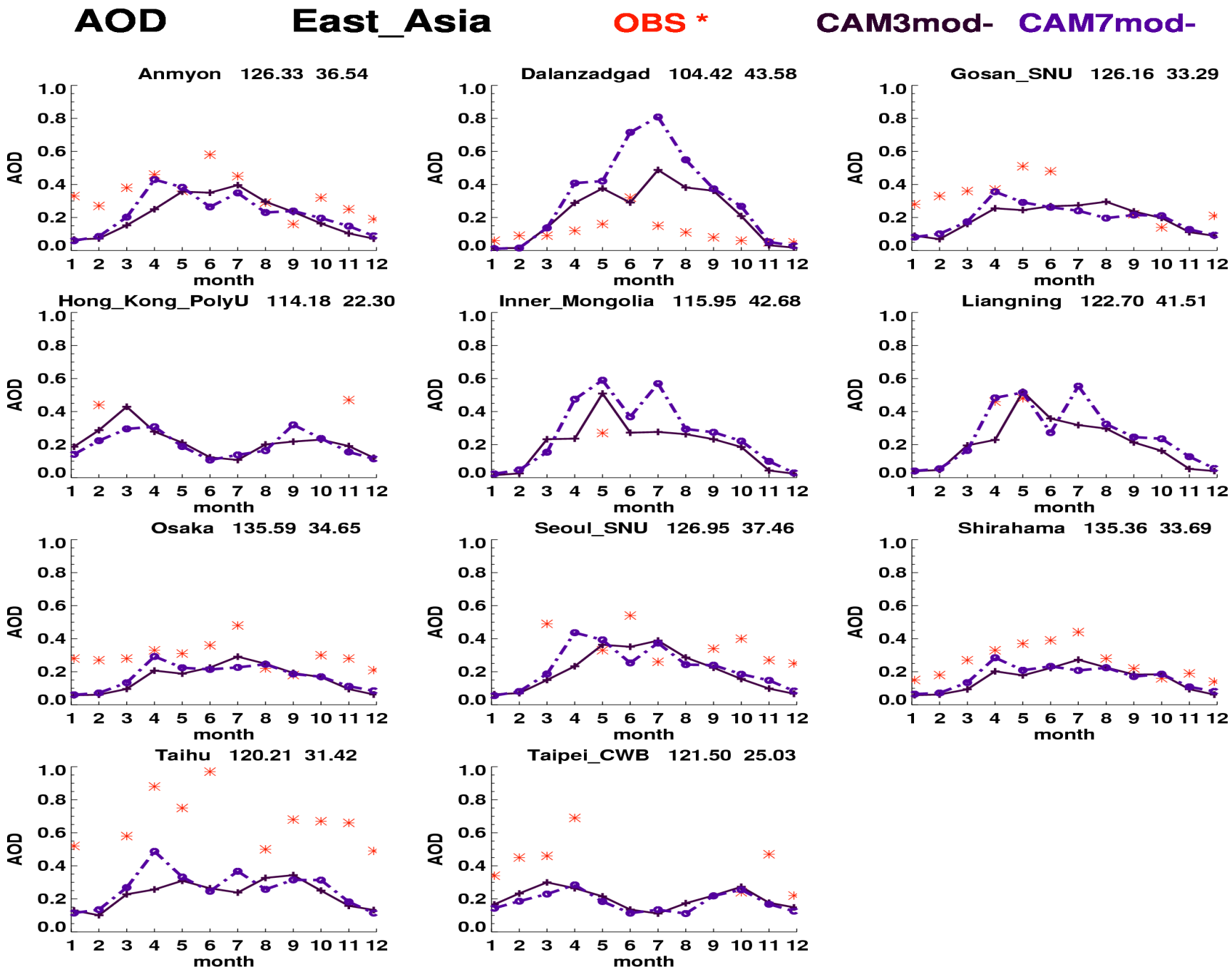
  - Anthropogenic emissions in developing counties
  - Biomass burning emissions (e.g., GFED)
  - Mineral dust and sea salt emissions
    - Dust: 1640 Tg/yr ± 50% (AEROCOM-A); 3200 Tg/yr (CAM5)
    - Sea salt: 6280 Tg/yr ± 200% (AEROCOM-A); 5000 Tg/yr (CAM5)

  - Primary organics from oceans

# Effect of Primary Emissions



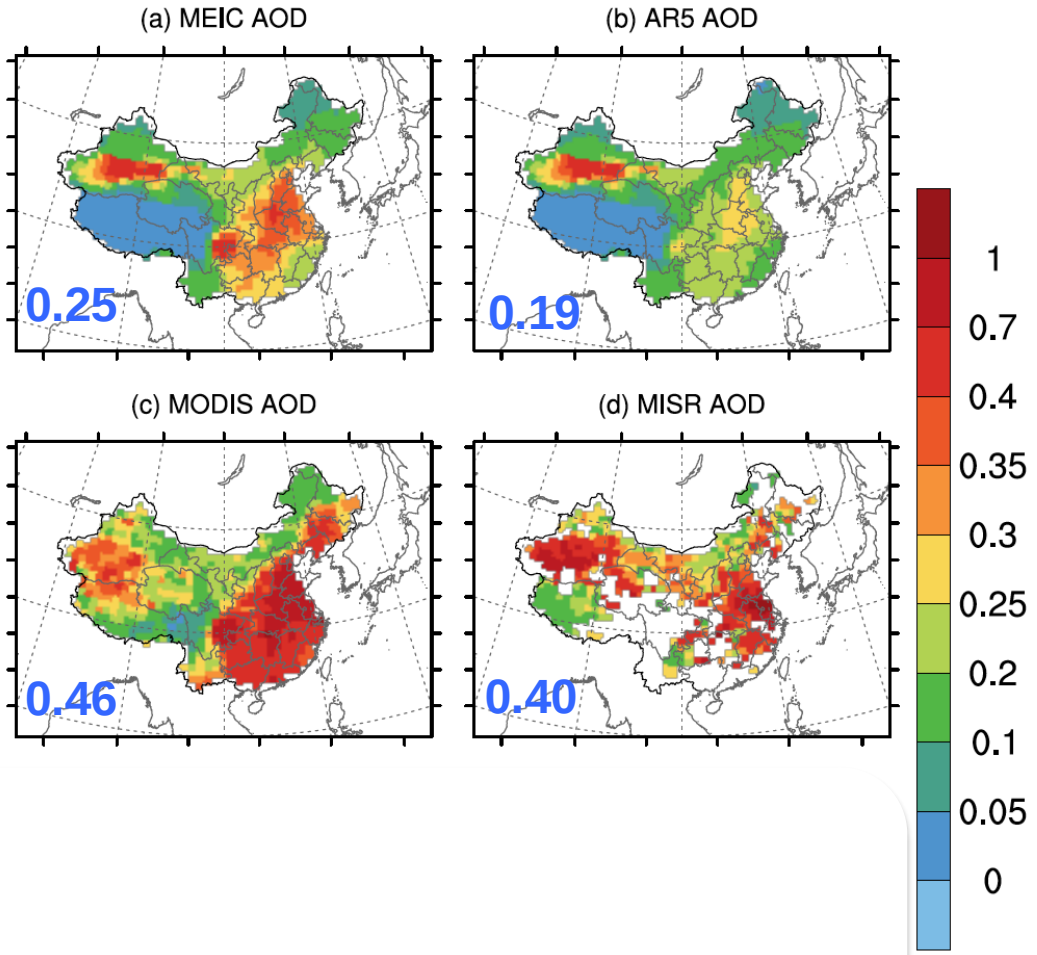
# Effect of Primary Emissions



# The emission accounts for 22%-28% of the modeled AOD low biases in eastern China

CAM5 AOD

Satellite AOD

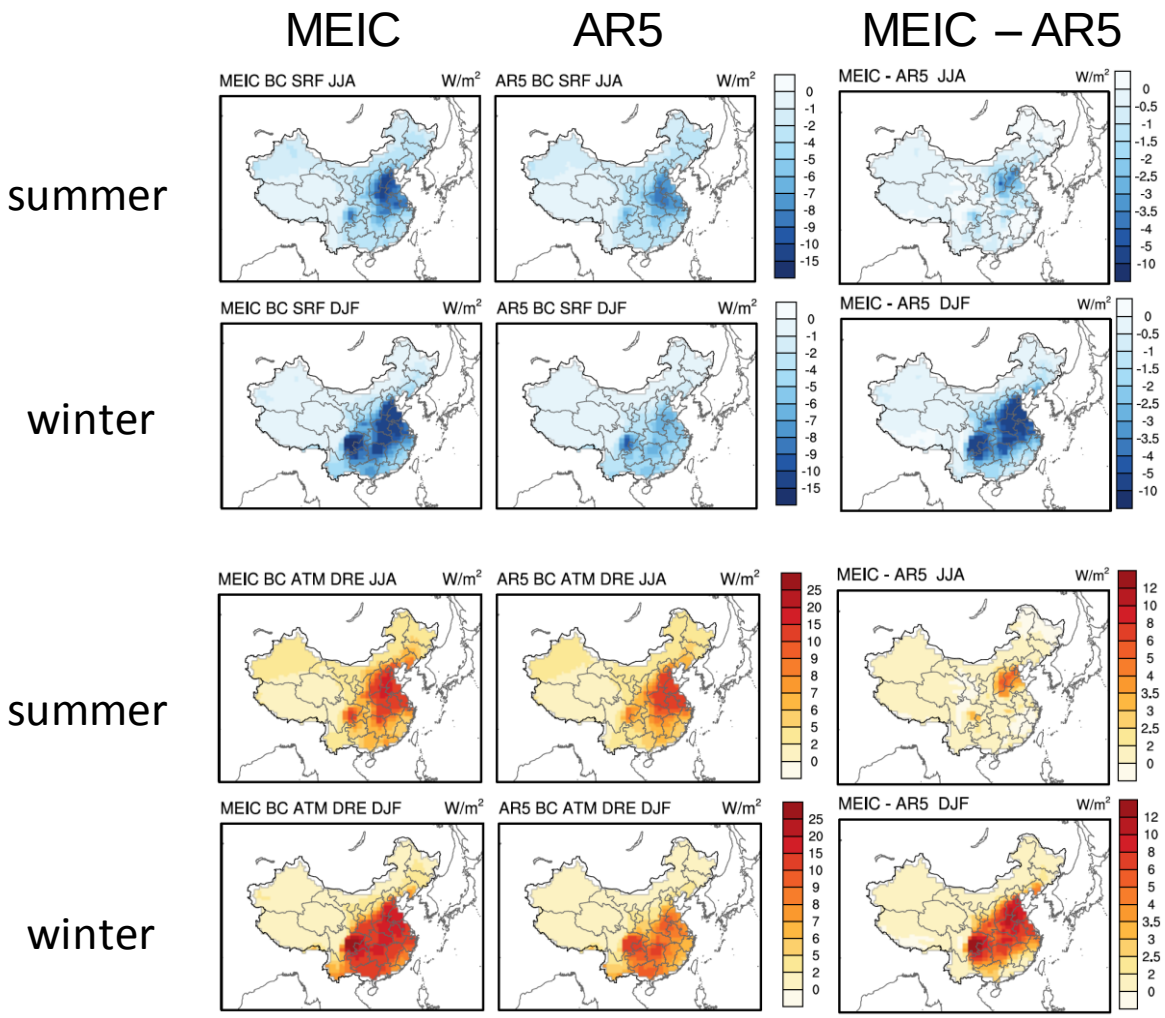



Averaged over 105-125 E

Fan et al. (2016)

*AOD is still underestimated by CAM5 with MEIC.*

# BC DRFs are doubled in winter due to the use of MEIC




First column (MEIC): seasonal change due to emission

Second column (AR5): seasonal change due to monsoon precipitation [Jiang et al., 2015]

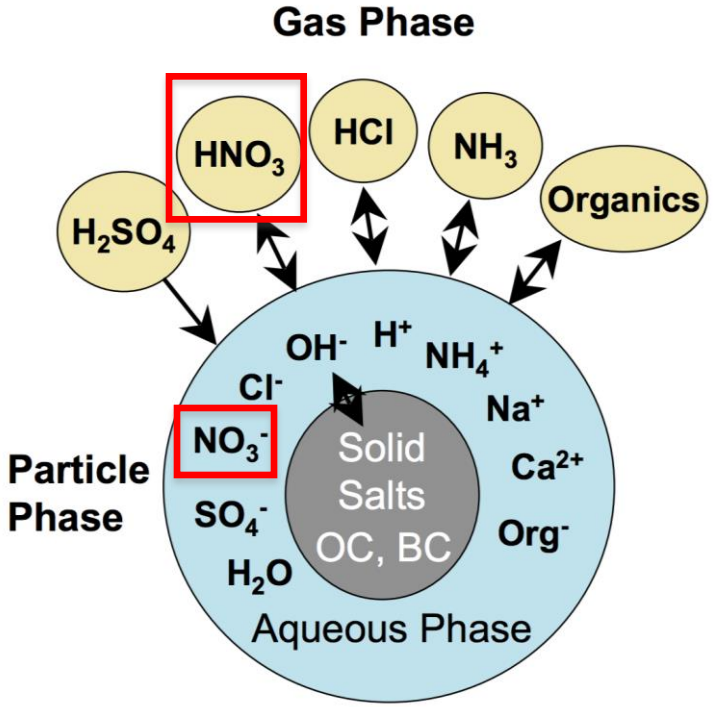
Third column (MEIC – AR5): BC DRF difference is largest in the winter.

# MOSAIC coupled in CAM5/MAM

- MOSAIC module is developed by Zaveri et al. [2008], which treats many processes during the evolution of aerosol particles, such as nucleation, gas-aerosol exchange, coagulation, wet/dry removal processes.

- In the version of MAM coupled with MOSAIC, gas-aerosol exchange is treated by MOSAIC. The remaining processes are still treated by MAM

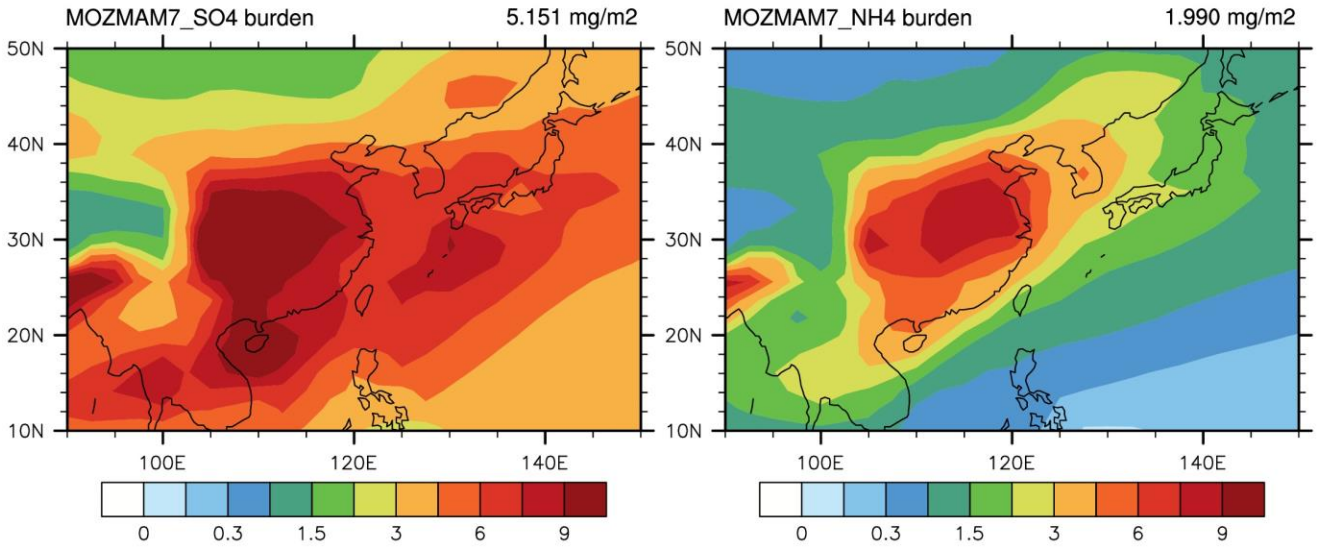
- Adaptive-step time-split Euler method is used for solving gas-aerosol exchange.




Source: presentation by Zaveri WRF tutorial, 2008

# Aerosol burdens modeled by MOSMAM7 – Jan.

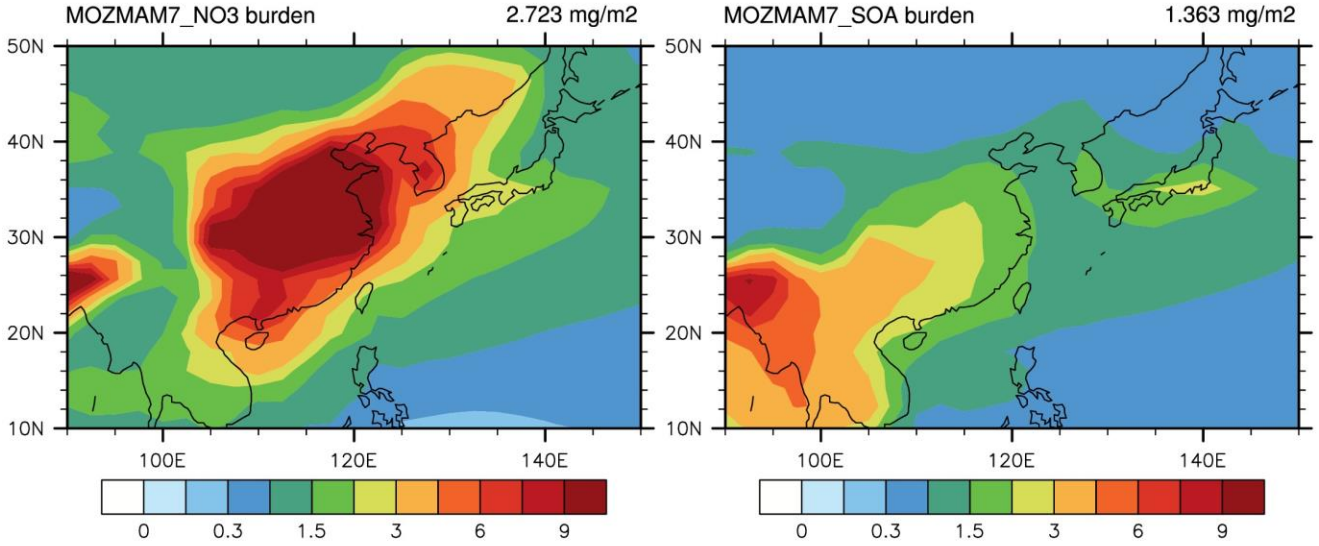
$SO_4$




$NH_4$



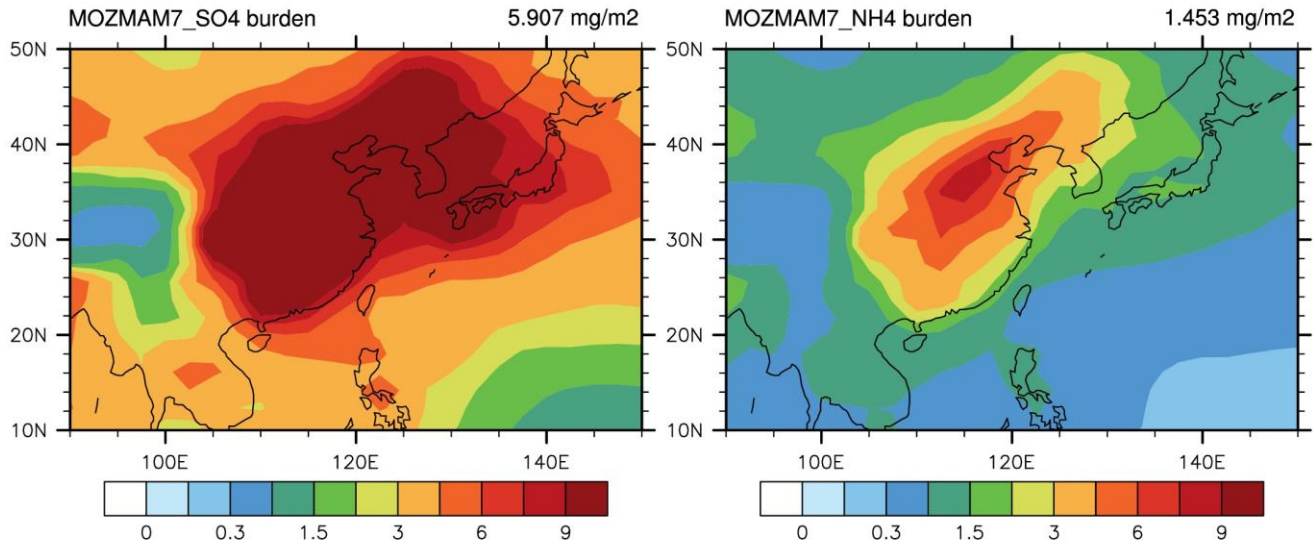

$NO_3$




SOA

# Aerosol burdens modeled by MOSMAM7 – July

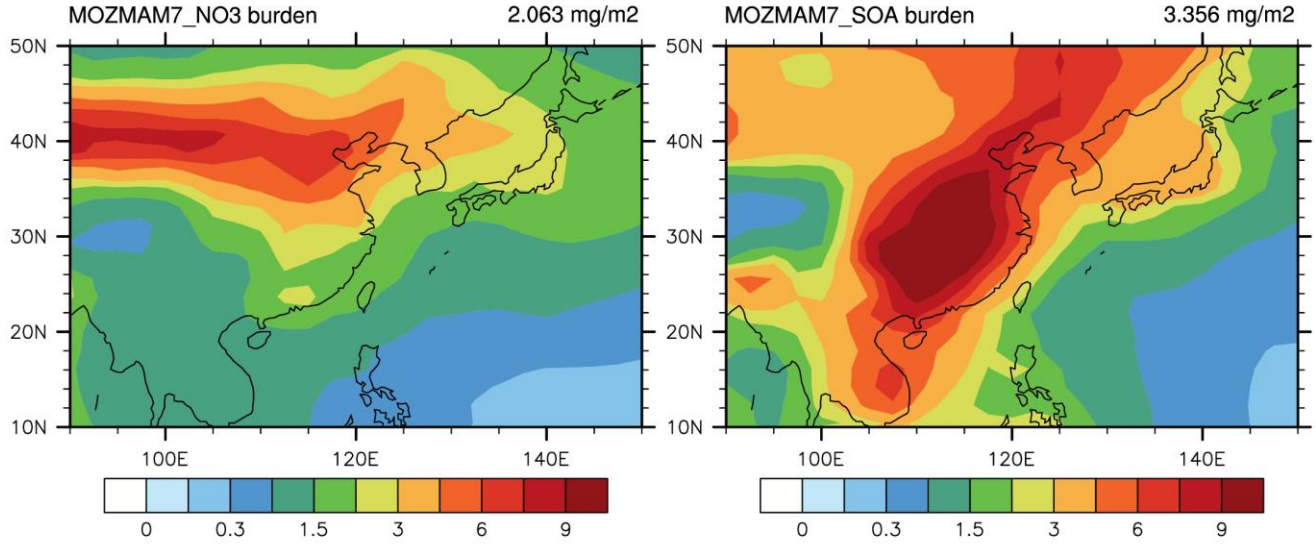
$SO_4$




$NH_4$



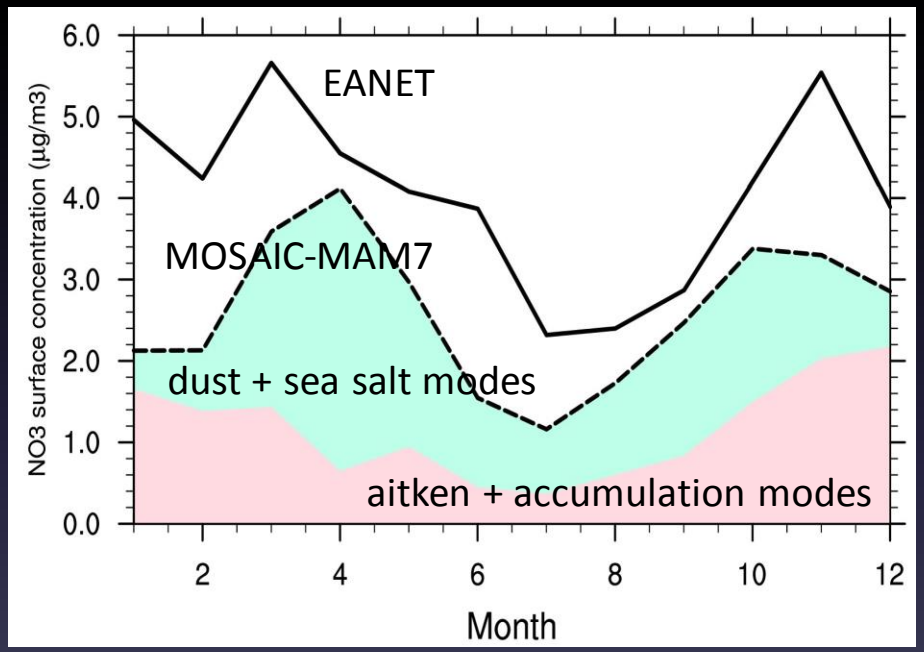

$NO_3$

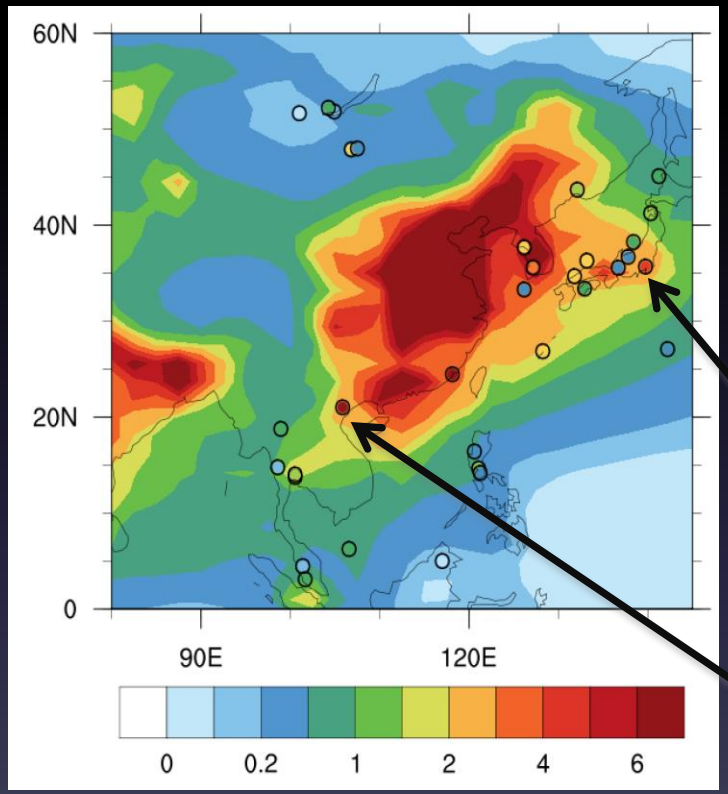



SOA

# Results – comparison against EANET

Tokyo




NO3 surface concentration ($\mu g/m^3$)

# Uncertainties in Aerosol Processes in GCMs

- Wet removal
  - Cloud water content, cloud fraction
  - Treatment of aerosol wet removal
  - Cloud microphysics in convective clouds

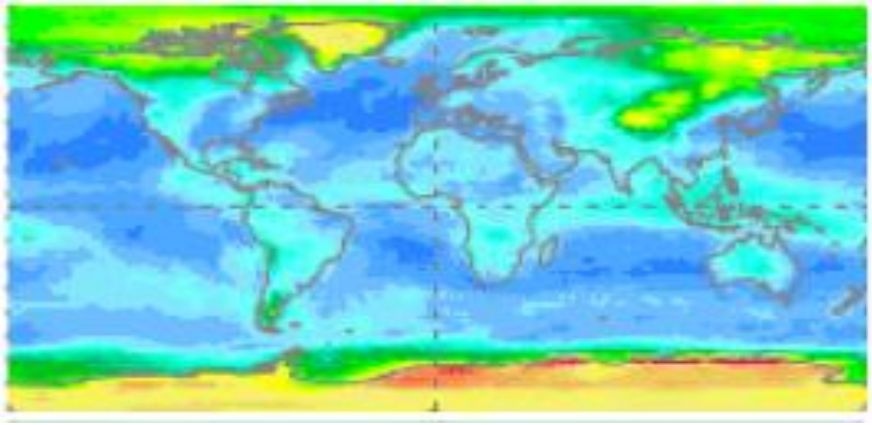
# Aerosol Models Have Particular Trouble Simulating Aerosol Beyond the Polar Front

- 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.

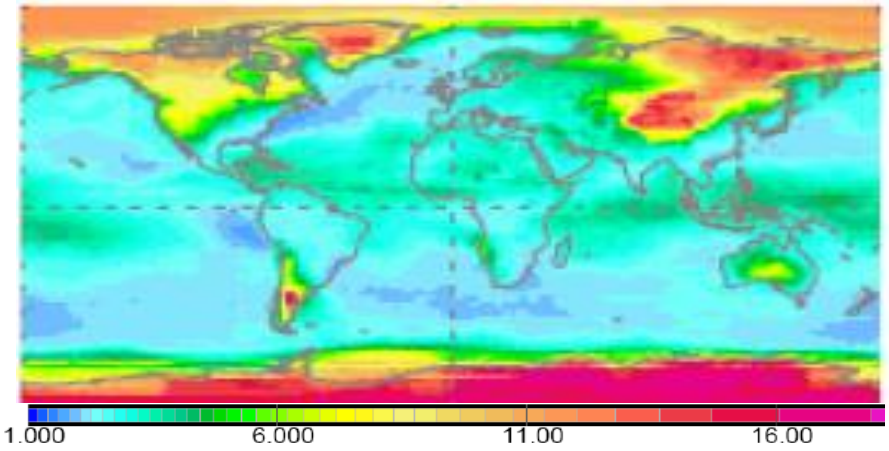
Major differences in poles

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

Aerosol Optical Depth

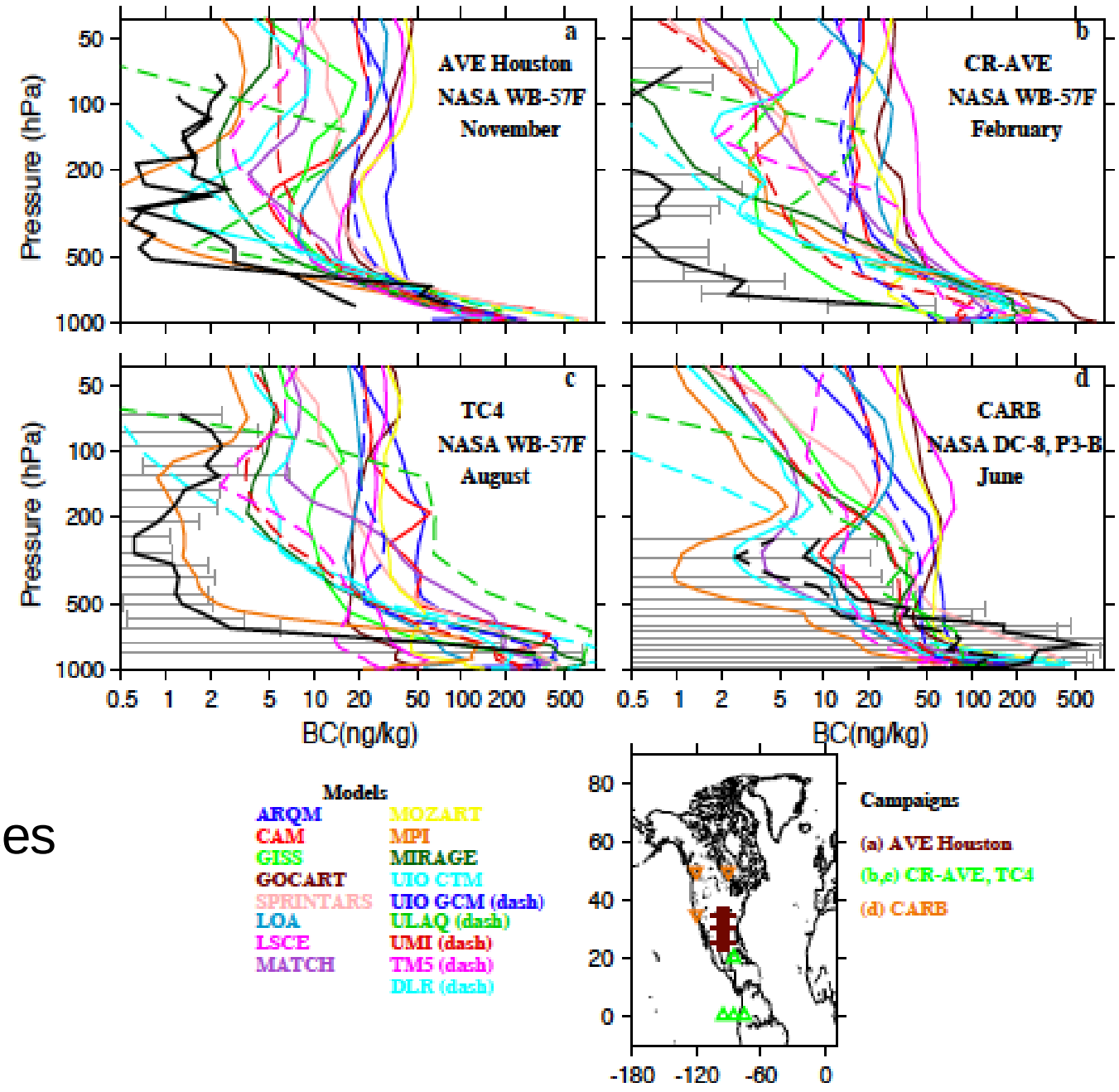


Aerosol Column Mass




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

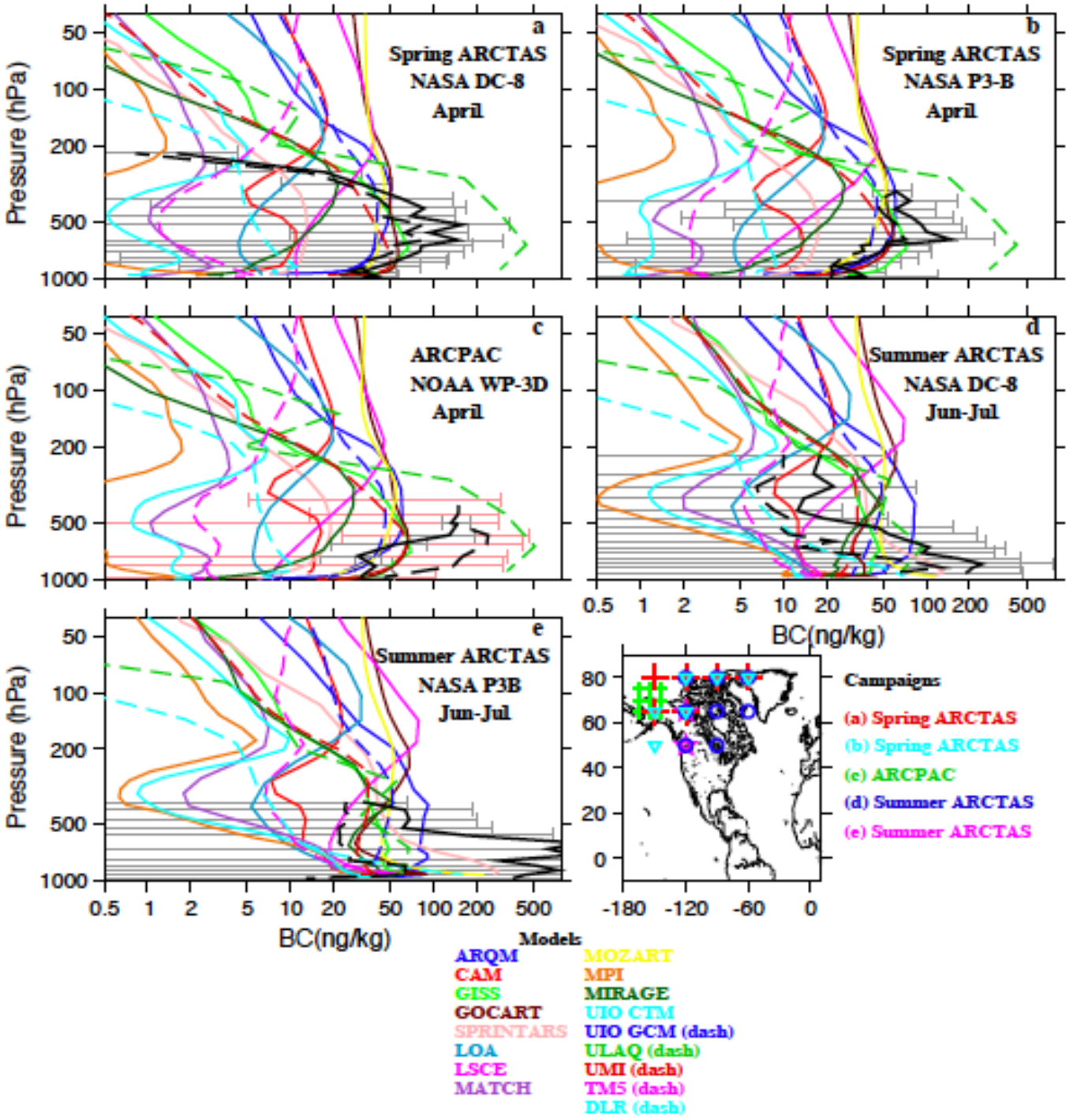
## BC compared with SP2 (tropics and midlat.)




Major differences in free troposphere

Koch et al. (2009)

# BC compared with SP2 (highlat.)




Koch et al. (2009)