Aerosol Representation in GCMs

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Questions :

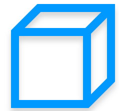
1) How are aerosol properties and processes represented in current GCMs? How do the models compare to each other?
2) What are the major assumptions/simplifications in the representations? What are the weaknesses in current representations?
3) Where are the trouble spots? Which types of aerosol, or which regions in which aerosols are not represented well, and/or simulated aerosols do not agree with 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

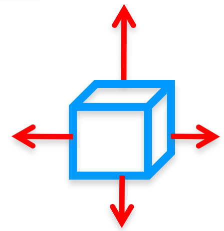
- Aerosol Representations in GCMs
  - Size representation
  - Processes (sources & sinks)
  - Properties (physical, chemical & optical)
- Uncertainties in Aerosol Processes and Properties in GCMs
  - Primary emissions
  - Secondary aerosol formation
  - Wet removal
- How Can Aerosol Representation in GCMs be Improved?

Host Models

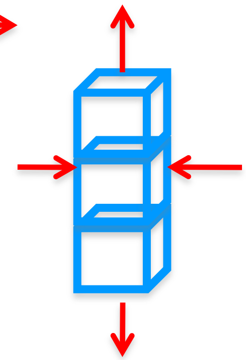
Box Model
0D, no transport, no external forcing



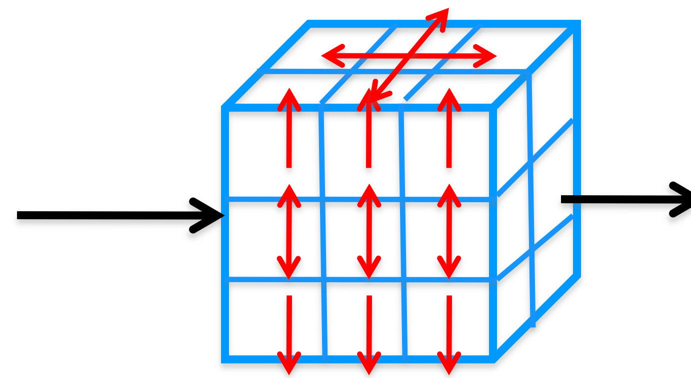
Parcel Model
0D, moved by prescribed external forcing



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

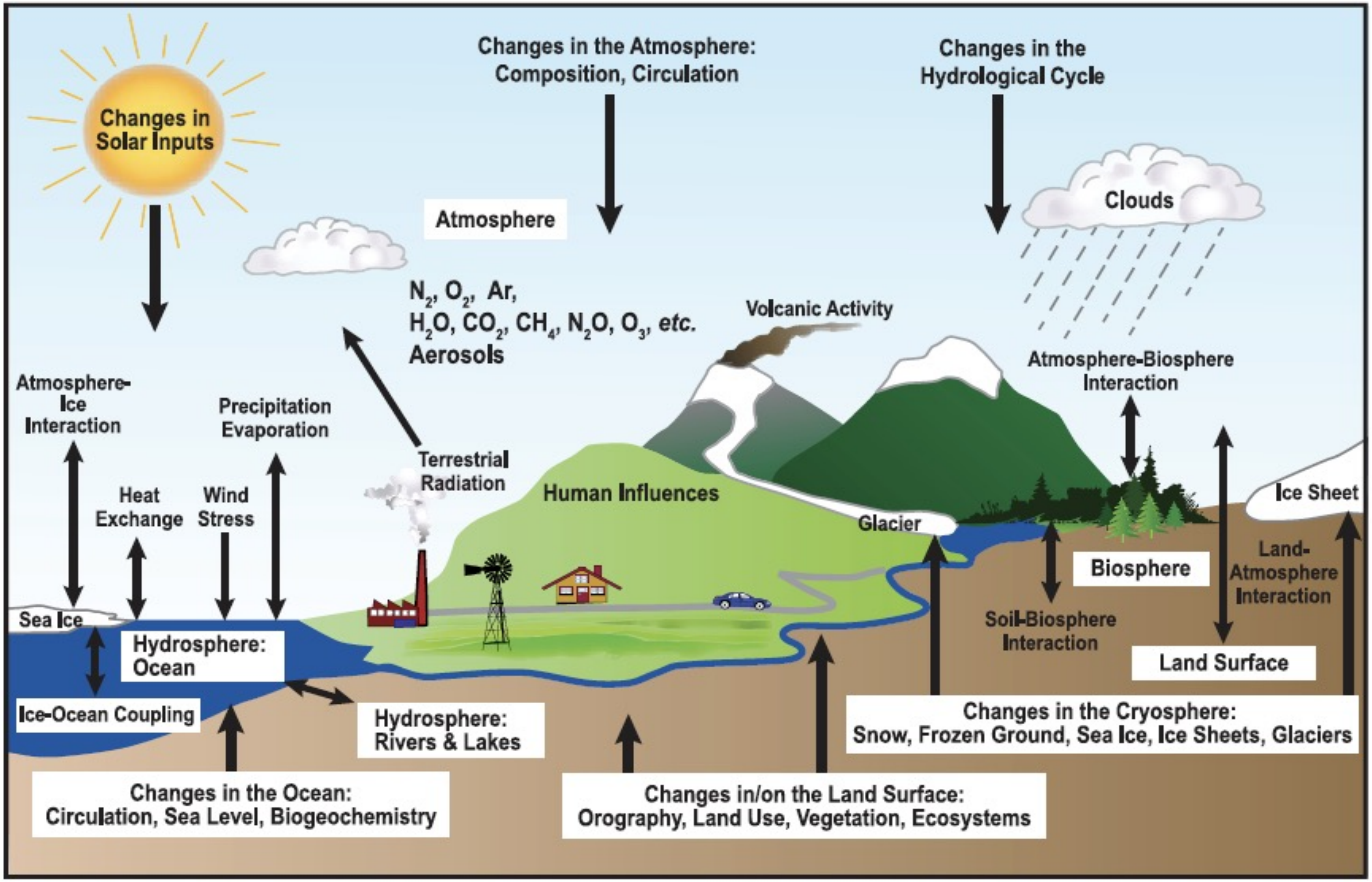


Regional Circulation Model (e.g., WRF)
3D, regional
Met-fields predicted with boundary conditions from GCMs or reanalysis data

Global Circulation Model (GCM)
3D, global, met-fields predicted, 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

Sea Salt

Fire Smoke

Dust

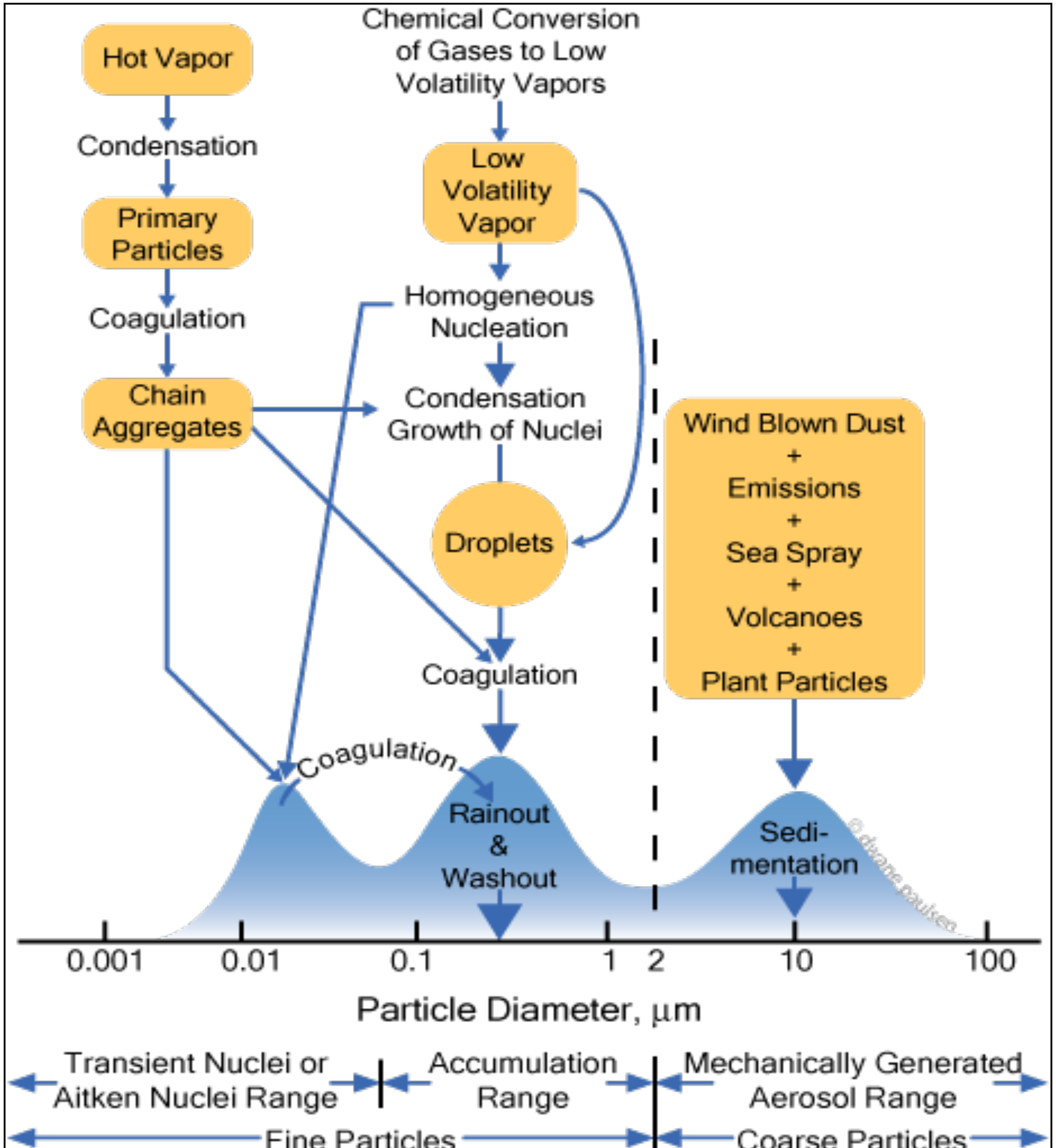
Secondary

Sulfate

Nitrate

Secondary Organics

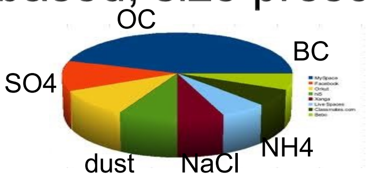
Aerosol Size and Composition in the Atmosphere

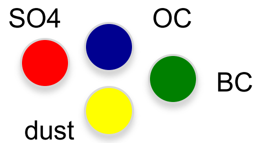


Aerosol Representation in GCMs

- **Bulk**

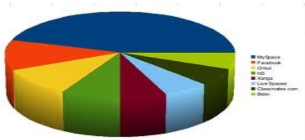
  Mass based, size prescribed, external mixture assumed, no aerosol microphysics

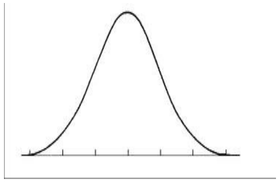


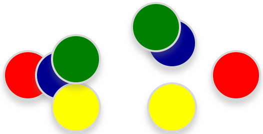


- **Moment-based (modal, 2-moment quadrature method of moments)**

  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

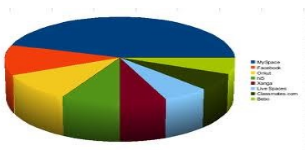


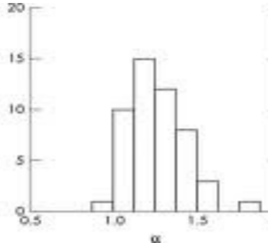


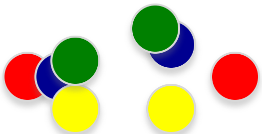


- **Sectional (bin) method**

  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 CESM1

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.

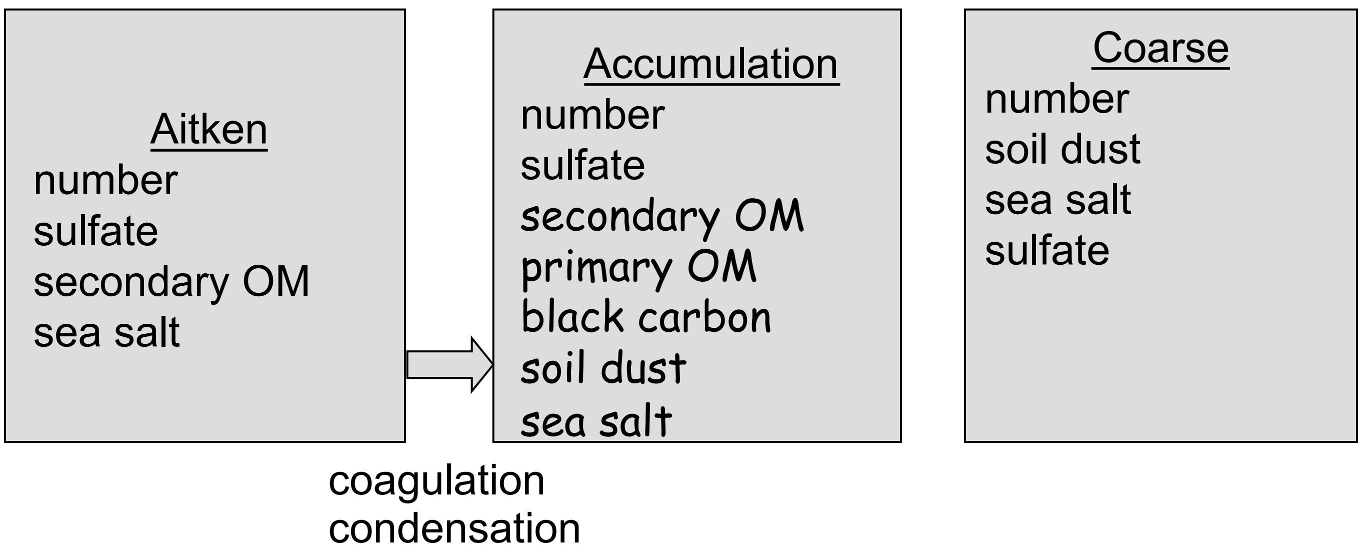
Total transported aerosol tracers: 31

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

Computer time is ~100% higher than BAM

Simplified 3-mode version of MAM in CESM1

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

4-mode version of MAM4 in CESM2/E3SM

Aitken
number
sulfate
secondary OM
sea salt

Accumulation
number
sulfate
secondary OM
primary OM
BC
soil dust
sea salt

Coarse
number
soil dust
sea salt
sulfate

coagulation
condensation

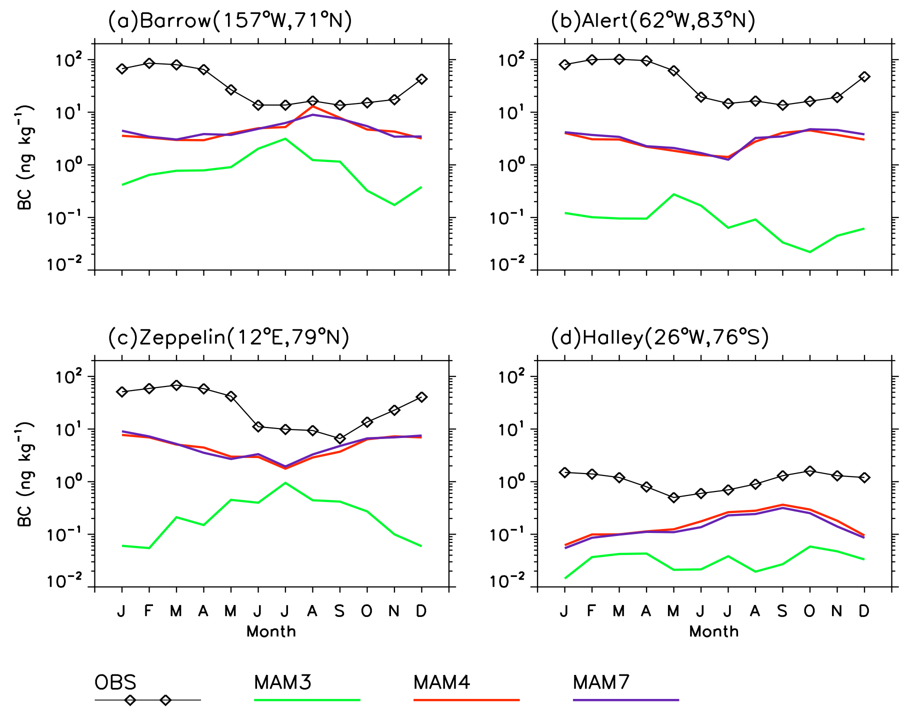
Primary Carbon
number
primary OM
BC

All modes log-normal with prescribed width.

Total transported aerosol tracers: **18**

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

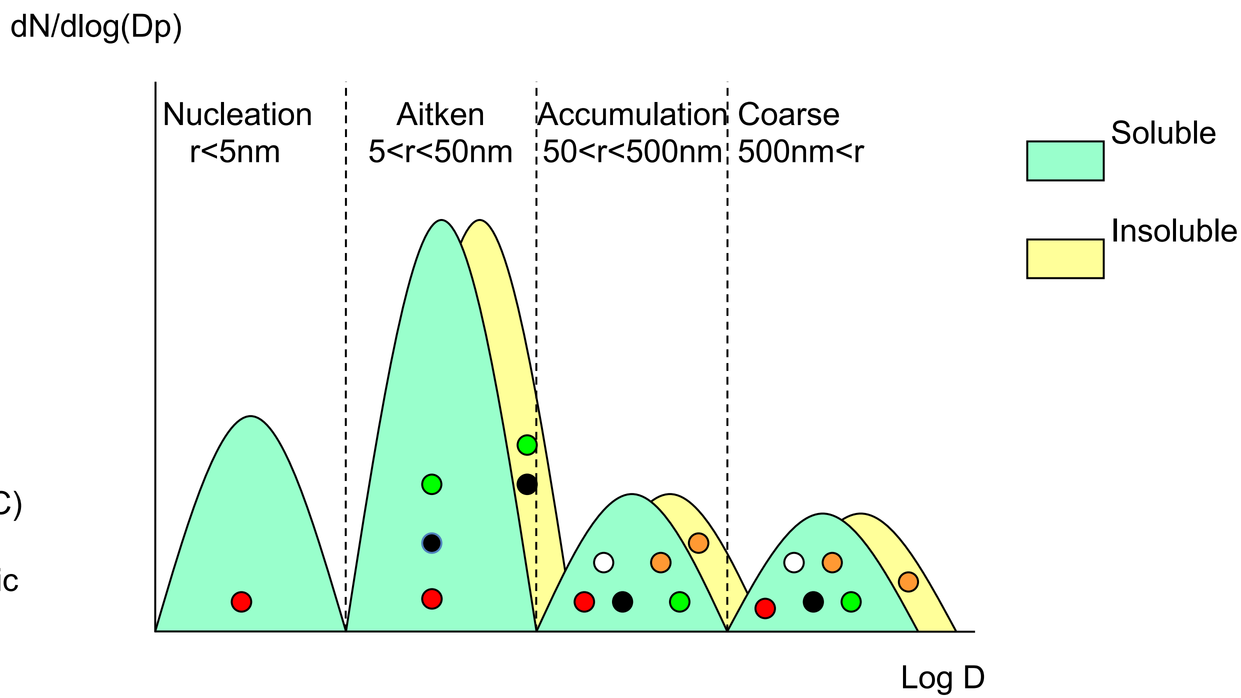
MAM4 significantly increases (and improves) BC concentration in Arctic compared to MAM3 (and agrees with MAM7). The remaining underestimation of BC concentration in Arctic in MAM4 is very likely due to wet scavenging by precipitation and/or emissions.

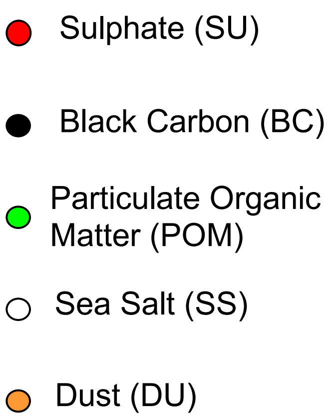


Adding a primary carbon mode in MAM4, and computer time is ~10% higher than MAM3

Comparison of model results (MAM3, MAM4, MAM7) with seasonal BC observations at surface in high latitudes

M7 (ECHAM-HAM)



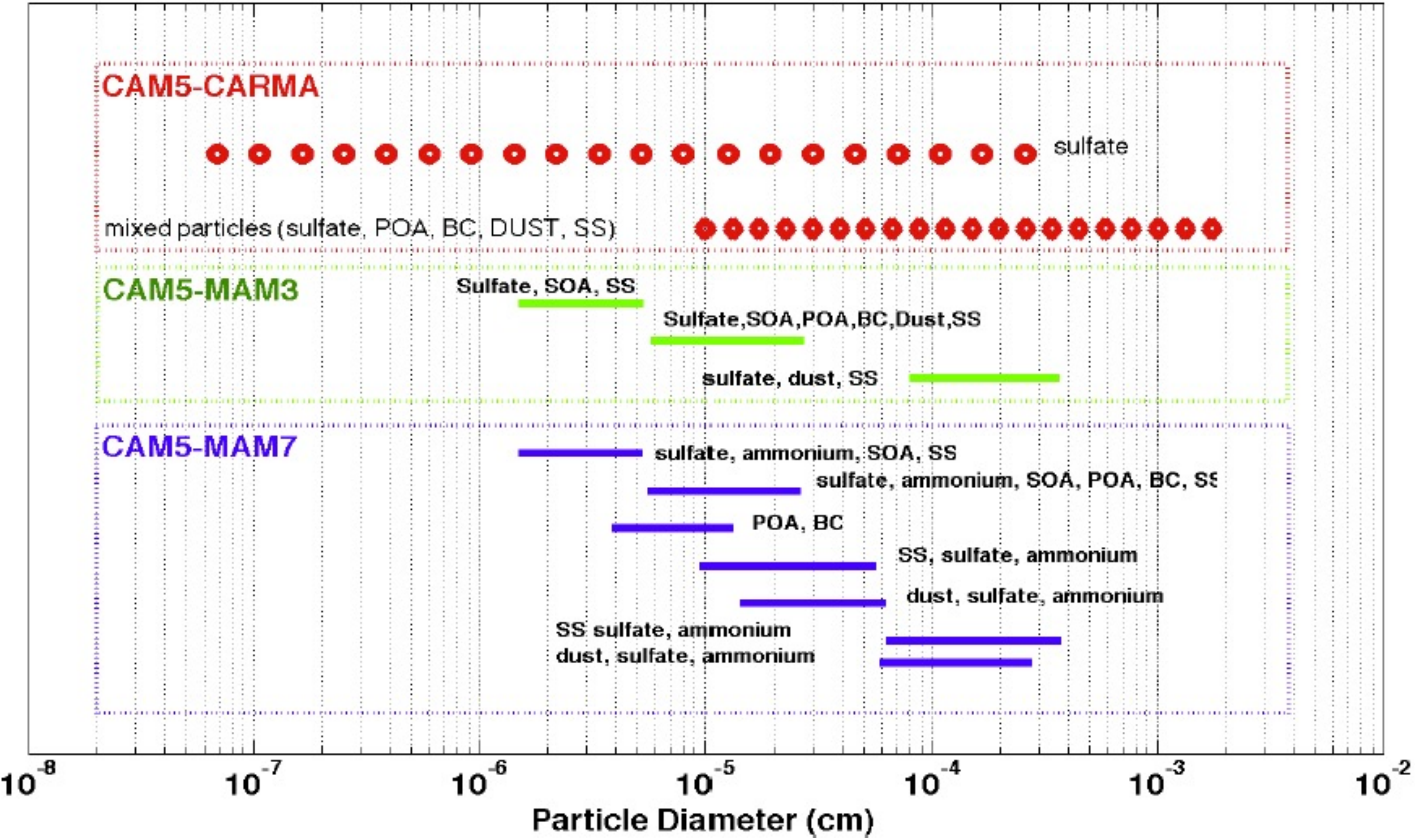


Predicted variables per mode:

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

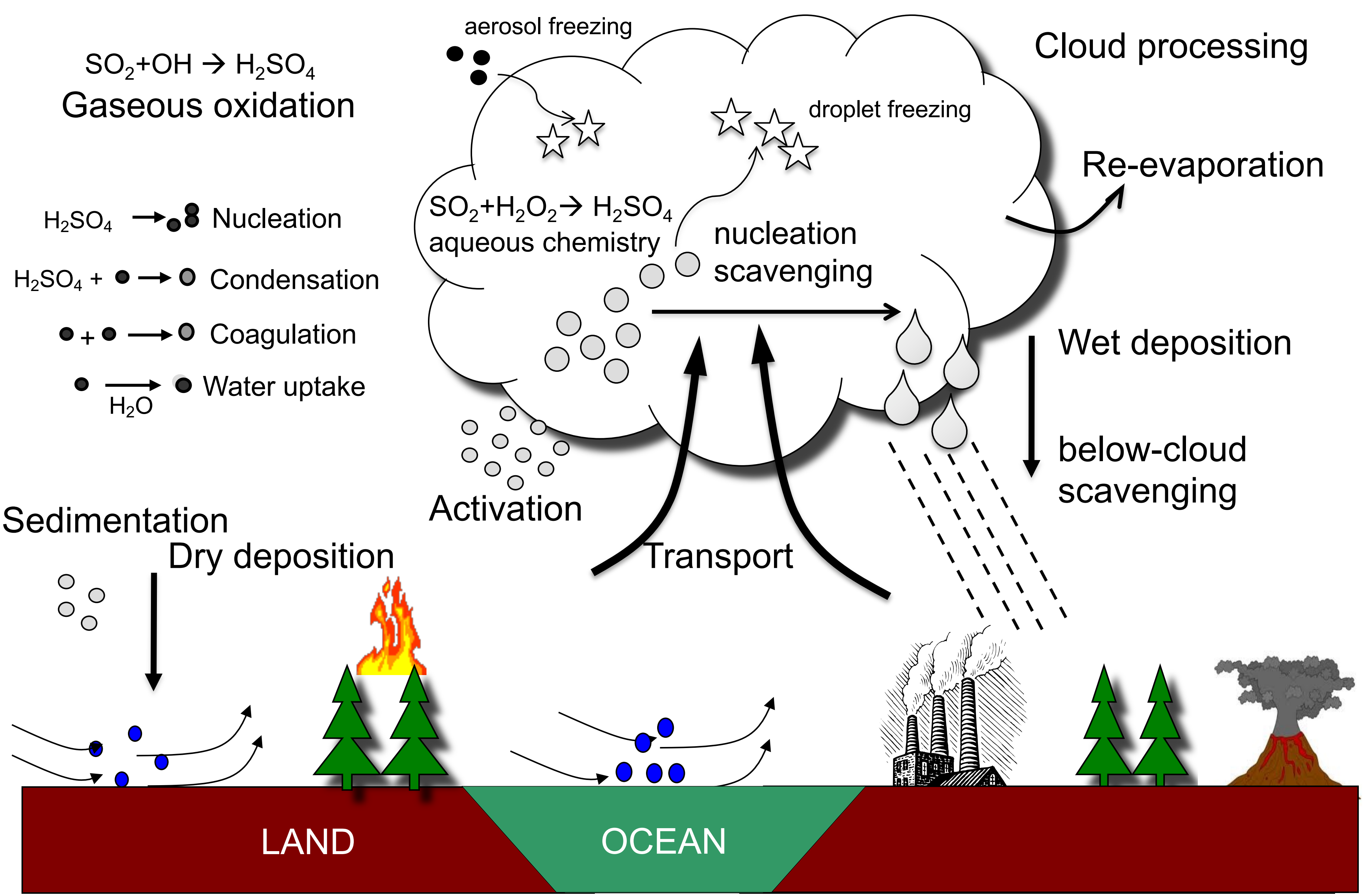
Courtecy of Declan O ʻDonnell

Sectional Aerosol Treatment in CESM-CAM5



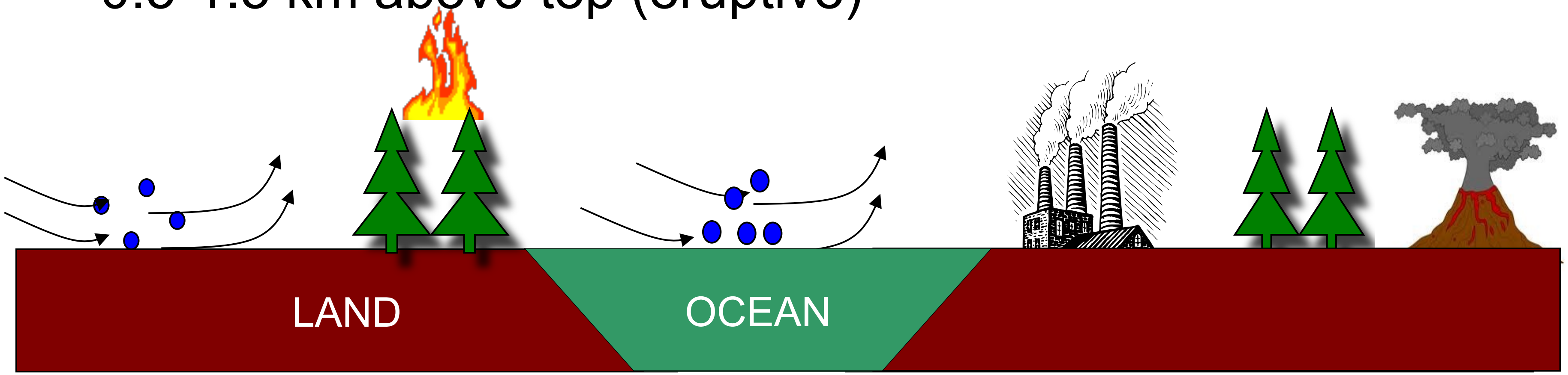
Pengfei Yu, 2015

Global Aerosol Cycles

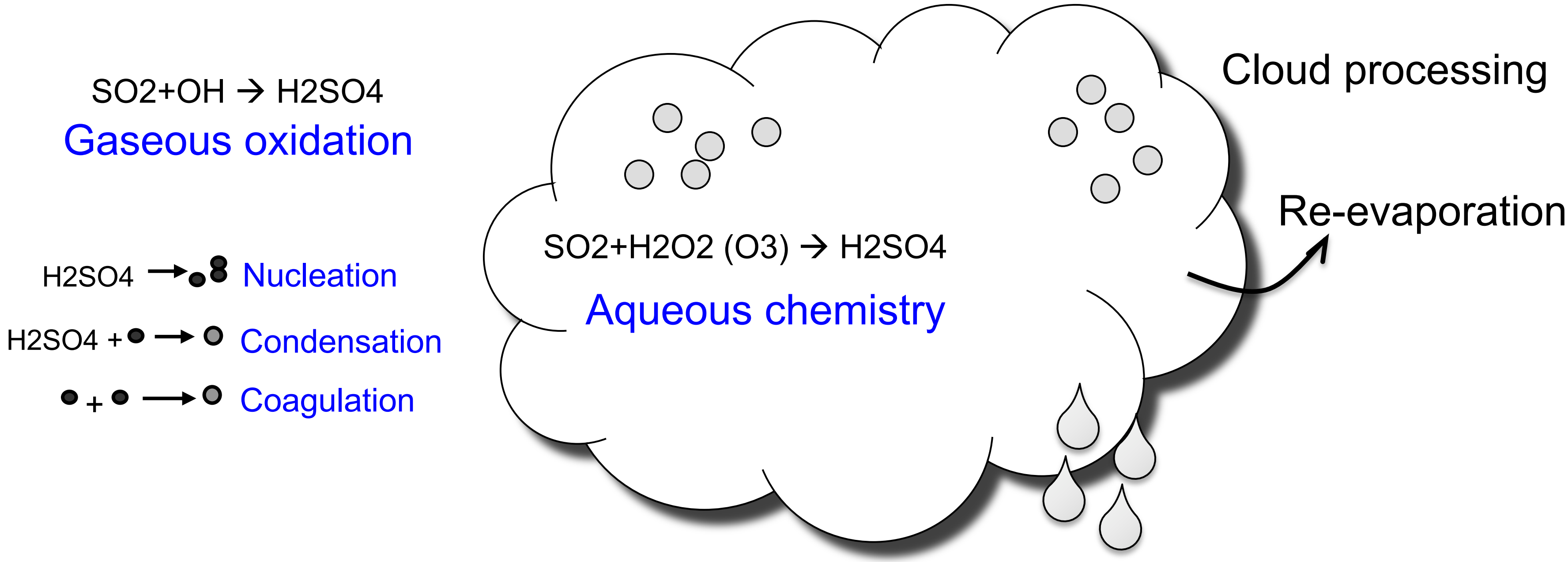


Aerosol Processes : Primary Emission

- 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:

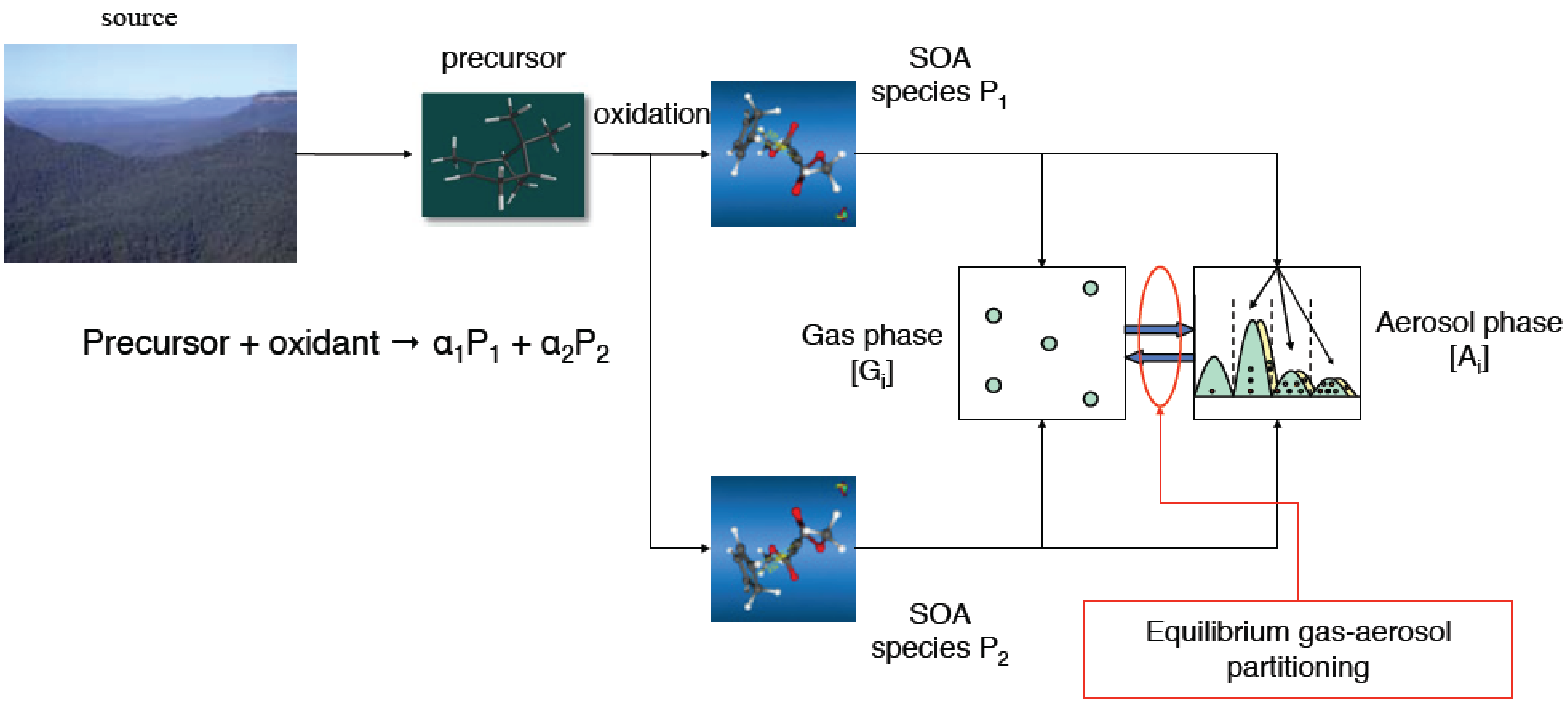
Prognostic SOA scheme with explicit gas/aerosol partitioning

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.

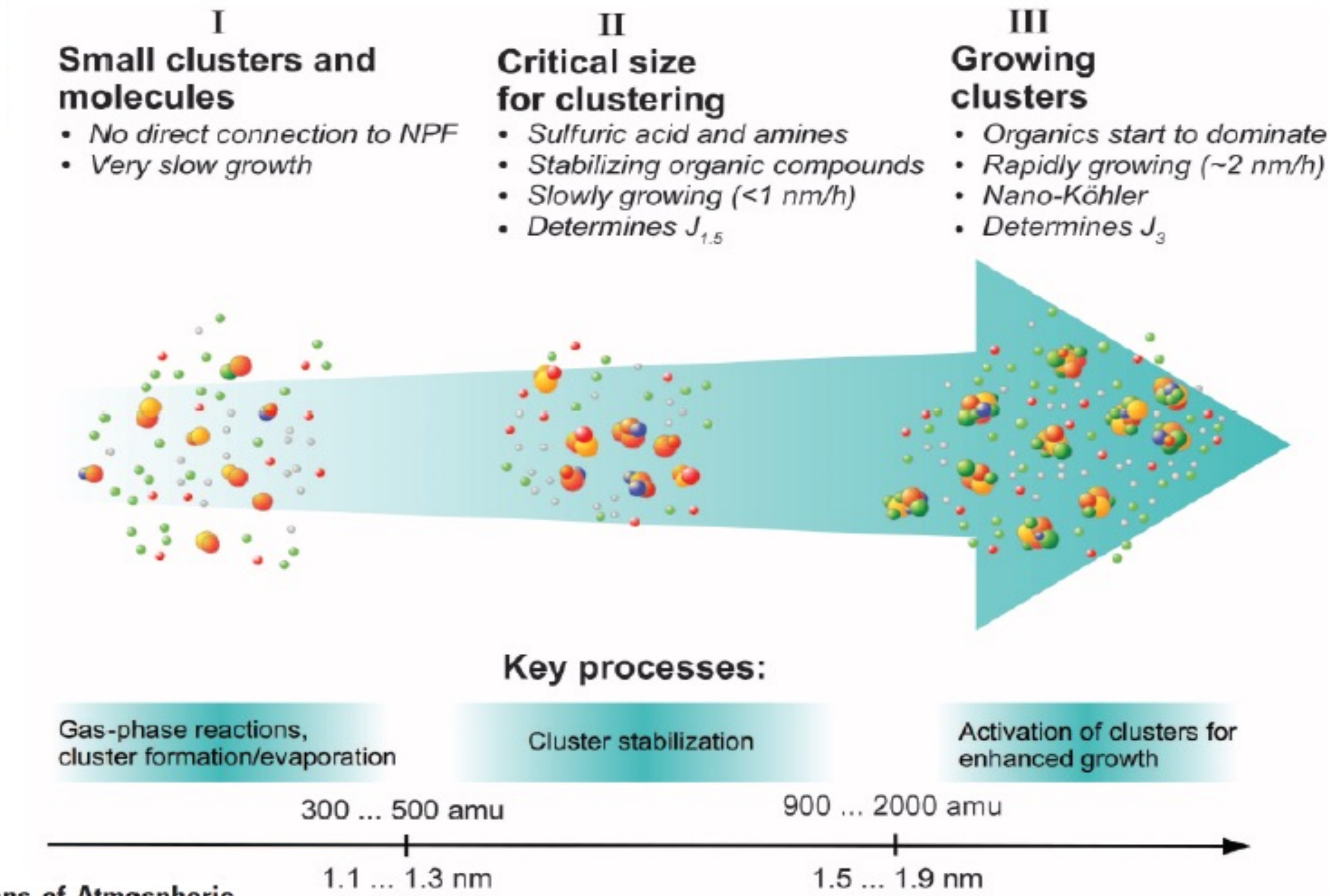
Two steps of more complexity : primary VOCs emission and oxidation in atmosphere to form SOA (g). explicit gas/aerosol partitioning of SOA (g) – ECHAM & GISS.

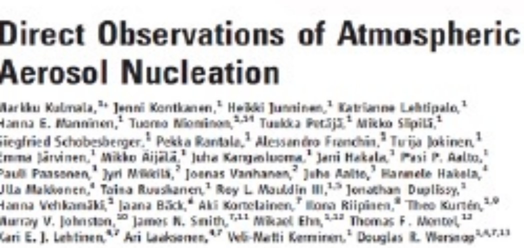
Multi-generational aging of organic vapors (VBS scheme) & treating SOA as non-volatile semi-solid (glassy) – CAM5/CAM6

SOA scheme in ECHAM-HAM2



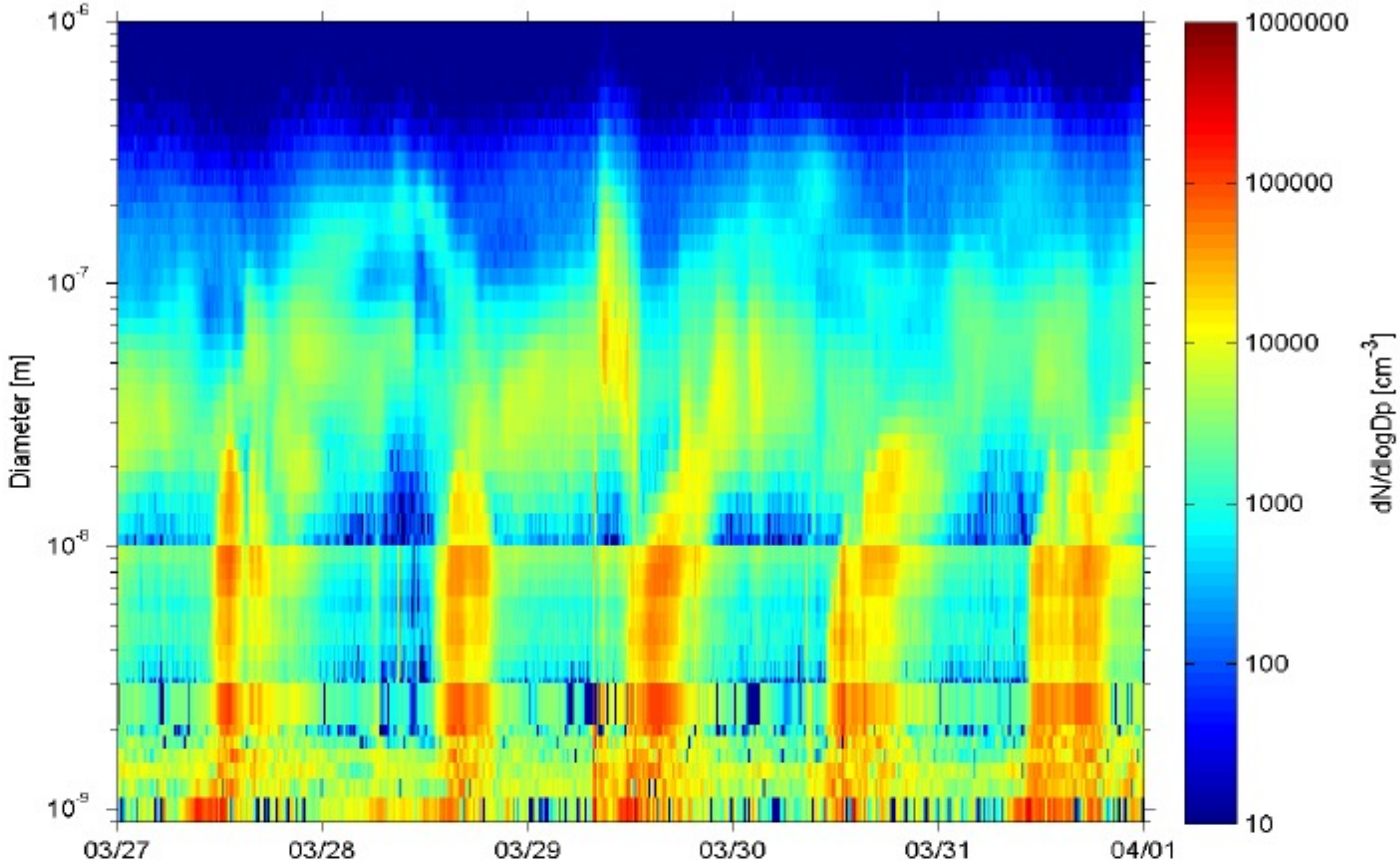
Aerosol Processes (Nucleation)





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

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-MAM4/7, ECHAM & GISS

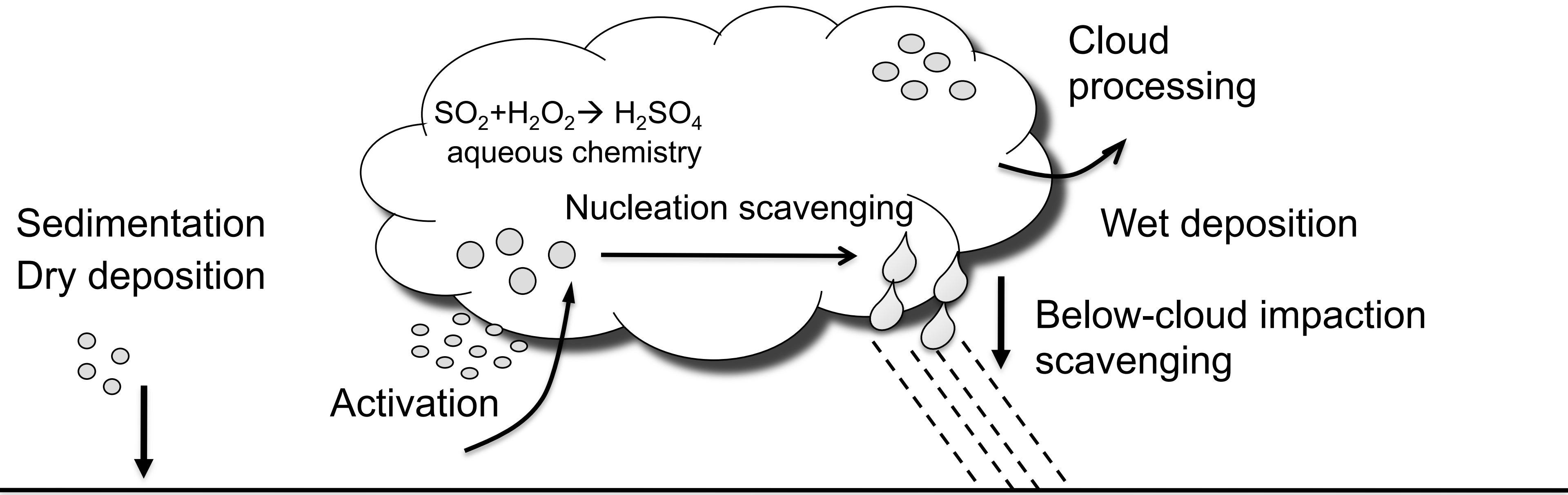
Aerosol Processes (Water Uptake)

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.

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

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

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

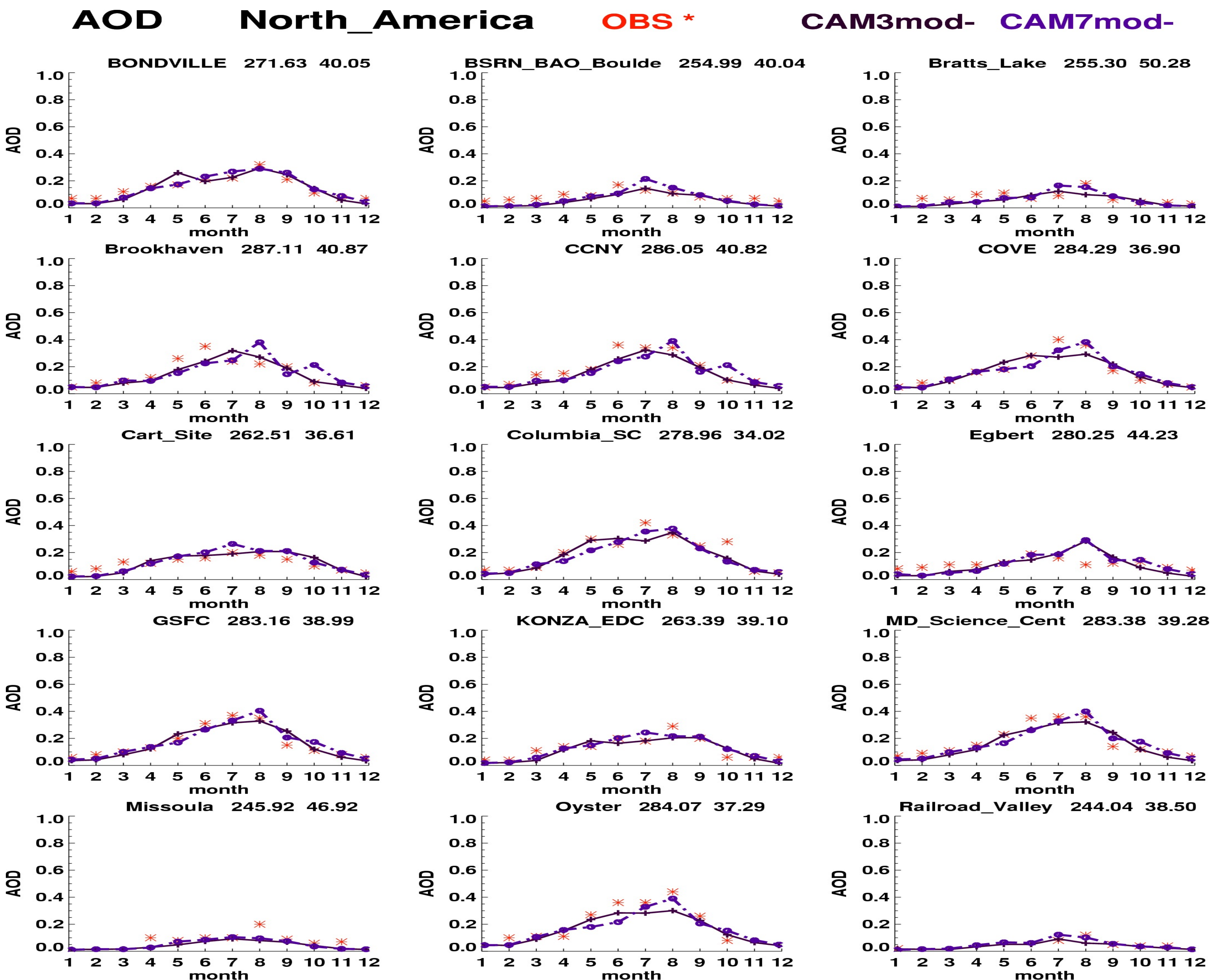
- Aerosol Representations in GCMs (CAM, GISS, ECHAM)
  - Size representation
  - Processes (sources, sinks)
  - Properties (physical, chemical, optical)
- Uncertainties in Aerosol Processes in GCMs
  - Primary emissions
  - Secondary aerosol formation
  - Wet removal

Uncertainties in Aerosol Processes in GCMs

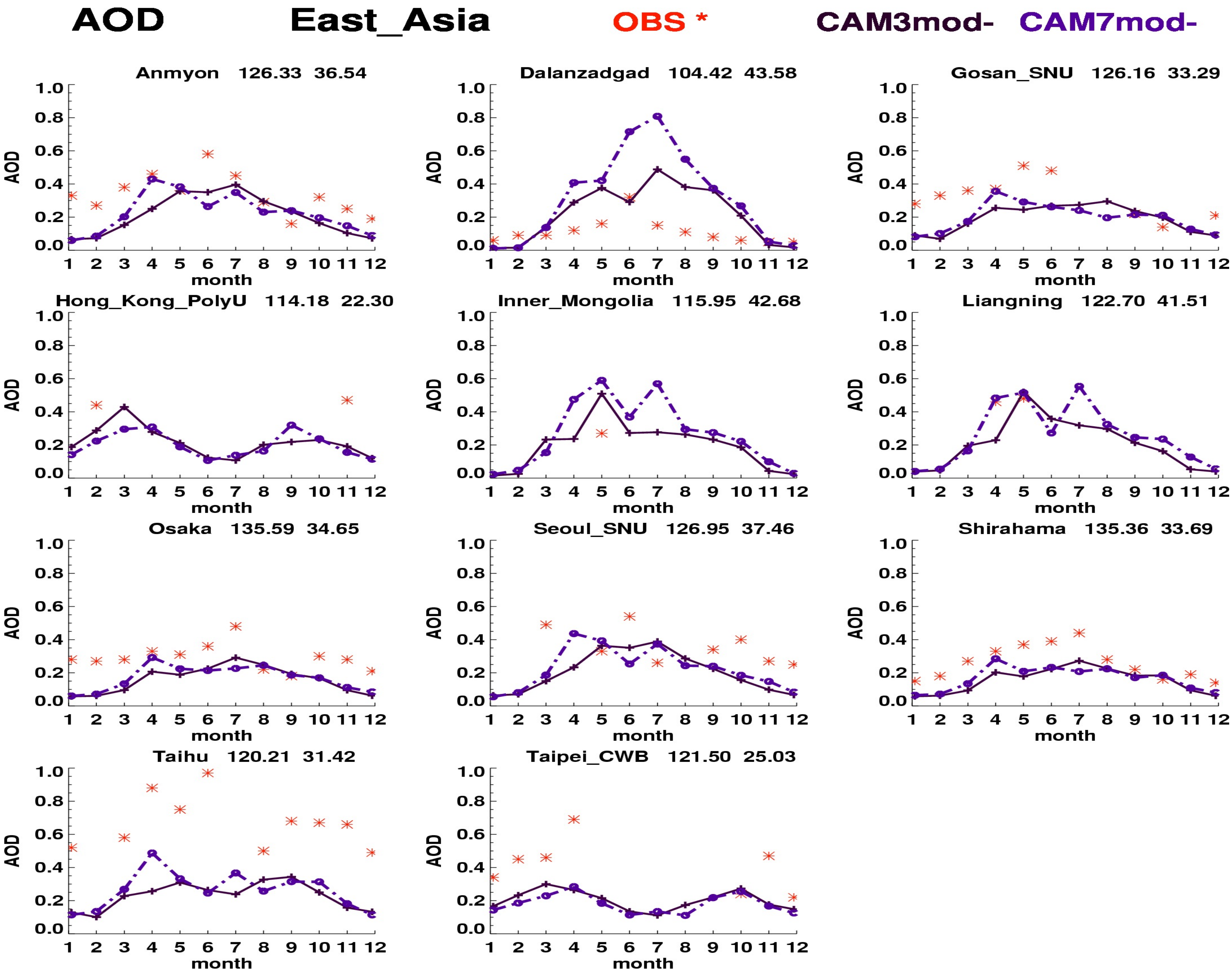
- Primary emissions: mass flux, size distribution, injection height

  - ➢ 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)

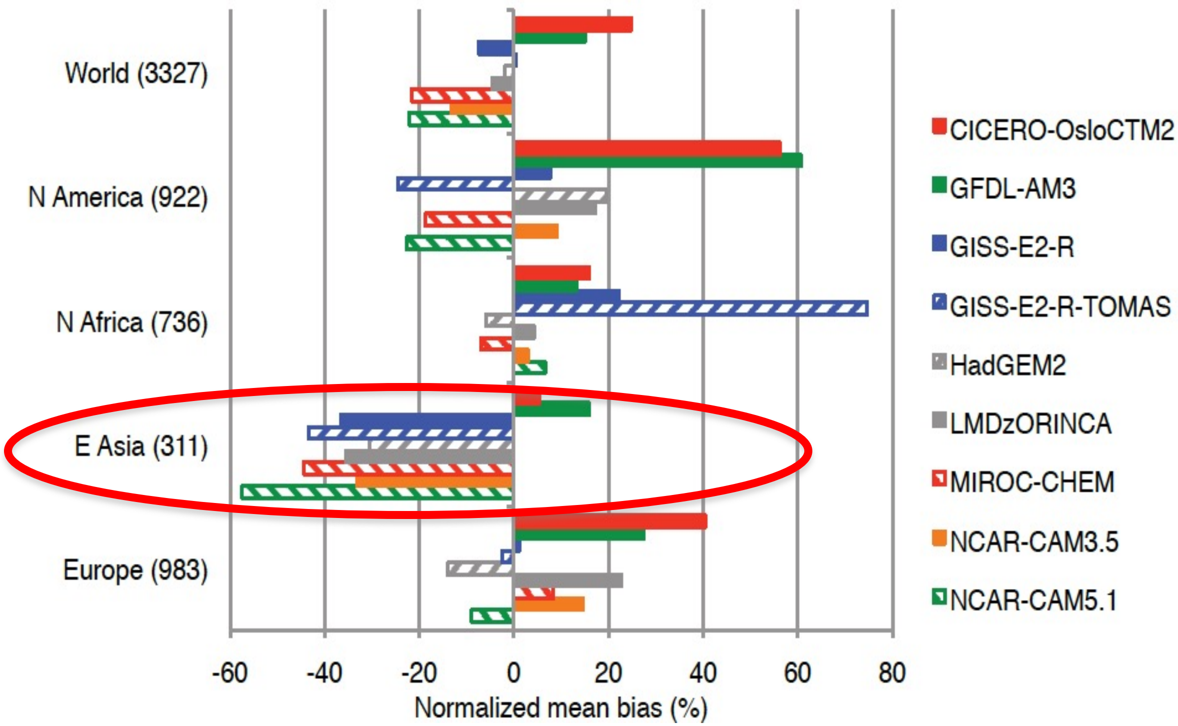
Effect of Primary Emissions



Effect of Primary Emissions



Underestimation of aerosols in East Asia

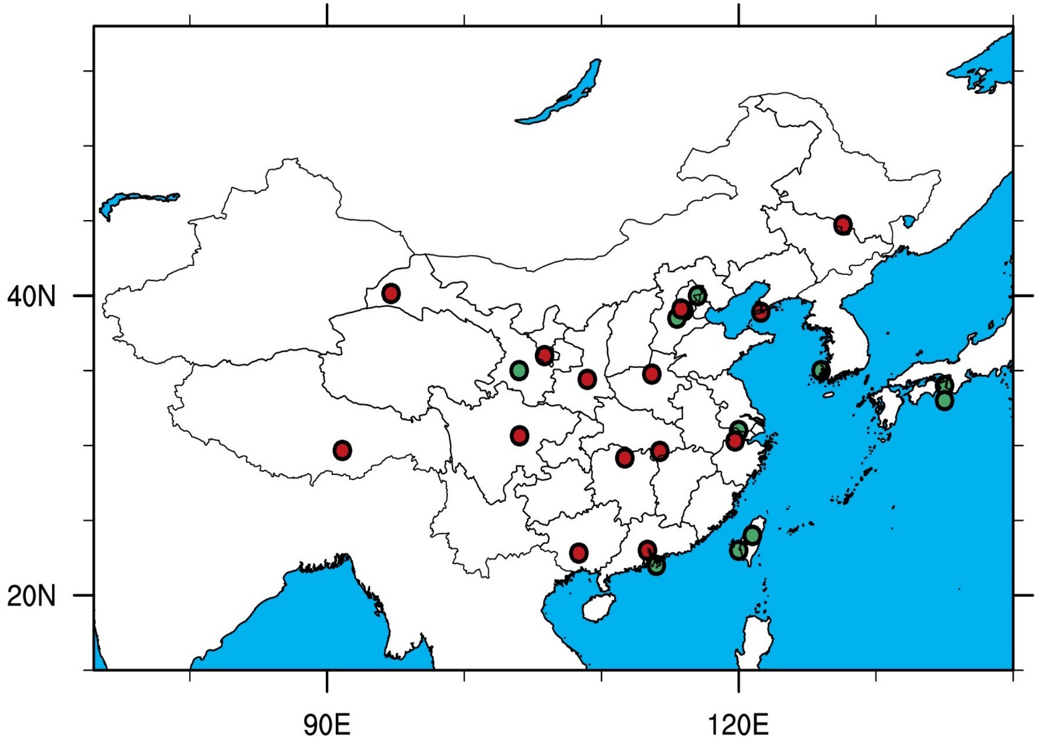


"Nearly all models show large negative biases over East Asia. The two models that do not show a large negative bias over East Asia show the largest positive biases over both Europe and North America, indicating they are systematically higher than the other models rather than matching East Asia observations better"

Comparison of Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) models with AERONET AOD

(Shindell et al. 2013)

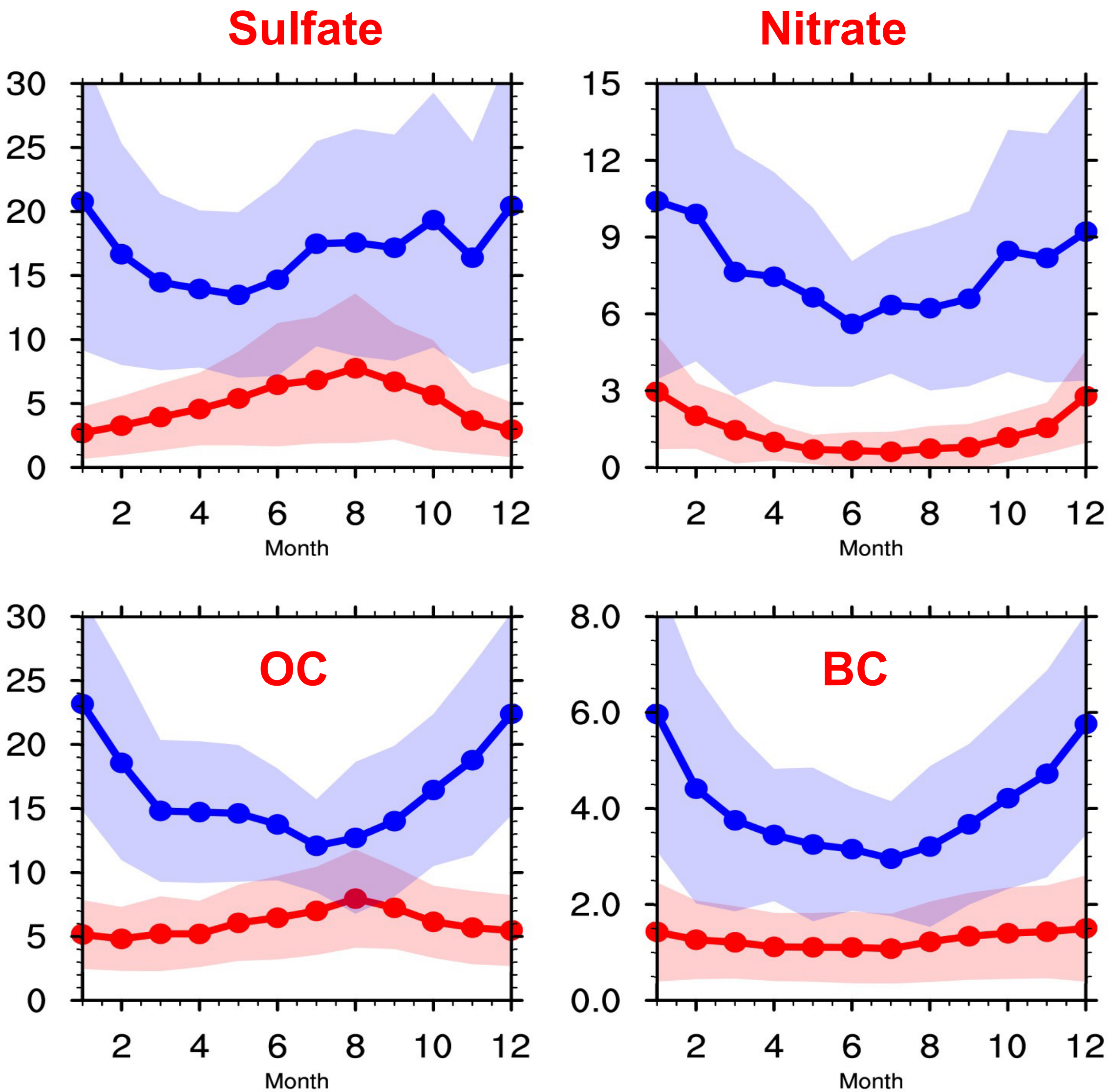
Long-term aerosol composition measurements used for model evaluation



- **Measured concentrations** at 14 CAWNET sites (**red circles**) of the China Meteorological Administration (CMA) Atmosphere Watch Network (**CAWNET**)

- **Measured AOD** from
  - ✧ AERONET sites (**green circles**) (Holben et al., 1998)
  - ✧ Satellite measurements (MODIS, MISR)

Simulated surface concentrations from ACCMIP models vs. measurements at CAWNET sites



Red: Multi-model mean conc.;

Blue: observed conc.;

Both simulated and measured concentrations are averaged over 14 CAWNET sites

Why are aerosols in East Asia underestimated in GCMs?

- Anthropogenic aerosol emissions underestimated?
- Aerosol processes under-represented or missing?
  - Nitrate
  - SOA
  - Dust-sulfate/nitrate chemistry interactions
- GCM model resolution too coarse? Subgrid variability

NCAR CAM5.2

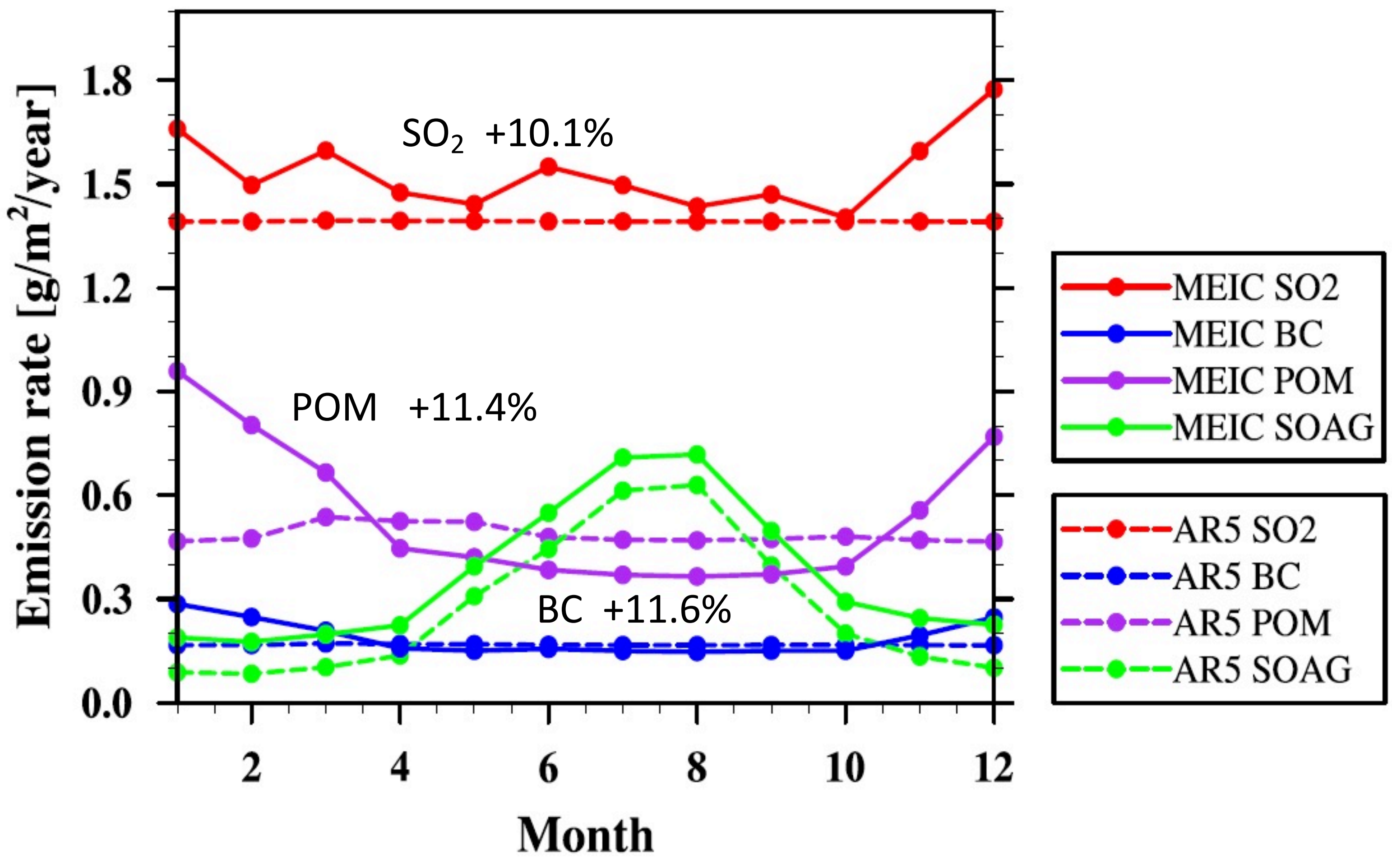
- Six-years simulations (2006~2011) nudged by ECMWF re-analysis data
- At $1.9^{\circ} \times 2.5^{\circ}$ resolution

New aerosol emission for China

- IPCC AR5 emission
  - update every 10 years
  - *no seasonal variation for anthropogenic aerosols*
  - horizontal resolution: 0.5^o x 0.5^o or model-dependent
  - anthropogenic, biogenic, and biomass burning aerosols

- Multi-scale Emission Inventory for China (MEIC)
  - technology-based
  - update every year
  - *seasonal variation*: monthly mean
  - horizontal resolution: 0.25^o x 0.25^o, 0.5^o x 0.5^o, 1^o x 1^o
  - anthropogenic aerosols only

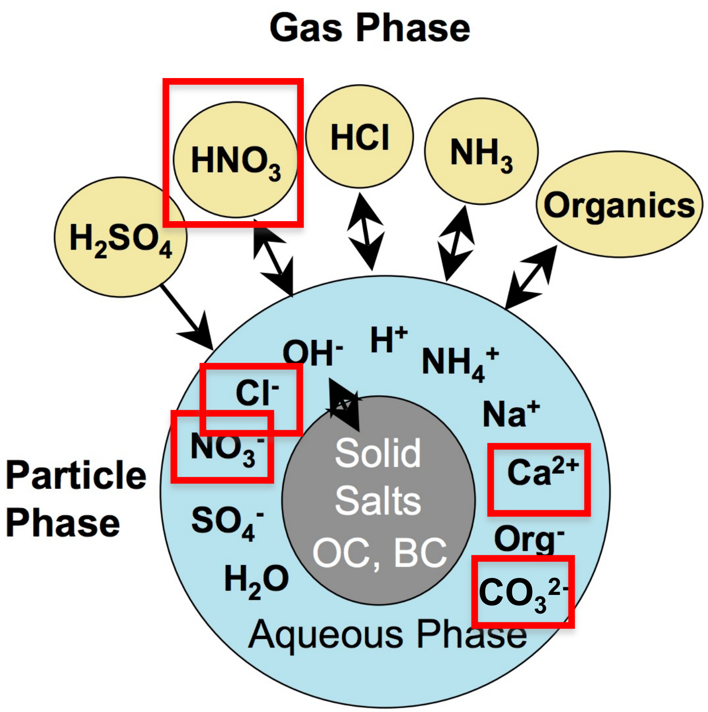
AR5 and MEIC emissions in East China



Nitrate aerosol in CAM5

- In order to treat NO3 aerosol, Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) module [Zaveri et al., 2008] is coupled with MAM4 and MAM7 (MOSAIC-MAM4/7)

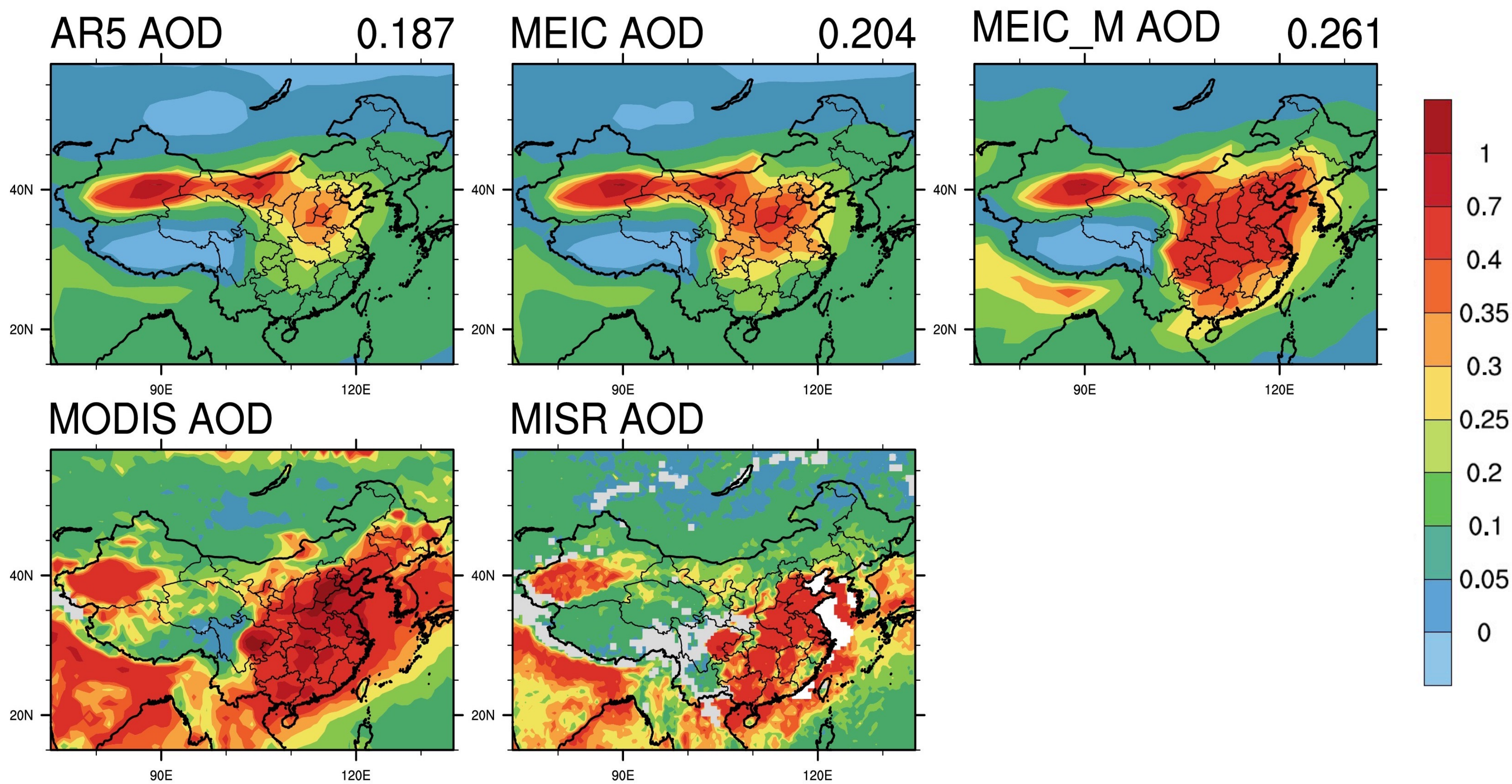
- In the version of MAM coupled with MOSAIC, *gas-aerosol exchange* is treated by MOSAIC. The remaining processes are still treated by MAM



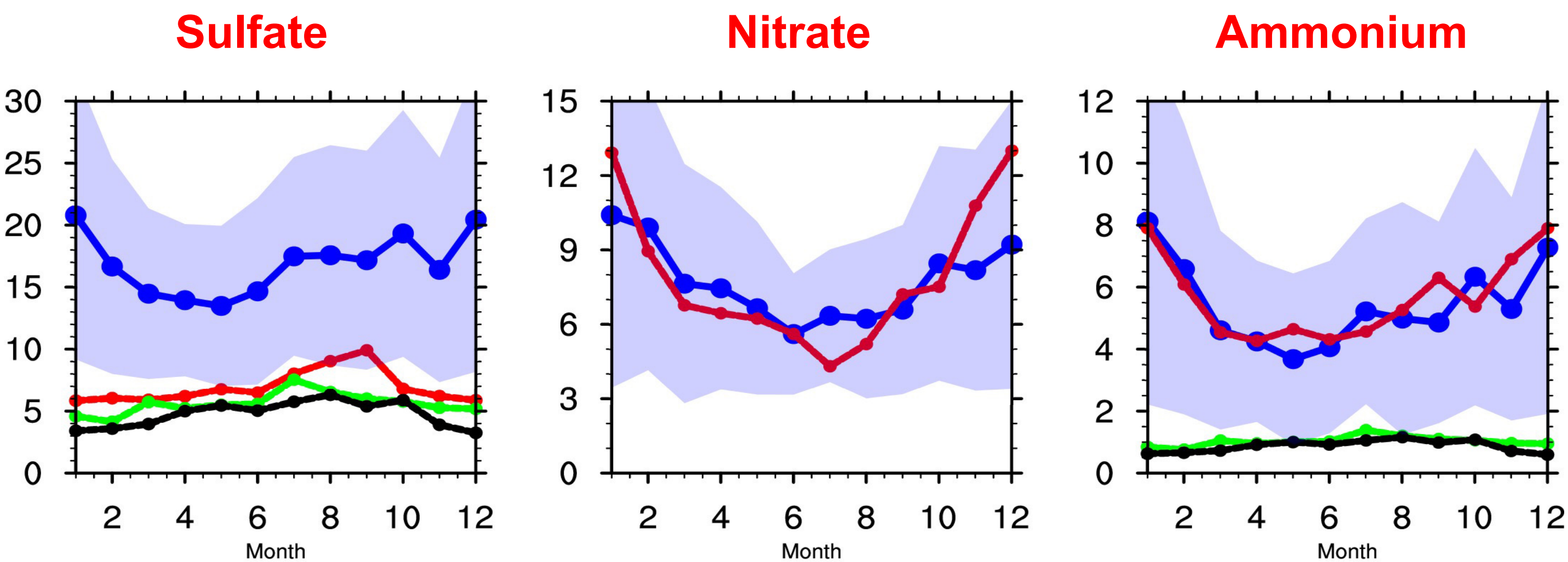
Source: presentation by Zaveri WRF tutorial, 2008

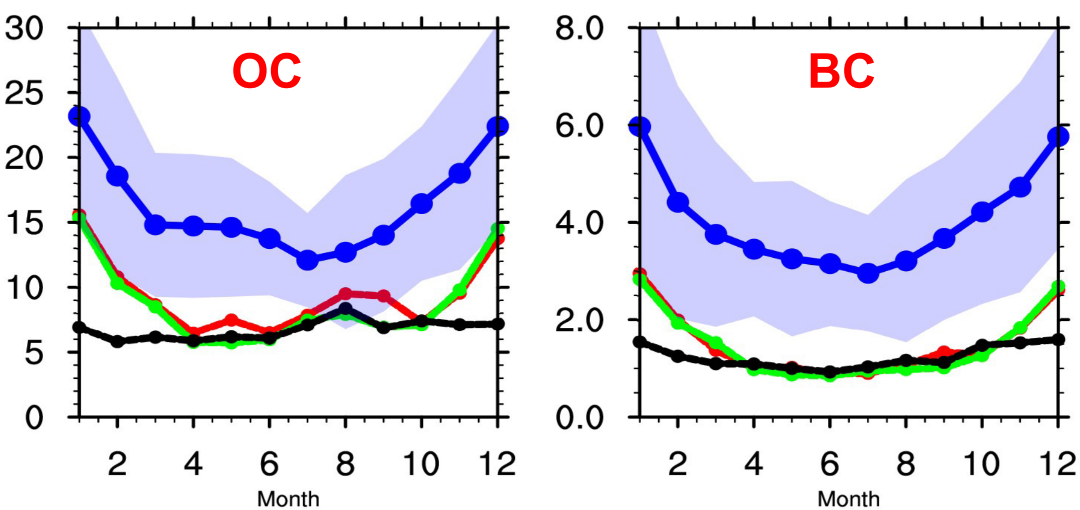
Lu et al. (2021), Zaveri et al. (2021)

Emission accounts for 16%-21%,
emission & nitrate account for 63%-86% of the modeled AOD low biases in eastern China



Simulated surface concentrations from CAM5 vs. measurements at CAWNET sites





Blue: CAM5 w/ AR5 emission
green: CAM5 w/ MEIC emission
Red: CAM5 w/ MEIC & nitrate

Blue: observed conc.

Uncertainties in Aerosol Processes in GCMs

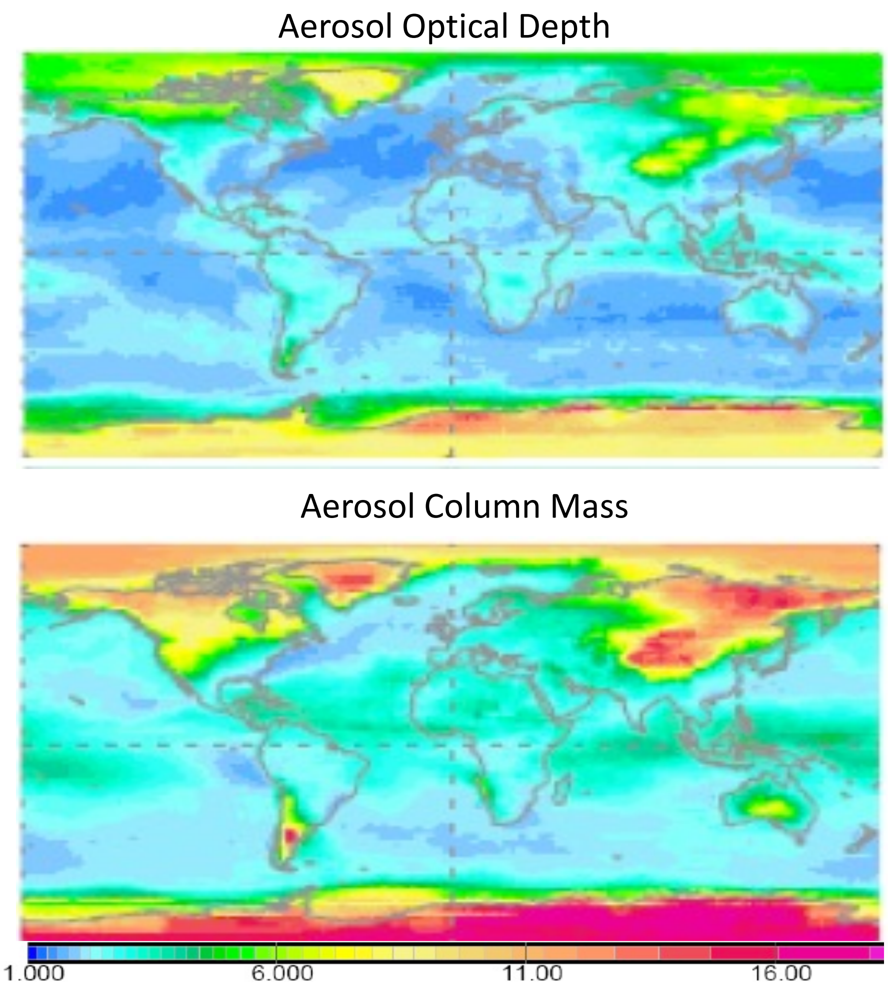
- Wet removal
  - Cloud water content, cloud fraction
  - Treatment of aerosol wet removal
  - Aerosol processes in convective clouds

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.

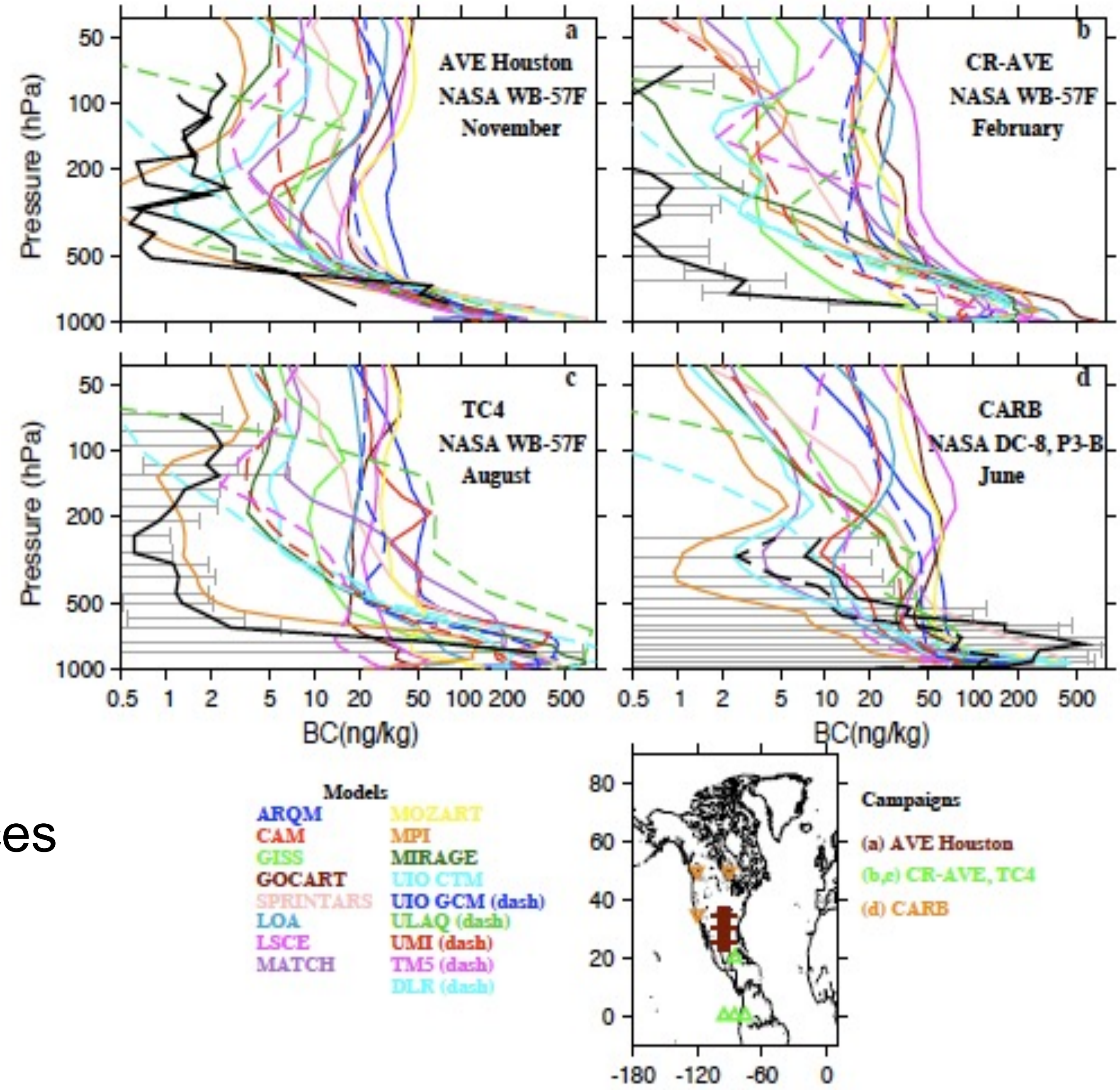
Major differences in poles

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



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

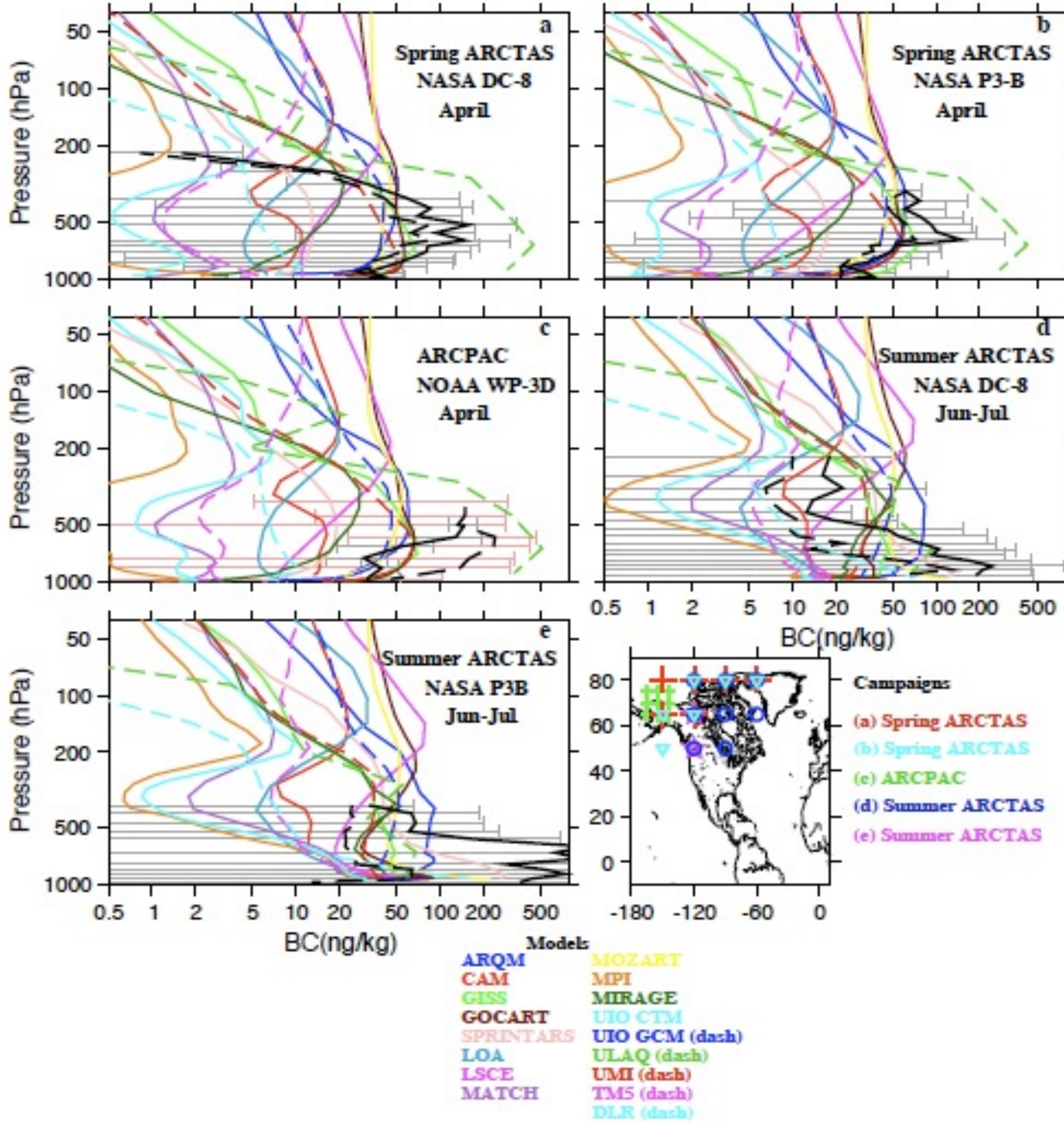
BC compared with SP2 (tropics and mid-lat.)



Major differences in free troposphere

Koch et al. (2009)

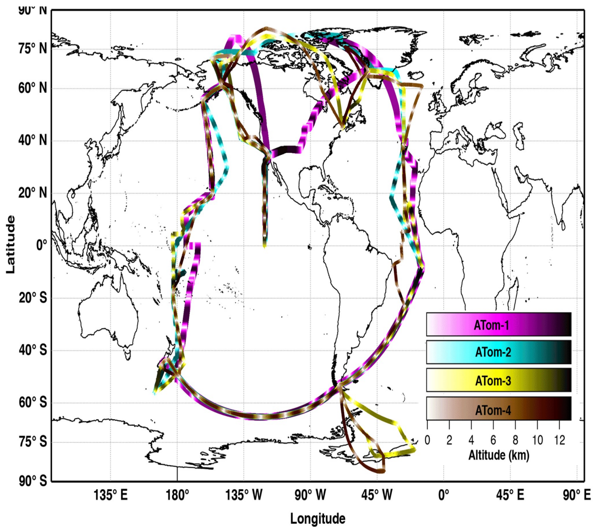
BC compared with SP2 (high-latitudes)



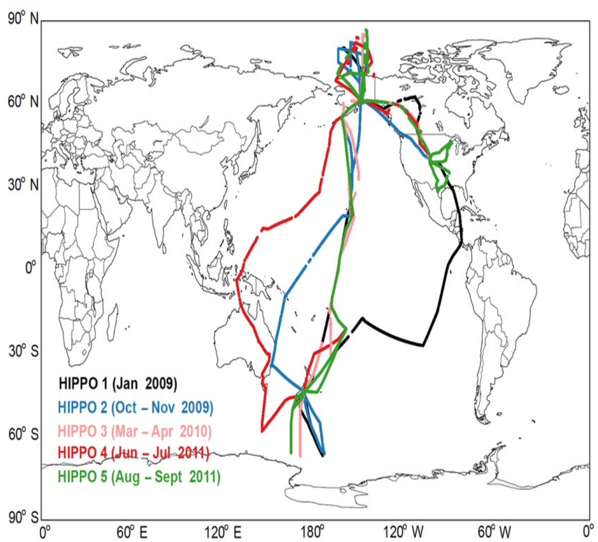
Koch et al. (2009)

Impact of convective processes in CESM2 on BC

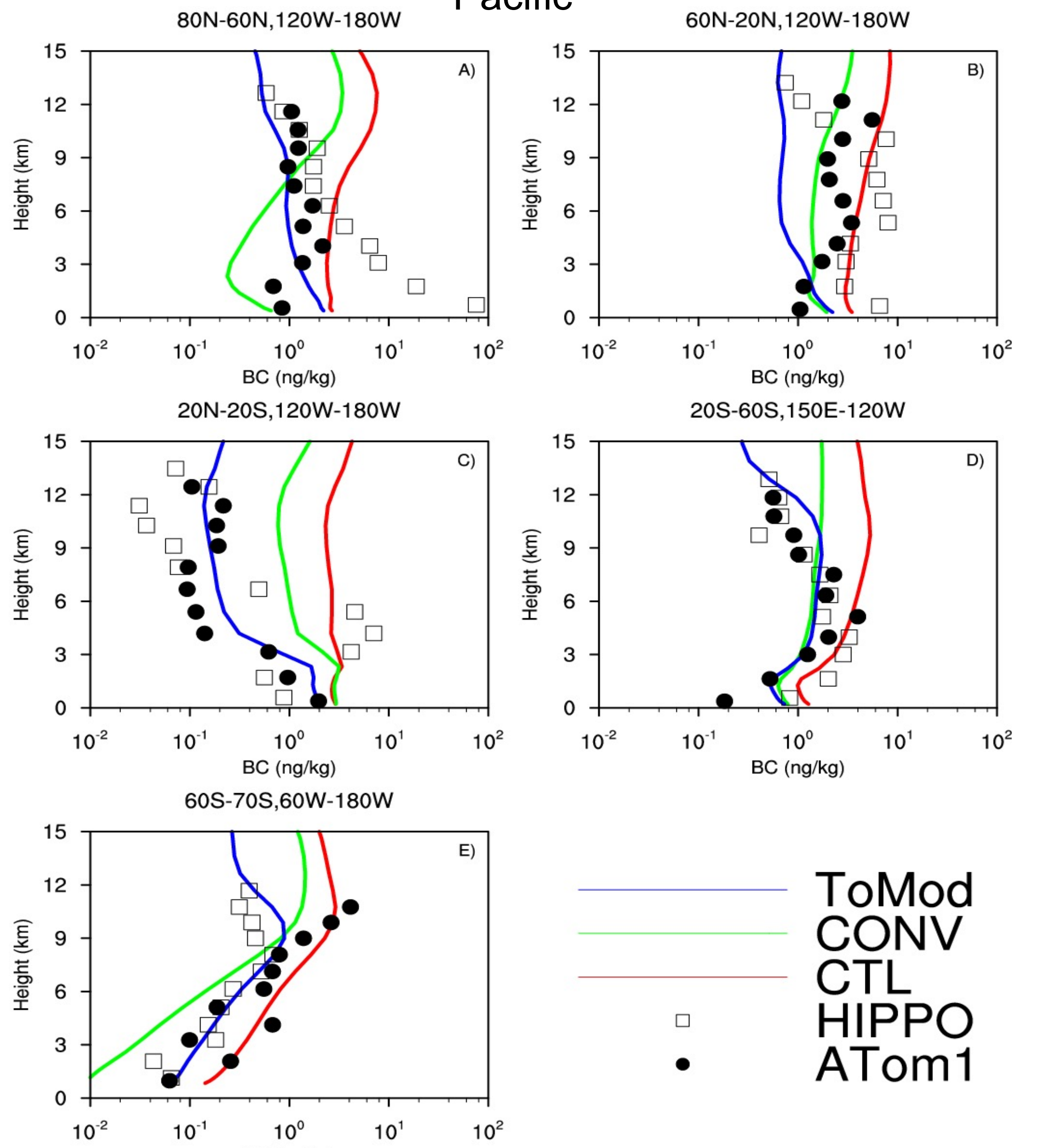
NASA-ATom



NSF-HIPPO



Pacific



Shan et al. (2021), Wang et al. (2013)

Outline

- Aerosol Representations in GCMs (CAM, GISS, ECHAM)
- Uncertainties in Aerosol Processes and Properties in GCMs
- How Can Aerosol Representation be Improved in GCMs?

How Can Aerosol Representation in GCMs be Improved?

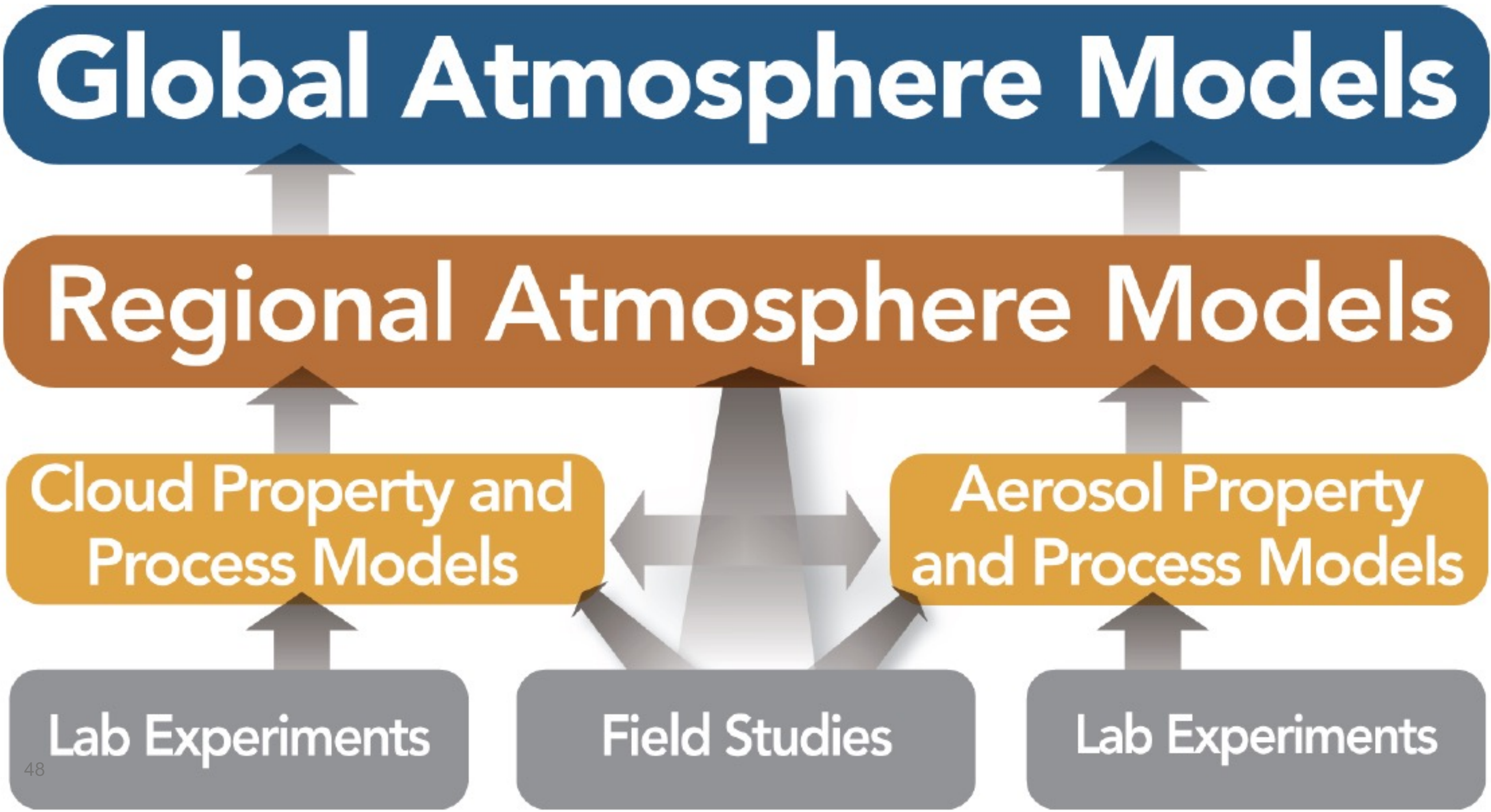
Processes :

- Improve primary emissions: flux, size distribution and injection heights
- Aerosol nucleation and growth (BL nucleation, role of organics)
- SOA production and evaporation
- Wet scavenging (cloud and precipitation in GCMs)

Properties :

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

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



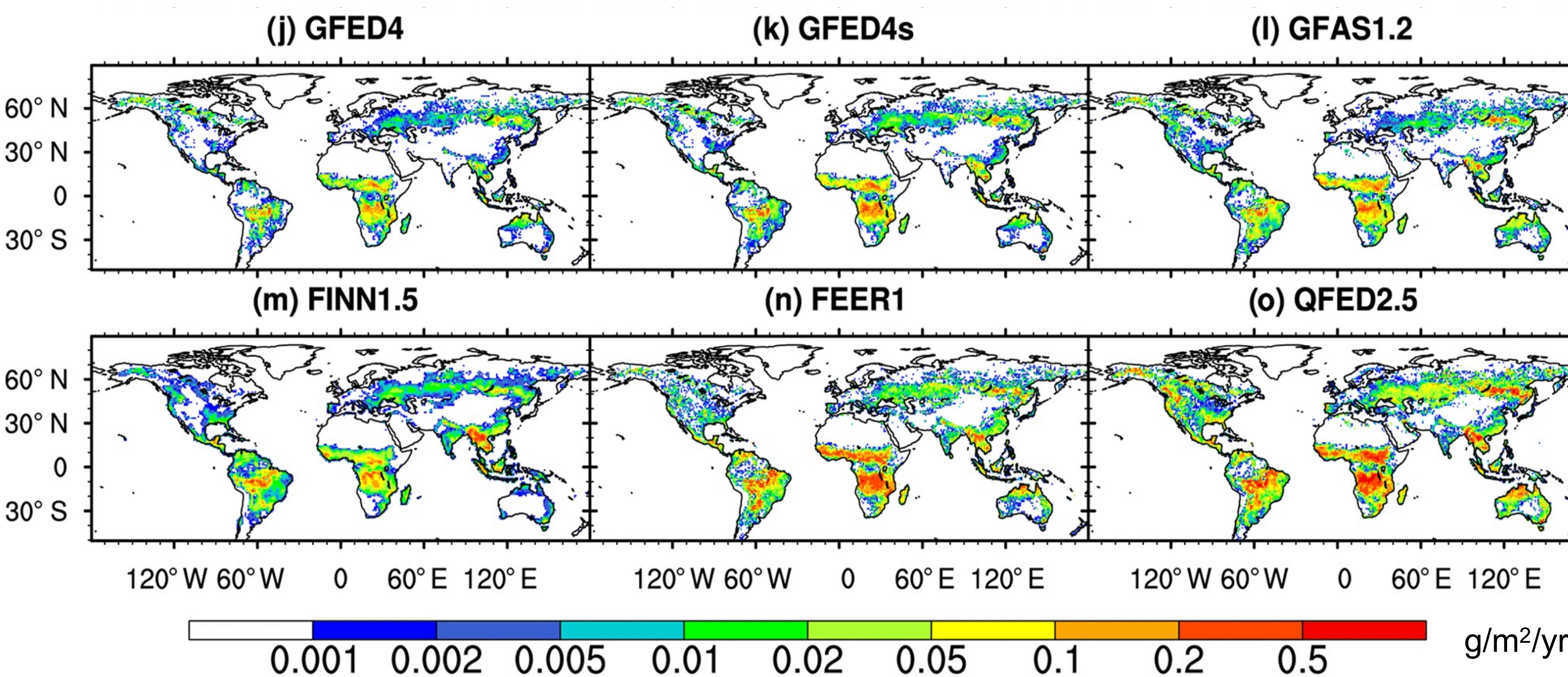
Thanks!

Book Chapter:

Liu, X., "Aerosols and Climate Effects",

In: *Fast Physics in Large Scale Atmospheric Models: Parameterization, Evaluation, and Observations* [Y. Liu, P. Kollias, L. Donner (eds.)], Wiley Publisher, in press, 2022.

Annual fire BC emissions from satellite-based products averaged over 2003-2008



Modified from Li et al. (2019)

Two types of fire emissions: burned area (BA)-based, like GFED and active fire or fire radiative power (FRP)-based emissions, like FINN and QFED.

Over India, small-size fires dominate, probably related to agriculture activity. FRP-based emission datasets perform better in capturing these small fires.