

Aerosol Representation in GCMs

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Thanks to

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

K. Zhang (MPI Meteorology)

P. Stier (Univ. Oxford)

S. Bauer (NASA GISS)

C. Chuang (LLNL)

DOE ASR Aerosol Lifecycle WG
October 11, 2010

Questions (by Jian Wang):

- 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)?

Host Models

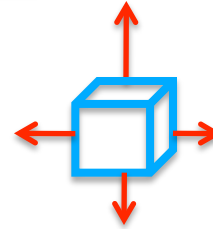
Box Model

0D, no transport, no external forcing



Parcel Model

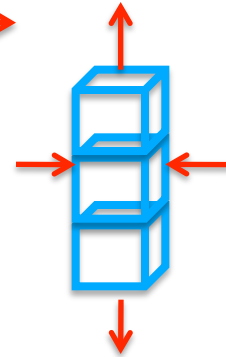
0D, moved by prescribed external forcing



Single Column Model (SCM)

1D, vertical transport

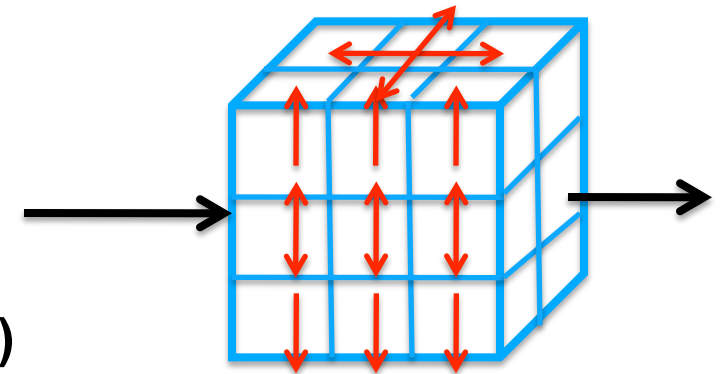
External forcings (e.g., [campaign](#))



Chemical Transport Model (CTM)

3D, regional or global

Met fields prescribed from GCMs or reanalysis,
[no feedbacks of aerosol & chem](#) on met fields



Regional Circulation Model (e.g., WRF-CHEM)

3D, regional

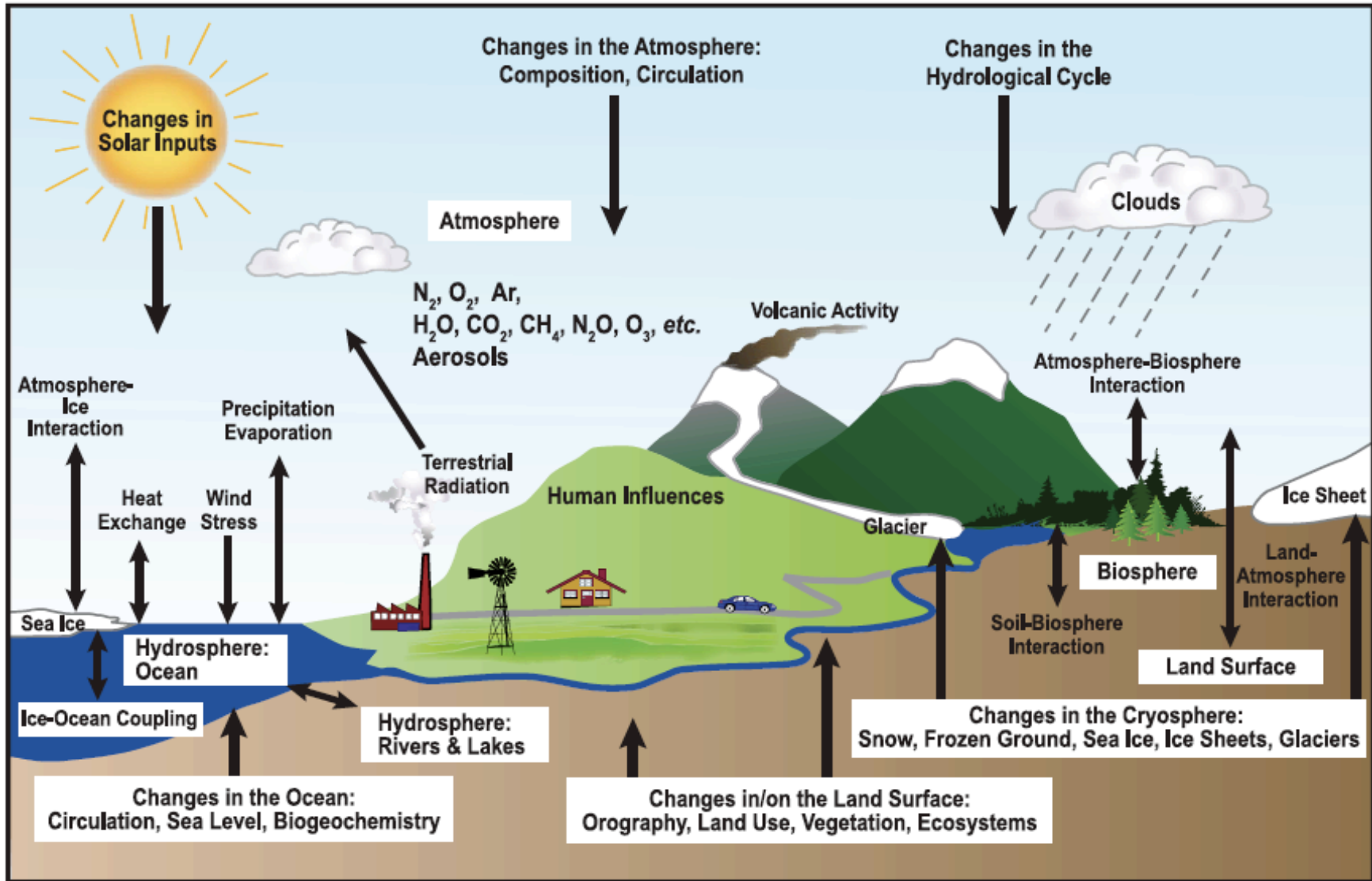
Met-fields predicted with boundary conditions from GCMs or reanalysis data

Global Circulation Model (GCM)

3D, global, met-fields predicted, [nudged](#) with reanalysis met-data,
[online or offline](#) aerosol

(Courtesy of P. Stier)

Components of the Climate System in GCMs

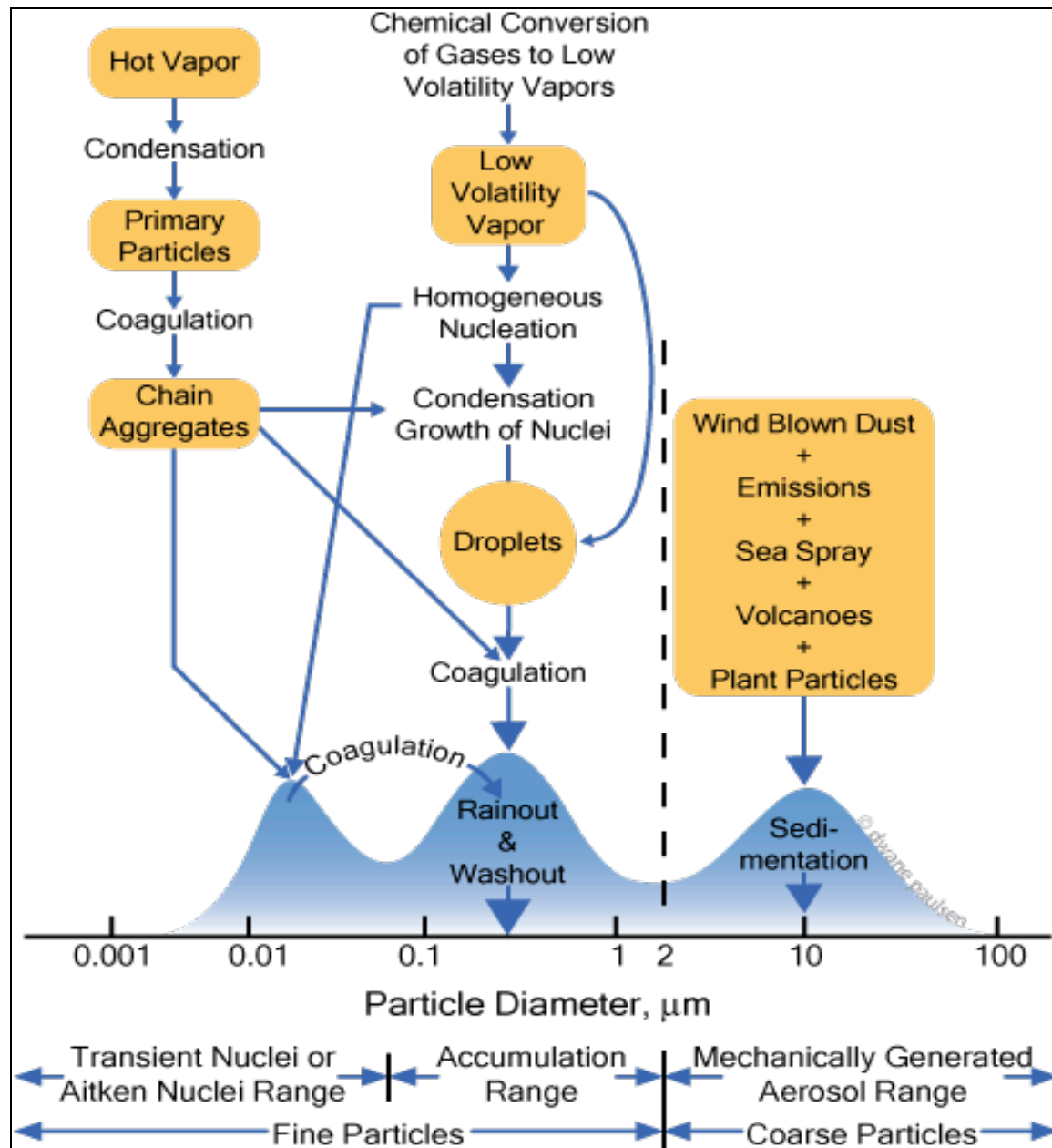


IPCC, 2007

Outline

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

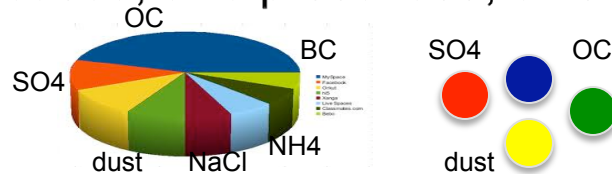
Aerosol Size and Composition in the Atmosphere



Aerosol Representation in GCMs

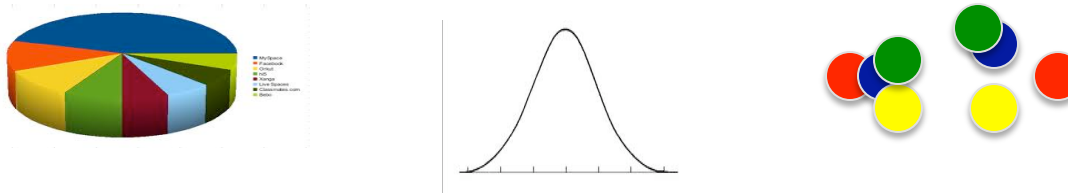
- **Bulk**

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



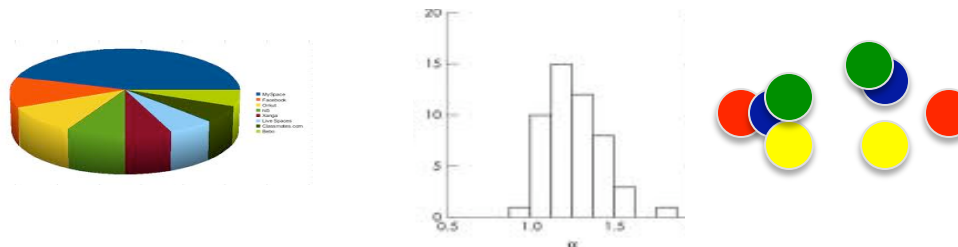
- **Moment-based (modal, 2-moment quadrature method of moments)**

Assumed functional form of size distributions (log-normal), predict evolution of size distribution by predicting mass (3rd moment) and number (0th 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**

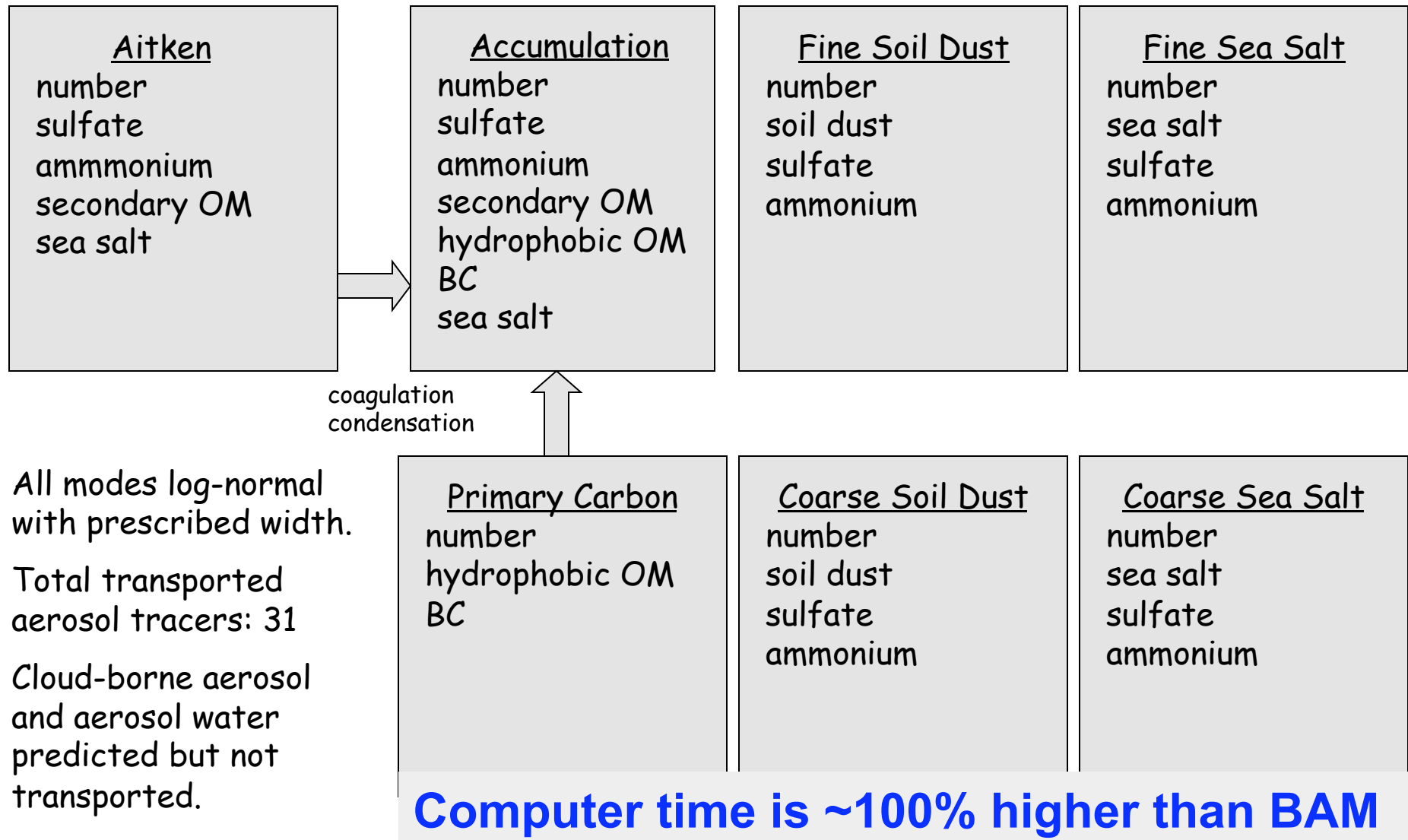
Split size distribution into bins, predict evolution of size distribution by predicting mass and number mixing ratio in each bins, aerosol microphysics



Bulk Aerosol Treatment in CAM3

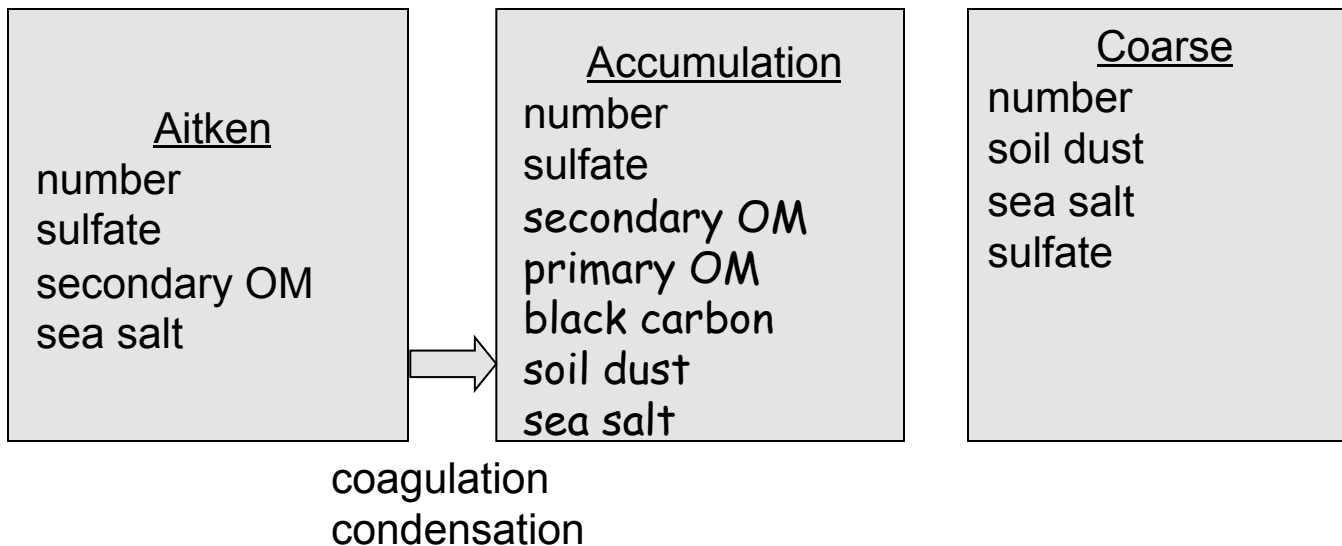
sulfate	hydrophobic black carbon	sea salt 1	soil dust 1
ammonium	hydrophobic organic carbon	sea salt 2	soil dust 2
nitrate	hydrophilic black carbon	sea salt 3	soil dust 3
secondary organic carbon	hydrophilic organic carbon	sea salt 4	soil dust 4

7-Mode Modal Aerosol Model (MAM) in CAM5



Simplified 3-mode version of MAM in CAM5

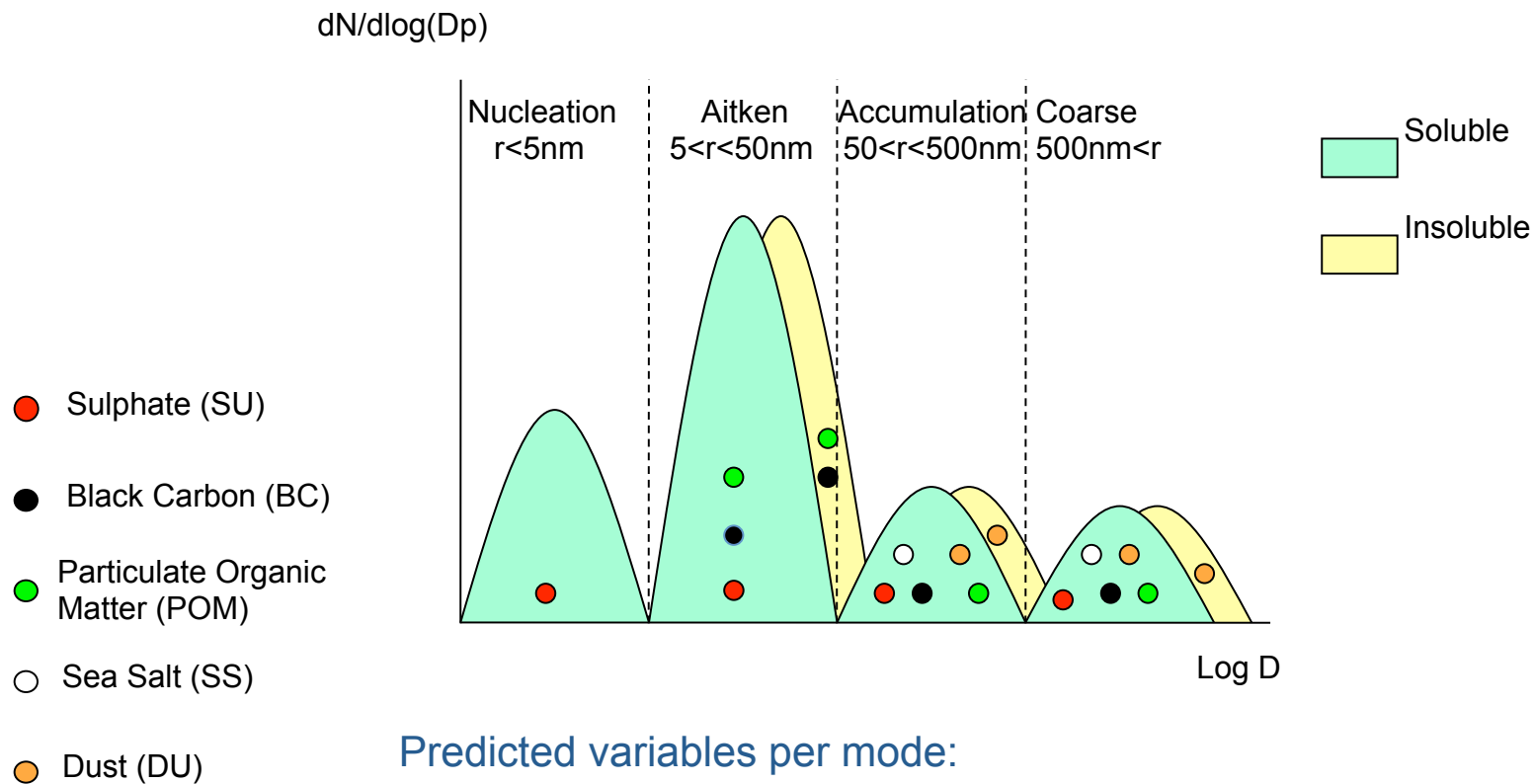
Assume primary carbon is internally mixed with secondary aerosol.
Sources of dust and seasalt are geographically separate
Assume ammonium neutralizes sulfate.



Total transported
aerosol tracers: 15

Computer time is 30% higher than BAM

Modal Aerosol Module (ECHAM-HAM)



Predicted variables per mode:

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

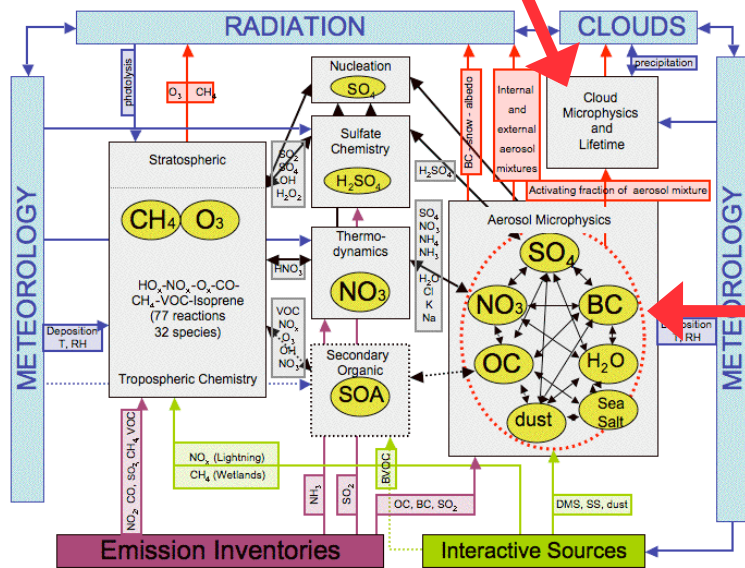
Courtesy of Declan O'Donnell

GISS-MATRIX (QMOM)

Droplet activation:

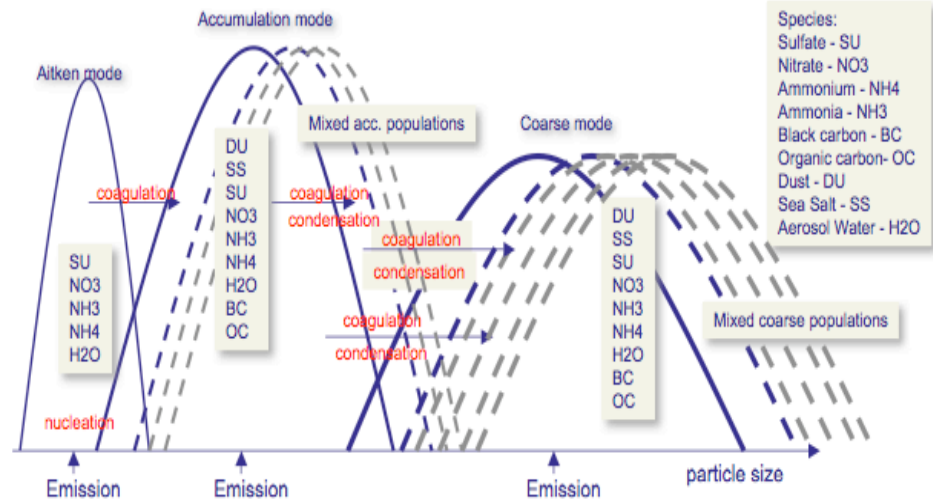
Abdul Razzak and Ghan (1998, 2000)

Cloud droplet nucleation follows prognostic treatment of Morrison et al. 2005, 2008



MATRIX

Aerosol Microphysical Model based on the Methods of Moments
Bauer et al. ACP 2008



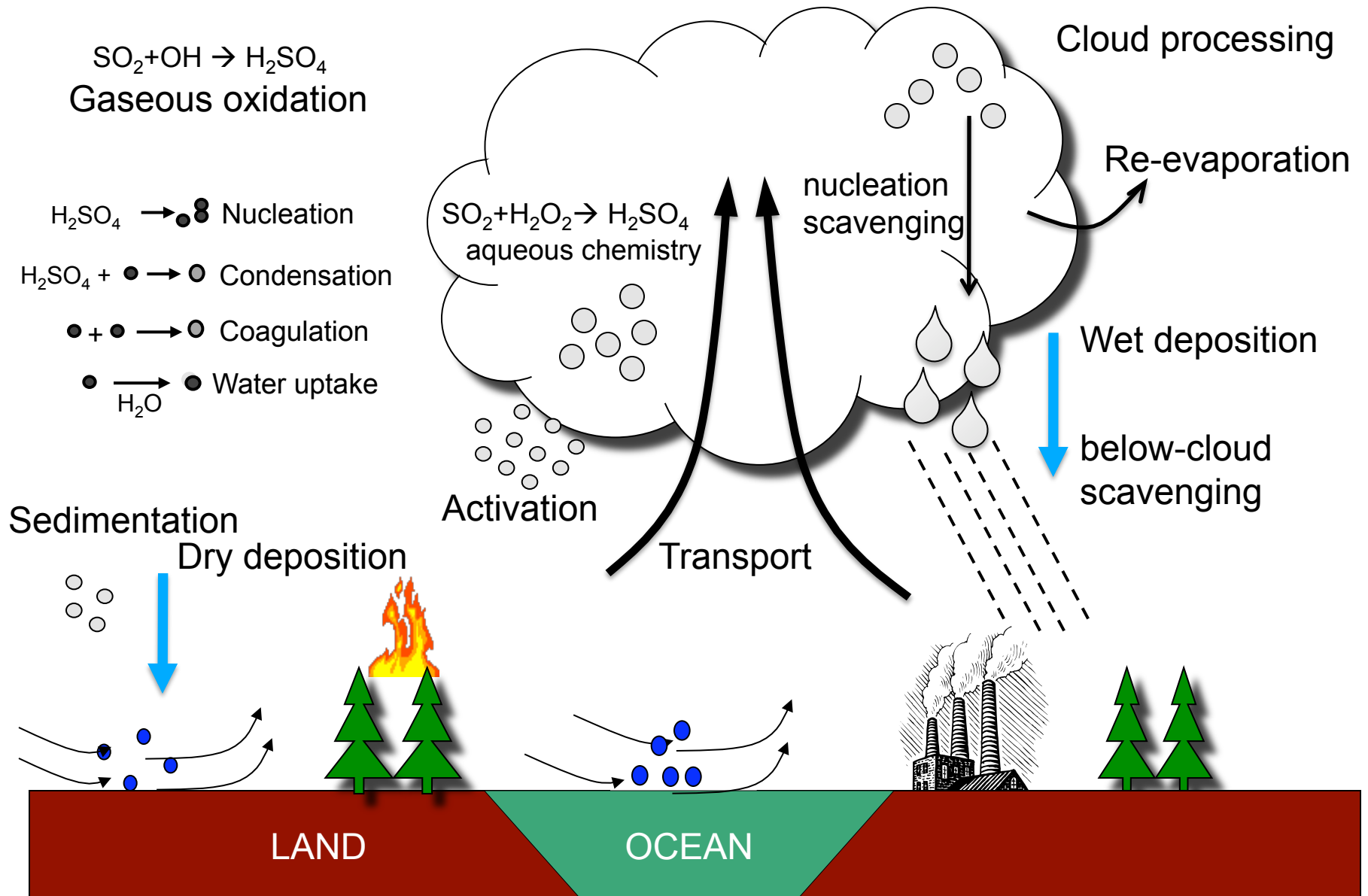
Aerosol Microphysics:

- 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

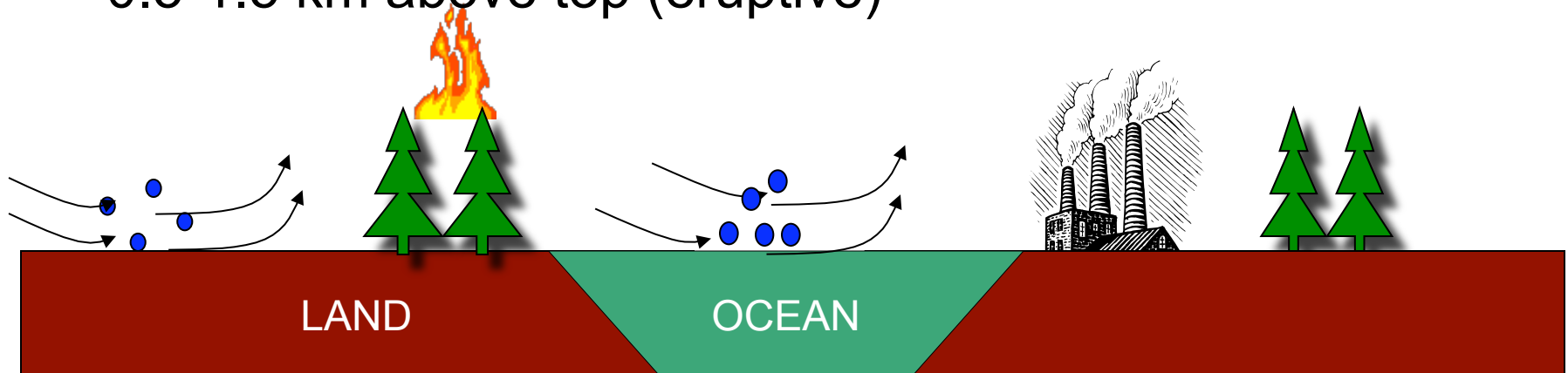
Courtesy of S. Bauer

Global Aerosol Cycles



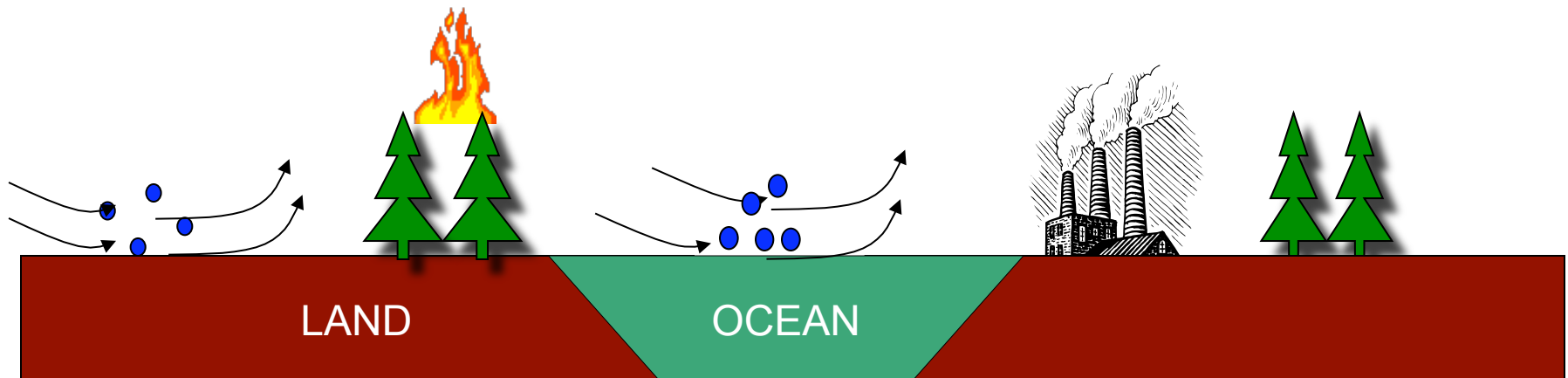
Aerosol Processes : Primary Emission

- **Offline** emission mass flux (for SO_2 , POA, BC, DMS): prescribed from inventory
- **Online** emission mass flux (for dust, sea salt, ocean POA): $f(u, r, \text{soil moisture or ocean concentrations})$
- **Injection Heights:**
 - Most emission fluxes applied at surface (lowest grid box), power plant $\text{SO}_2 \sim 100\text{-}300 \text{ m}$;
 - Biomass burning applied an injection height profile;
 - Volcanic emission at $2/3\text{-}1/1$ of volcano top (continuous) and $0.5\text{-}1.5 \text{ km}$ above top (eruptive)

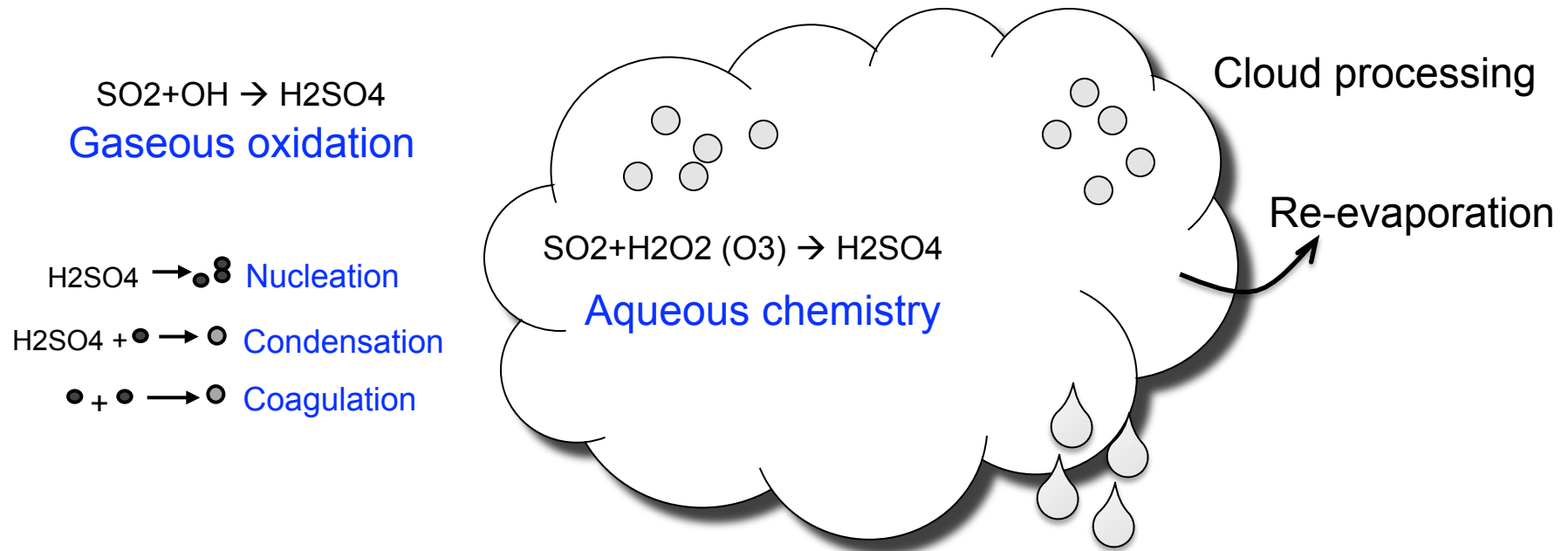


Aerosol Processes : Primary Emission

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



Aerosol Processes (Secondary SO₄ Formation)



All models: include gas and aqueous phase SO₂ chemistry

Bulk models: assume instantaneous conversion of H₂SO₄ (g) to sulfate, no nucleation/condensation/coagulation

Modal (bin) models:

Nucleation of H₂SO₄/NH₃/H₂O : form new particles

Condensation of H₂SO₄/NH₃/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**.

Aerosol Processes (Nucleation)

CAM5: Ternary $\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$ nucleation in MAM7 (Merikanto et al, 2007)
Binary $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ 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 $1.0 \times 10^{-6} \text{ s}^{-1}$

GISS: Ternary $\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$ nucleation (Napari et al., 2002)
Binary $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation (Jaecker-Voirol and
Mirabel, 1989; Vehkamaki et al. 2002)

ECHAM:

Old: Binary $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ 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 ($\sim 1 \text{ nm}$) to
Aitken mode size

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

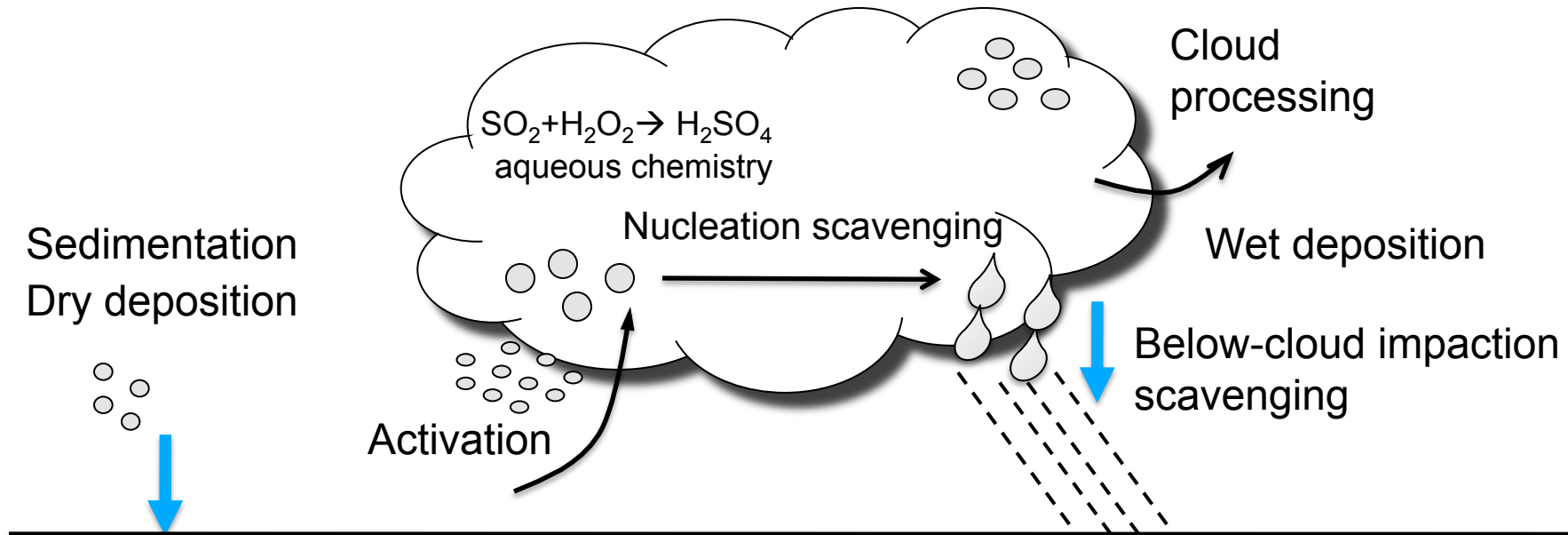
Newer Approaches:

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-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.

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

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, 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 refractive index and wet effective radius or look-up tables
- **Hygroscopicity**
 - volume average of K from components in each mode

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 - ❑ 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 countries**
 - **Biomass burning** emissions
 - **Mineral dust and sea salt** emissions
 - **Primary organics** from oceans

Effect of Primary Emissions

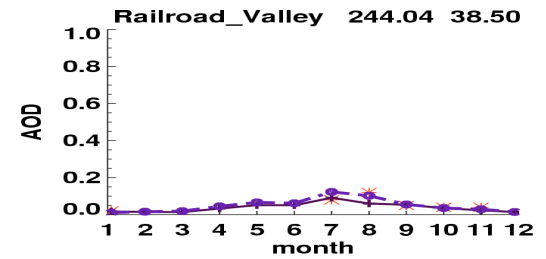
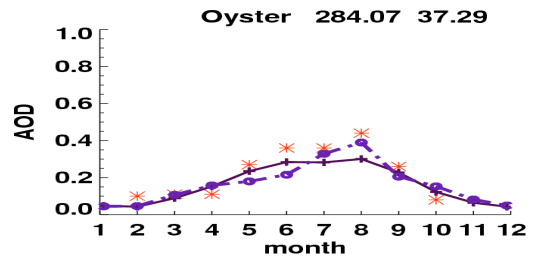
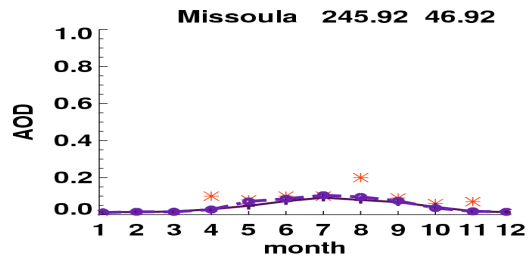
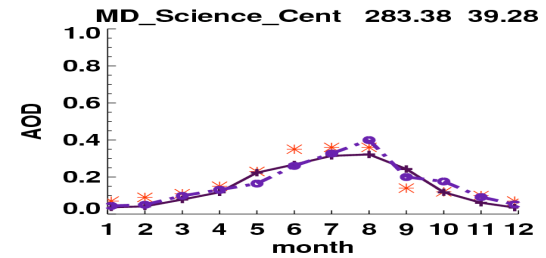
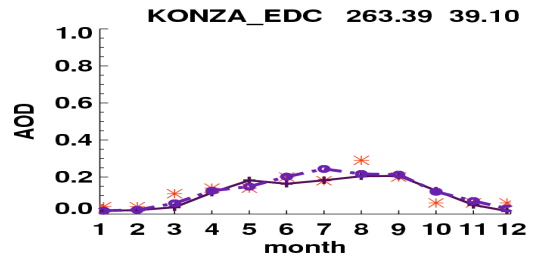
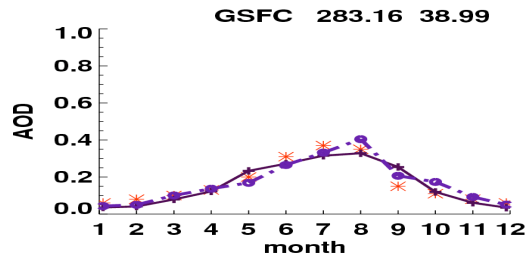
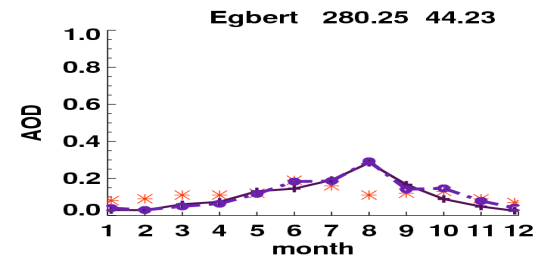
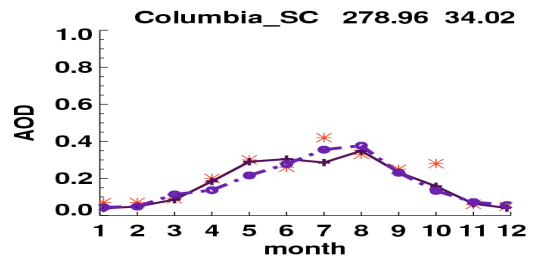
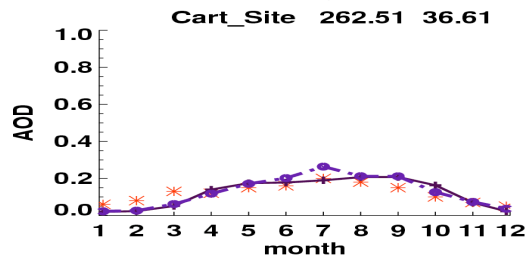
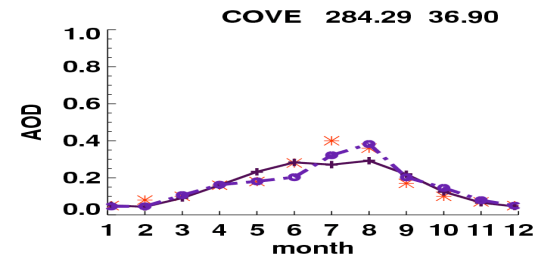
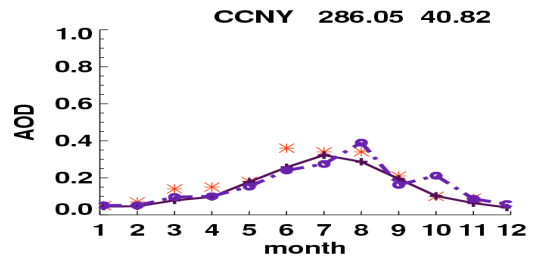
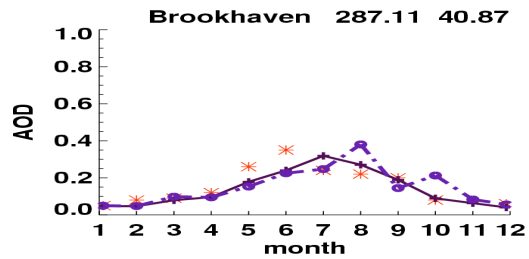
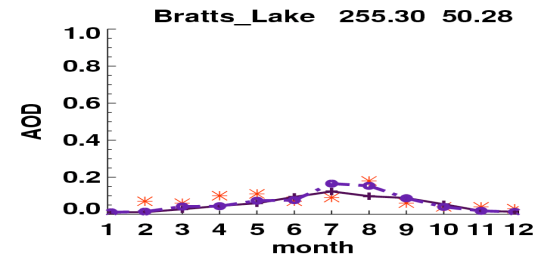
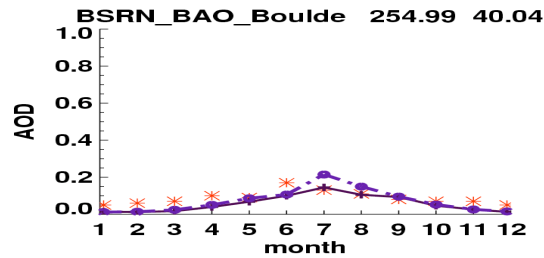
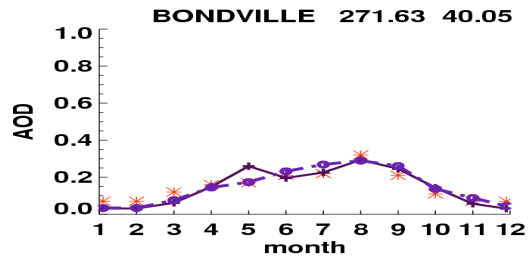
AOD

North_America

OBS *

CAM3mod-

CAM7mod-



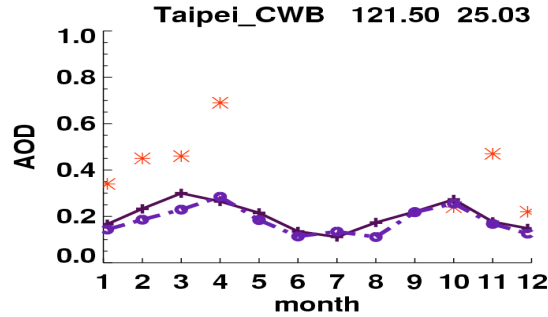
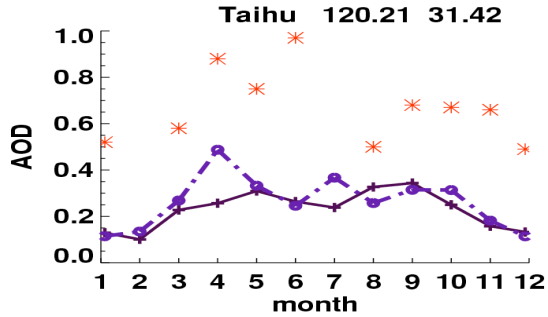
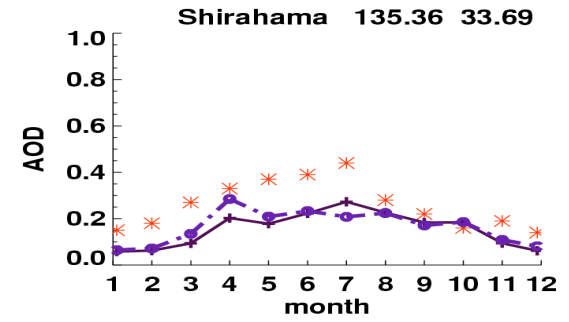
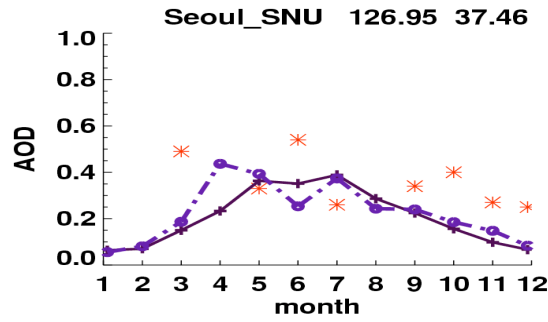
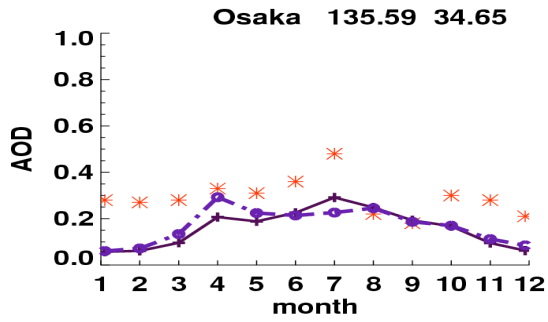
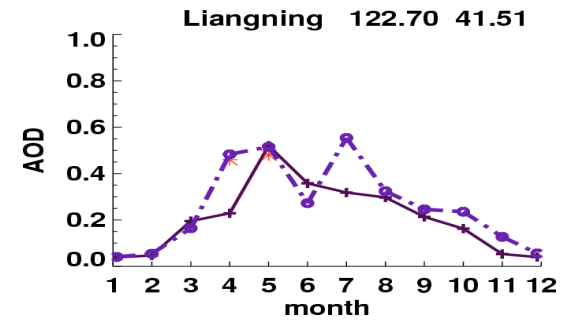
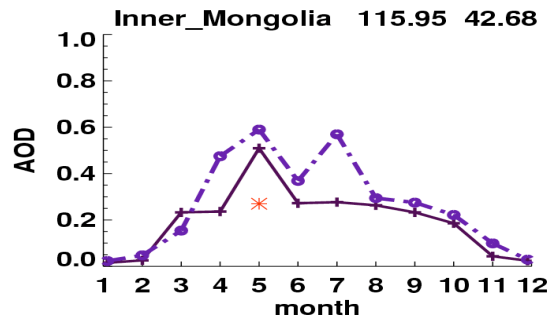
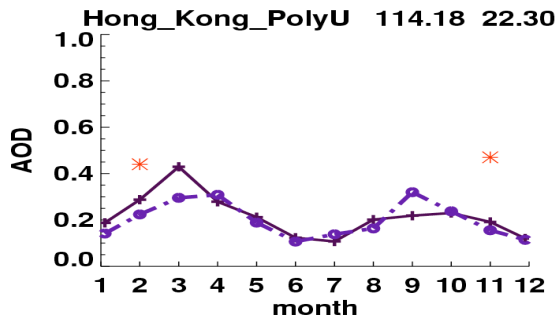
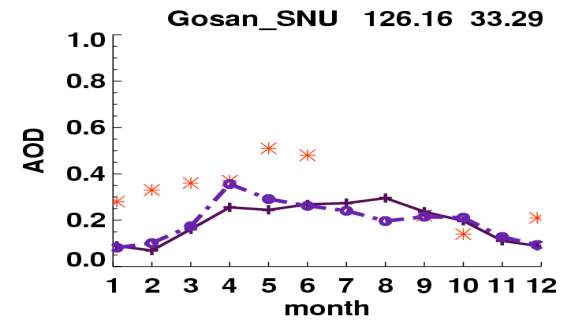
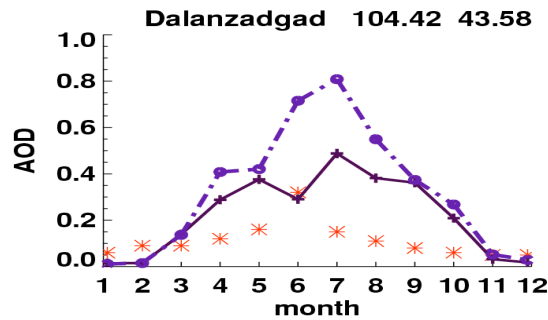
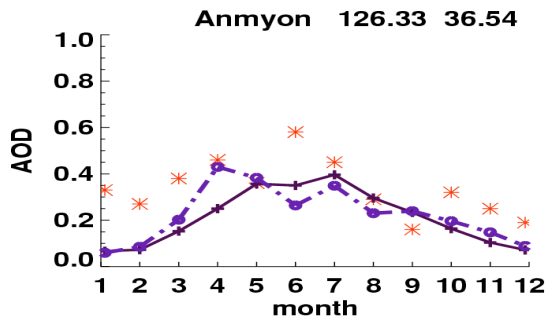
Effect of Primary Emissions

AOD

East_Asia

OBS *

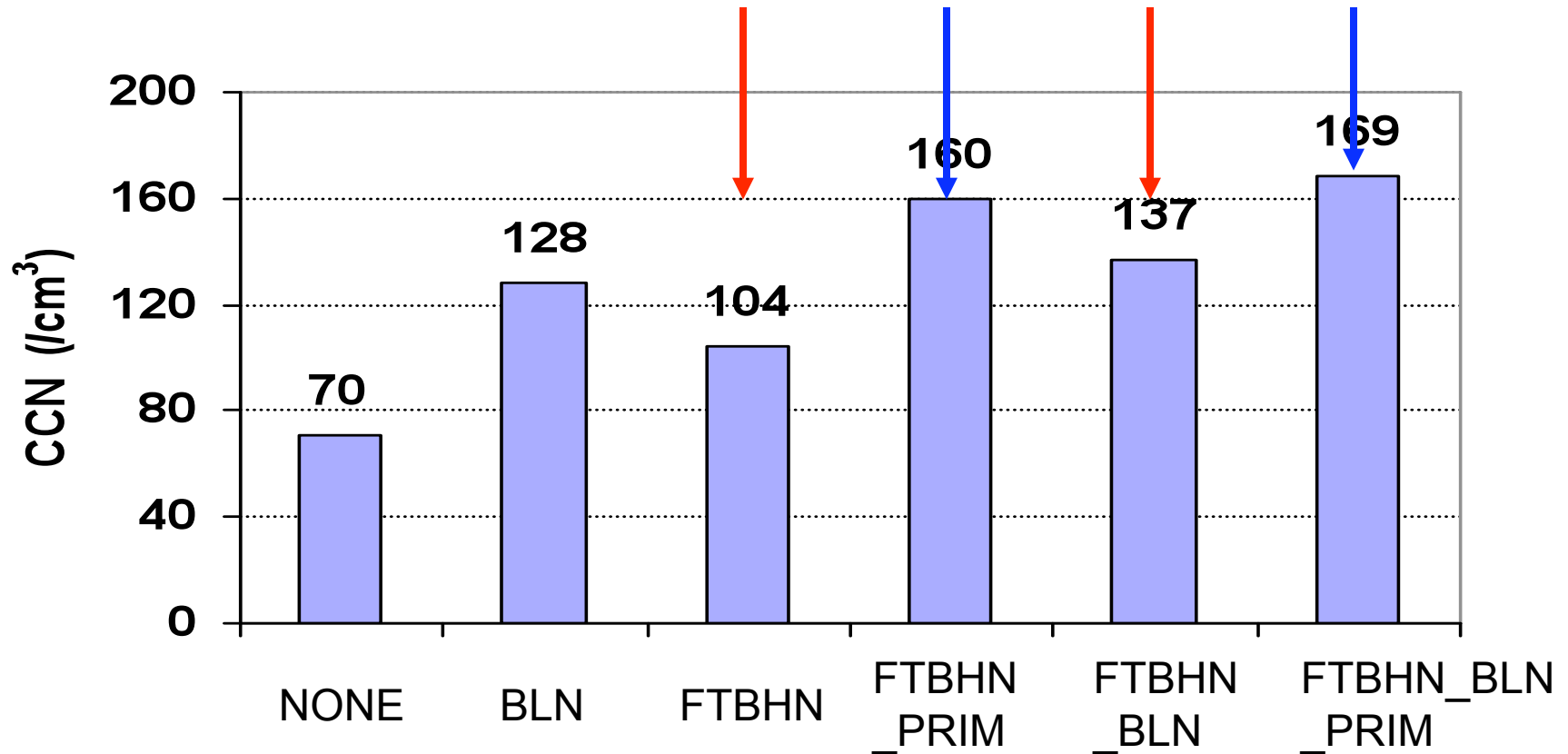
CAM3mod- CAM7mod-



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

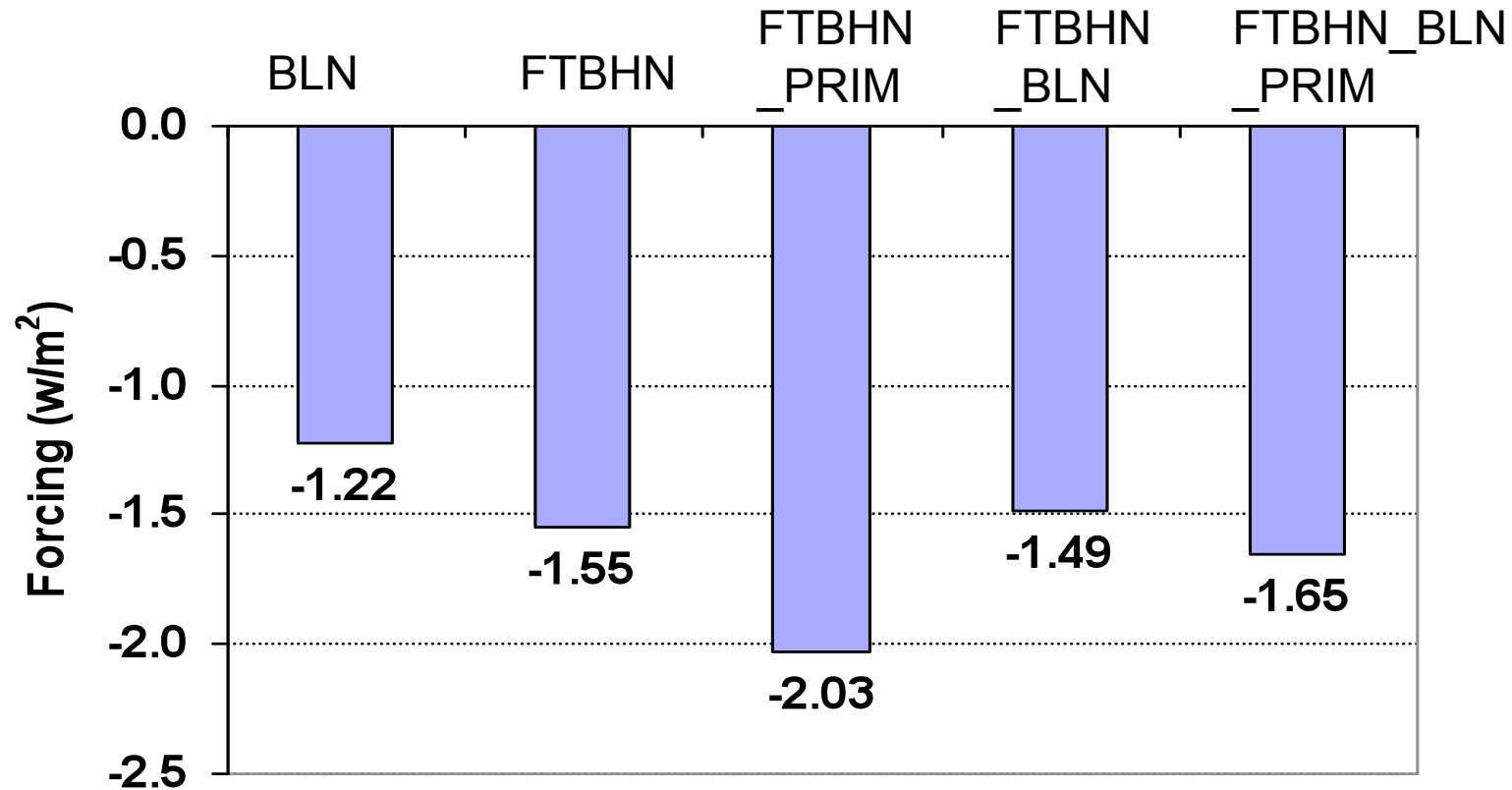
CCN CONCENTRATION IN THE BOUNDARY LAYER (930 hPa)



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/m².

Effect of the new SOA scheme

Original

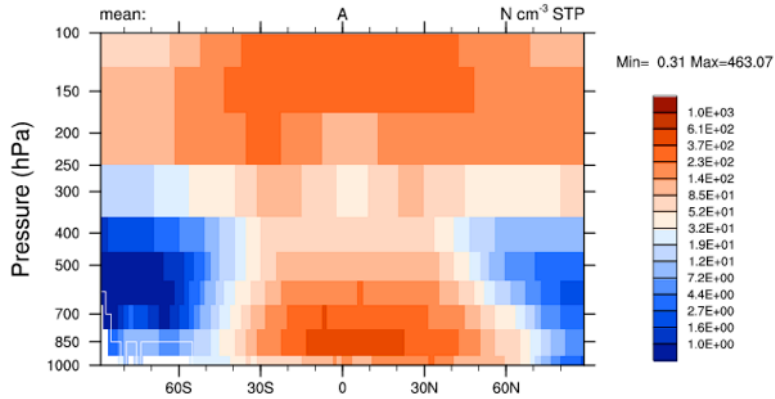
“organic carbon” refers to POA plus SOA formed by assuming a fixed 15% SOA yield from the monoterpene emissions estimates of Guenther et al. (1995), with immediate non-volatile SOA production.

New

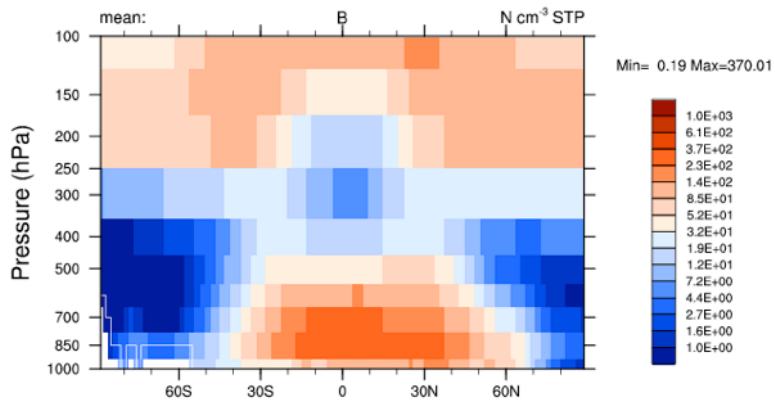
Prognostic SOA scheme with explicit gas/liquid partitioning

(courtesy of K. Zhang, ECHAM-HAM)

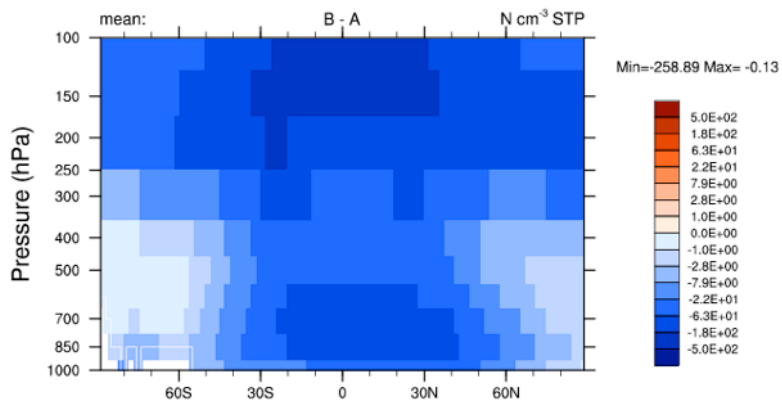
aerosol number (soluble accumulation mode)
zonal mean, unit: N cm^{-3} STP



A: CTRL (with explicit SOA)



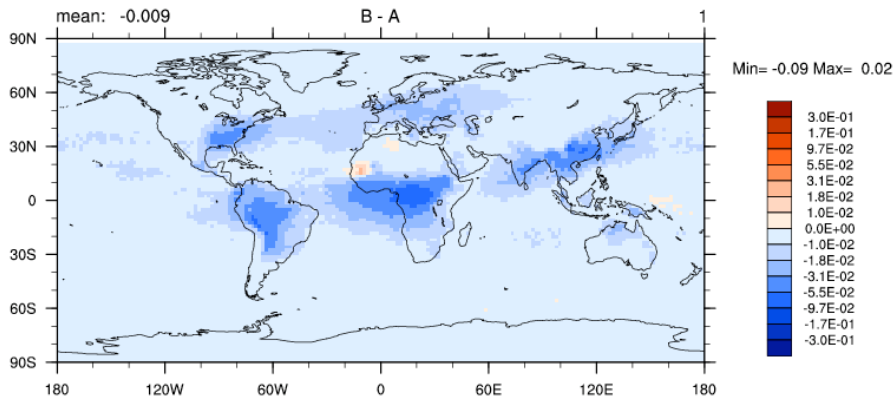
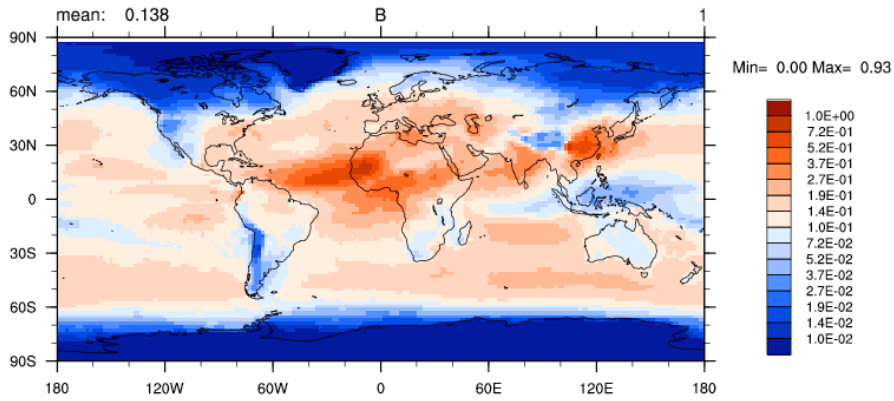
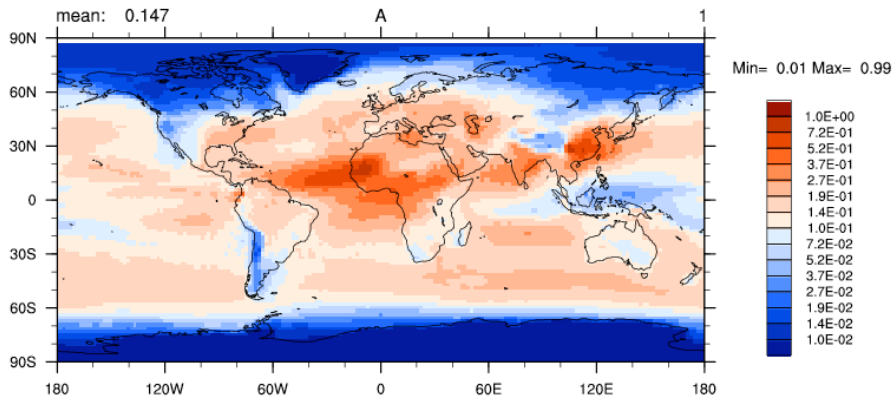
B: without explicit SOA



B - A

much more accumulation mode
particles in the upper atmosphere

Aerosol Optical Depth (AOD)
unit: 1



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
- extremely sensitive to high RH

Jacobson et al. JGR-1996

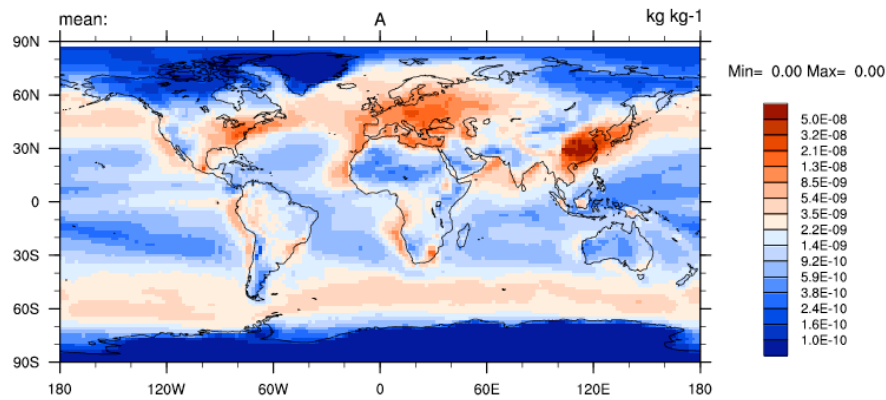
New: K-Köhler theory based scheme

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

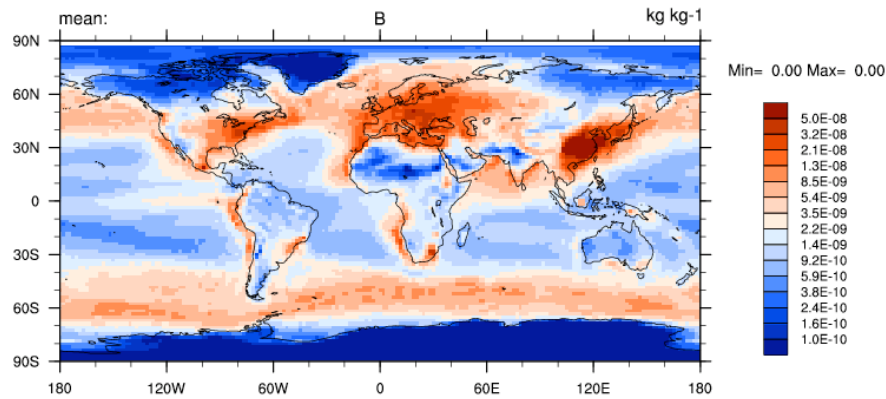
Petters and Kreidenweis ACP-2007

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

aerosol water content (soluble accumulation mode)
unit: kg kg⁻¹

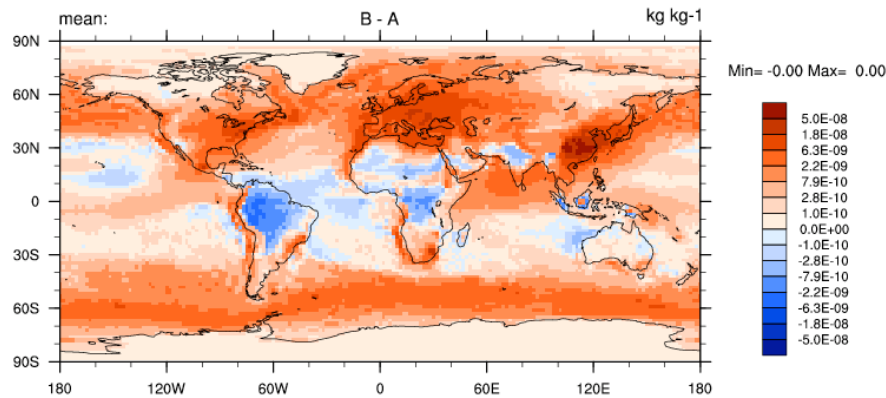


K method (new)



ZSR method

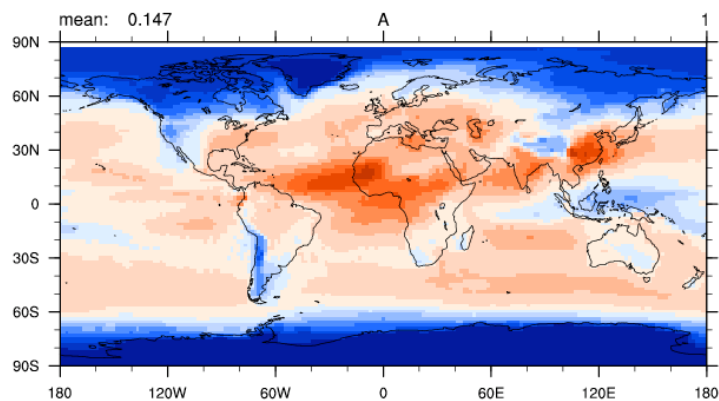
The ZSR based method produces much **larger** aerosol water-uptake over both the ocean and the industrial area



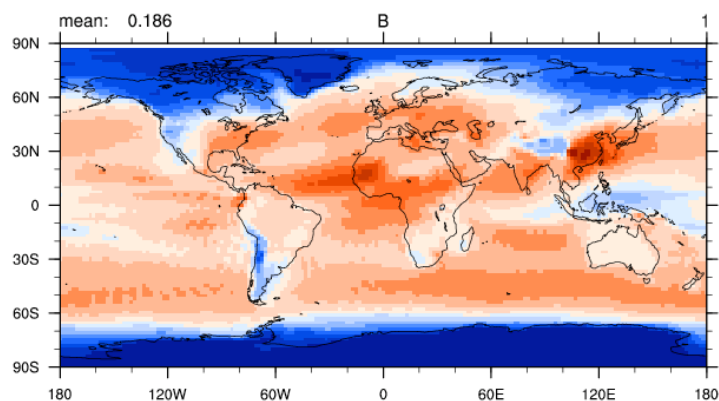
Difference (ZSR - K)

Aerosol Optical Depth (AOD)

unit: 1

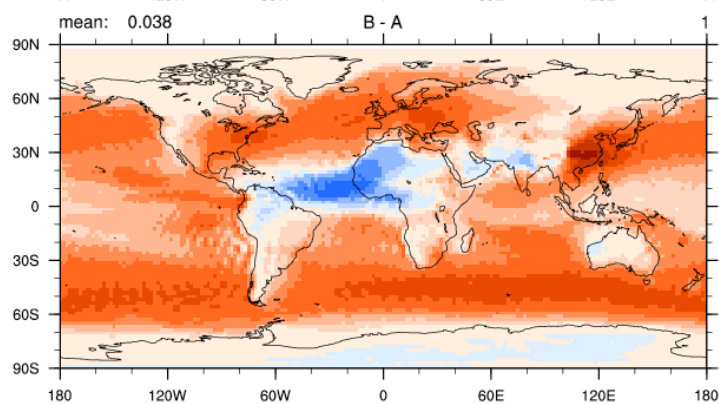


K method (new)



ZSR method

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



Difference (ZSR - K)

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

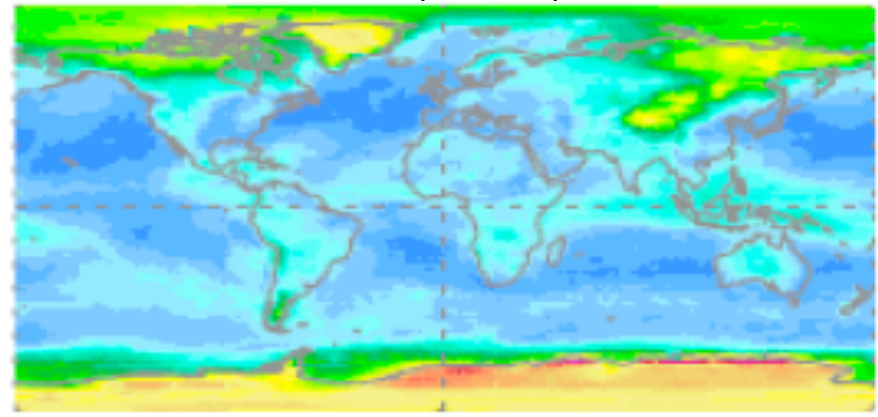
Aerosol Models Have Particular Trouble Simulating Aerosol Beyond the Polar Front

- 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.

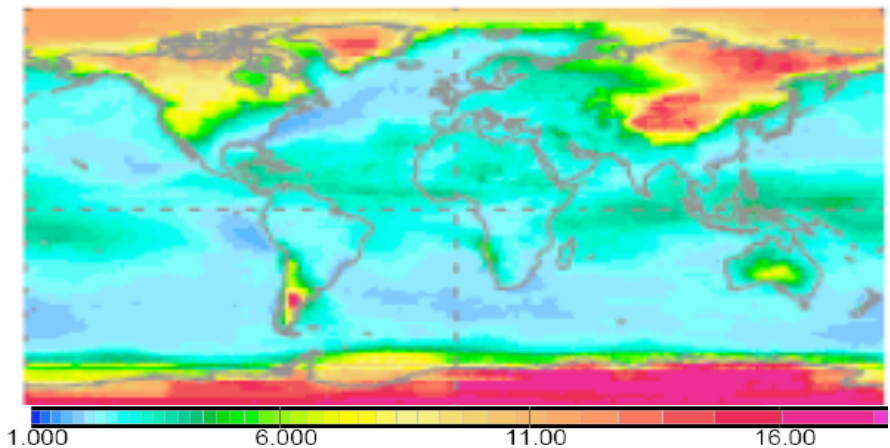
Major differences in **poles**

Max/Min of Central 2/3 of 16 Models

Aerosol Optical Depth

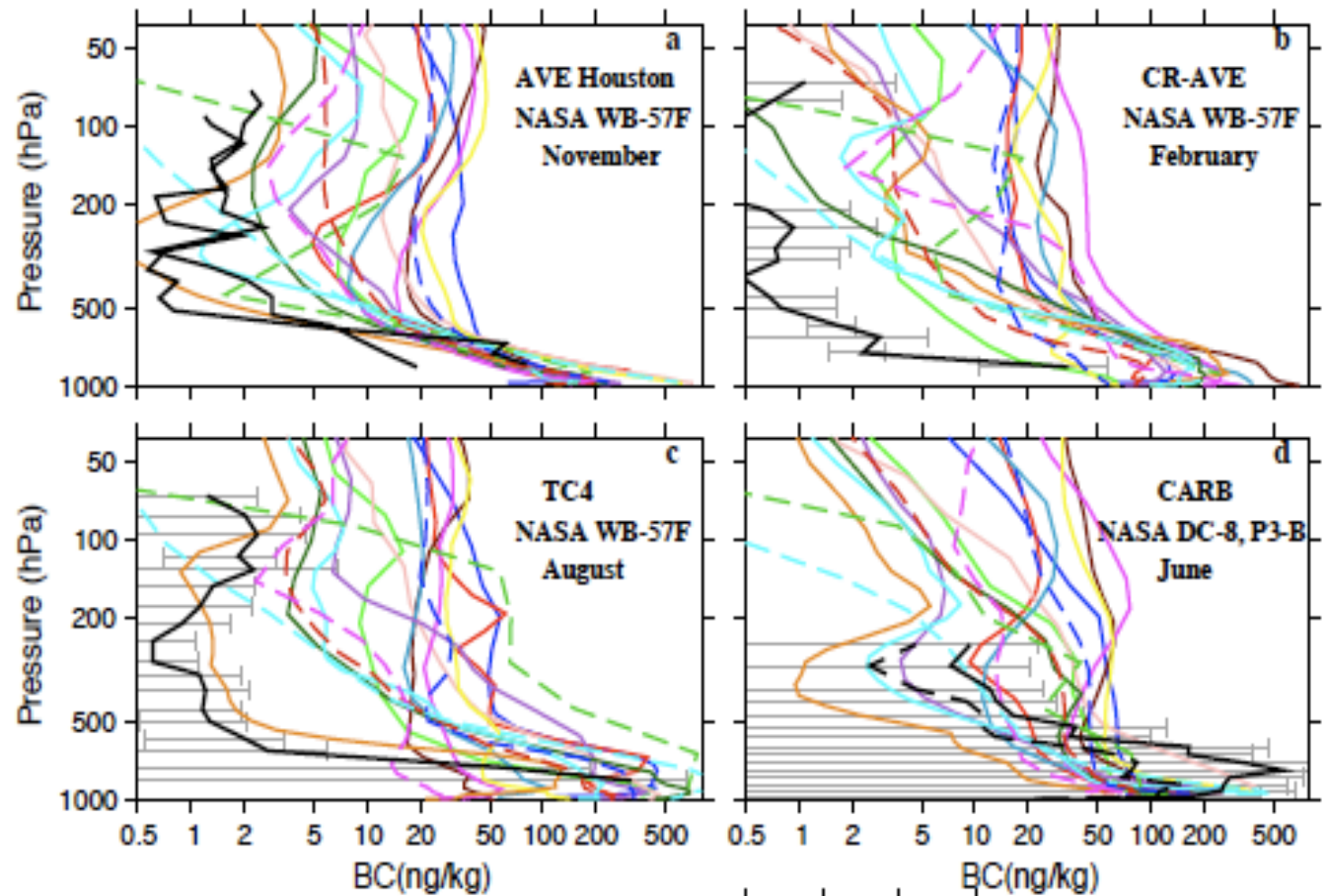


Aerosol Column Mass

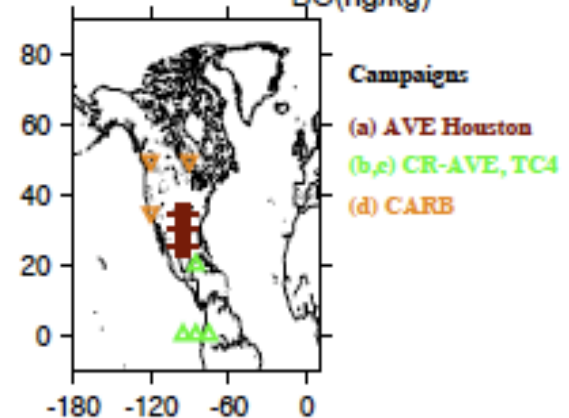


Kinne et al., An AeroCom initial assessment. *Atmos. Chem. & Phys.*, 2006.

BC compared with SP2 (tropics and midlat.)



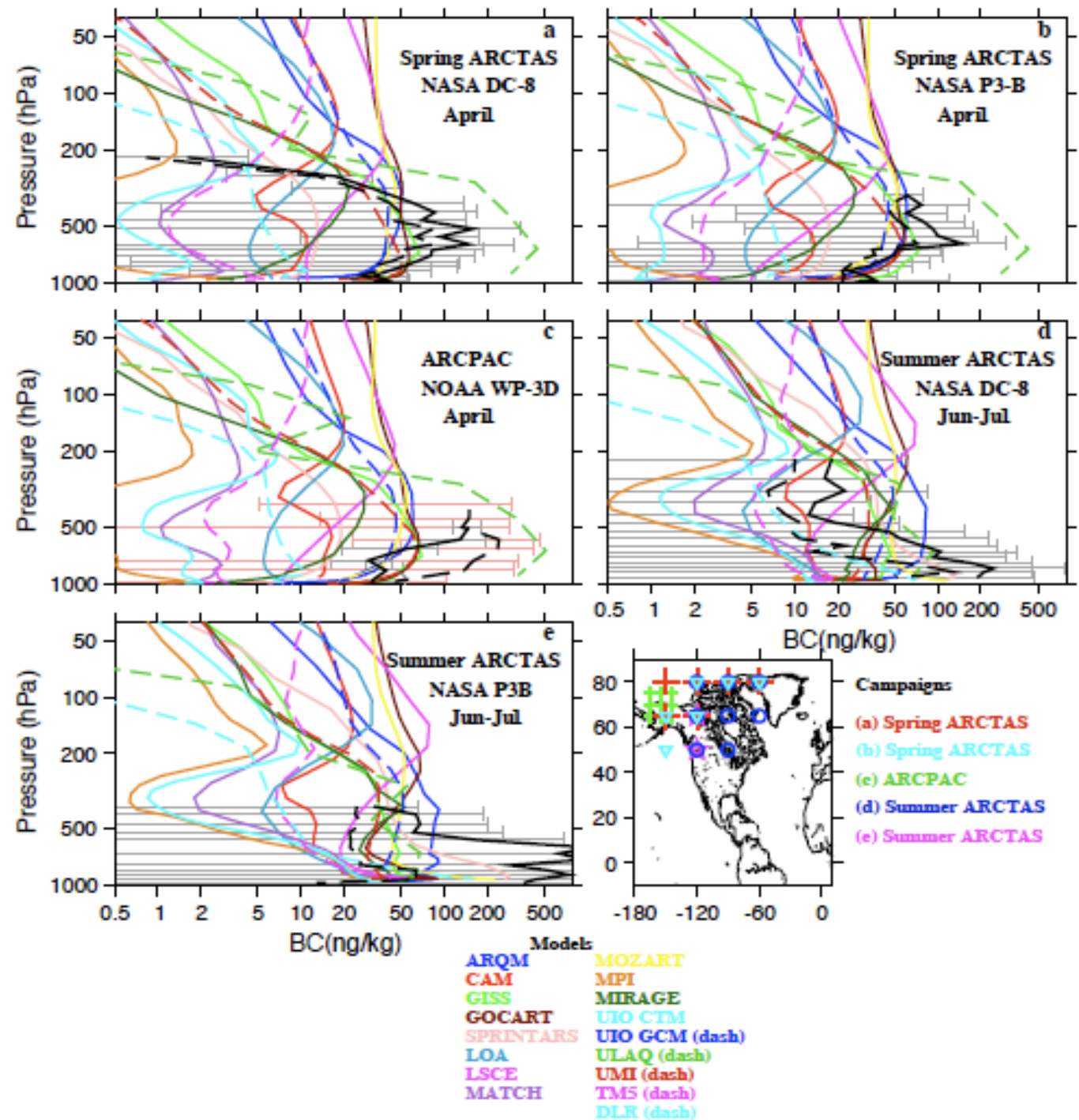
- Models**
- ARQM
 - CAM
 - GISS
 - GOCART
 - SPRINTARS
 - LOA
 - LSCE
 - MATCH
 - MOZART
 - MPI
 - MIRAGE
 - UIO CTM
 - UIO GCM (dash)
 - ULAQ (dash)
 - UMI (dash)
 - TM5 (dash)
 - DLR (dash)



Major differences in free troposphere

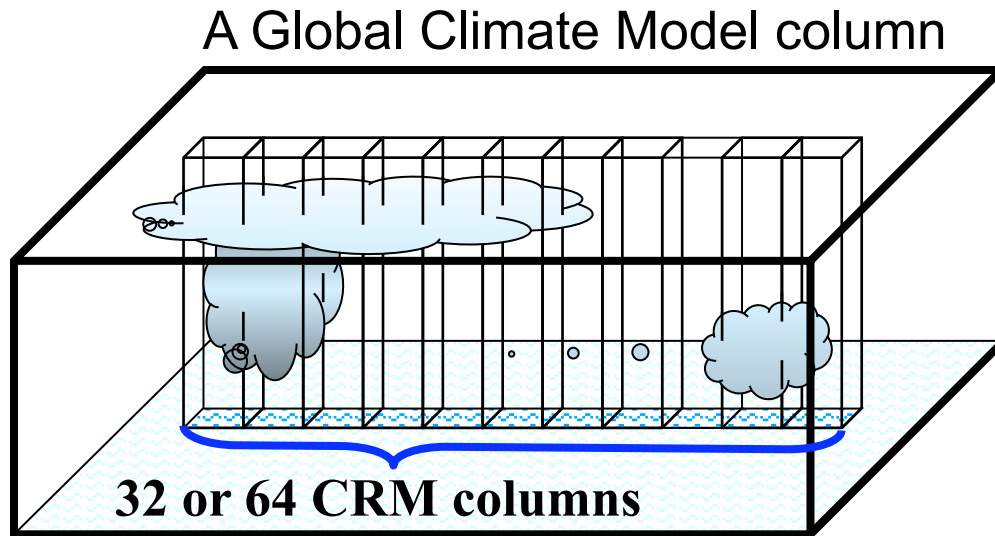
Koch et al. (2009)

BC compared with SP2 (highlat.)



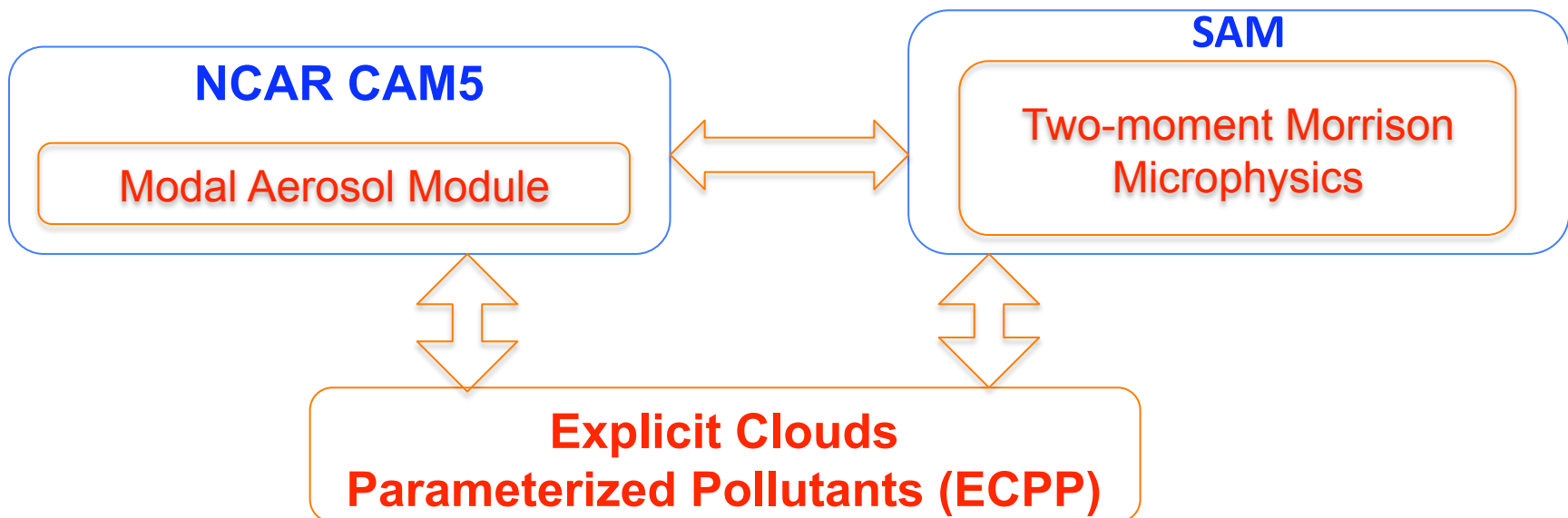
Koch et al. (2009)

Multiscale Modeling Framework Approach (MMF) (Superparameterization)

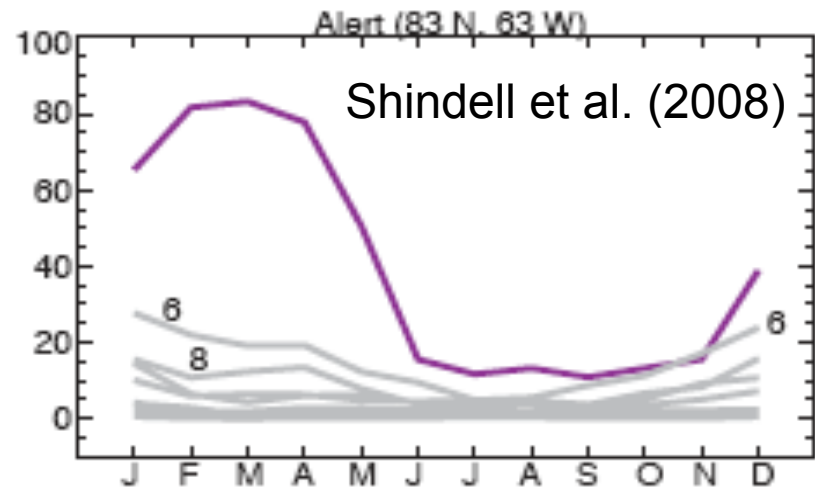
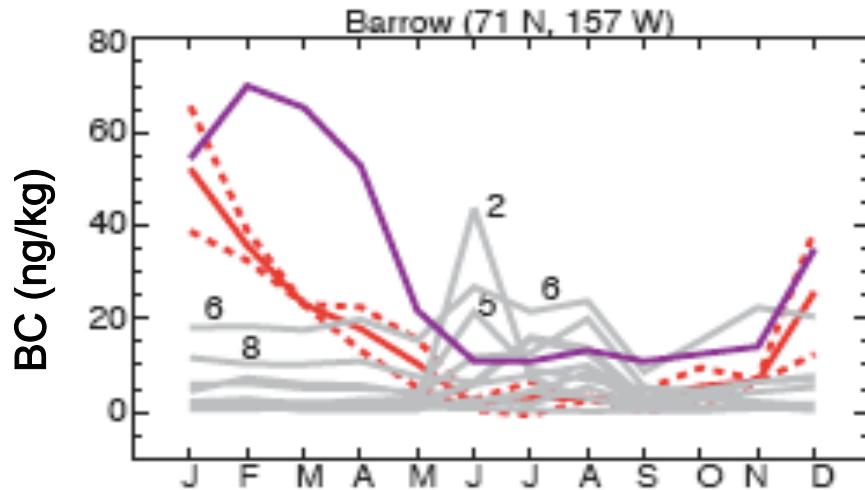
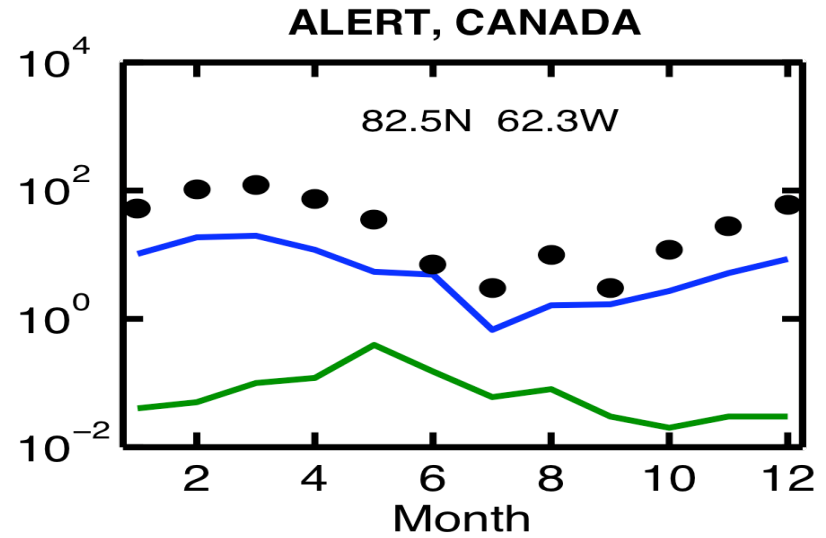
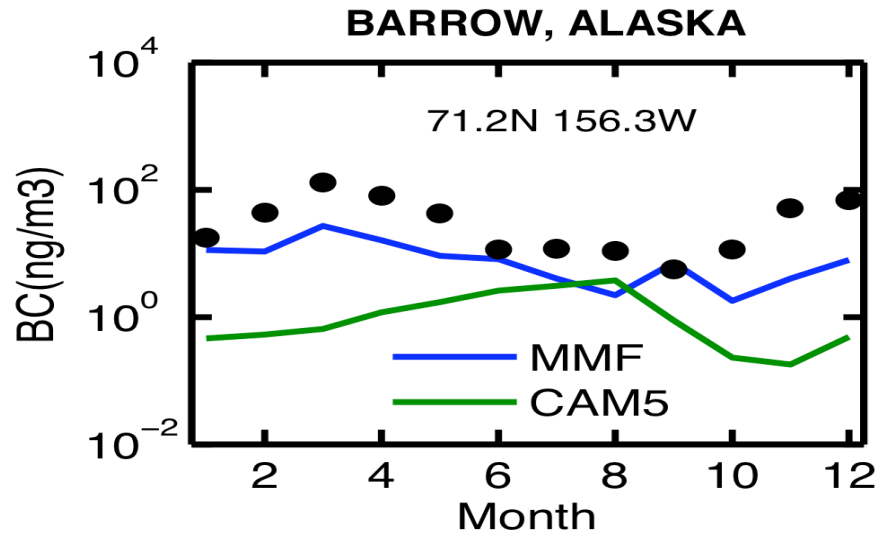


Grabowski, 2001;
Khairoutdinov and
Randall, 2001.

The MMF approach permits
explicit simulations of deep
convective clouds.



Monthly BC concentrations



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 :

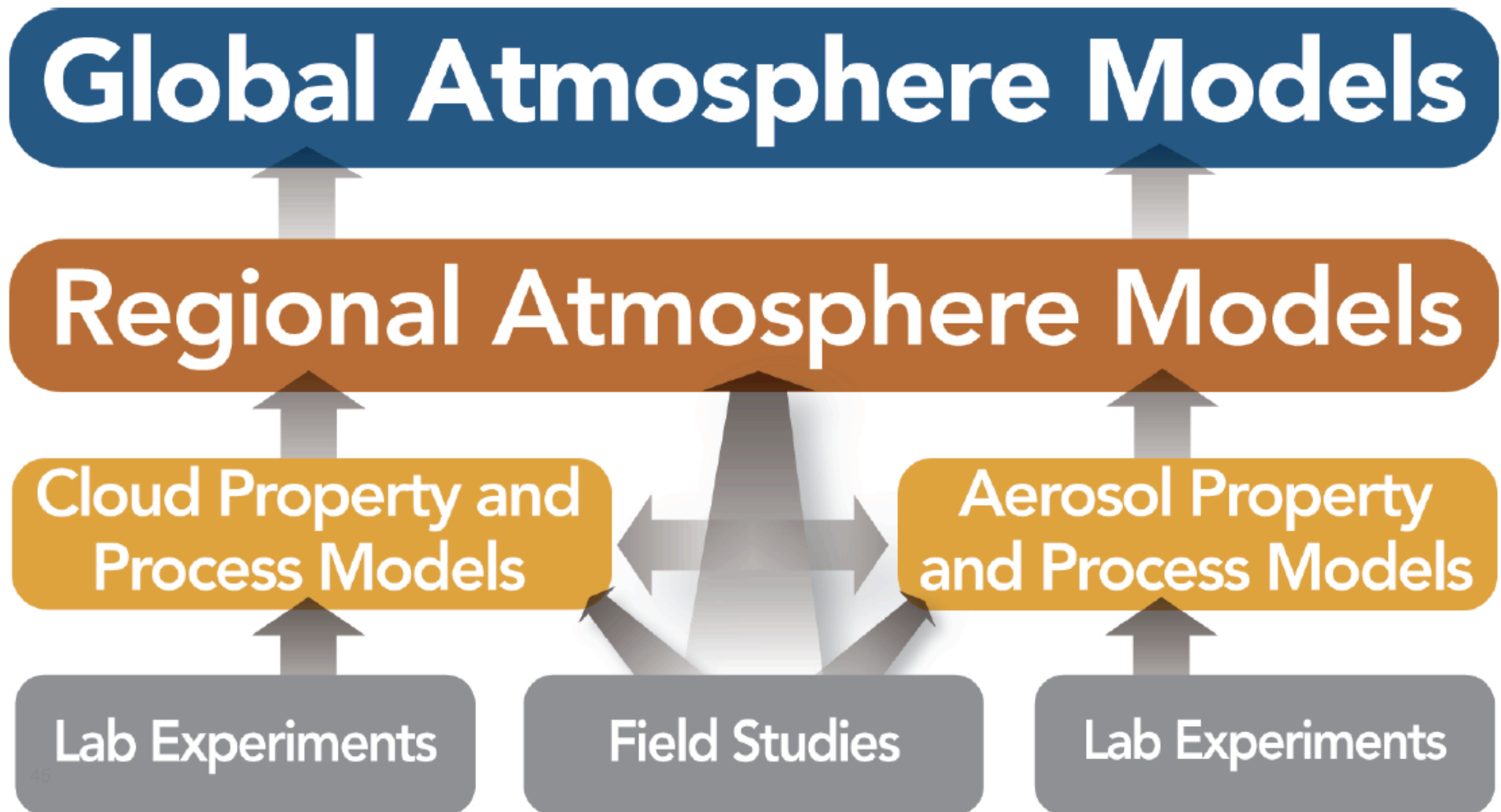
- Improve primary emissions: types and regions, size distribution and injection heights
- Aerosol nucleation and growth (BL nucleation, role of organics)
- SOA production
- HNO_3 and water uptake (MOSAIC)
- Wet scavenging (cloud and precipitation in GCMs, link to CAPI & CLWG)

Properties :

- Refractive index (dust)
- Hygroscopicity of organics and mineral dust
- Mixing state

Road Map from Process Studies to GCMs

(Ghan and Schwartz, BAMS, 2007)



THANKS!