Aerosol Representation in GCMs

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DOE ASR Aerosol Lifecycle WG
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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

Changes in the Atmosphere: Composition, Circulation

Changes in the Hydrological Cycle

Atmosphere-Biosphere Interaction

Atmosphere

Volcanic Activity

N₂, O₂, Ar,
H₂O, CO₂, CH₄, N₂O, O₃, etc.
Aerosols

Clouds

Changes in the Cryosphere:
Snow, Frozen Ground, Sea Ice, Ice Sheets, Glaciers

Changes in the Ocean:
Circulation, Sea Level, Biogeochemistry

Changes in/on the Land Surface:
Orography, Land Use, Vegetation, Ecosystems

IPCC, 2007
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 (3\textsuperscript{rd} 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

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
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)

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

SO₂ + OH → H₂SO₄
Gaseous oxidation

H₂SO₄ → Nucleation

H₂SO₄ + → Condensation

+ → Coagulation

H₂O → Water uptake

SO₂ + H₂O₂ → H₂SO₄
aqueous chemistry

SO₂ + H₂O₂ → H₂SO₄
Gaseous oxidation

Cloud processing

Re-evaporation

Wet deposition

below-cloud scavenging

Sedimentation

Activation

Dry deposition

Transport

LAND

OCEAN
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 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 the smaller emission size than biomass burning and biofuel.
Aerosol Processes (Secondary $\text{SO}_4^2-$ Formation)

$\text{SO}_2 + \text{OH} \rightarrow \text{H}_2\text{SO}_4$

Gaseous oxidation

$\text{H}_2\text{SO}_4 \rightarrow \bullet$ Nucleation

$\text{H}_2\text{SO}_4 + \bullet \rightarrow \bullet$ Condensation

$\bullet + \bullet \rightarrow \bullet$ Coagulation

$\text{SO}_2 + \text{H}_2\text{O}_2 (\text{O}_3) \rightarrow \text{H}_2\text{SO}_4$

Aqueous chemistry

Cloud processing

Re-evaporation

All models: include gas and aqueous phase $\text{SO}_2$ chemistry

Bulk models: assume instantaneous conversion of $\text{H}_2\text{SO}_4$ (g) to sulfate, no nucleation/condensation/coagulation

Modal (bin) models:

Nucleation of $\text{H}_2\text{SO}_4/\text{NH}_3/\text{H}_2\text{O}$: form new particles

Condensation of $\text{H}_2\text{SO}_4/\text{NH}_3/\text{SOA}$: 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 H$_2$SO$_4$-NH$_3$-H$_2$O nucleation in MAM7 (Merikanto et al., 2007) 
Binary H$_2$SO$_4$-H$_2$O nucleation in MAM3 (Vehkamaki et al. 2002); 
Boundary layer nucleation: empirical 1$^{st}$ order nucleation rate in H$_2$SO$_4$ (Sihto et al., 2006) with the rate coefficient of 1.0x10$^{-6}$ s$^{-1}$

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

**ECHAM:**
Old: Binary H$_2$SO$_4$-H$_2$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 (~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

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.

\[ F_d = C \rho_a v_d \]
\[ v_d = v_g + \frac{1}{r_a + r_s} \]

**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 (c₀) assumed

Improved models:
- **CAM5**: predicting aerosols in cloud water (through activation, aqueous chemistry, diffusion, and evaporation); size dependent of c₀

**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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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
  - Primary organics from oceans
Effect of Primary Emissions

AOD North_America OBS CAM3mod- CAM7mod-

BONDVILLE 271.63 40.05
Brookhaven 287.11 40.87
Cart_Site 262.51 36.61
GSFC 283.16 38.99
Missoula 245.92 46.92

BSRN_BAO_Boule 254.99 40.04
CCNY 286.05 40.82
Columbia_SC 278.96 34.02
KONZA_EDC 263.39 39.10

Bratts_Lake 255.30 50.28
COVE 284.29 36.90
Egbert 280.25 44.23
MD_Science_Cent 283.38 39.28
Oyster 284.07 37.29
Railroad_Valley 244.04 38.50
Effect of Primary Emissions

AOD | East_Asia | OBS | CAM3mod- | CAM7mod-

Anmyon 126.33 36.54

Dalanzadgad 104.42 43.58

Gosan_SNU 126.16 33.29

Hong_Kong_PolyU 114.18 22.30

Inner_Mongolla 115.95 42.68

Liangning 122.70 41.51

Osaka 135.59 34.65

Seoul_SNU 126.95 37.46

Shirahama 135.36 33.69

Taihu 120.21 31.42

Taipei_CWB 121.50 25.03
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)

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<th>FTBHN</th>
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</tbody>
</table>

Wang & Penner (2008)
The forcing from various treatments of aerosol nucleation ranges from -1.22 to -2.03 W/m².

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

(courtesy of K. Zhang, ECHAM-HAM)
much more accumulation mode particles in the upper atmosphere

A: CTRL (with explicit SOA)

B: without explicit SOA

B - A
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: Köhler theory based scheme
- can easily be applied for non-electrolytes (e.g. organic specie)
- a hygroscopicity parameter $\kappa$ 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 $K$

Courtesy of K. Zhang (ECHAM-HAM)
The ZSR based method produces much larger aerosol water-uptake over both the ocean and the industrial area.
with the new scheme, global mean AOD decreases by 0.04 (~25%)
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

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)
The MMF approach permits explicit simulations of deep convective clouds. 


The MMF approach permits explicit simulations of deep convective clouds.

NCAR CAM5
Modal Aerosol Module

SAM
Two-moment Morrison Microphysics

Explicit Clouds Parameterized Pollutants (ECPP)
Monthly BC concentrations

BARROW, ALASKA

71.2N 156.3W

BC (ng/m3)

Month

ALERT, CANADA

82.5N 62.3W

BC (ng/m3)

Month

Barrow (71 N, 157 W)

BC (ng/kg)

Alert (83 N, 63 W)

Shindell et al. (2008)
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!