Atmosphere Modeling IV, Chemistry and Aerosols

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CAM-chem Liaison: Simone Tilmes
WACCM Liaison: Mike Mills
Chemistry and aerosols interact with the climate system, 
-> need to be described well in climate models
Poor air quality is a major health issue

Health Burden of Global Air Pollution is Enormous

(7+ million premature deaths due to air pollution per year !!)
Simulation of atmospheric composition requires many components.
Chemistry-Climate Interactions in CESM2: CAM6

Atmospheric Chemistry
- Prescribed ozone/oxids
- Modal Aerosol Model
- Prescribed strat. Aerosols

Emissions
- Anthropogenic
- Biomass burning
- Biogenic

Fire emits

Prescribed Nitrogen deposition

Black carbon deposition

Land/Ocean Biosphere

Radiation
- Clouds (indirect)
- Dynamics
- Water vapor

Snow/Ice
Chemistry-Climate Interactions in CESM2: CAM-chem or WACCM
CAM6 vs CAM-chem

Same atmosphere, physics, resolution
Different chemistry and aerosols -> emissions and coupling

• **CAM6**: Aerosols are calculated, using simple chemistry ("fixed" oxidants) (prescribed: N₂, O₂, H₂O, O₃, OH, NO₃, HO₂; chemically active: H₂O₂, H₂SO₄, SO₂, DMS, SOAG)

  **Limited interactions between Chemistry and Climate**
  -> prescribed fields have to be derived using chemistry-climate simulations
  – Prescribed ozone is used for radiative calculations
  – Prescribed oxidants is used for aerosol formation
  – Prescribed methane oxidation rates
  – Prescribed stratospheric aerosols
  – Prescribed nitrogen deposition
  – Simplified secondary organic aerosol description
Modeling Chemistry-Climate Interactions in CESM2

Surface emissions and concentrations

• emissions: anthropogenic, biogenic, biomass burning, ocean, soil, volcanoes
• surface concentrations (greenhouse gases)

Chemical mechanism: important for chemistry and aerosol production

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).
Example: NOx Emissions

Anthropogenic + biomass burning + ships: kg(N)/year
Modeling Chemistry-Climate Interactions in CESM2

- Greenhouse gases are prescribed as monthly fields of CO$_2$, CH$_4$, O$_3$, N$_2$O, CFCs as lower boundary conditions. All CFCs can be combined to create effective CFC emissions.
Modeling Chemistry-Climate Interactions in CESM2

Surface emissions and concentrations

- emission: anthropogenic, biogenic, biomass burning, ocean, soil, volcanoes
- surface concentrations (greenhouse gases)

Chemical mechanism: important for chemistry and aerosol production

- WACCM and CAMchem: 483 reactions and 231 solution species
- CAM6: 6 chemical reactions and 25 solution species (much simpler)

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).
## Modeling Chemistry-Climate Interactions in CESM2

<table>
<thead>
<tr>
<th>Mechanism (pre-processor code)</th>
<th>Model: Chemistry Description</th>
<th>#Species</th>
<th>#Reactions</th>
</tr>
</thead>
<tbody>
<tr>
<td>TSMLT1 (pp_waccm_tsmlt_mam4)</td>
<td>WACCM: Troposphere, stratosphere, mesosphere, and lower thermosphere</td>
<td>231 solution, 2 invariant</td>
<td>583 (433 kinetic, 150 photolysis)</td>
</tr>
<tr>
<td>TS1 (pp_trop_strat_mam4_vbs)</td>
<td>CAM-chem: Troposphere and stratosphere</td>
<td>221 solution, 3 invariant</td>
<td>528 (405 kinetic, 123 photolysis)</td>
</tr>
<tr>
<td>MA (pp_waccm_ma_mam4)</td>
<td>WACCM: Middle atmosphere (stratosphere, mesosphere, and lower thermosphere)</td>
<td>98 solution, 2 invariant</td>
<td>298 (207 kinetic, 91 photolysis)</td>
</tr>
<tr>
<td>MAD (pp_waccm_mad_mam4)</td>
<td>WACCM: Middle atmosphere plus D-region ion chemistry</td>
<td>135 solution, 2 invariant</td>
<td>593 (489 kinetic, 104 photolysis)</td>
</tr>
<tr>
<td>SC (pp_waccm_sc_mam4)</td>
<td>WACCM: Specified chemistry</td>
<td>29 solution, 8 invariant</td>
<td>12 (11 kinetic, 1 photolysis)</td>
</tr>
<tr>
<td>CAM</td>
<td>CAM: Aerosol chemistry</td>
<td>25 solution, 7 invariant</td>
<td>7 (6 kinetic, 1 photolysis)</td>
</tr>
</tbody>
</table>
Tropospheric Ozone Chemistry

Young et al., Elementa, 2017
Models treat each process as a separate module

**Chemistry:**
gas-phase kinetics, photolysis, aerosol formation, heterogeneous reactions

**Transport:**
Advection, boundary layer mixing, diffusion

**Cloud Processes:**
Convective transport, Aqueous chemistry, Wet deposition

**Vertically-distributed Emissions**
Aircraft (NO, BC), Lightning NO

**Surface Emissions**
Anthropogenic, Biomass burning

**Biogenic Emissions**

**Dry Deposition**

**Land / Ocean / Snow / Ice**
Solution for each chemical species $i$

$$\frac{\partial c(i)}{\partial t} = Production(i) - Loss(i) = E_i + C_i + A_i + T_i + W_i + D_i$$

- For each compound, at each timestep, the change in concentration is the sum of the change in concentration for each process:
  - $E_i$: Emissions
  - $C_i$: Gas-phase-Chemistry
  - $A_i$: Aerosol-processes
  - $T_i$: Advection + Diffusion
  - $W_i$: Cloud-processes (wet deposition)
  - $D_i$: Dry deposition

- For compounds with short lifetimes the order of operators can affect results
Modal Aerosol Model (MAM4)

Representation of:
- Sulfates,
- Black Carbon
- Organic Carbon, Organic Matter (OC, SOA),
- Mineral Dust and Sea-Salt

Liu et al., 2016
Secondary Organic Aerosol Description in WACCM and CAM-chem

**Simplified Chemistry (CAM6):**
- SOAG (oxygenated VOCs) derived from fixed mass yields
- no interactions with land

**Comprehensive Chemistry:**
- SOAG formation derived from VOCs using Volatility Basis Set (VBS) description
- 5 volatility bins
- Interactive with land emissions
- more physical approach

Modified after C. Heald, MIT Cambridge
Modeling Chemistry-Climate Interactions in CESM2

Surface concentrations and emissions

- surface concentrations (greenhouse gases)
- emission: anthropogenic, biogenic, biomass burning, ocean, soil, volcanoes

Chemical mechanism: important for chemistry and aerosol production

- WACCM and CAMchem: 417 reactions and 220 solution species
- CAM6: 6 chemical reactions and 26 solution species (much simpler)

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).
Dry Deposition

$$V_d = \frac{1}{R_a + R_b + R_c}$$

- $R_a$: Resistance of dynamic sublayer
- $R_b$: Resistance of interfacial sublayer
- $R_c$: Resistance of vegetation sublayer

- Resistance of laminar sub-layer:
  - Resistance of wet surface
  - Resistance of stomata
  - Resistance of dry surface

Varies with surface type (vegetation, ocean, etc.)
Key component of ozone budget
Important for sticky and soluble gases: HNO$_3$, CO, OVOCs, etc.
Important processes for simulating Aerosols

Surface concentrations and emissions
Chemical mechanism: important for chemistry and aerosol production

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).

- Removal is modeled as a simple first-order loss process

\[
X_{\text{iscav}} = X_i \times F \times (1 - \exp(-\lambda \Delta t))
\]

- \(X_{\text{iscav}}\) is the species mass (in kg) of \(X_i\) scavenged in time
- \(F\) is the fraction of the grid box from which tracer is being removed, and \(\lambda\) is the loss rate.

Compsets define the specifics of emissions, chemistry, and deposition!
CAM-chem – Spectral Element

CAM6-chem with Spectral Element and Regional Refinement is running with ~14 km over U.S. (~1° elsewhere)

Current science goals:
• Studying air quality and health impacts in U.S.
• Evaluating importance of greater chemical complexity vs. higher horizontal resolution
AMWG Diagnostic Package includes WACCM and Chemistry diagnostics

**Chemistry Set Description**
1. Tables / Chemistry of ANN global budgets
2. Vertical Contour Plots contour plots of DJF, MAM, JJA, SON and ANN zonal means
3. Ozone Climatology Comparisons Profiles, Seasonal Cycle and Taylor Diagram
4. Column O3 and CO lon/lat Comparisons to satellite data
5. Vertical Profile Profiles Comparisons to NOAA Aircraft observations
6. Vertical Profile Profiles Comparisons to Emmons Aircraft climatology
7. Surface observation Scatter Plot Comparisons to IMROVE

**WACCM Set Description**
1. Tables of regional min, max, means
2. Seasonal cycle line plots of SP, SM, EQ, NM, NP zonal means (vertical log scale)
3. Vertical seasonal cycle plots of SP, SM, EQ, NM, NP zonal means (vertical log scale)
4. Vertical contour plots of JUN, DEC, DJF, MAM, JJA, SON and ANN zonal means (vertical log scale)
5. Horizontal contour plots of JUL, AUG, JJA, DJF and ANN zonal means
## User Support: CAM-Chem Wiki page

https://wiki.ucar.edu/display/camchem/Home

| **Advanced Changes**            | • Data Assimilation  
|                                 | • Online Air-Sea Interface for Soluble Species  
|                                 | • Updating Gas-phase Chemistry  
|                                 | • Tagging CO and simple tracers  
|                                 | • Clone a Case  
|                                 | • Create a Branch  
|                                 | • Biogenic Emission Options (MEGAN)  
| **Model Component Descriptions** | • Wet Deposition  
|                                 | • Dry Deposition  
|                                 | • Gas-phase Chemistry  
|                                 | • Emission Inventories  
|                                 | • Aerosols  
| **Processing**                  | • Pre-processing  
|                                 | • Using CAM-chem output  
|                                 | • Automated CESM diagnostic package  
|                                 | • GitHub Tutorial  
| **User Community**              | • Current Users/Projects  
|                                 | • Chemistry-Climate Working Group Publications  
|                                 | • UCAR Publications  |
WACCM and CAM-Chem Customer Support

CGD Forum: [http://bb.cgd.ucar.edu/](http://bb.cgd.ucar.edu/)

Mike Mills  
WACCM Liaison  
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(303) 497-1425

Simone Tilmes  
CAM-Chem Liaison  
tilmes@ucar.edu  
(303) 497-1445
Extras
New Secondary Organic Aerosol approach in CESM2
CAM-chem and WACCM

Simplis=c ways of treating the complex SOA lifecycle

More physical approach
Direct coupling to biogenic emissions changes from MEGAN
-> couples SOA formation to land use and climate change

-> VBS (volatility bin scheme) only works in full chemistry version at this point
Biogenic, anthropogenic and biomass burning VOC, SIVOC

chem. Prod. 294 Tg/yr

+ Oxidants
Glyoxal uptake

Oxygenated VOC (gas)
0.38 Tg

dry
44 Tg/yr
wet
108 Tg/yr

Net gas-particle partitioning
142 Tg/yr

SOA
1.04 Tg

dry
12 Tg/yr
wet
72 Tg/yr

VBS Budgets 1995-2010

Lifetime: 4.5 years

J SOA
57 Tg/yr

Values very close to observational estimates!

Most of it from biogenic emissions
-> strongly dependent on MEGAN emissions

44 Tg/yr

chem. Prod. + Oxidants
Glyoxal uptake

Values very close to observational estimates!
• Gases and aerosols are transported in stratosphere-troposphere exchange
• Impact of halogen loading on stratospheric ozone (ozone hole)
• Impact on climate (importance of very short-lived species)

-> local changes on short time scales are important