Global aerosol microphysics modeling: Implications of new particle formation & growth for aerosol-climate simulations

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IPCC models have so far included only a simple representation of aerosols when simulating climate effects.

Only mass of aerosol components is advected quantity: (e.g., sulphate, black carbon, dust, sea-salt mass)

For size-dependent processes: An assumed size distribution

Direct aerosol forcing: Use composition-dependent mass scattering efficiency (or assume a fixed size distribution)

Indirect forcing: Use empirical cloud drop—aerosol relations,

New particle formation not included

Important aerosol types (e.g. organics, nitrate) omitted.

External mixtures only considered in optical properties.
Processes control size & composition

- Emission of gas phase precursors
- In-air oxidation
- Low volatility gas phase oxidation products
- Nucleation
- Coagulation
- Condensation
- Emission of primary particles
- In-cloud oxidation
- Activation of CCN
- Uptake to clouds
- Cloud processing
- Dry Deposition
- Wet Deposition

 Particle Size

~1.E-9 m

~50.E-6 m
Composite of CDN-aerosol observations from many sites

No single relationship fits observed CDN vs aerosol number. Different regions have different particle types, size distbtn, etc. IPCC models use of different relations must cause part of large “model uncertainty” in estimated 1st indirect aerosol forcing

From Ramanathan, Crutzen et al (2001)
Nucleation and CDN

Nucleation is an important source of CCN. Mass-only predictions cannot capture new particle formation and growth to CCN sizes.
Global Model of Aerosol Processes (GLOMAP)

Global CTM forced by 6-hourly ECMWF winds
Usually run at T42L31 (2.8°x2.8°) resolution
Sectional aerosol scheme: 20 bins, 3 nm – 20 μm
Modal scheme: 7 or 4 log-normal modes
Chemistry usually driven by offline oxidants, now coupled to CTM chemistry
Aerosol transport, new particle formation, growth by coagulation, condensation, cloud processing.
Wet and dry deposition of gases & aerosol particles

Emissions of DMS $\rightarrow$ SO$_2$ $\rightarrow$ H$_2$SO$_4$; monoterpenes $\rightarrow$ biogenic SOA
Primary emissions of sea salt, dust, black & organic carbon (fossil and biofuels, vegetation fires)
Nucleation via binary homogeneous nucleation of H$_2$SO$_4$-H$_2$O and also now implemented boundary layer nucleation mechanism

Spracklen et al. (ACP, 2005a,b, 2006, 2007)
Monthly mean global fields

Using model size distribution and the mechanistic CDN scheme of Nenes and Seinfeld (2003)

Total aerosol

Max supersaturation

Cloud drop number (w=0.3 m/s)
Exploring the scatter in model CDN-aerosol
Variability in predicted CDN

Percent of days that exceed 15th & 85th percentile
Global CDN prediction based on single-region CDN-aerosol relation

Use model output to generate CDN-aerosol empirical fit
Use the fit to calculate global CDN
Calculate % difference compared to mechanistic CDN scheme

75% more CDN in S. Ocean using mechanistic scheme than predicted from CDN-aerosol relation over the Atlantic
Why is new particle formation & growth is important

- SO2 emissions regionally different potential to form CCN
- Impact of DMS on CCN controlled by new particle formation
- 1st indirect effect: change in cloud albedo 1850-2000

Global aerosol models now simulate aerosol microphysics:

- Can resolve size distribution & size-dependent composition
- AEROCOM modellers to evaluate particle size.
- Utilize GAW, ARM, EMEP, EUSAAR data records from CPCs, CCN, DMPSs, Aerosol Mass Spectrometers etc
- Also use field campaign climatologies.
Regional export potential of SO2 emissions

SO\textsubscript{4} mass

CN no. conc.

CCN no. conc.
Regional CCN potential of SO2 emissions

<table>
<thead>
<tr>
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<th>N. America</th>
<th>Europe</th>
<th>E. Asia</th>
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</thead>
<tbody>
<tr>
<td>SO(_4) production efficiency(^1)</td>
<td>0.42</td>
<td>0.35</td>
<td>0.39</td>
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<tr>
<td>SO(_4) lifetime (days)</td>
<td>3.2</td>
<td>4.7</td>
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<td>SO(_4) burden potential(^2)</td>
<td>0.77</td>
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<td>CCN potential(^3)</td>
<td>0.4</td>
<td>0.13</td>
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<tr>
<td>CCN climate potential(^4)</td>
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<td>0.07</td>
<td>0.06</td>
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<tr>
<td>SO(_4) export(^5)</td>
<td>0.34</td>
<td>0.61</td>
<td>0.26</td>
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<tr>
<td>CCN export</td>
<td>0.68</td>
<td>0.82</td>
<td>0.90</td>
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Manktelow et al (in prep., 2008)
GLOMAP sensitivity simulations confirm that DMS is the cause of observed annual CCN cycle at Cape Grim, Tazmania.
Spatial impact of DMS on CCN strongly spatially inhomogeneous

Non-local effect of DMS on CCN concentration

Low increase in CCN in 50-65S despite highest DMS emissions
Highest increase in CCN in 30-50S (>50 cm^{-3}, 70-100%)

Korhonen et al (2008, JGR)
Sensitivity experiments in GLOMAP reveal controlling processes in remote CCN production.

By switching off various processes we find that >90% of DMS-derived CCN in the Southern Ocean originate from the free troposphere. We find that growth of ultrafine sea spray is unimportant for CCN.
Kulmala et al., have showed that new particle formation in boundary layer is given by

$$J_3 = A[H_2SO_4] \exp\left( B \frac{CS'}{[H_2SO_4]} \right)$$

Spracklen et al. (2006)
Enhancement of CCN with BL nucleation

Ratio of March-May CCN (1%) with NPF ($A=2\times10^{-6}$ s$^{-1}$) : CCN without NPF

Simulated ratio of global mean (grey shading) and regional mean CCN (Europe, Finland, Boreal Asia) with to without NPF. Error bars show sensitivity to increasing secondary organic aerosol by a factor 5. The x-axis shows sensitivity to varying nucleation rate [Spracklen et al, GRL, 2008]

New particle formation increases global mean BL CCN concentrations by 5-50%.
Global increase in CDNC 16% in 1850 and 14% in 2000. However, there are large regional differences!
According to Twomey, the change in cloud droplet number results in a change in cloud albedo $R_c$

$$\Delta R_c = \frac{R_c(1 - R_c)}{3} \ln \left( \frac{CDNC(2000)}{CDNC(1850)} \right)$$
The resulting change in cloud albedo with when particle formation is included:

- Arctic: -24.3%
- North temperate zone: -9.6%
- North tropics: -6.7%
- South tropics: 19.4%
- South temperate zone: 25.8%
- Antarctic: 94.5%

- Global average difference only -3%
- However, a large north-south contrast in results
GLOMAP CN being evaluated against observations at GAW and ARM sites

Ratio of simulated annual mean CN concentration with BL new particle formation: without

Model: Binary Homogeneous nucleation. Shading shows primary particle number emission varied by a factor 8
Model: BL New Particle Formation ($A = 2 \times 10^{-6} \text{ s}^{-1}$, $M=1$) + primary particles
GLOMAP CCN being evaluated against a range of worldwide observations

Note: map shows CCN at 0.2% supersaturations. Coloured circles show observations at range of supersaturations.
GLOMAP CCN being evaluated against a range of worldwide observations

GLOMAP-mode vs. observations

GLOMAP-bin vs. observations

GLOMAP-bin vs. -mode

SS<0.25%
SS<0.5%
SS<0.75%
SS>0.75%

0–90E
90E–180E
180W–90W
90W–0W
GLOMAP size distributions being evaluated against European DMPS observations

Hyytiala, Finland

Hohenpeissenberg, Germany

BHN

\[ A = 2 \times 10^{-7} \text{ s}^{-1} \]

\[ A = 2 \times 10^{-6} \text{ s}^{-1} \]

\[ A = 2 \times 10^{-5} \text{ s}^{-1} \]

\[ A = 2 \times 10^{-5} \text{ s}^{-1}, \text{SOA} \times 5 \]
Conclusions

GLOMAP aerosol microphysics model simulates new particle formation and processes which control growth to CCN.

Simulated AOD, CN, CCN, mass and size agree quite well with observations giving confidence for model predictions.

DMS impact on CCN mainly via UT binary nucleation and subsequent growth & entrainment into MBL.

Boundary layer nucleation enhances cloud droplet number concentrations significantly both in 1850 and 2000.

Simulating BLN enhances Southern Hemisphere CDN change and reduces Northern Hemisphere CDN change.

UKCA aerosol-chemistry-climate model now developed with GLOMAP aerosol microphysics via modal scheme in UM.

UKCA will more realistically simulate aerosol-climate effects.