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

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National Centre for Atmospheric Science IPCC models have so far included only a simple representation of aerosols when simulating climate effects.

- Only <u>mass</u> 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



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

Nucleation and CDN





nucleation mode

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 μ m 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 SO₂ \rightarrow H₂SO₄; monoterpenes \rightarrow biogenic SOA

Primary emissions of sea salt, dust,

black & organic carbon (fossil and biofuels, vegetation fires)

Nucleation via binary homogeneous nucleation of H₂SO4-H₂O 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)





Variability in predicted CDN

PDF of CDN being > 85th Percentile CDN (w = 0.15ms-1)



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 CCN potential of SO2 emissions







	N.Americ a	Europe	E. Asia
SO ₄ production efficiency ¹	0.42	0.35	0.39
SO ₄ lifetime (days)	3.2	4.7	2.7
SO_4 burden potential ²	0.77	0.93	0.64
CCN potential ³	0.4	0.13	0.19
CCN climate potential ⁴	0.12	0.07	0.06
SO ₄ export ⁵	0.34	0.61	0.26
CCN export	0.68	0.82	0.90



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



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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





Enhancement of CCN with BL nucleation



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%.



Cloud droplet number concentrations



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 *Rc*

$$\Delta Rc = \frac{Rc(1-Rc)}{3} \ln \left(\frac{CDNC(2000)}{CDNC(1850)} \right)$$

dRC with BLPF, Rc=0.35, w=0.4



dRC without BLPF, Rc=0.35, w=0.4



Effect of boundary layer particle formation to the change in cloud albedo

The resulting change in cloud albedo with when particle formation is included:



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- Global average difference only -3%
- However, a large north-south contrast in results

GLOMAP CN being evaluated against observations at GAW and ARM sites





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 size distributions being evaluated against European DMPS observations





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