

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

Graham Mann¹, Ken Carslaw¹, Dominick Spracklen¹,
Kirsty Pringle², Hannele Korhonen³, Joonas Merikanto¹,
Paul Manktelow¹, Martyn Chipperfield¹

1: School of Earth & Environment, University of Leeds, U.K.

2: now Max Planck Institute for Chemistry, Mainz, Germany

3: now University of Kuppio, Finland



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)

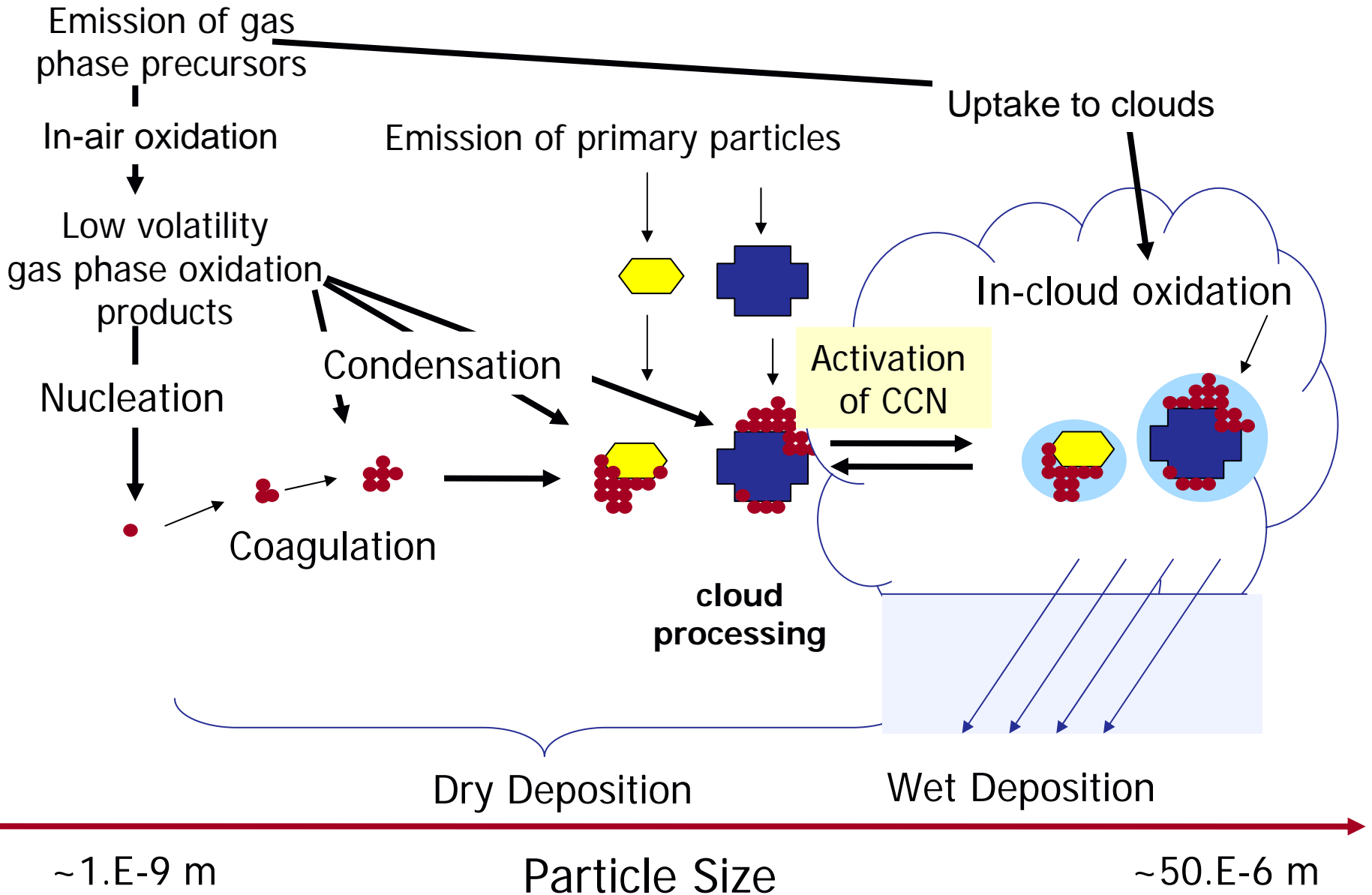
Indirect forcing: Use empirical cloud drop—aerosol relations,

New particle formation not included

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

External mixtures only considered in optical properties.

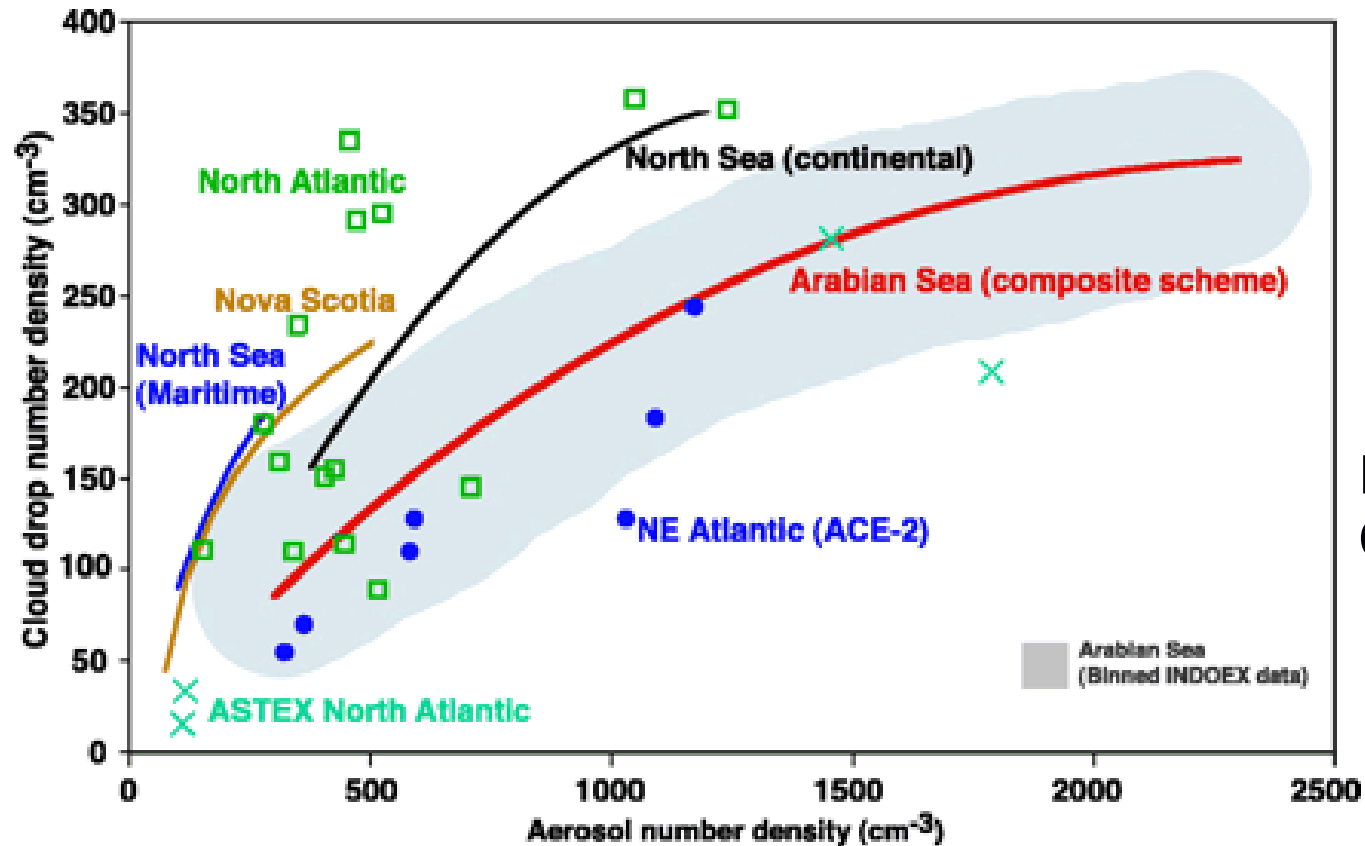
Processes control size & composition



Composite of CDN-aerosol observations from many sites

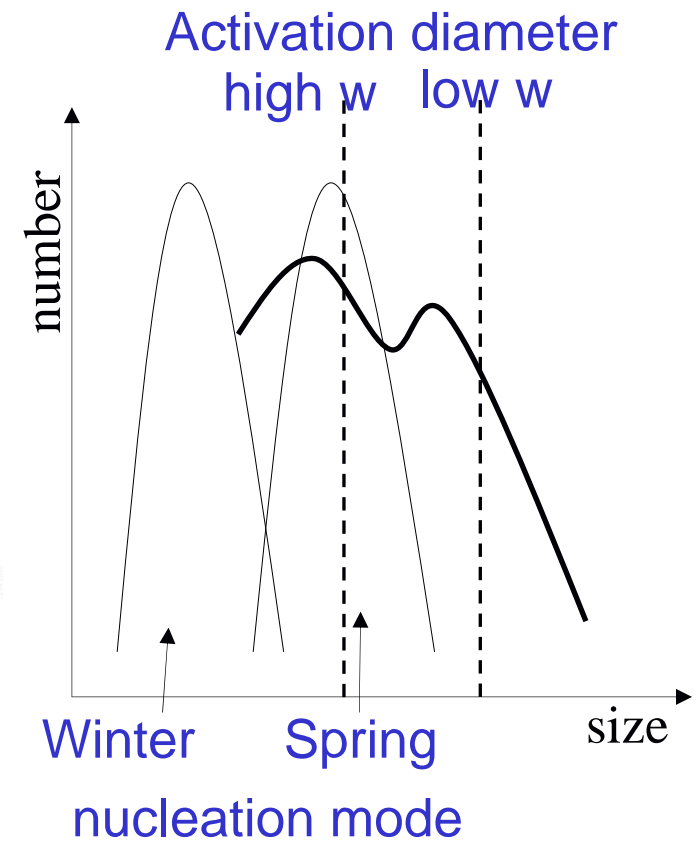
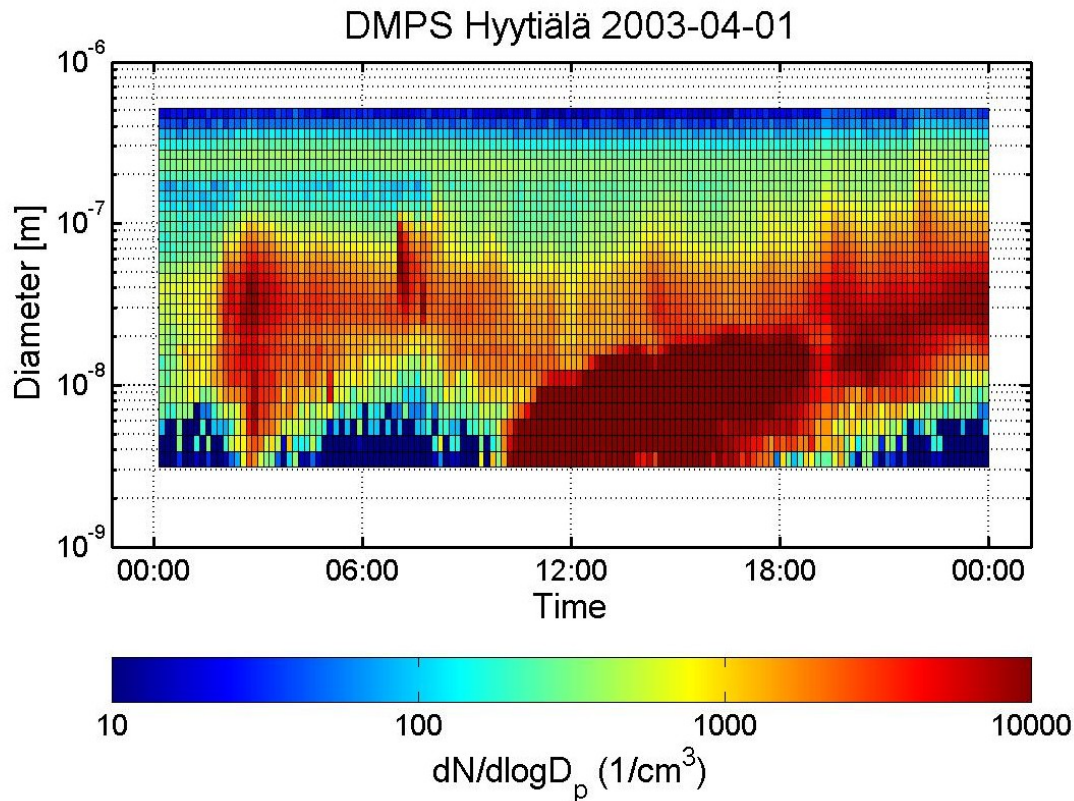


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From Ramanathan, Crutzen et al (2001)

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



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)



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Global CTM forced by 6-hourly ECMWF winds

Usually run at T42L31 ($2.8^\circ \times 2.8^\circ$) 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.

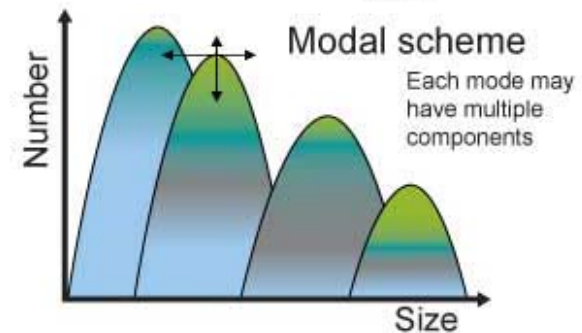
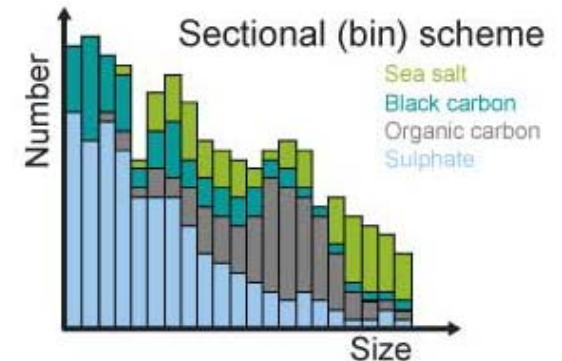
Wet and dry deposition of gases & aerosol particles

Emissions of DMS \rightarrow $\text{SO}_2 \rightarrow \text{H}_2\text{SO}_4$; monoterpenes \rightarrow biogenic SOA

Primary emissions of sea salt, dust,
black & organic carbon (fossil and biofuels, vegetation fires)

Nucleation via binary homogeneous nucleation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$
and also now implemented boundary layer nucleation mechanism

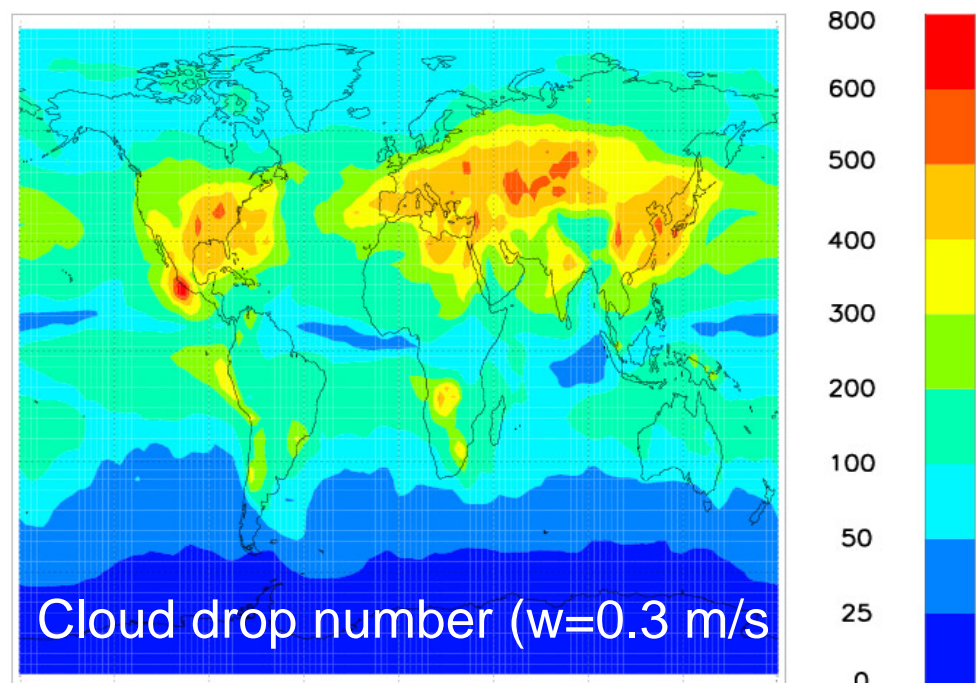
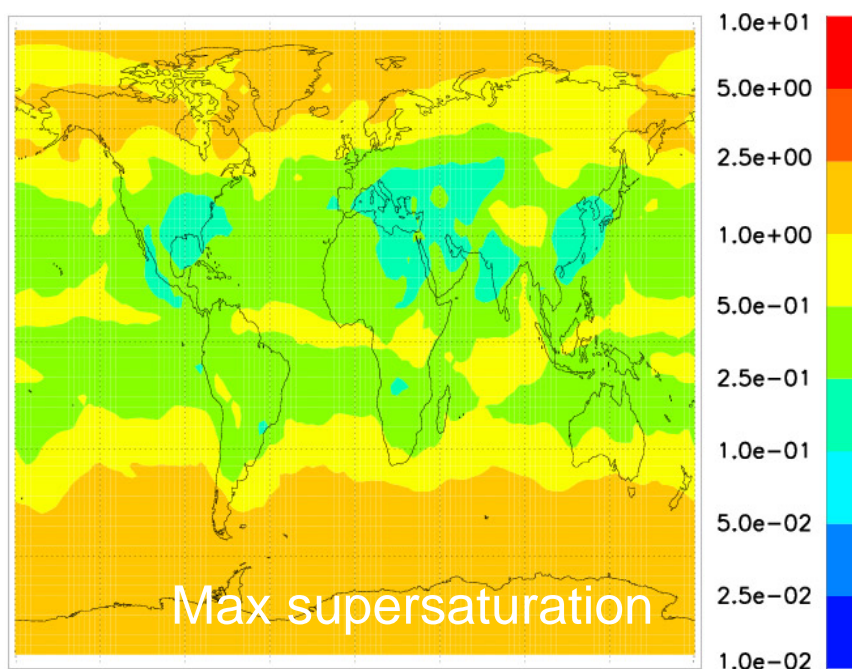
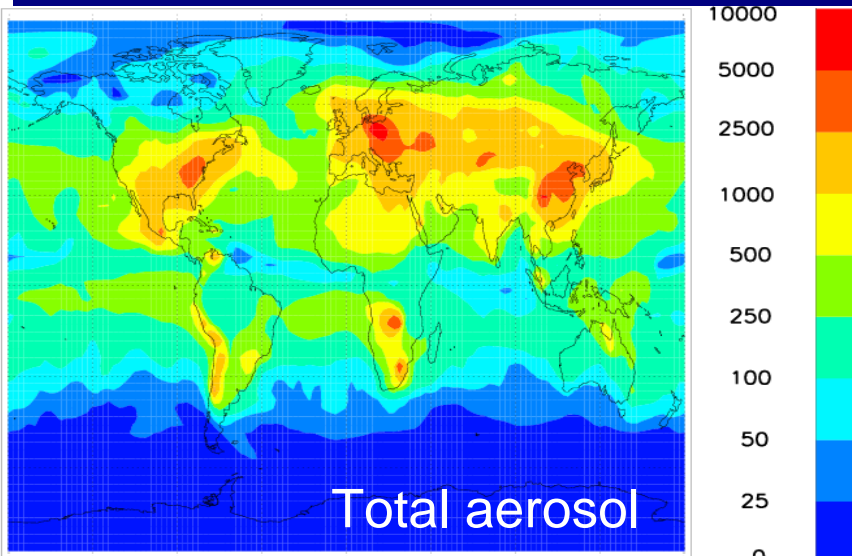
Spracklen et al. (ACP, 2005a,b, 2006, 2007)



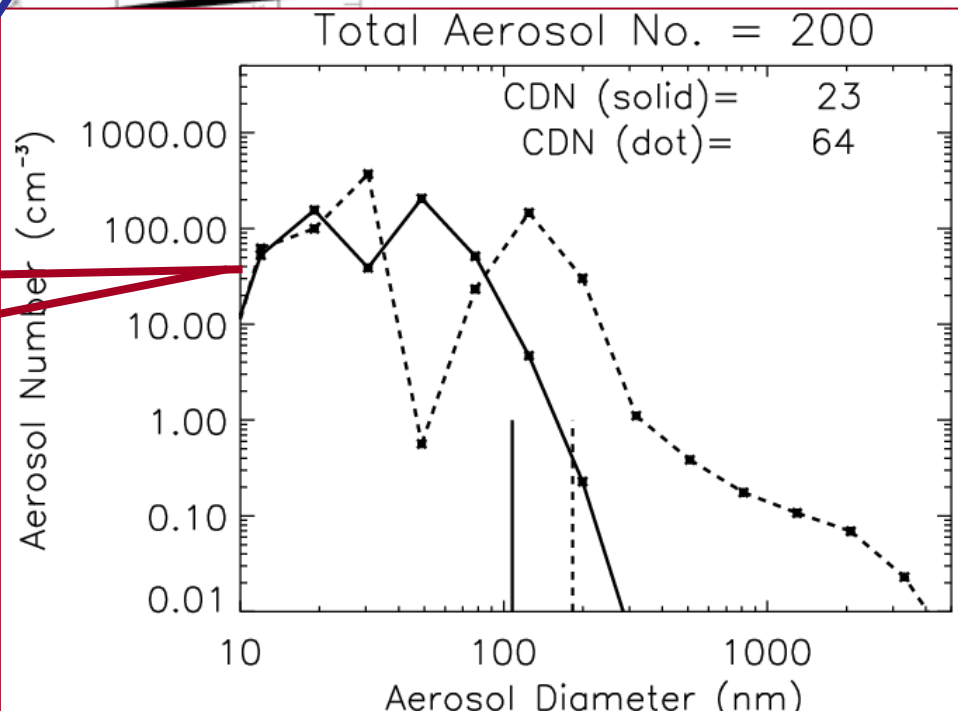
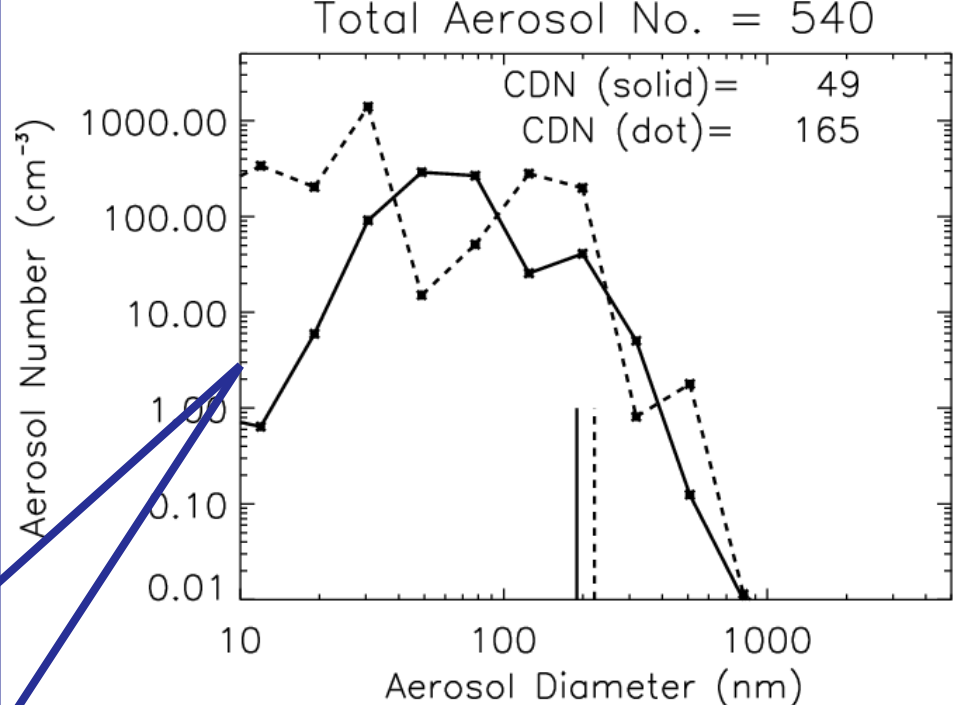
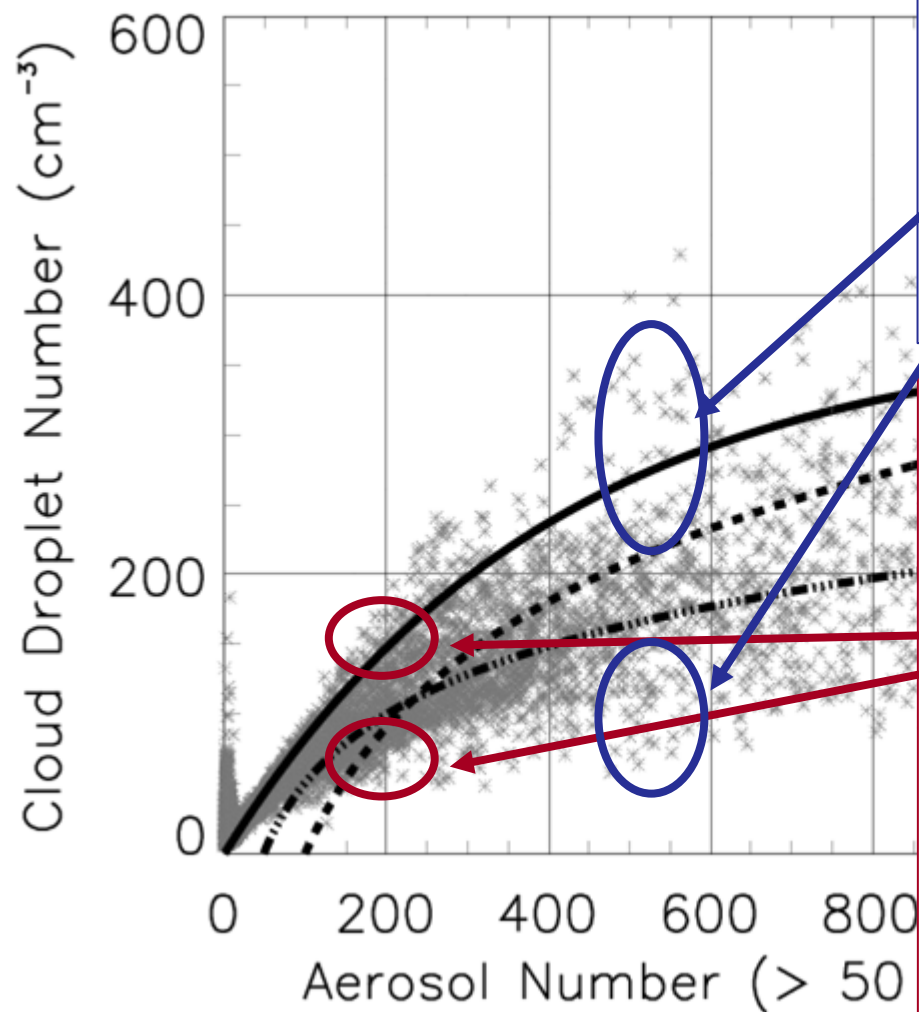
Monthly mean global fields



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



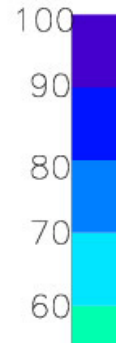
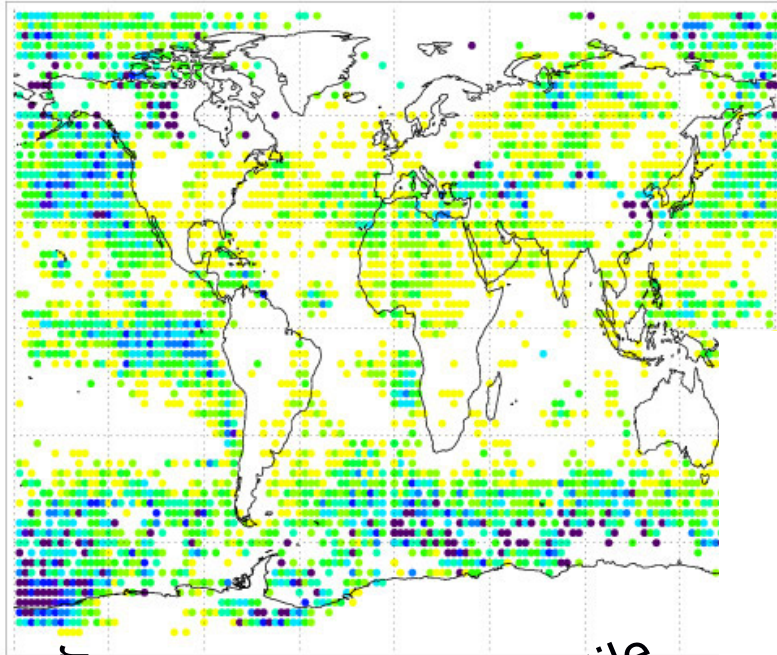
Exploring the scatter in model CDN-aerosol



Variability in predicted CDN

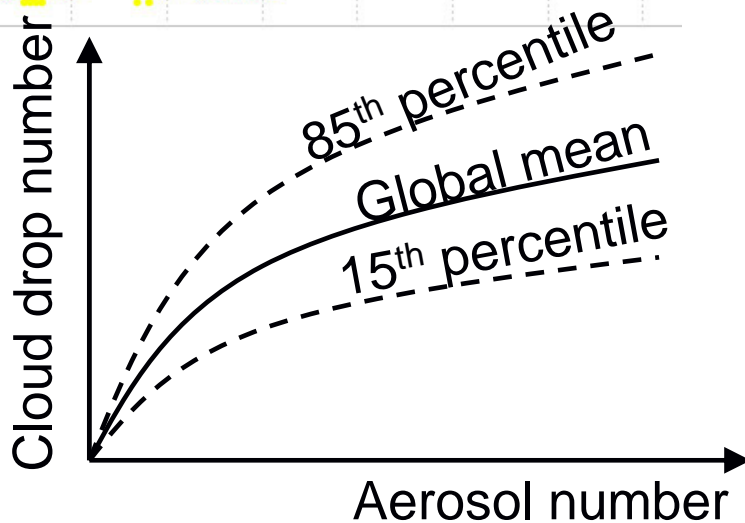
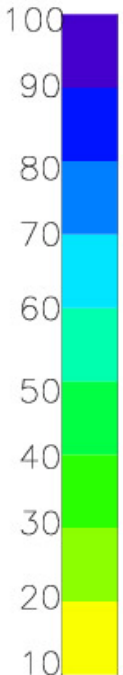
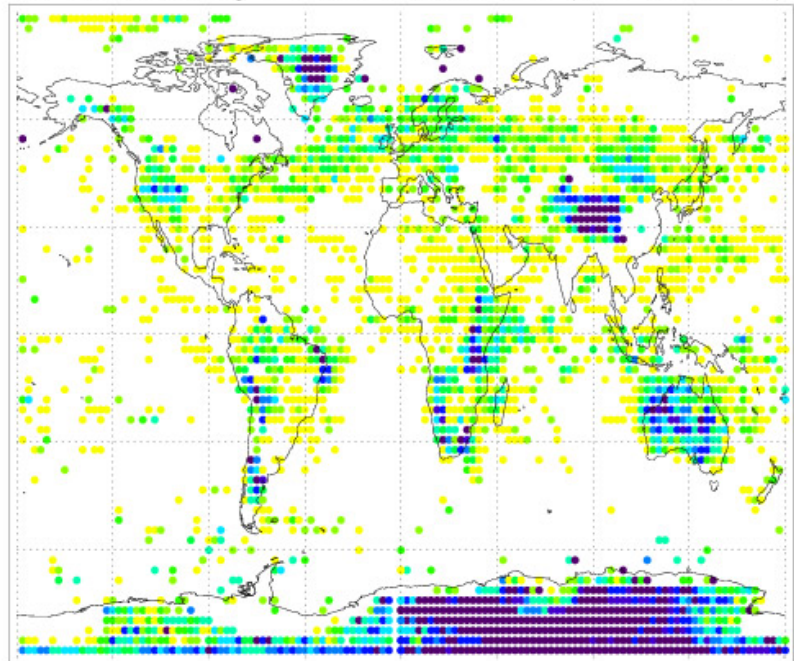


PDF of CDN being > 85th Percentile CDN ($w = 0.15\text{ms}^{-1}$)



Percent of days that exceed
15th & 85th percentile

PDF of CDN being < 15th Percentile CDN ($w = 0.15\text{ms}^{-1}$)



Global CDN prediction based on single-region CDN-aerosol relation



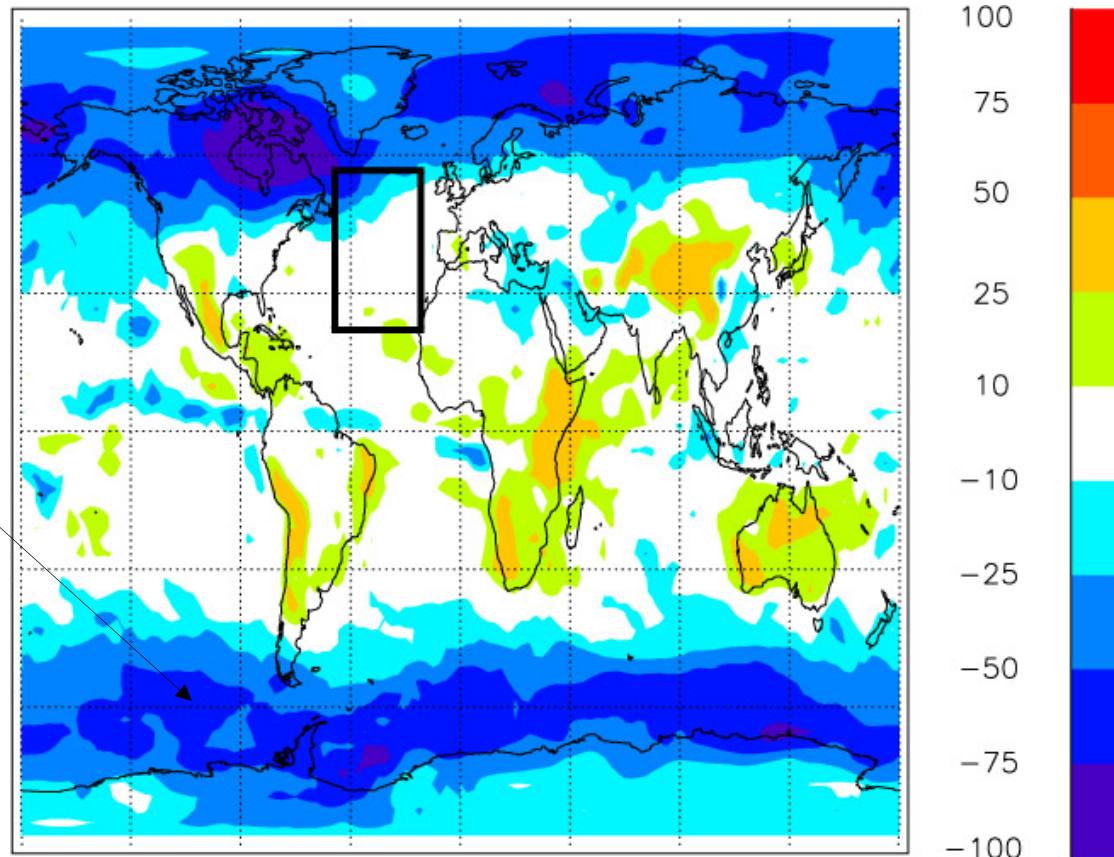
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Use model output to generate CDN-aerosol empirical fit

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 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



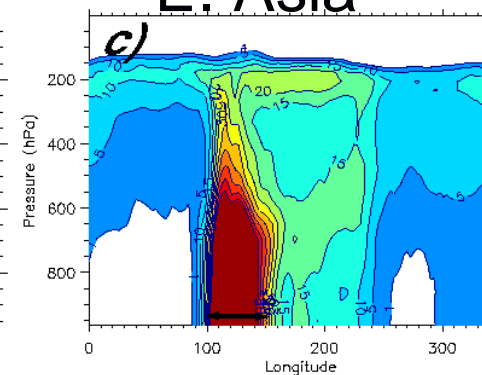
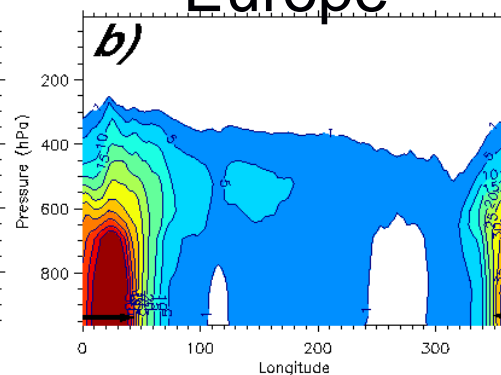
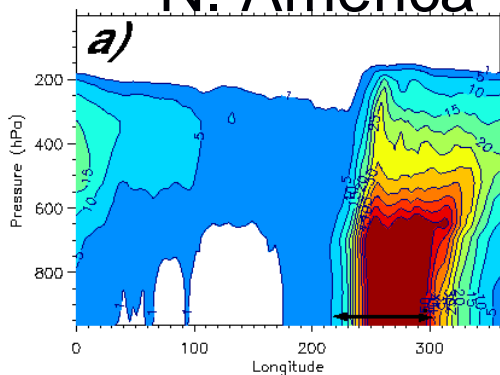
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N. America

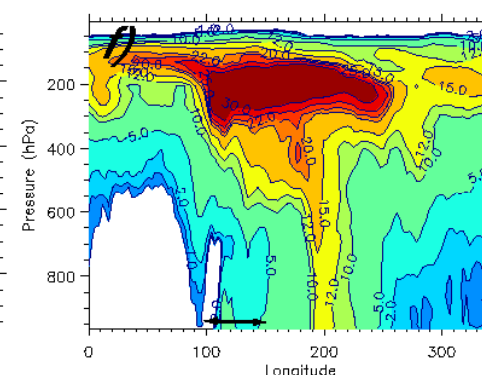
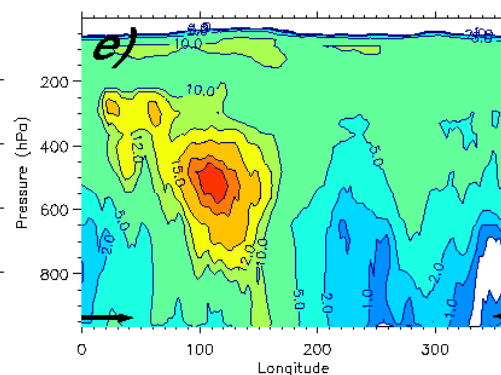
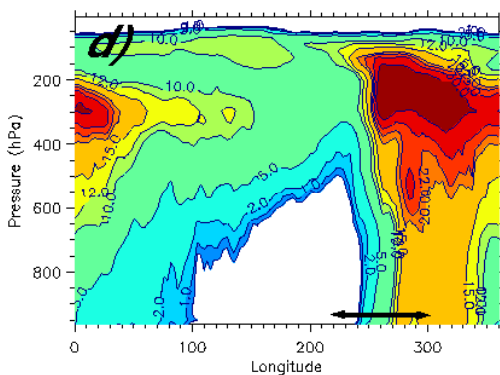
Europe

E. Asia

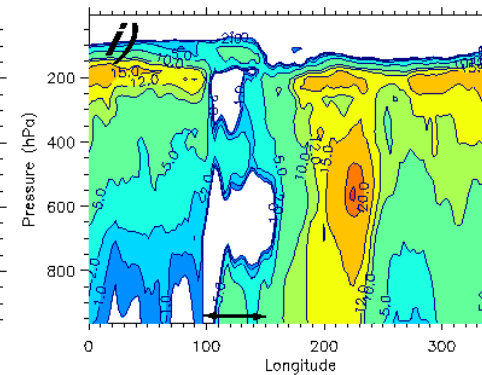
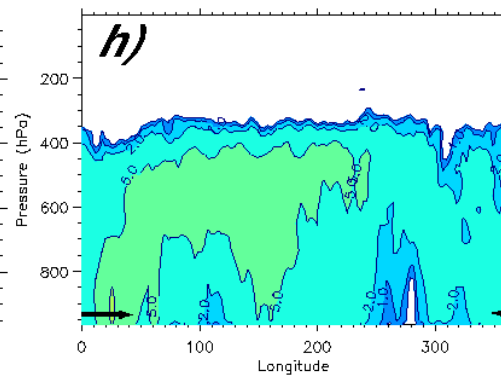
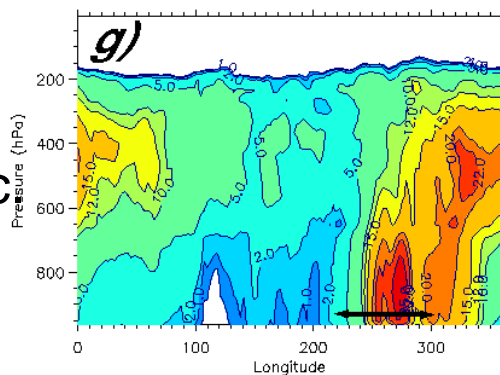
SO₄ mass



CN no. conc.



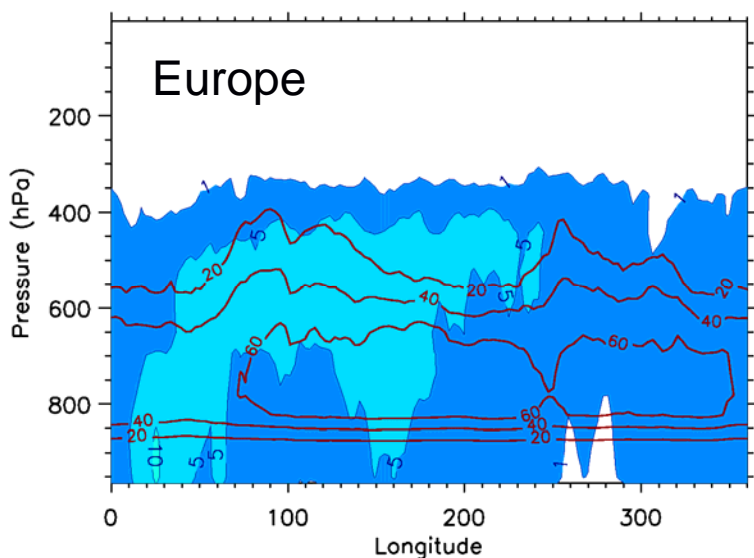
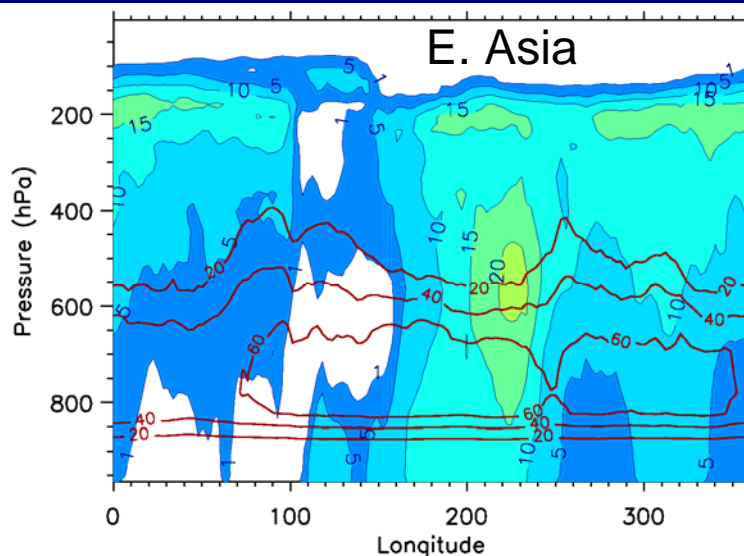
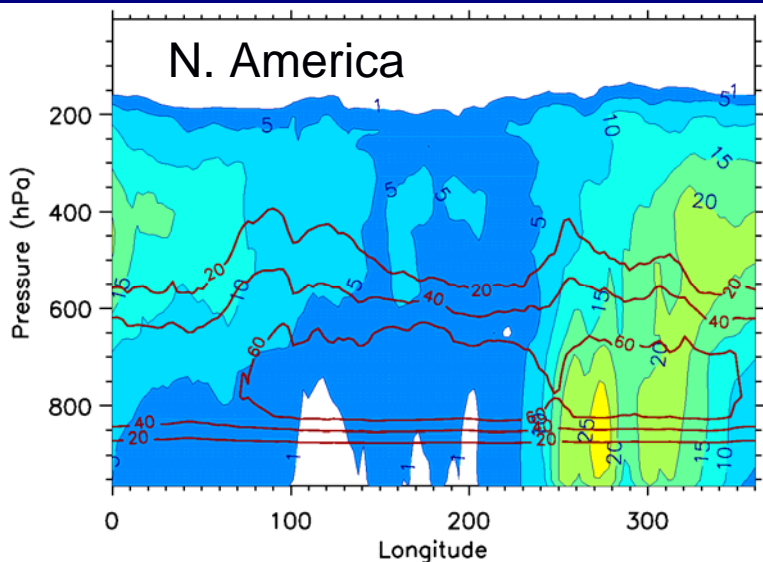
CCN no. conc



Regional CCN potential of SO₂ emissions



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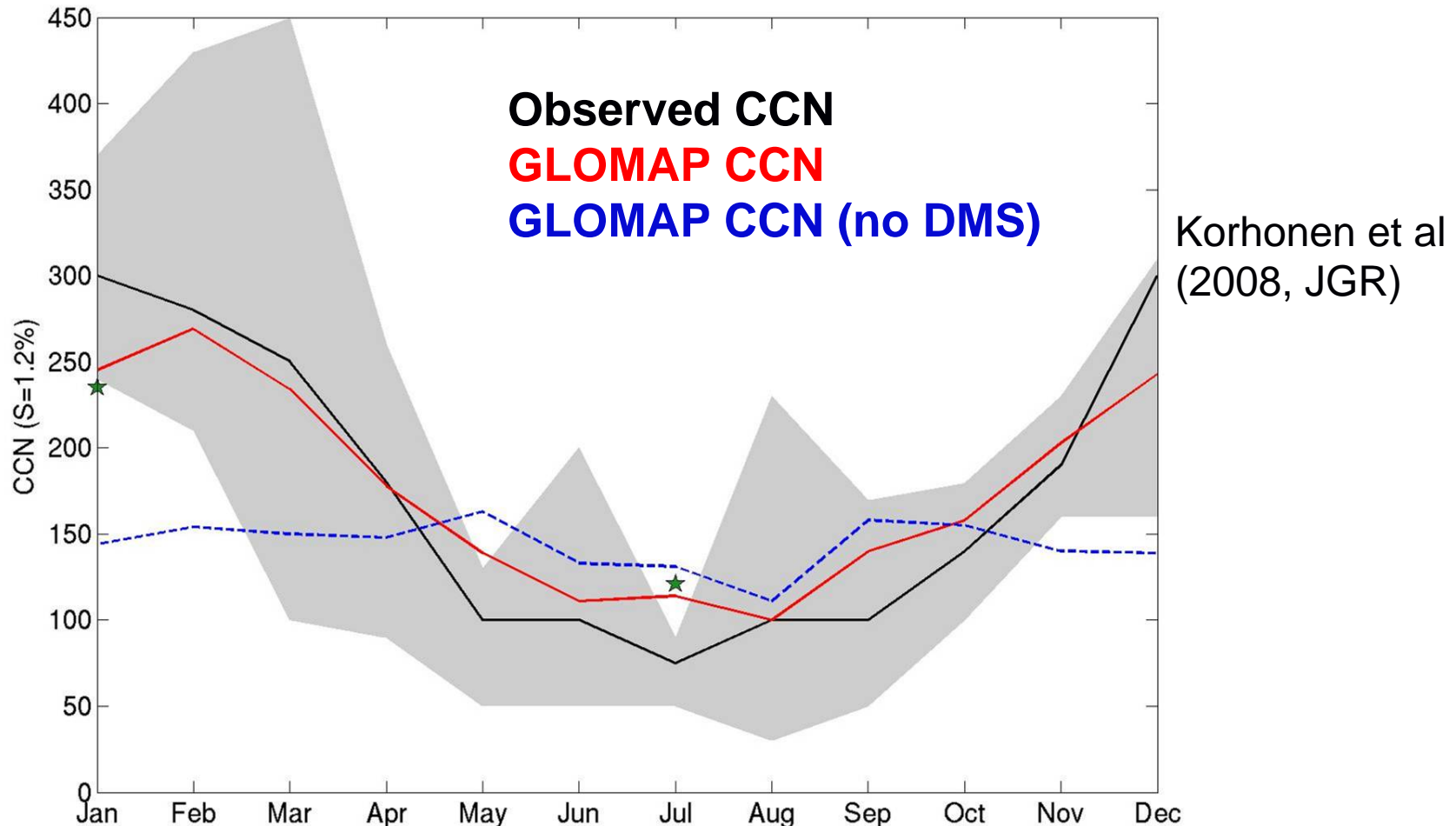
	N.Americ a	Europe	E. Asia
SO ₄ production efficiency ¹	0.42	0.35	0.39
SO ₄ lifetime (days)	3.2	4.7	2.7
SO ₄ burden potential ²	0.77	0.93	0.64
CCN potential ³	0.4	0.13	0.19
CCN climate potential ⁴	0.12	0.07	0.06
SO ₄ export ⁵	0.34	0.61	0.26
CCN export	0.68	0.82	0.90

Manktelow et al (in prep., 2008)

DMS controls annual CCN cycle at Cape Grim



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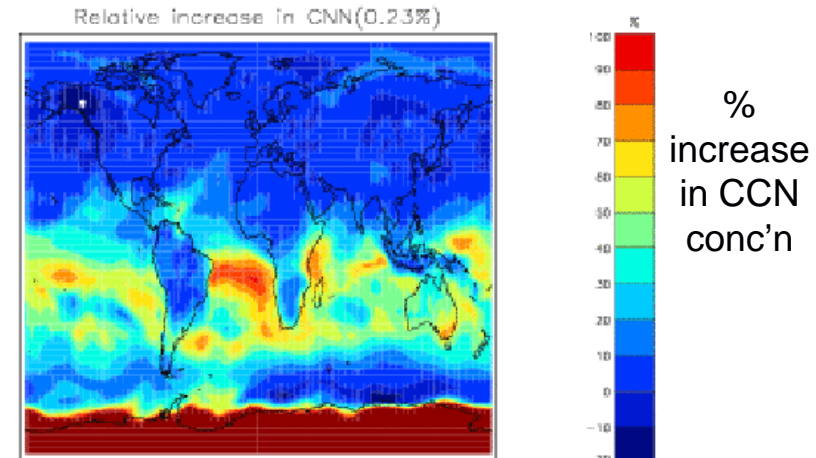
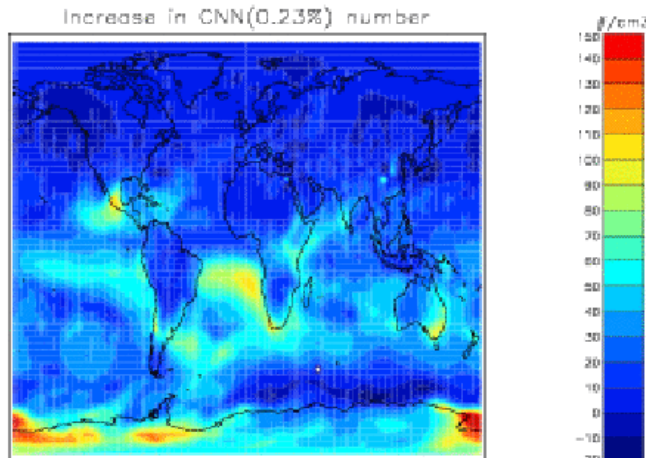
GLOMAP sensitivity simulations confirm that DMS is the cause of observed annual CCN cycle at Cape Grim, Tasmania.

Spatial impact of DMS on CCN strongly spatially inhomogeneous

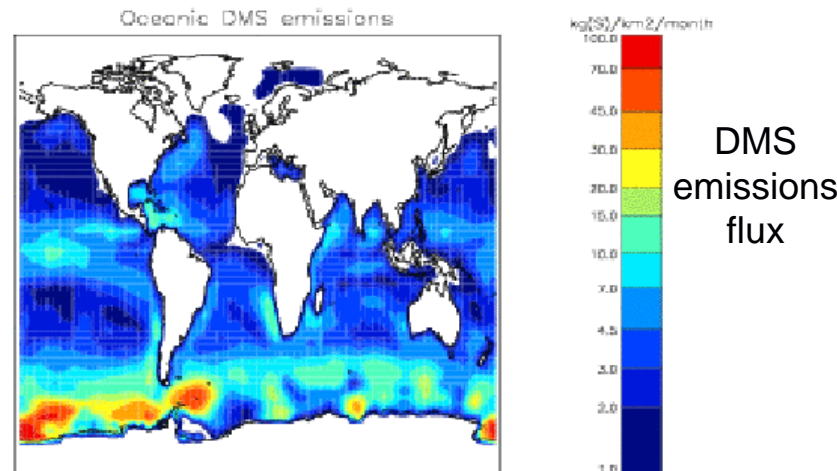
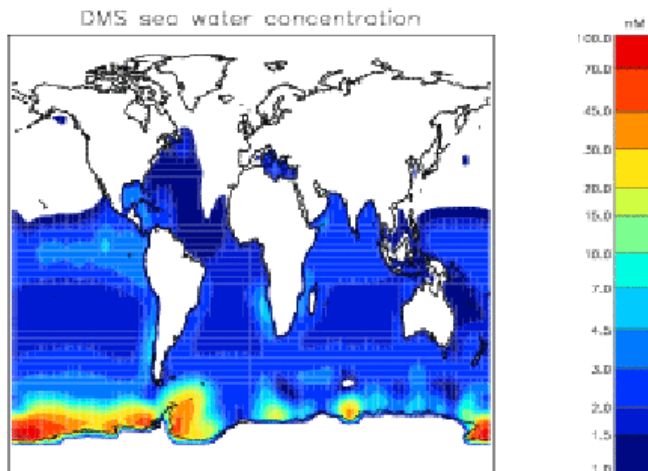


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Absolute increase in CCN concentration



DMS seawater concentration



Non-local effect of DMS on CCN concentration

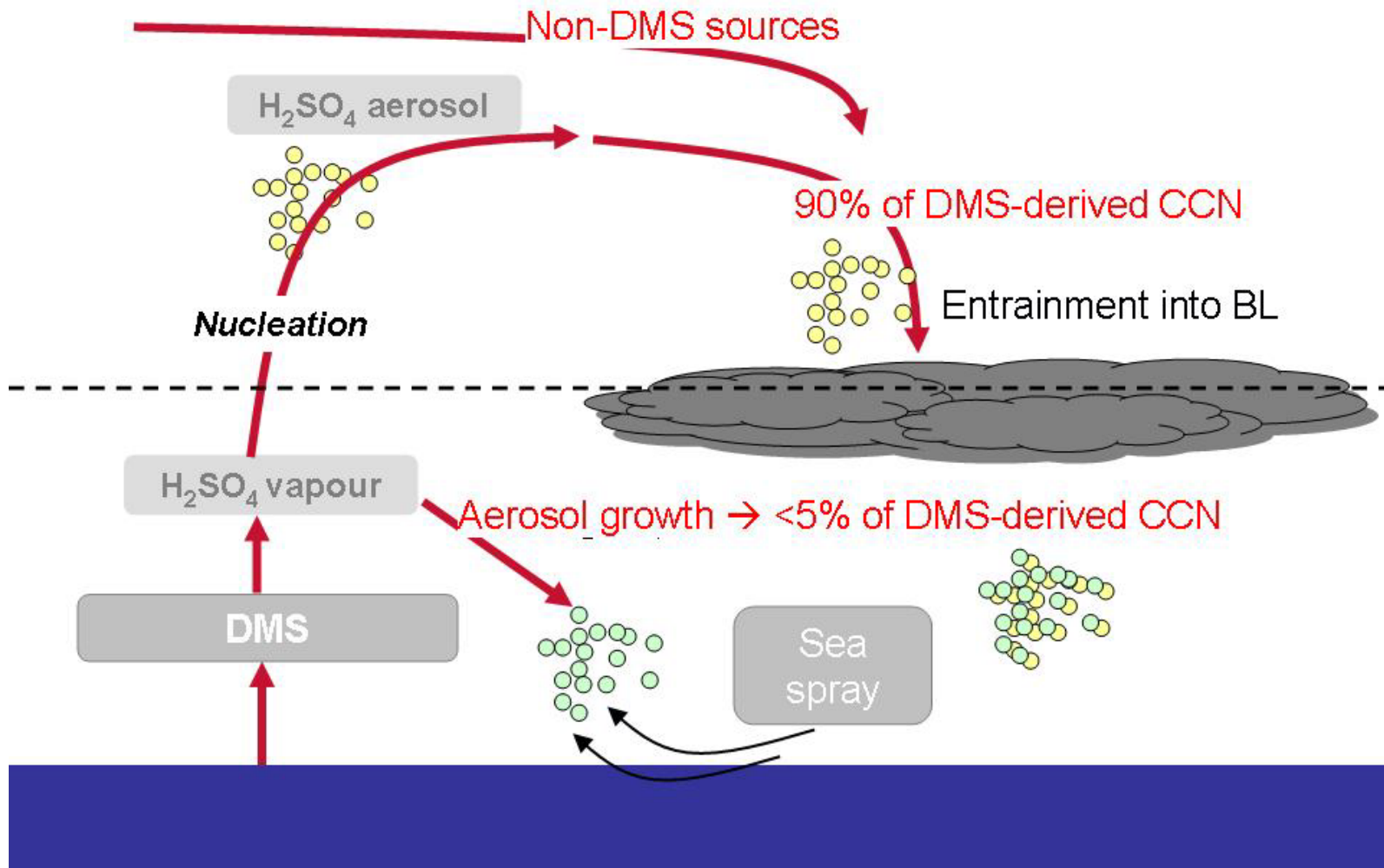
Low increase in CCN in 50-65S despite highest DMS emissions
Highest increase in CCN in 30-50S (>+50 cm⁻³, +70-100%)

Korhonen et al
(2008, JGR)

Sensitivity experiments in GLOMAP reveal controlling processes in remote CCN production



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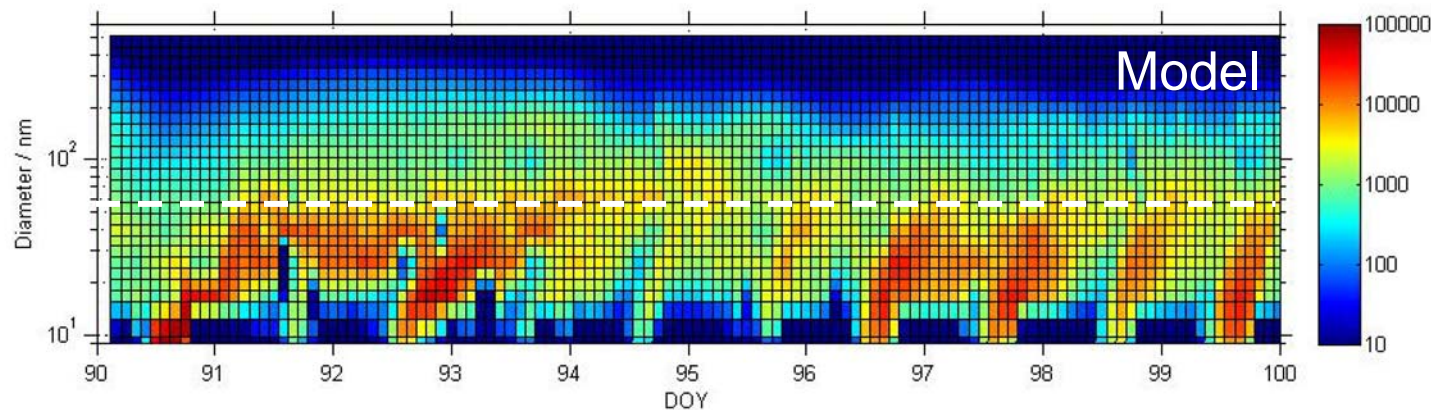
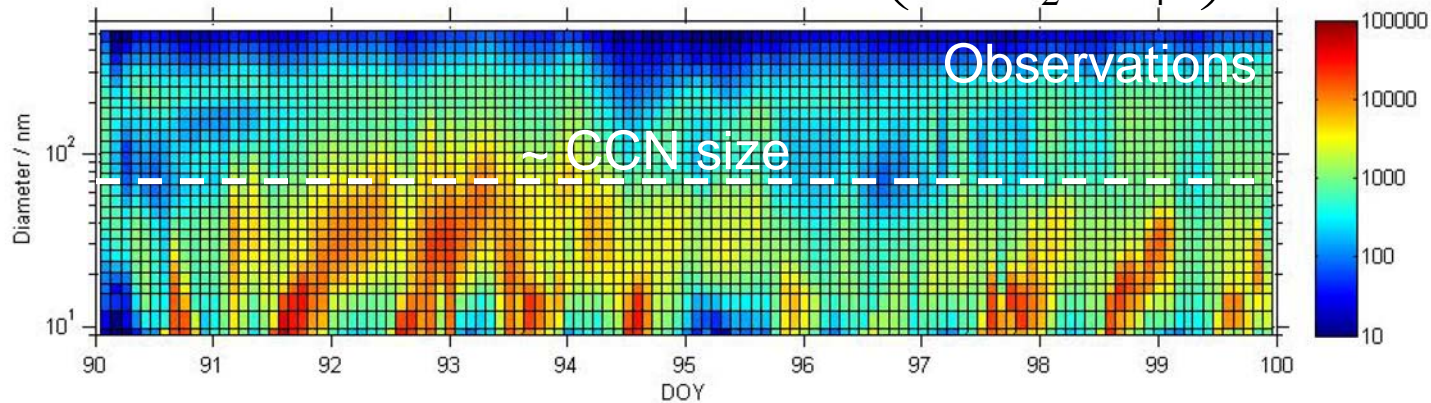
By switching off various processes we find that >90% of DMS-derived CCN in the Southern Ocean originate from the free troposphere.

We find that growth of ultrafine sea spray is unimportant for CCN

Growth of particles from nm to cloud nuclei

Kulmala et al., have showed that new particle formation in boundary layer is given by

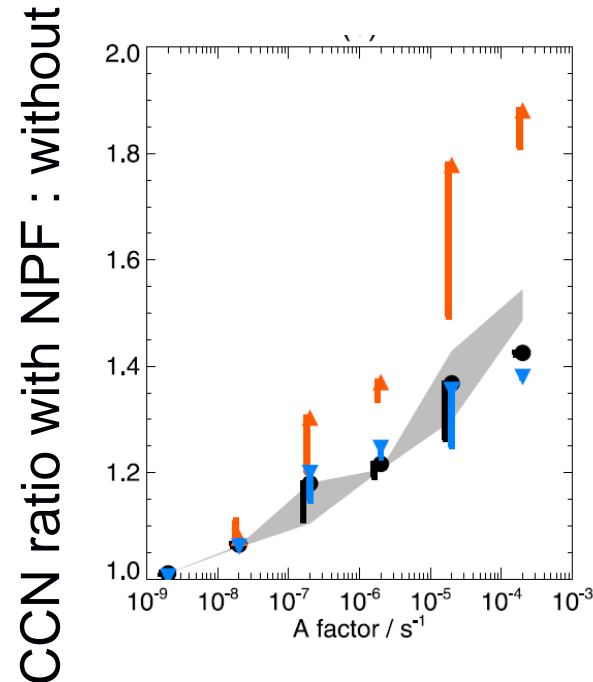
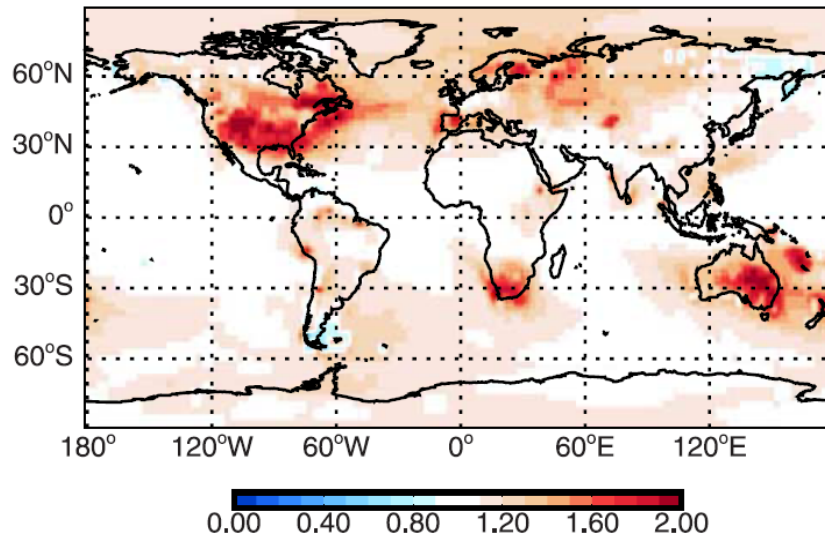
$$J_3 = A[\text{H}_2\text{SO}_4] \exp\left(B \frac{CS'}{[\text{H}_2\text{SO}_4]}\right)$$



Spracklen
et al (2006)

Enhancement of CCN with BL nucleation

Ratio of March-May CCN (1%) with NPF ($A=2 \times 10^{-6} \text{ s}^{-1}$) : CCN without NPF



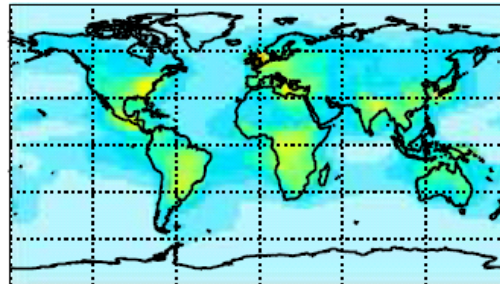
Simulated ratio of global mean (grey shading) and regional mean CCN (Europe, Finland, Boreal Asia) with to without NPF. Error bars show sensitivity to increasing secondary organic aerosol by a factor 5. The x-axis shows sensitivity to varying nucleation rate [Spracklen et al, GRL, 2008]

New particle formation increases global mean BL CCN concentrations by 5-50%.

Cloud droplet number concentrations

1850

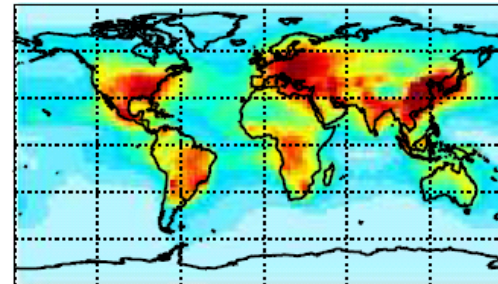
CDNC with BLPF, year 1850, $w=0.4$



0 200 400 600 800 cm⁻³

2000

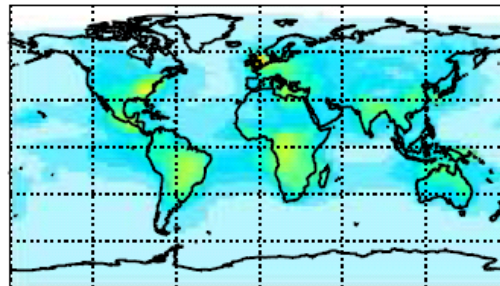
CDNC with BLPF, year 2000, $w=0.4$



0 200 400 600 800 cm⁻³

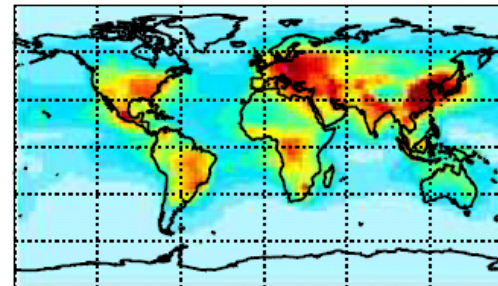
With particle formation

CDNC without BLPF, year 1850, $w=0.4$



0 200 400 600 800 cm⁻³

CDNC without BLPF, year 2000, $w=0.4$



0 200 400 600 800 cm⁻³

Without particle formation

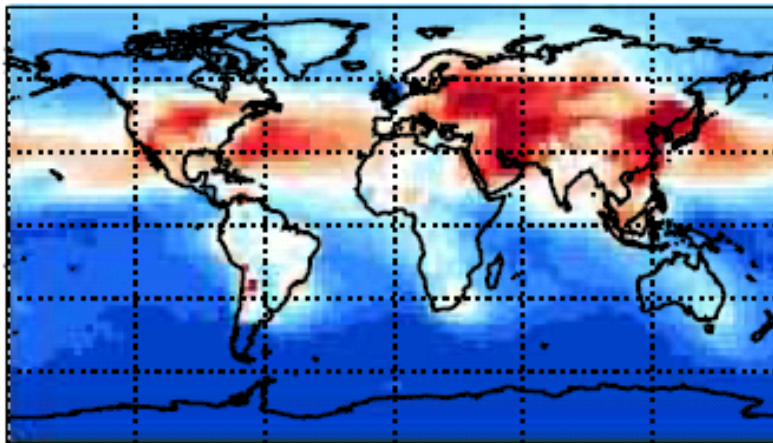
Global increase in CDNC 16% in 1850 and 14% in 2000.
However, there are large regional differences!

Cloud droplet number and cloud albedo

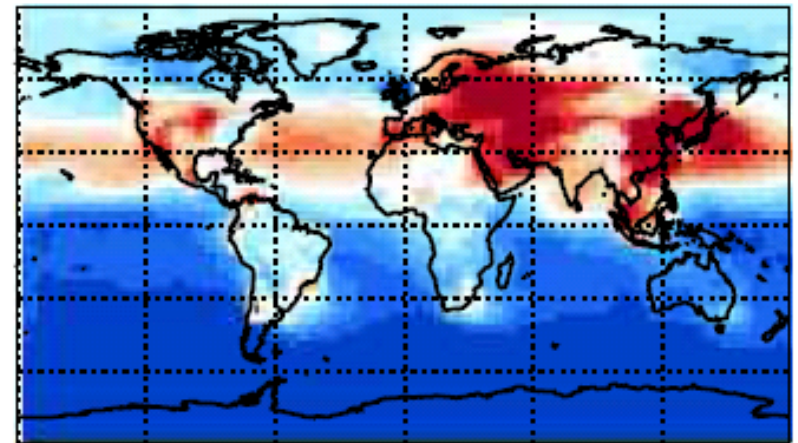
According to Twomey, the change in cloud droplet number results in a change in cloud albedo R_c

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

dRC with BLPF, $R_c=0.35$, $w=0.4$

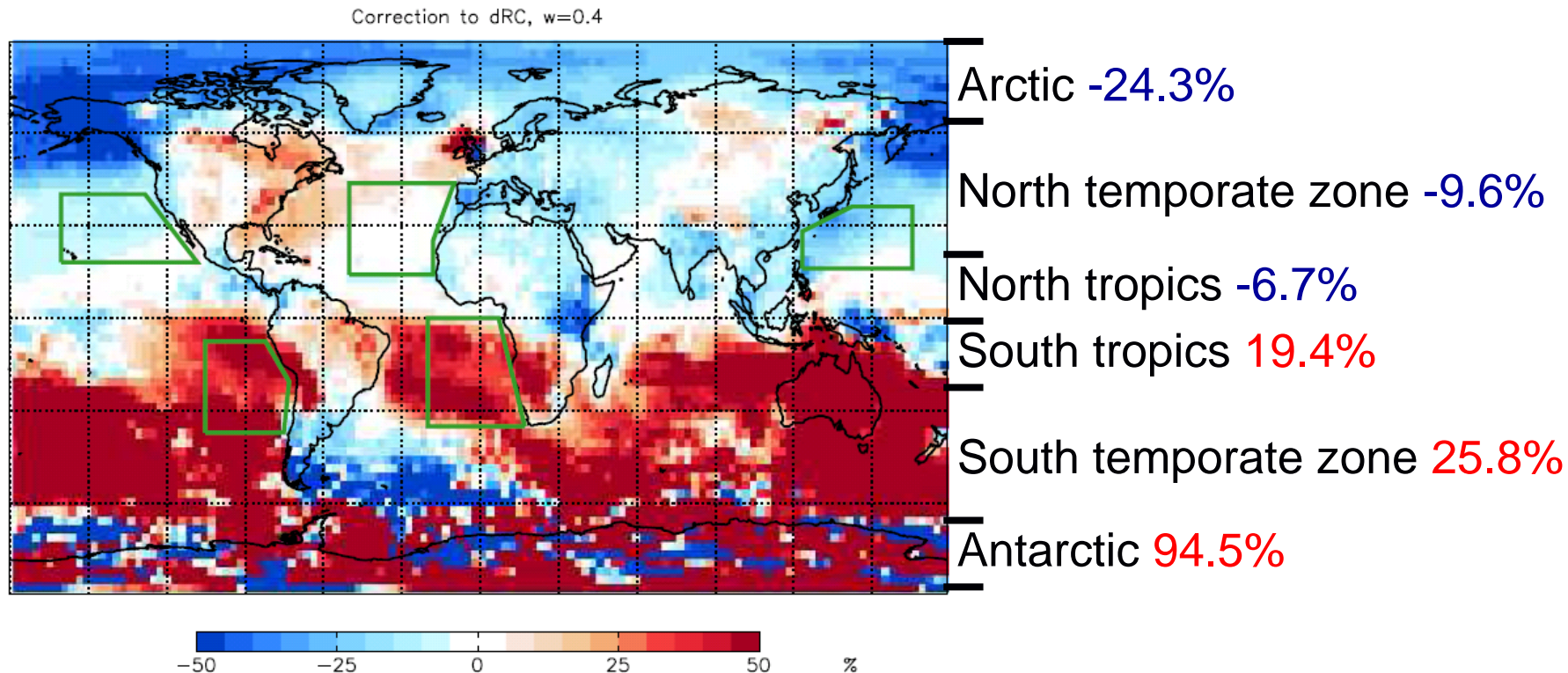


dRC without BLPF, $R_c=0.35$, $w=0.4$



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

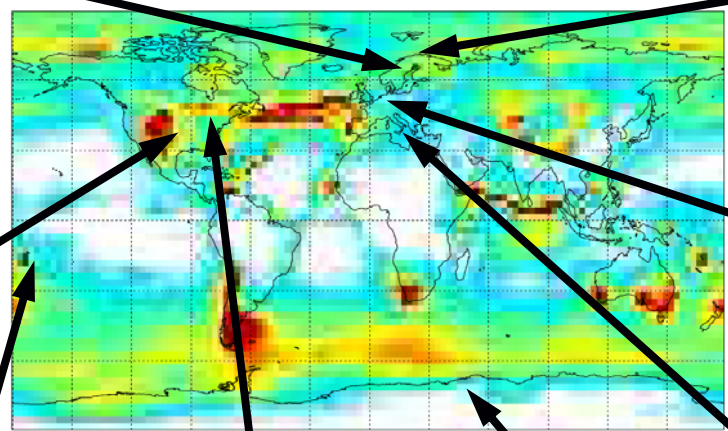
The resulting change in cloud albedo with when particle formation is included:



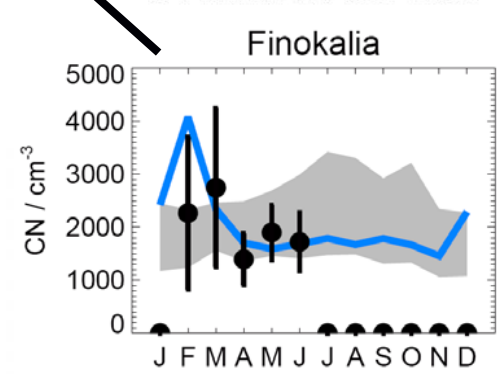
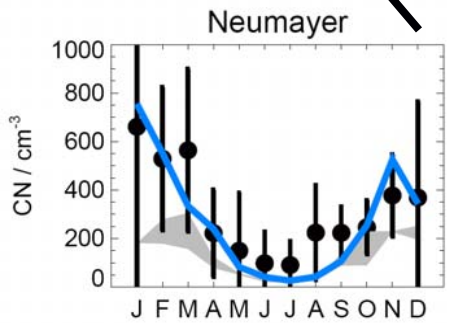
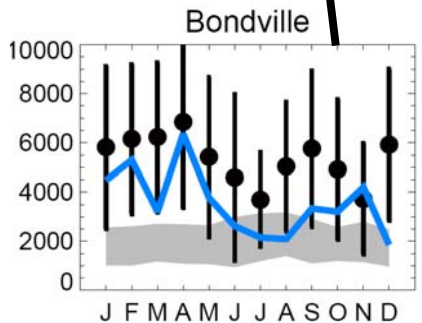
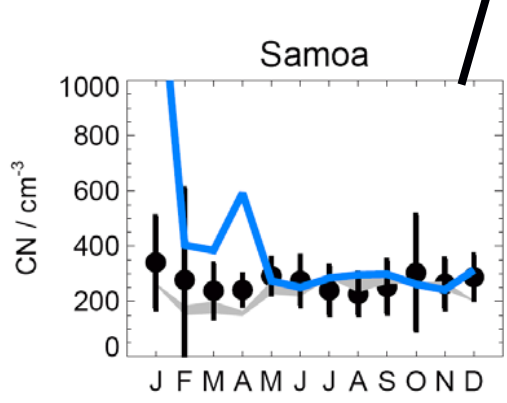
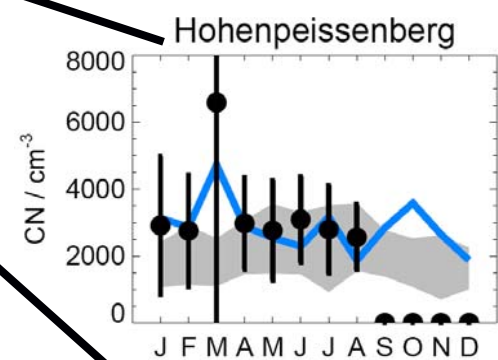
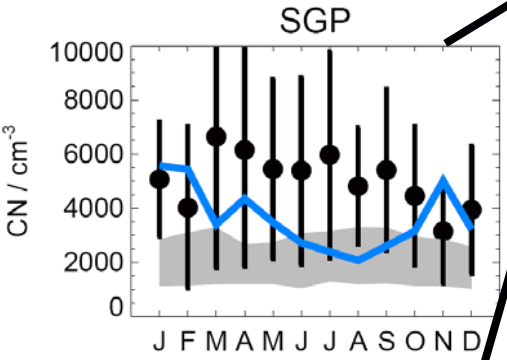
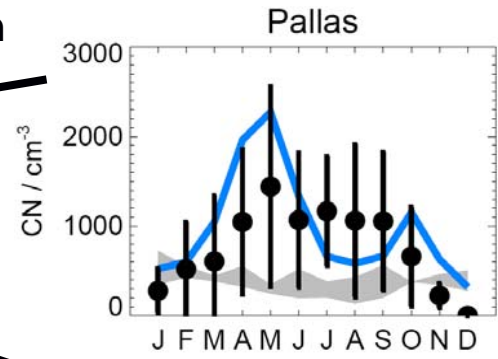
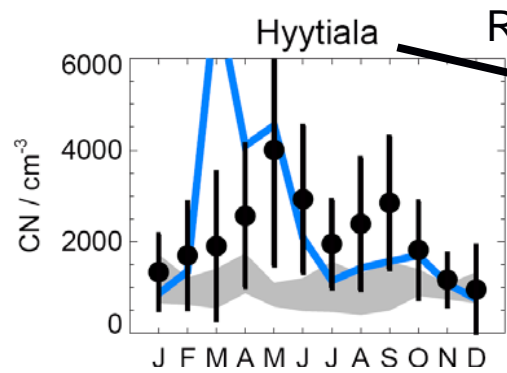
- Global average difference only -3%
- However, a large north-south contrast in results

GLOMAP CN being evaluated against observations at GAW and ARM sites

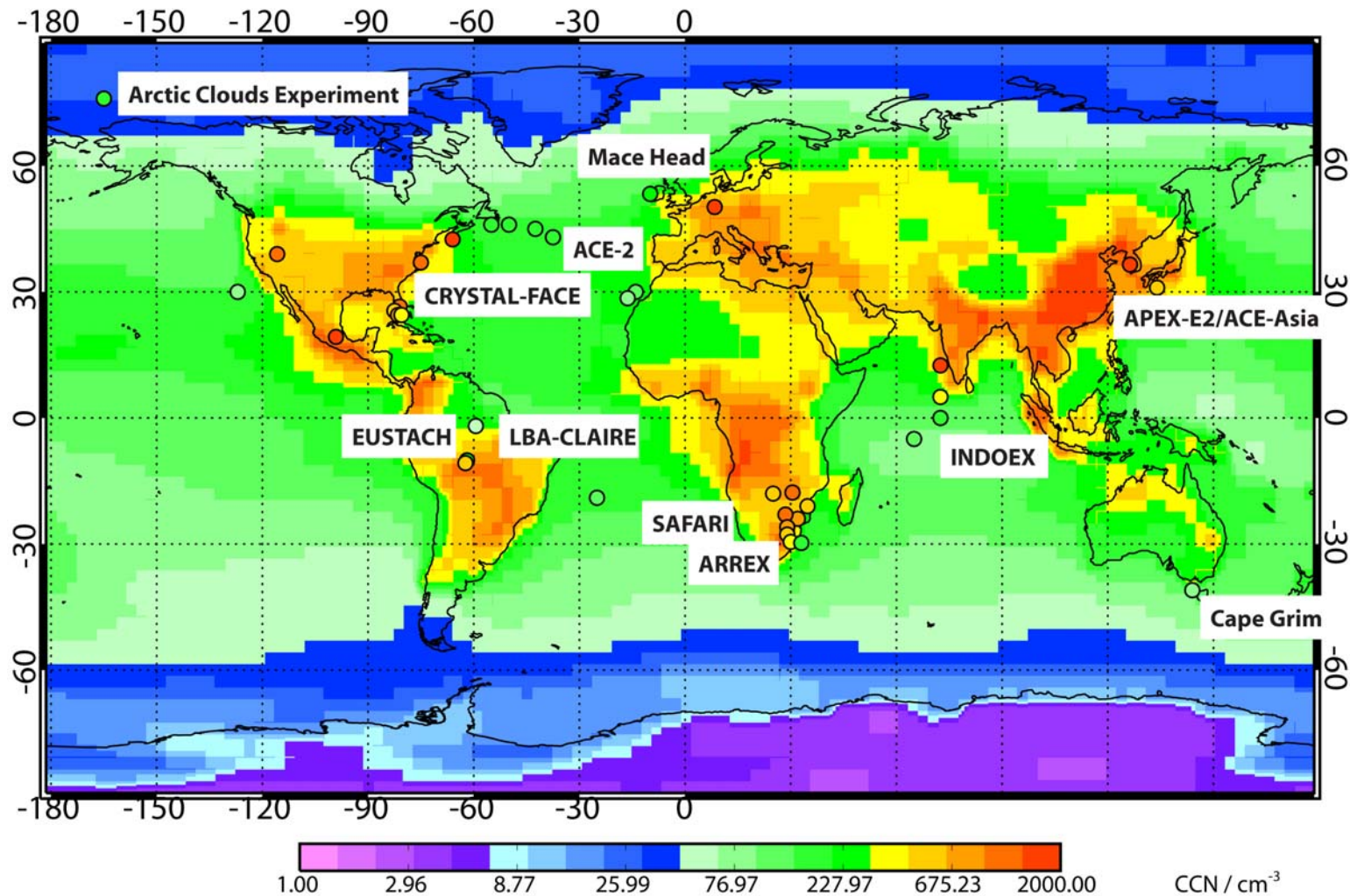
Ratio of simulated annual mean CN concentration with BL new particle formation: without



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



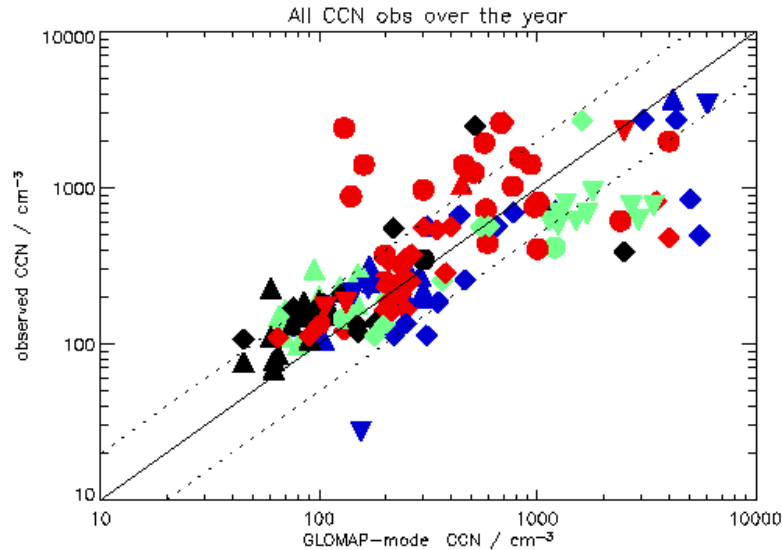
GLOMAP CCN being evaluated against a range of worldwide observations



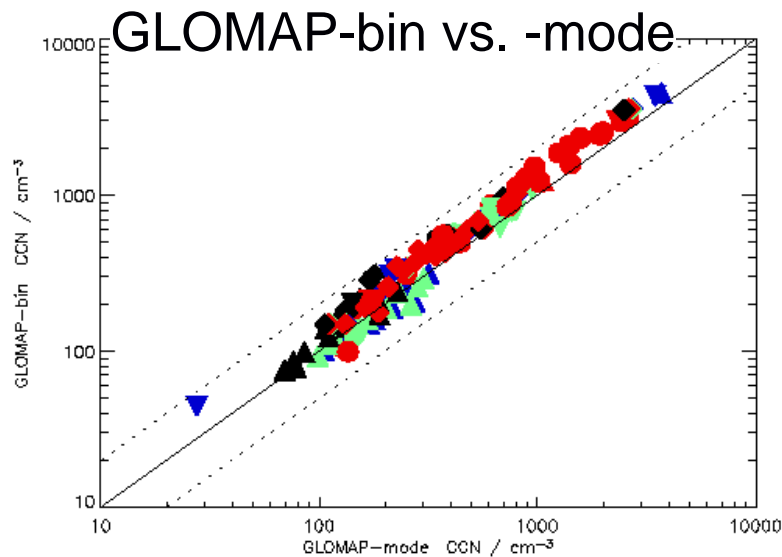
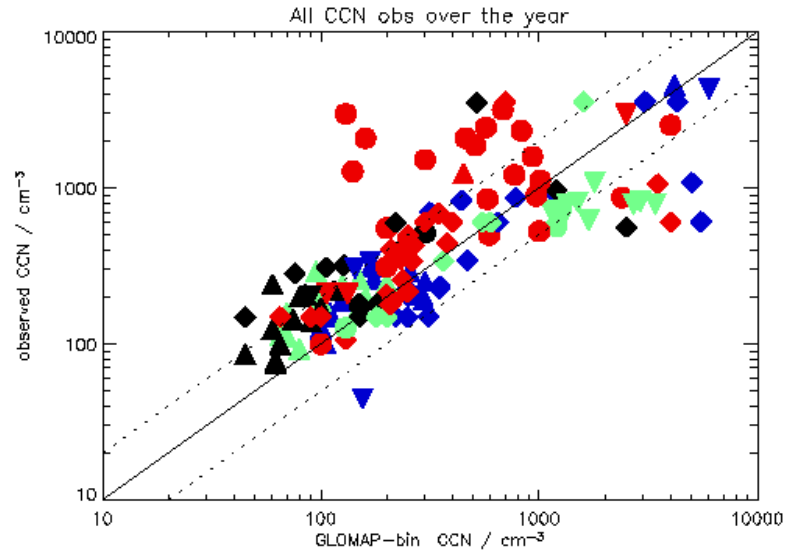
Note: map shows CCN at 0.2% supersaturations.
Coloured circles show observations at range of supersaturations

GLOMAP CCN being evaluated against a range of worldwide observations

GLOMAP-mode vs. observations



GLOMAP-bin vs. observations

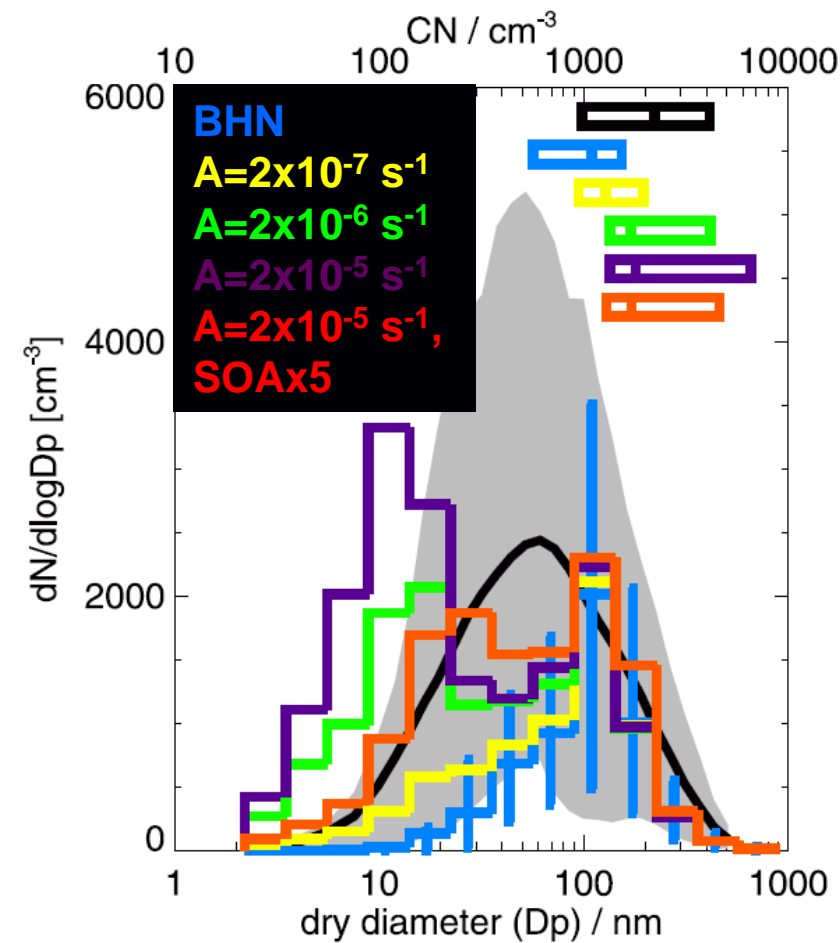


- SS < 0.25% (black)
- SS < 0.5% (red)
- SS < 0.75% (green)
- SS > 0.75% (blue)
- 0-90E (circle)
- 90E-180E (triangle up)
- 180W-90W (triangle down)
- 90W-0W (diamond)

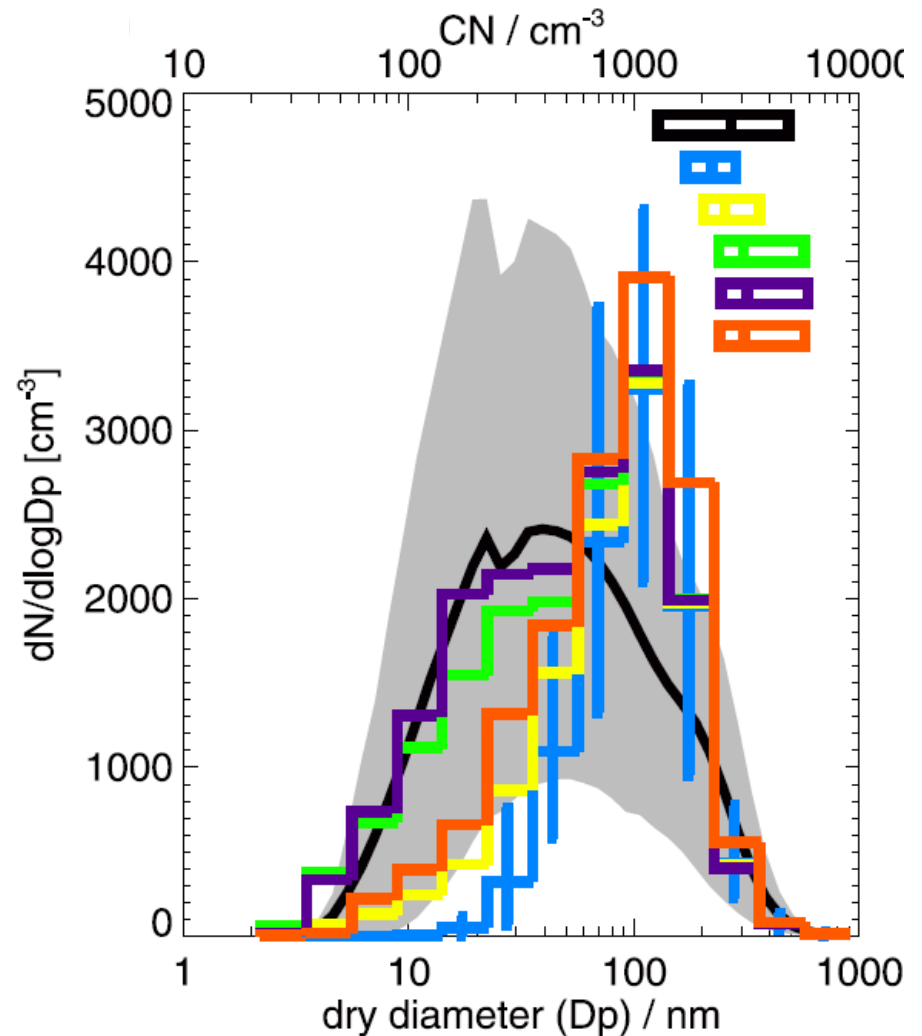
Here, model CCN are calculated using threshold diameter derived from instrument supersaturation (Kohler theory)

GLOMAP size distributions being evaluated against European DMPS observations

Hyytiala, Finland



Hohenpeissenberg, Germany





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