



Understanding the sources of uncertainty in global aerosol

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With input from Graham Mann Dom Spracklen, Philip Stier,
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NERC AEROS project (2010-2013)

Aerosol model **R**obustness and **S**ensitivity study for improved climate and air quality prediction (Leeds and Oxford)

Robustness: A prediction that doesn't change fundamentally in the face of model uncertainty

Use new statistical tools and uncertainty analysis techniques to quantify the sources of global aerosol model uncertainty at the process level.

Follow-up project NERC 2013-2016

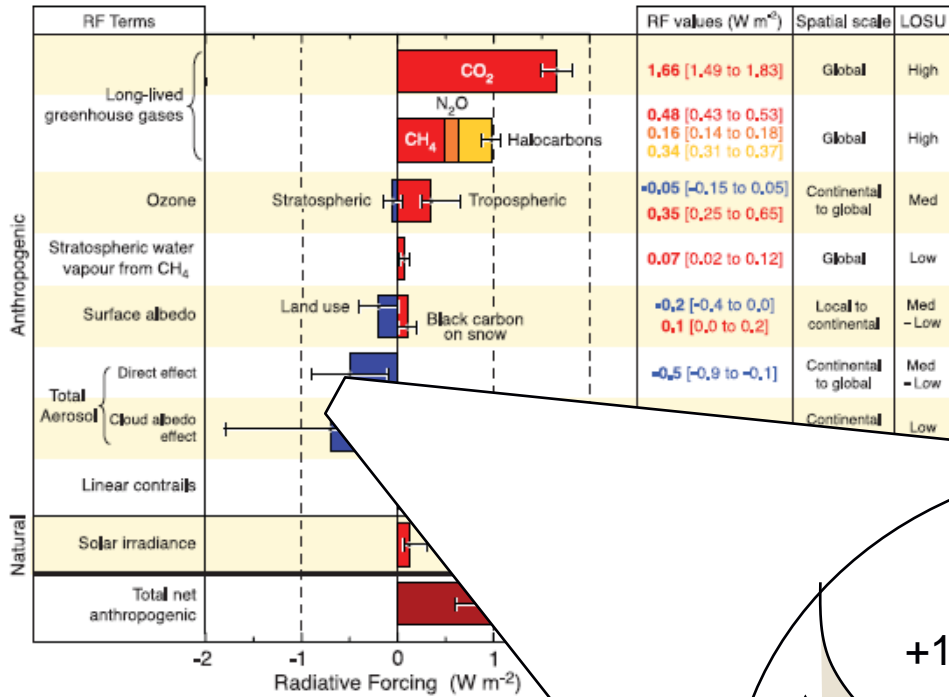
Global **A**erosol **S**ynthesis and **S**cience project (GASSP!)

Leeds, Oxford, Manchester + 10 data partners + Met Office

What do we mean by “uncertainty” of the model?



RADIATIVE FORCING COMPONENTS



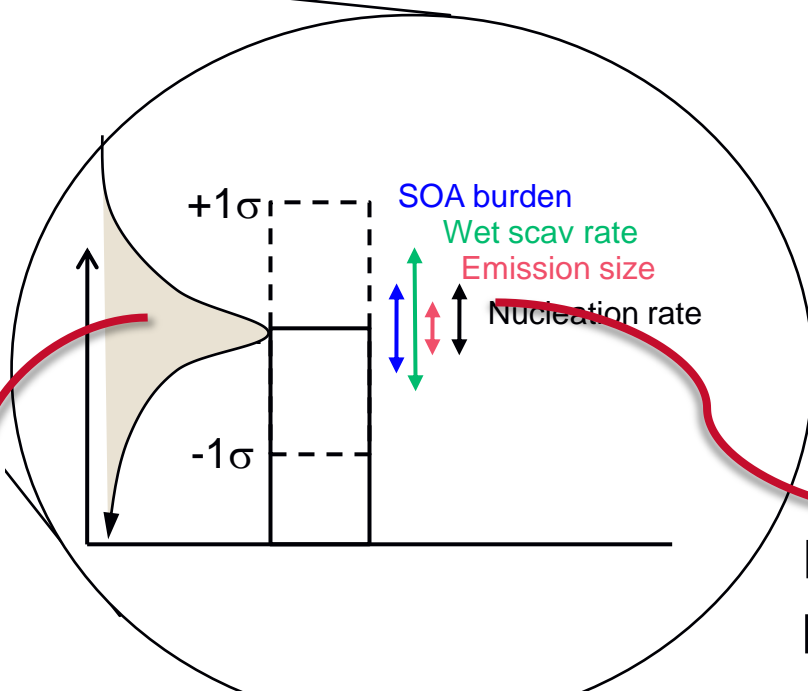
Model Intercomparison Projects focus on *diversity*

Uncertainty attributable to processes would be a valuable addition

Observations

Best model

“Real” discrepancy

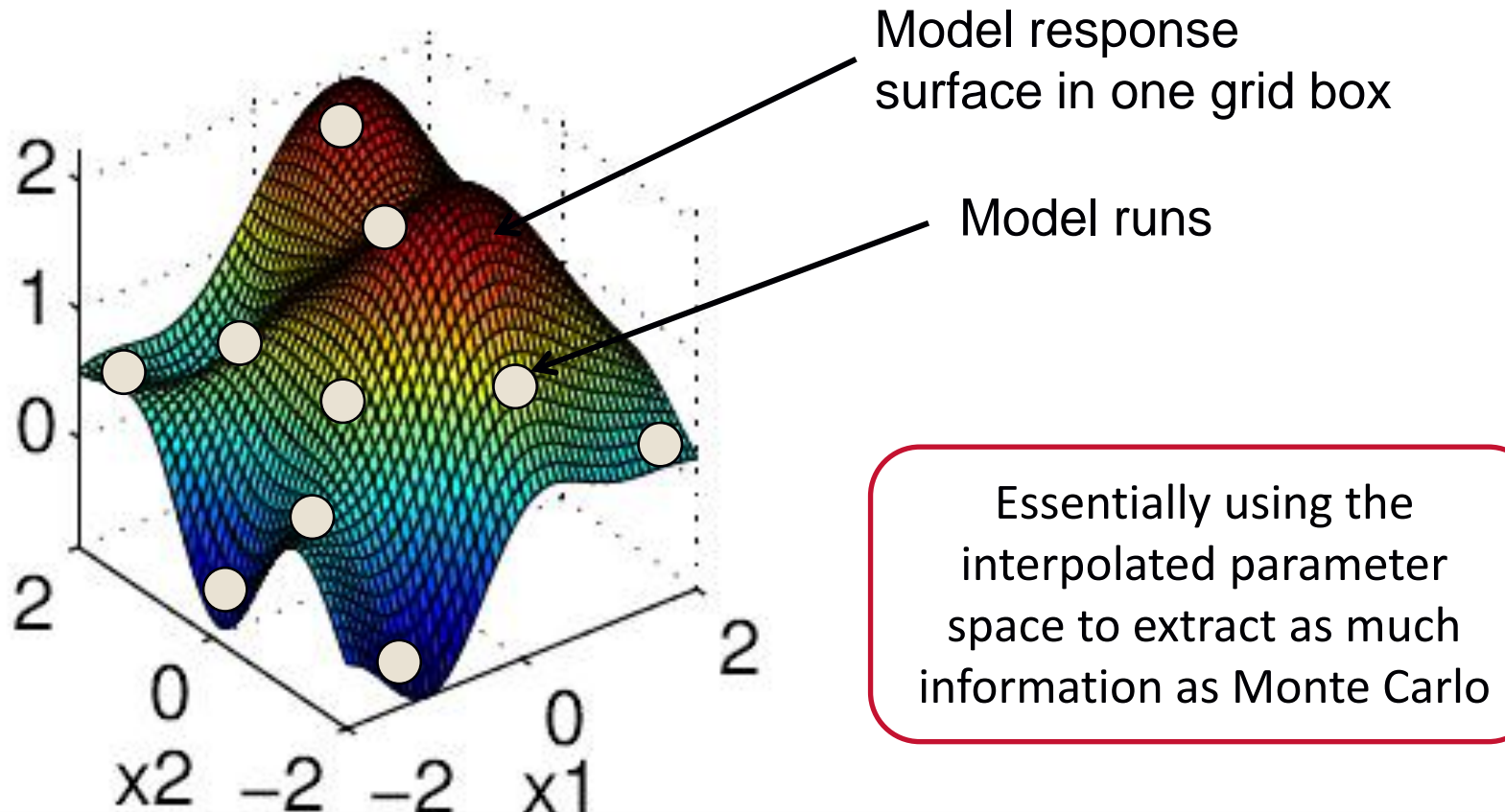


Development path

Using an emulator to do Monte Carlo on the cheap



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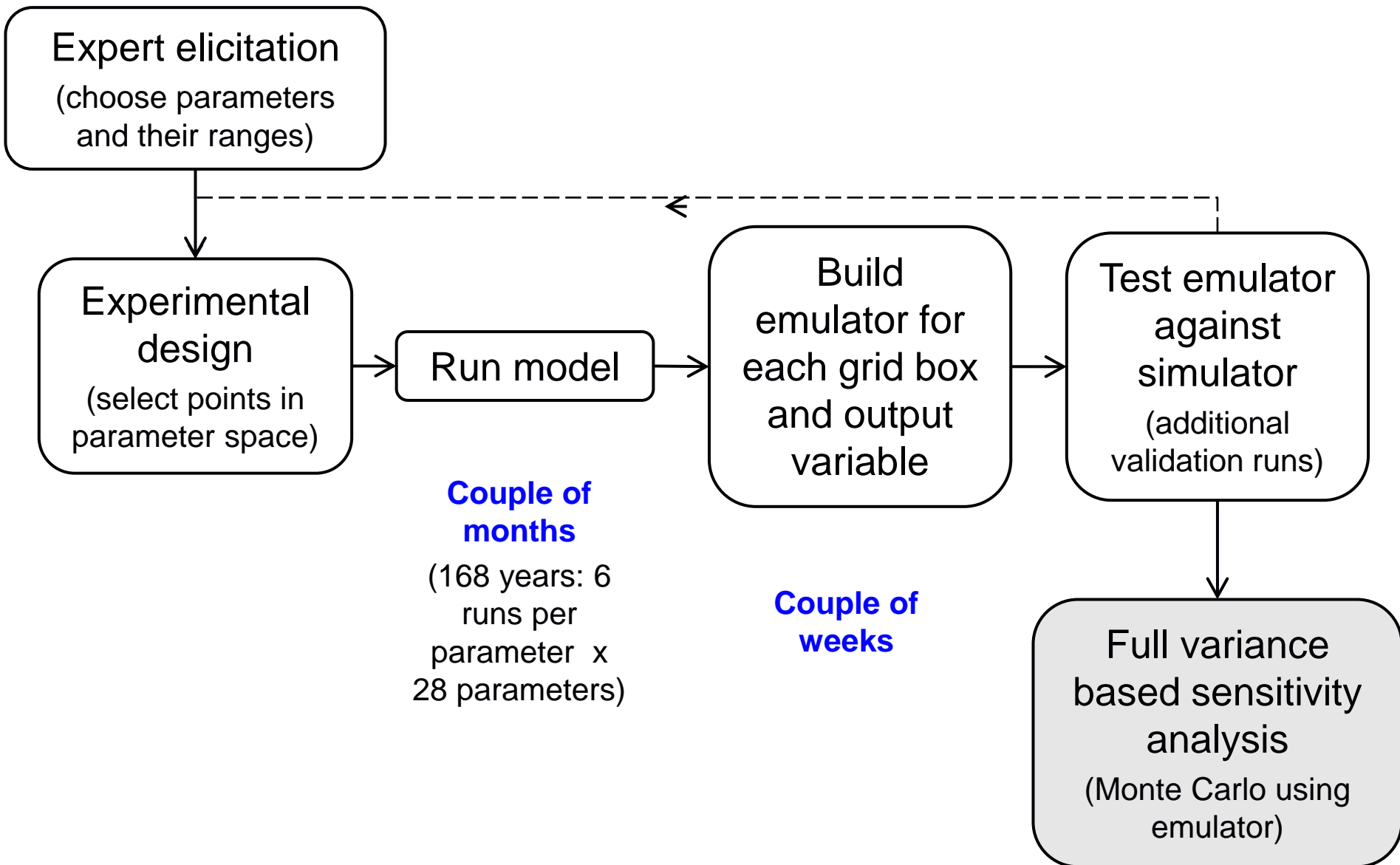


Oakley, J. and O'Hagan, A.: Probabilistic sensitivity analysis of complex models: a Bayesian approach, *J. Roy. Stat. Soc. B*, 66, 751–769, 2004.

Lee, L.A. et al., Emulation of a complex global aerosol model to quantify sensitivity to uncertain parameters, *ACP* 2011.



The process of model emulation



The Global Model of Aerosol Processes (GLOMAP-mode)



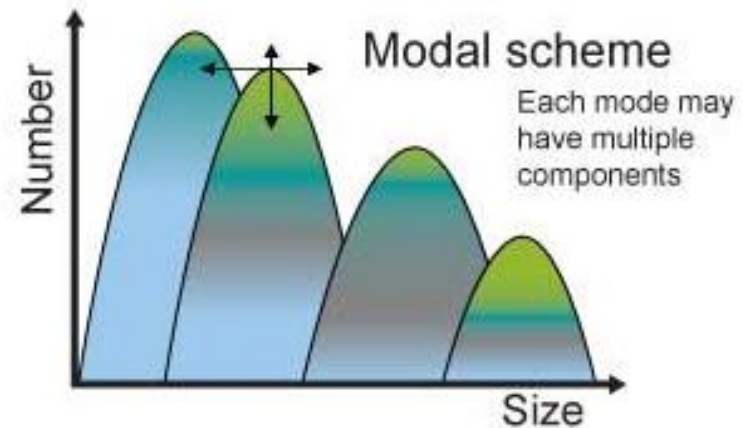
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Global aerosol microphysics model within a 3D offline CTM forced by ECMWF winds

- Usually run at T42L31 ($2.8^\circ \times 2.8^\circ$) resolution
- Modal scheme: 7 log-normal modes
- Aerosol transport, new particle formation, growth by coagulation, condensation, cloud processing.
- Wet and dry deposition of gases & aerosol particles
- Chemistry can be driven by offline oxidants
- 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/biofuel/biomass)

Not a GCM, so no aerosol feedbacks

Model description in Mann et al. (GMD, 2010)





Perturbed parameters 1/2

Parameter	Lower	Upper
BCOC mass emission rate (fossil fuel)	0.5	2.0
BCOC mass emission rate (biomass burning)	0.25	4.0
BCOC mass emission rate (biofuel)	0.25	4.0
Sea spray mass flux (coarse/acc)	0.2x	5.0x
SO ₂ emission flux (anthropogenic)	0.6x	1.5x
SO ₂ emission flux (volcanic)	0.5x	2.0x
Biogenic monoterpene production of SOA	5 Tg/a	360Tg/a
Anthropogenic VOC production of SOA	3Tg/a	160Tg/a
DMS mass flux	0.5x	3.0x
BCOC mode diameter (fossil fuel)	30 nm	80 nm
BCOC mode diameter (biomass burning)	50 nm	200 nm
BCOC mode diameter (biofuel)	50 nm	200 nm
Subgrid conversion of SO ₂ to SO ₄ ("primary SO ₄ ")	0%	1%
Mode diameter of "primary SO ₄ "	20 nm	100 nm

Particle and precursor gas emission rates

Properties of emitted particles



Perturbed parameters 2/2

Parameter	Lower	Upper
BL nucleation rate $k[\text{H}_2\text{SO}_4]$	1E-10	2E-04
FT nucleation rate (BHN)	x0.01	X10
Ageing "rate" from insol to sol (monolayer)	0.3	5
Modal width (accumulation)	1.2	1.8
Modal width (Aitken)	1.2	1.8
Mode separation diameter (nucleation/Aitken)	9nm	20nm
Mode separation diameter (Aitken/accumulation)	x1.5	x3

Microphysical rates

Model "structural choices"

Cloud drop activation dry diameter	30	100
Reaction $\text{SO}_2 + \text{O}_3$ in cloud water (clean)	pH=4	pH=6.5
Reaction $\text{SO}_2 + \text{O}_3$ in cloud water (polluted)	pH=3.5	pH=5

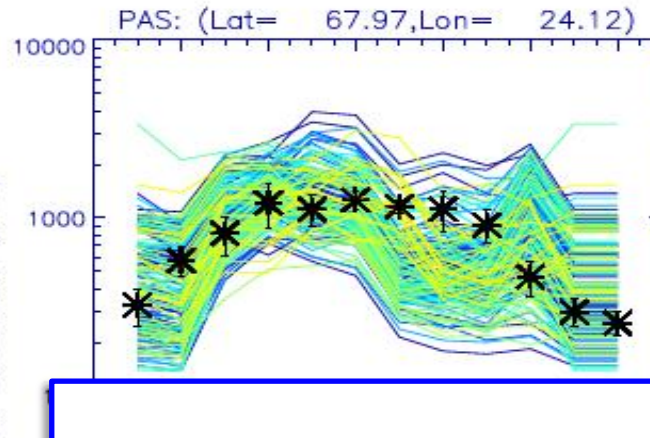
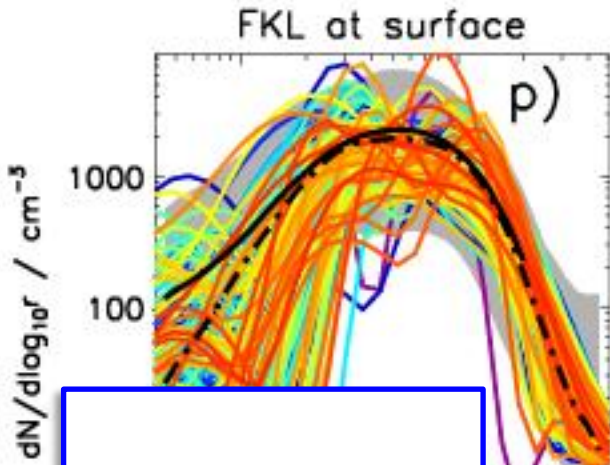
Cloud processing

Nucleation scavenging dry D (above activation)	0	100
Nucleation scavenging fraction ($T > -15\text{C}$)	0.2	0.99
Dry deposition velocity (Aitken)	x0.5	X2.0
Dry deposition velocity (accumulation)	X0.1	X10.0

Dry and wet deposition



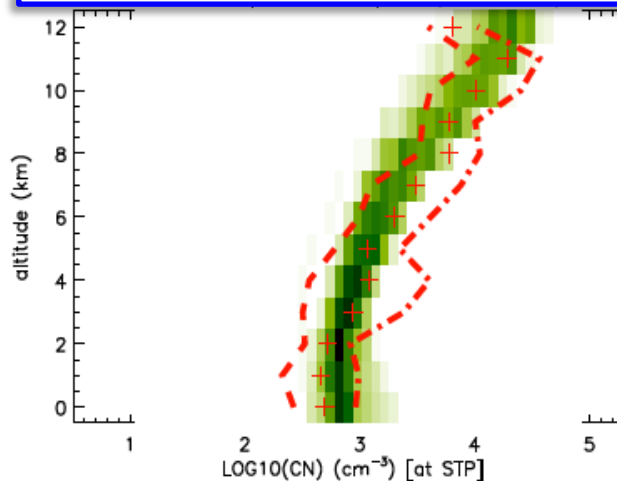
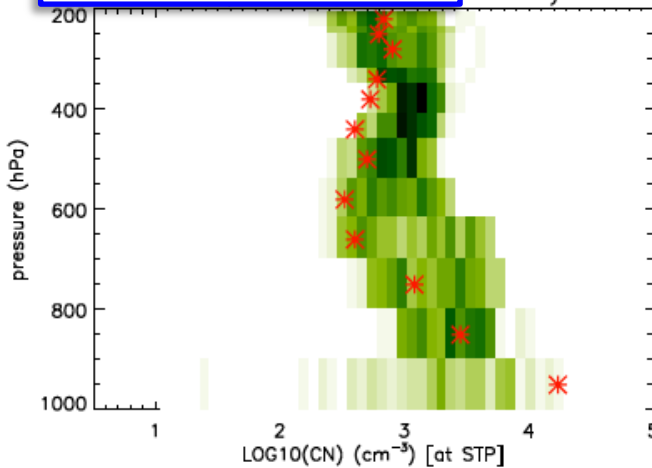
Raw model results in each grid box



Emulator

(each quantity in each grid box)

Continuous output across whole of parameter space



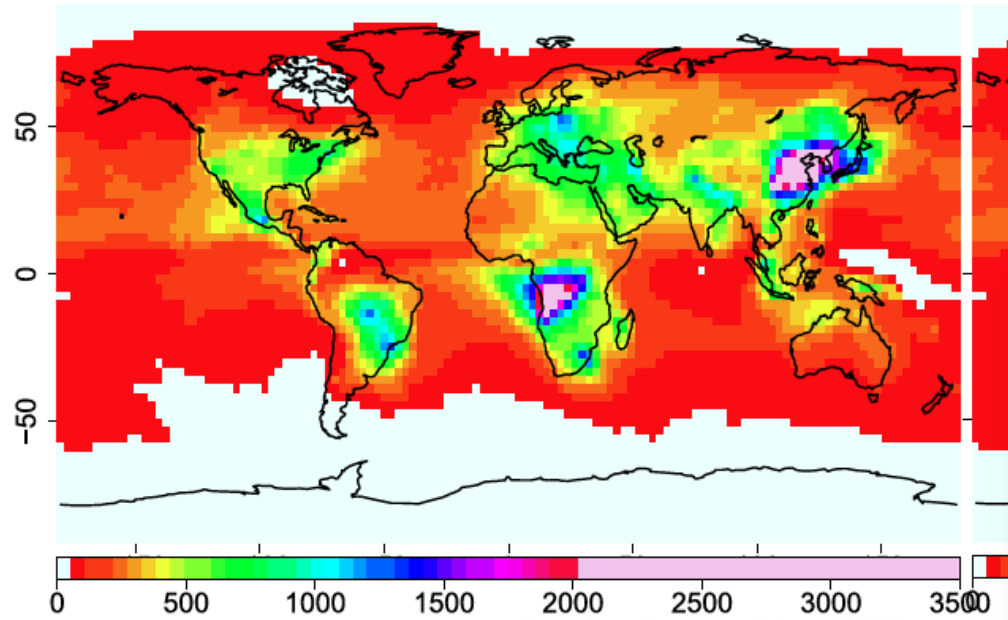
168 annual model runs covering 28-dimensional parameter space

Far from sufficient for Monte Carlo

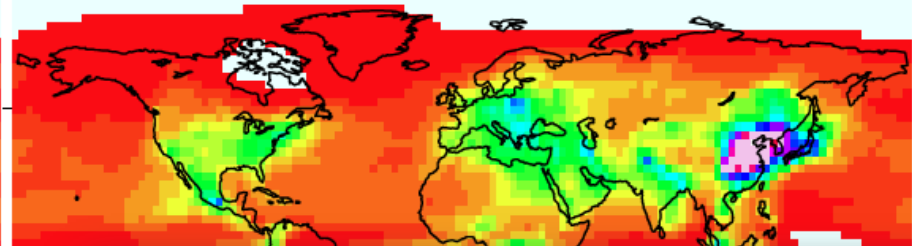


Validate the emulator

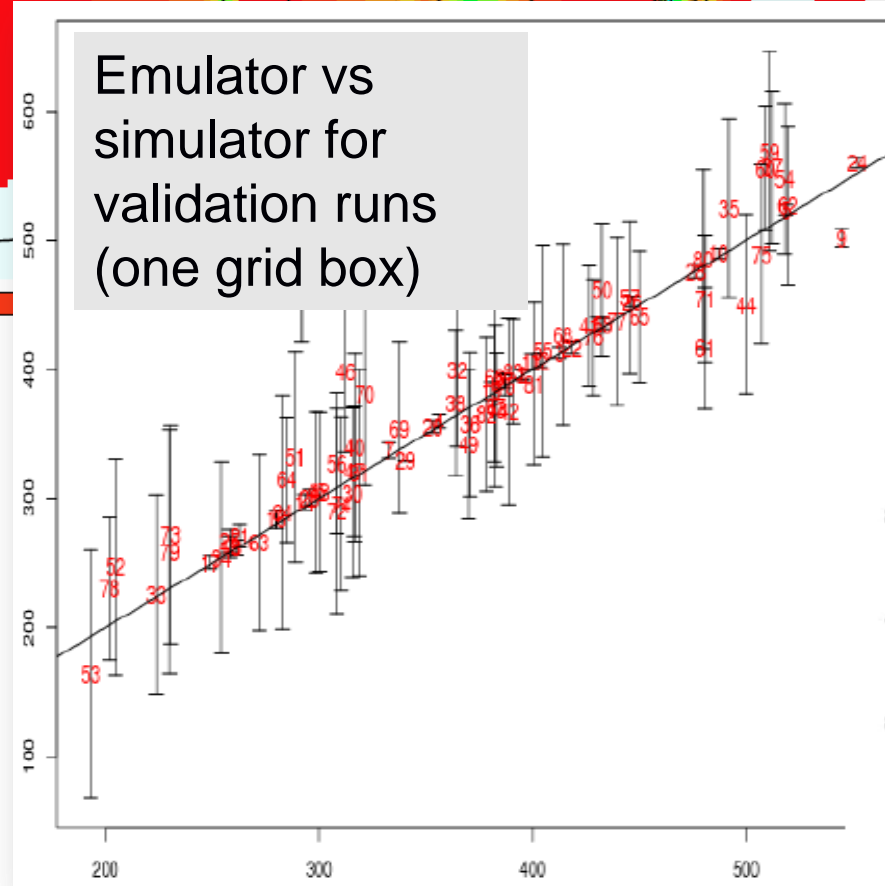
Emulator estimated CCN (cm⁻³)



GLOMAP estimated CCN (cm⁻³)



Emulator vs simulator for validation runs (one grid box)



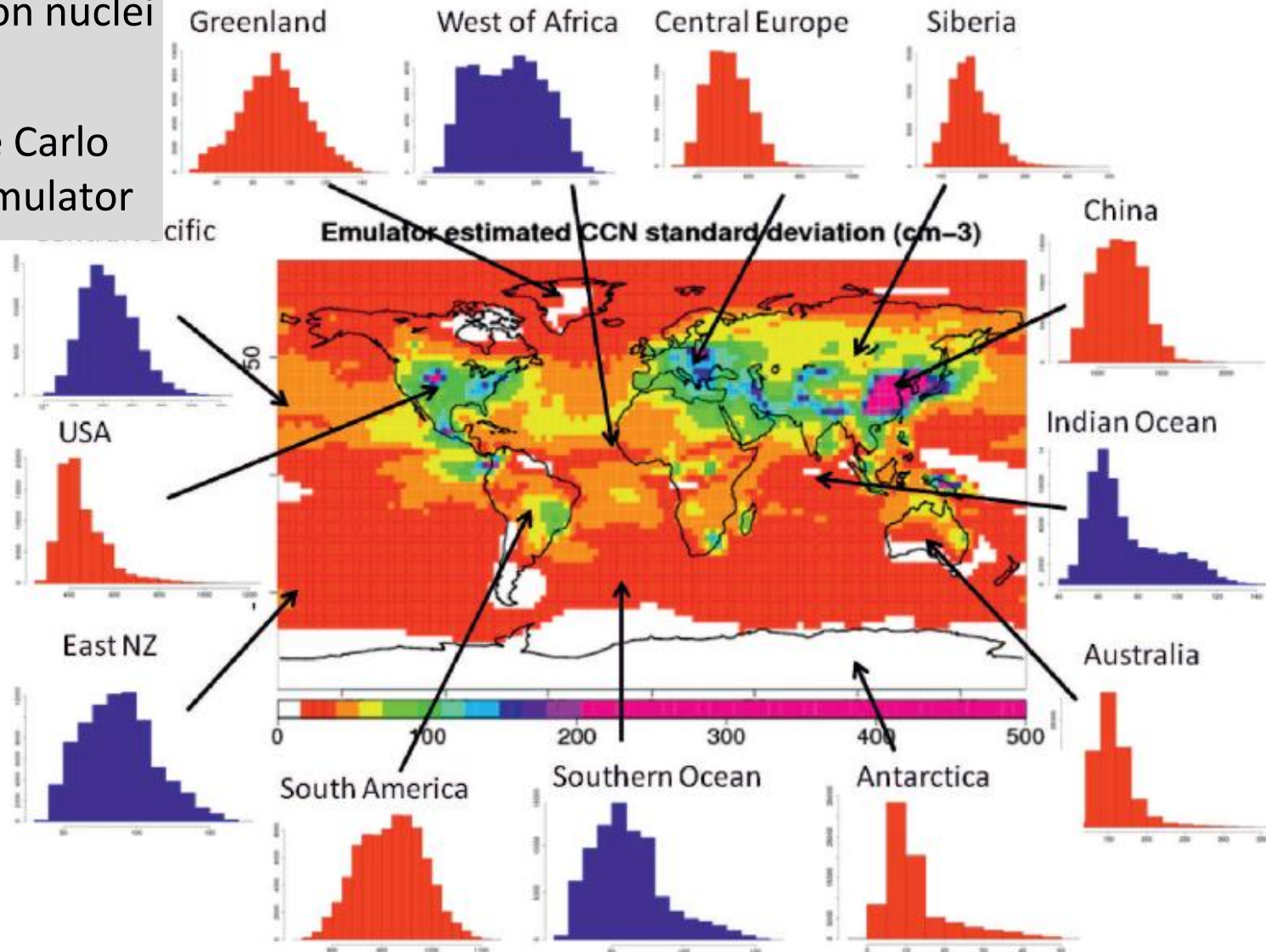
How uncertain is global aerosol?

PDFs of CCN concentration in every grid box



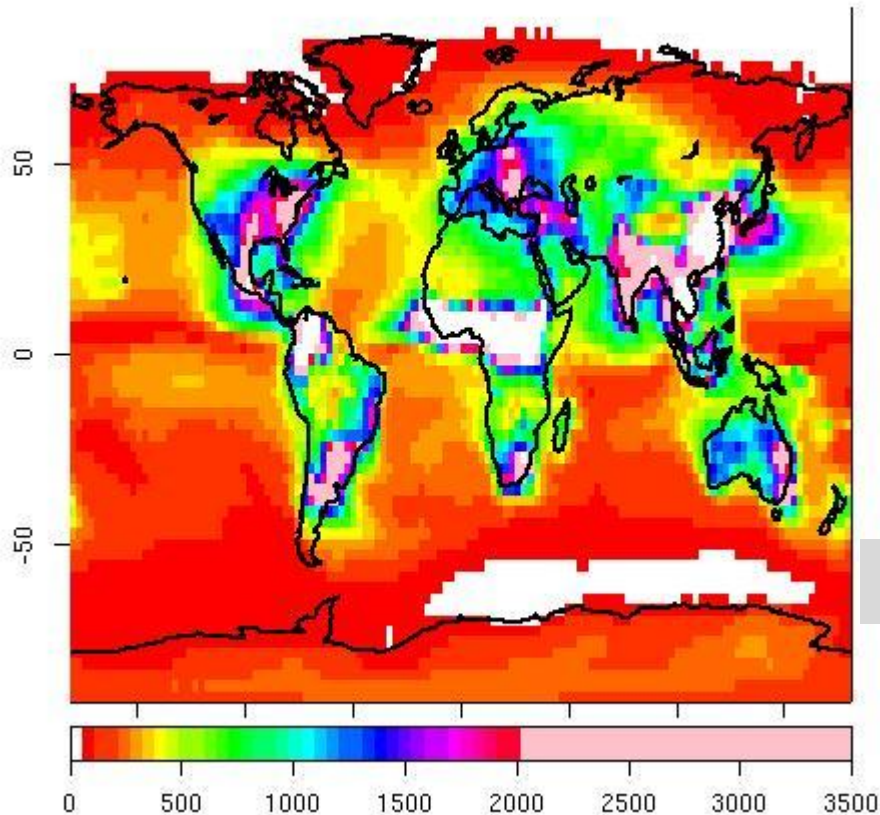
Cloud condensation nuclei (CCN)

Based on a Monte Carlo sampling of the emulator



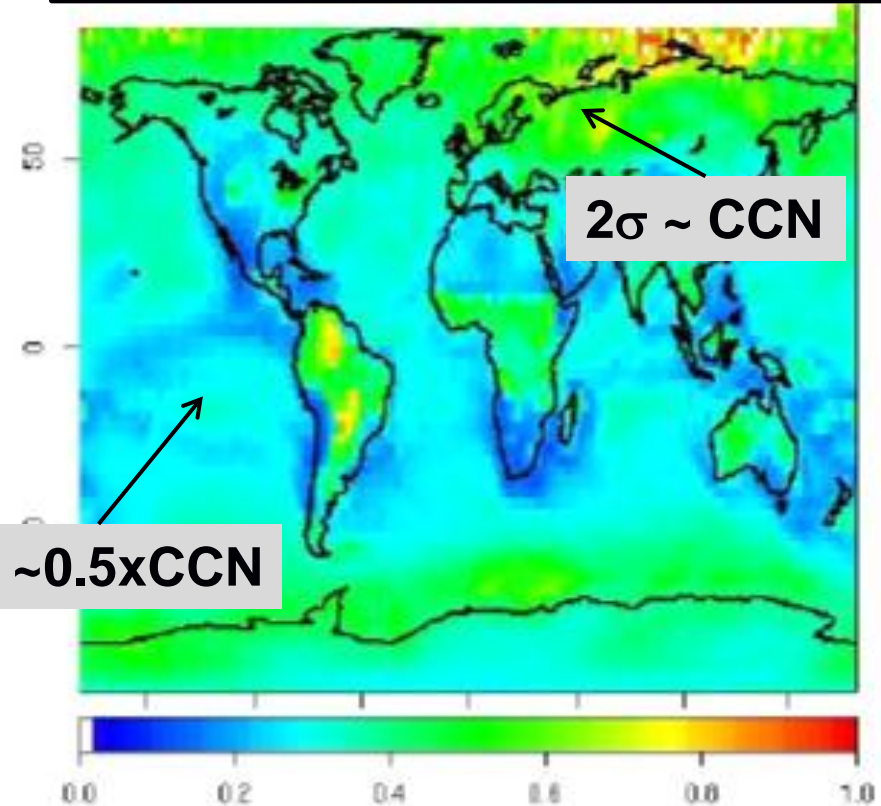
CCN and associated uncertainty

Emulator estimated CCN



CCN concentration / cm^{-3}

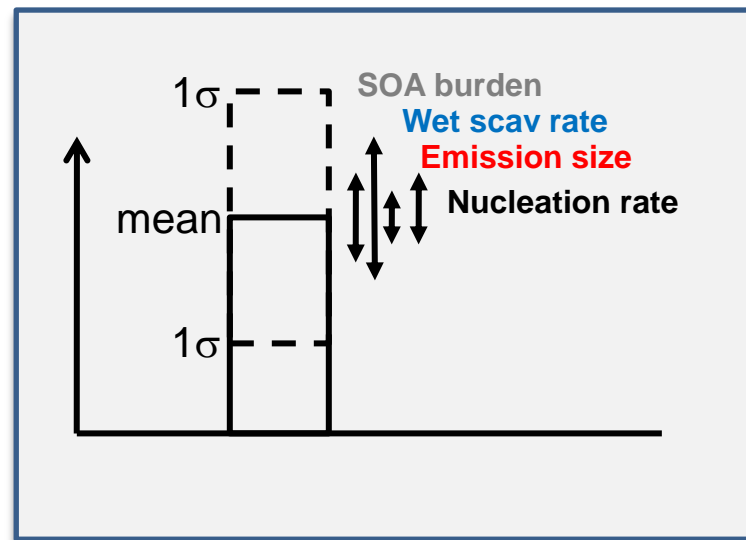
Emulator estimated coefficient of variation (σ/CCN)



(σ/CCN)

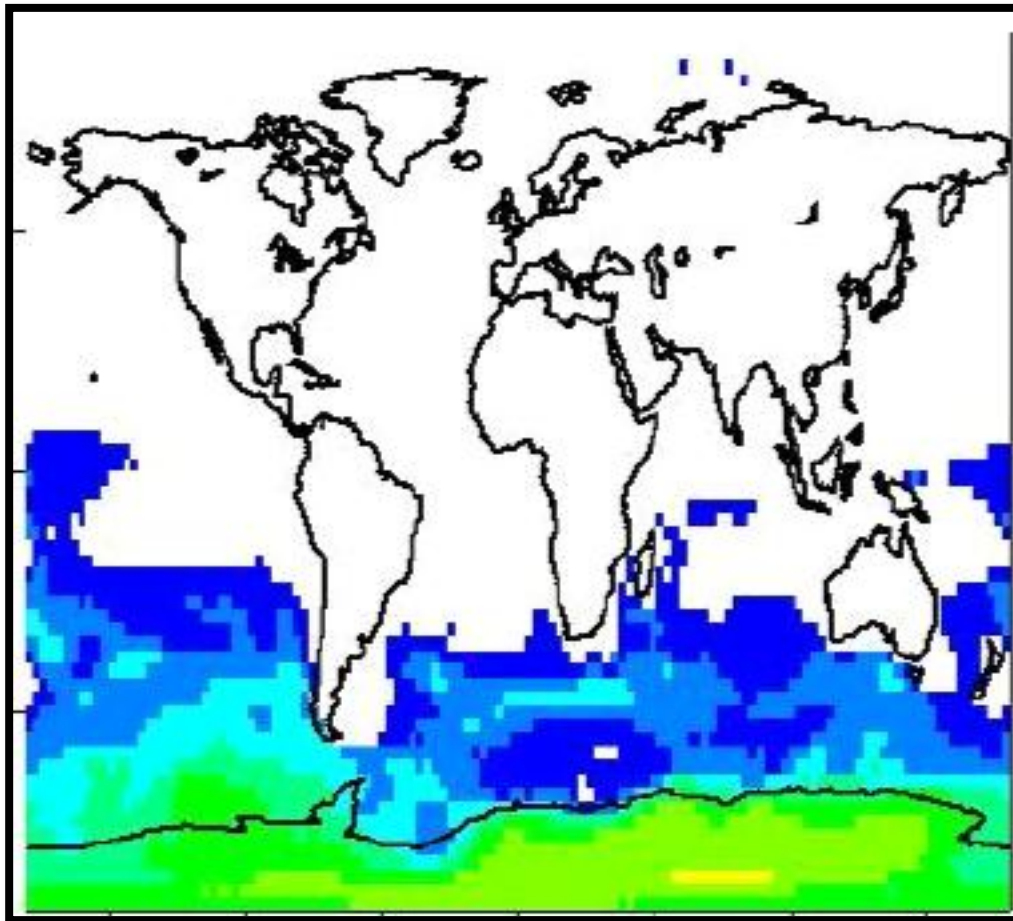
January

How much do the different parameters contribute to the uncertainty?

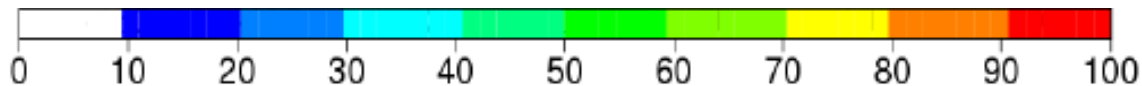


Contributions to CCN variance

Percent variance due to DMS flux



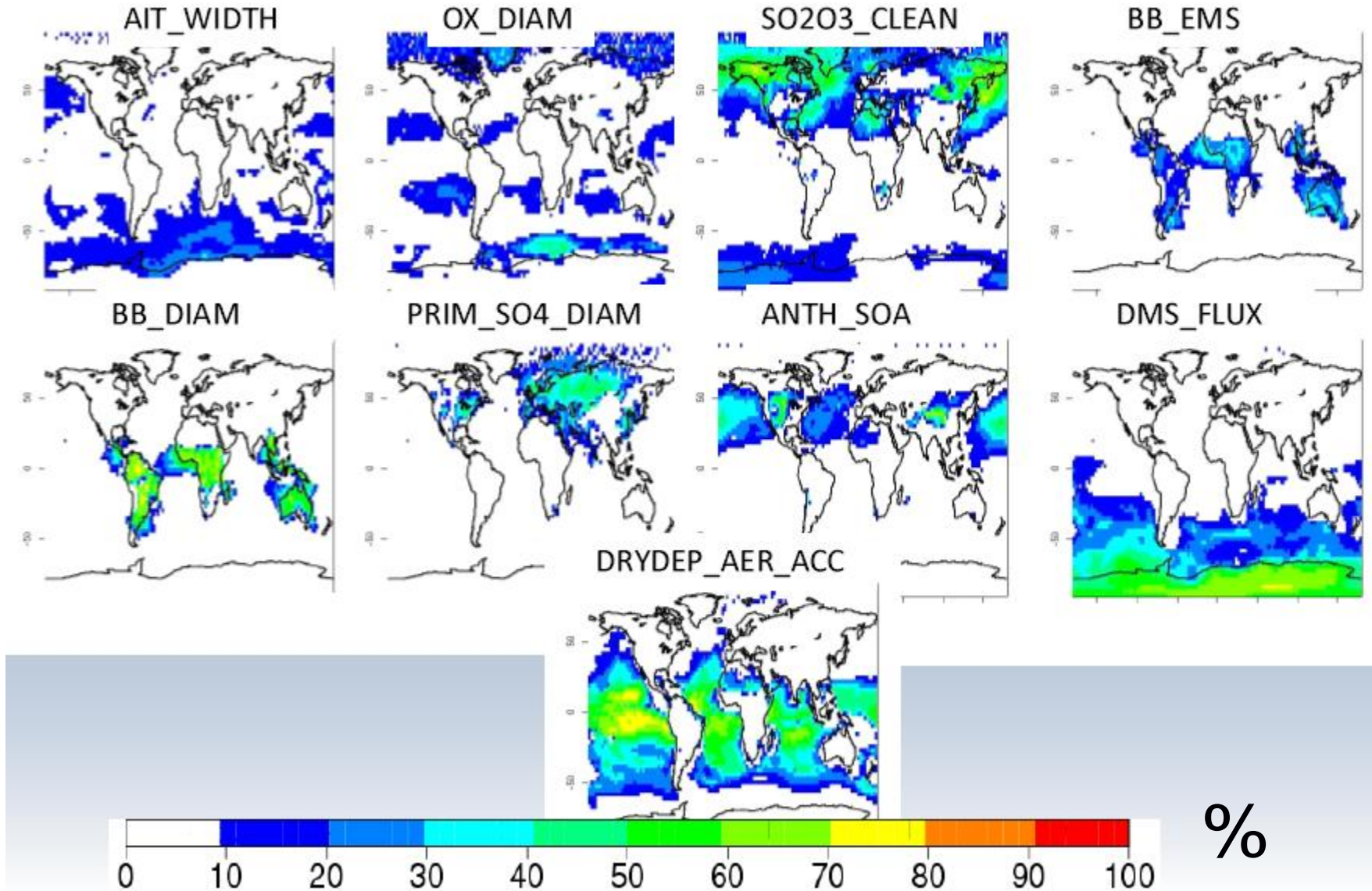
Meaning: If we knew the *DMS flux* precisely, this is the reduction in CCN variance we would achieve



Contributions to CCN variance January



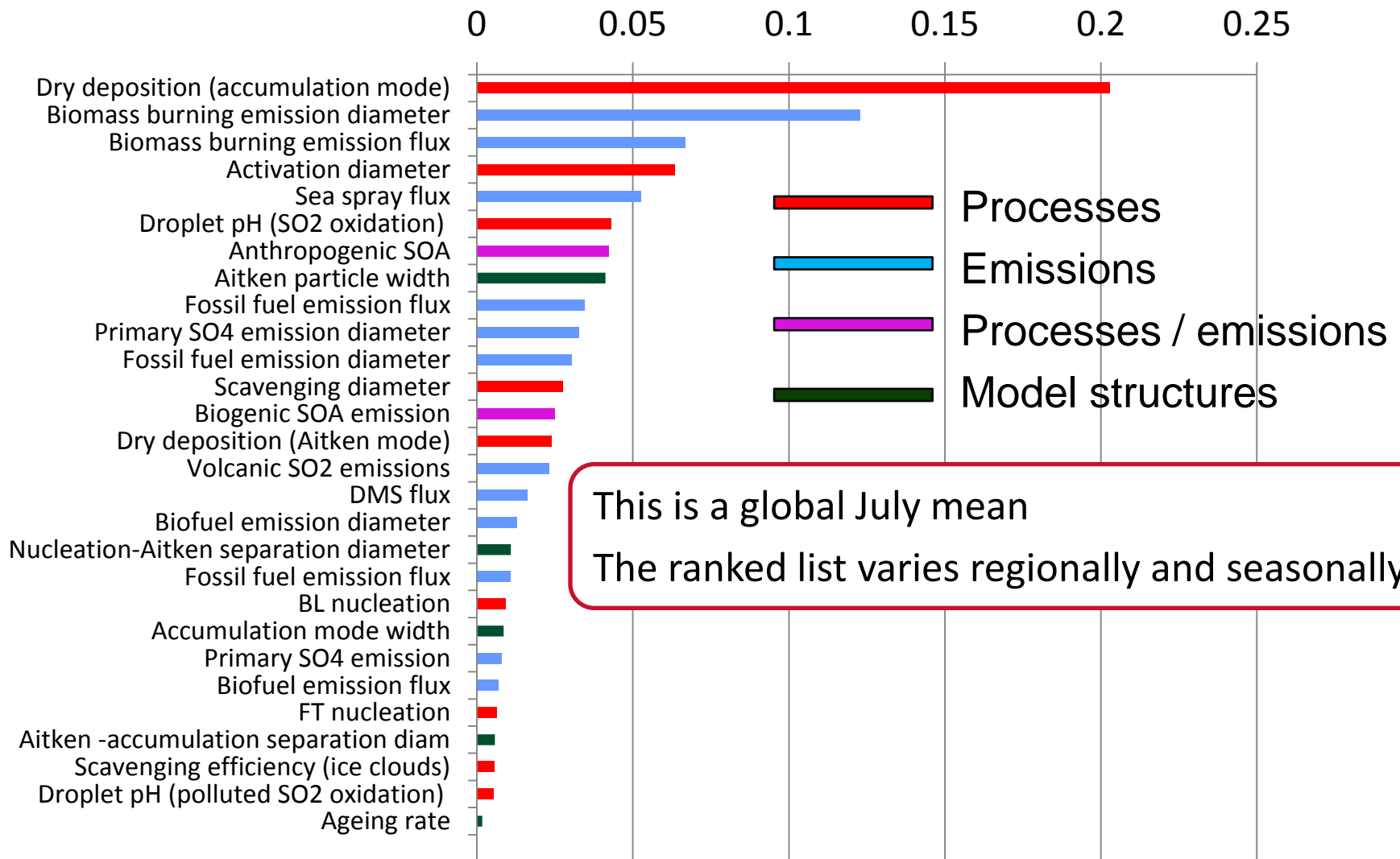
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Ranked CCN uncertainty



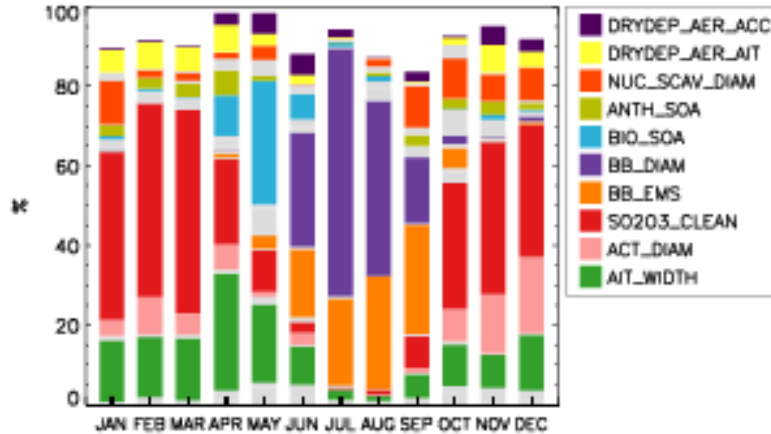
Global mean of grid-box fractional variance



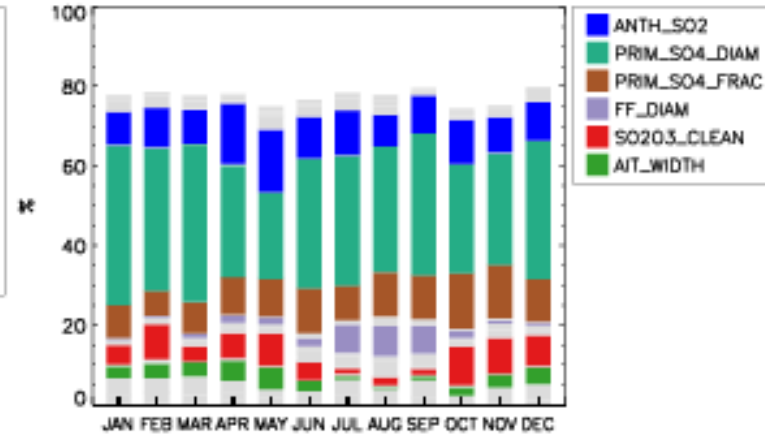


Regional/seasonal sensitivities

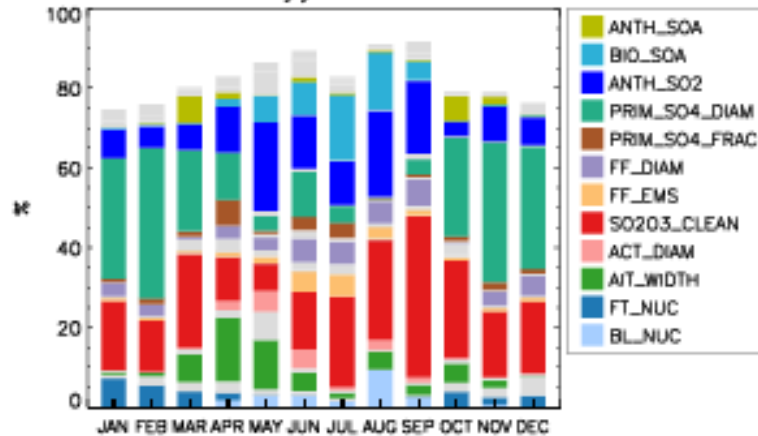
Barrow



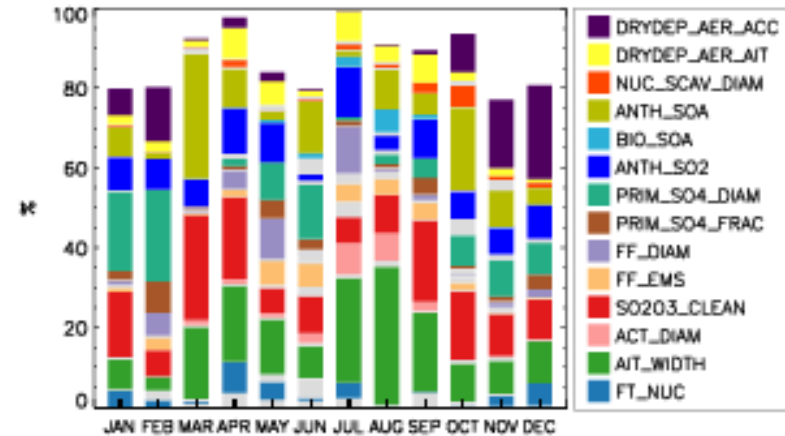
Bondville



Hyytiälä



Mace Head





Indirect forcing

Forcing depends on the ***change*** in aerosol properties between pre-industrial and present day

Some factors may “cancel out”

Forcing depends non-linearly on CCN

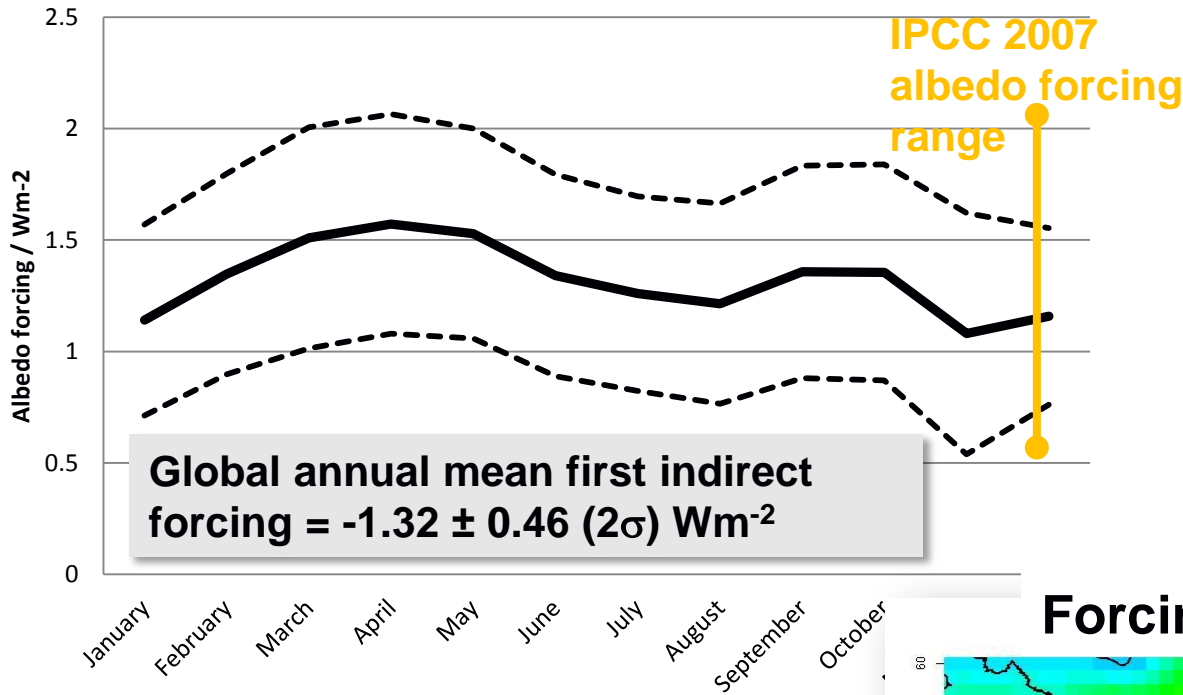
Repeat the 168 runs with pre-industrial emissions but same meteorology

Emulate change in effective radius → forcing



Uncertainty in indirect forcing

Expected forcing +/- 2*standard deviation

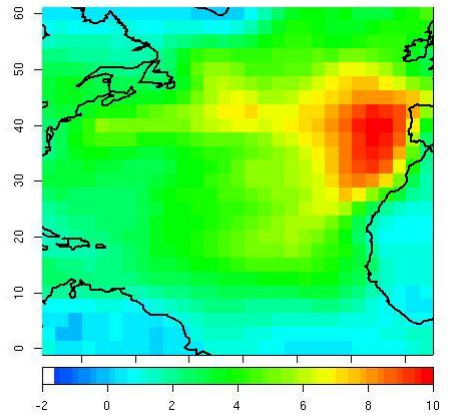


Global annual mean first indirect forcing = $-1.32 \pm 0.46 (2\sigma) \text{ Wm}^{-2}$

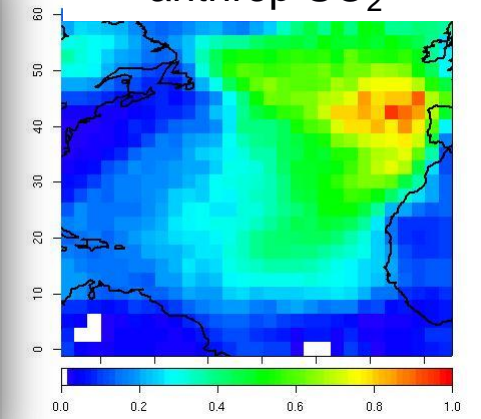
Top 5 contributions to albedo forcing uncertainty (variance)

- 24.3% Degassing volcanic SO₂
- 18.1% Anthropogenic SO₂
- 12.3% DMS flux
- 11.2% Anthropogenic SOA
- 10.8% Accumulation mode width

Forcing



Uncertainty due to anthrop SO₂





Future work

Global Aerosol Synthesis and Science Project (GASSP!)



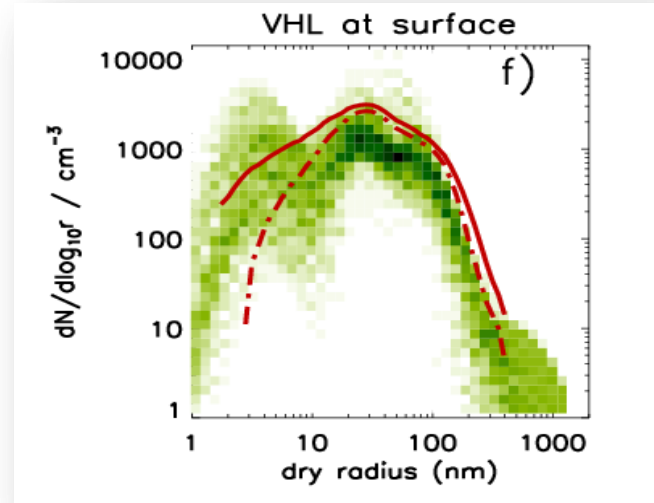
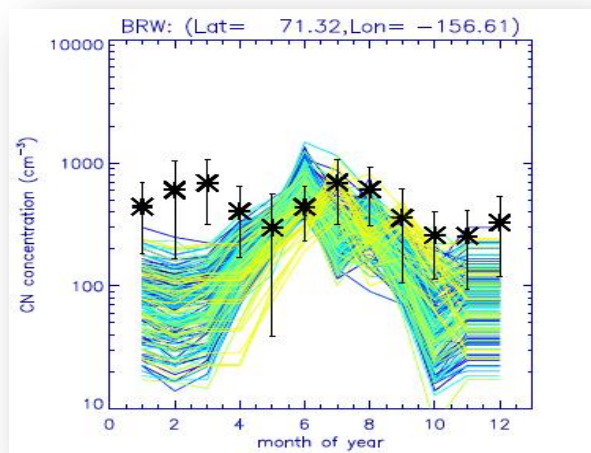
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Leeds/Oxford + 10 data partners

Synthesis of global aerosol microphysics data

Use model sensitivity and uncertainty statistics to:

- Identify plausible, implausible and **best models**
- Identify **structural weaknesses** from remaining bias and inconsistencies between datasets
- Inform **observational strategies**
- **Indirect forcing** most consistent with modern aerosol measurements

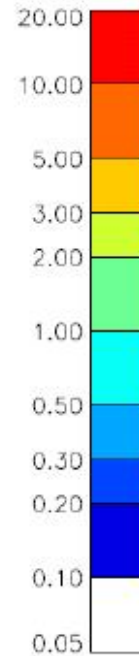
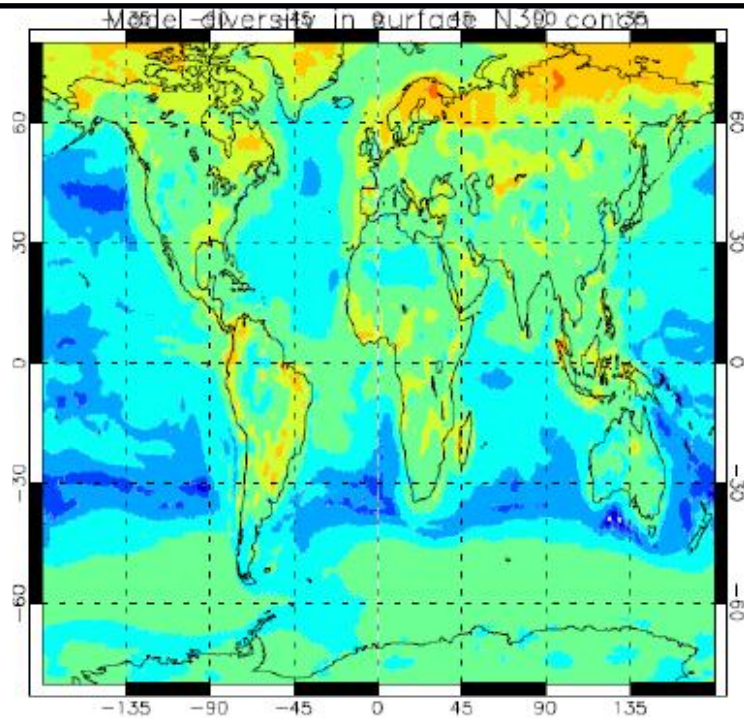


Parametric uncertainty versus multi-model diversity

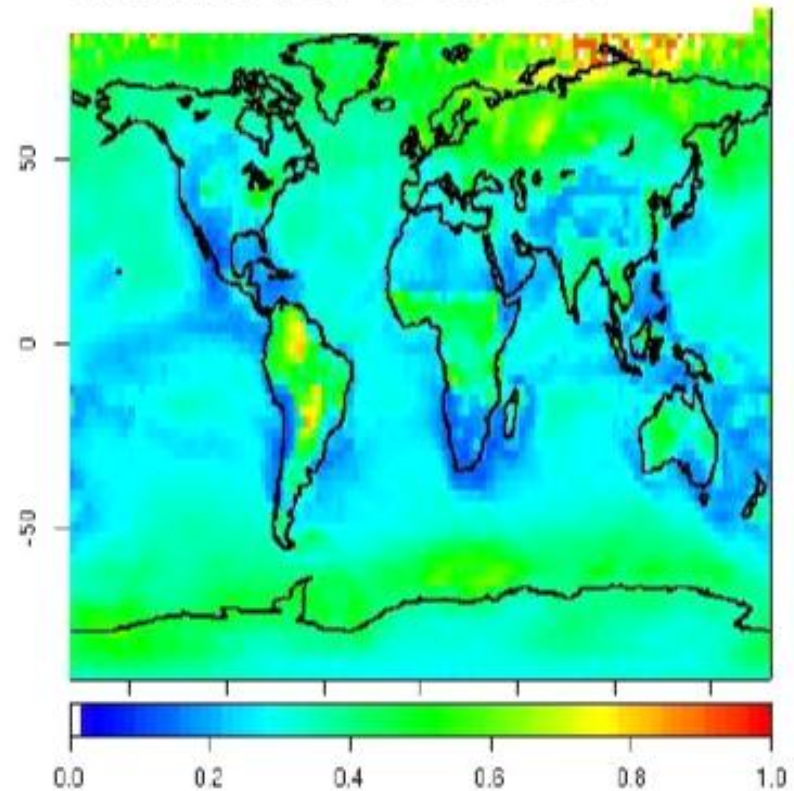


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AEROCOM 12-model diversity in N30
 $= (P75-P25)/P50$



GLOMAP coefficient of variation
 (σ/CCN)



AEROCOM data from Graham Mann



Summary

- Can use model emulation to calculate a Monte Carlo-level of information: global 3-D fields of *aerosol variance*, *variance contributions*, and *probability distributions* for all parameters
- CCN parametric uncertainty 1σ varies between 30 and 80%
- Uncertainty in BC is similar to CCN, but fewer important parameters
- Parametric uncertainty in indirect effect (due to emissions and aerosol processes) is **-1.32 ± 0.46 (2σ) Wm^{-2}** (vs IPCC 0.3-1.8 Wm^{-2} range)
- Parameters controlling indirect forcing are not the same ones controlling present day CCN \Rightarrow new strategy for model evaluation
- ~40% of parametric uncertainty in indirect effect from natural emissions

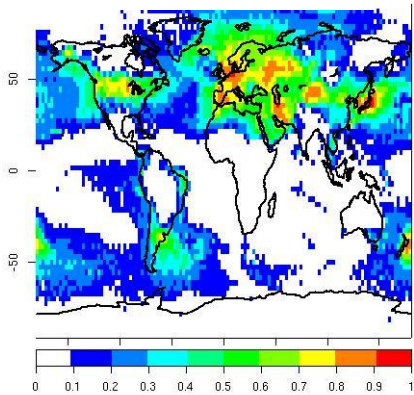
Happy to initiate collaborations to exploit all the data!

Lee, L.A. et al., Emulation of a complex global aerosol model to quantify sensitivity to uncertain parameters, ACP 2011.

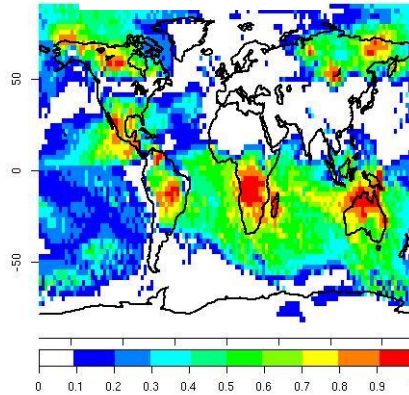
Lee L.A. et al., Mapping the uncertainty in global CCN using emulation, ACPD, 2012.

Black carbon

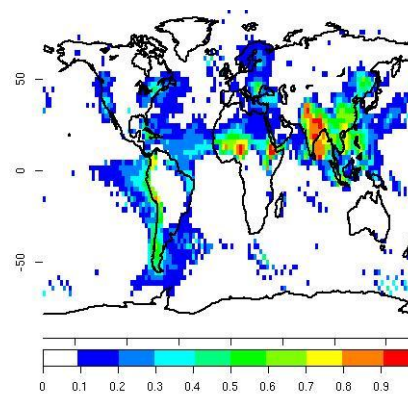
Fossil fuel



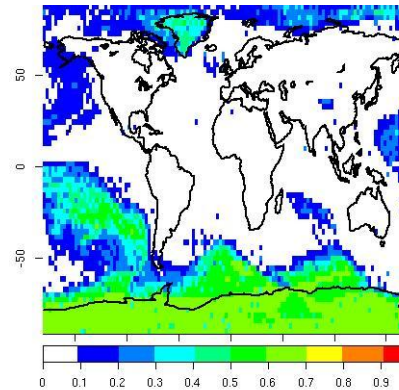
Biomass



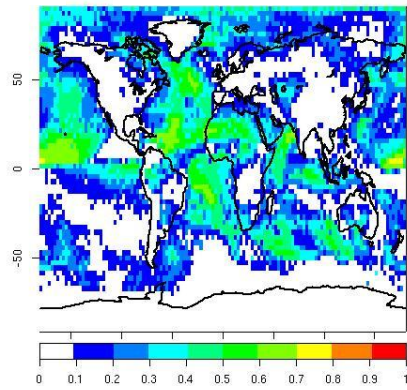
Biofuel



Activation diameter



Dry dep



BC mass is much simpler to model than CCN

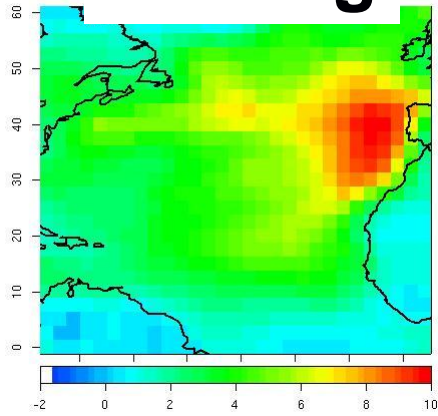
Uncertainty dominated by 5 parameters: *emission factors and removal processes*

North Atlantic forcing variances (February)

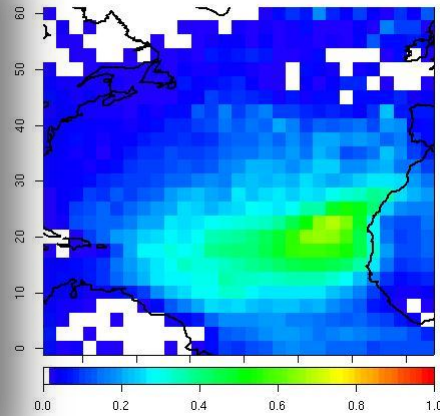


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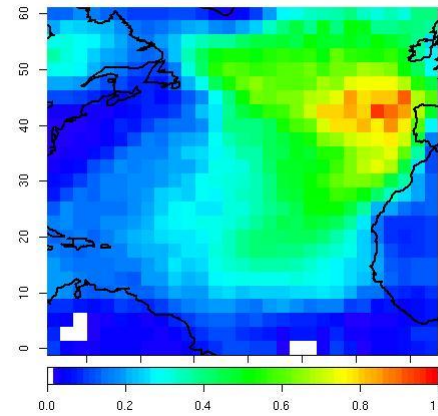
Forcing



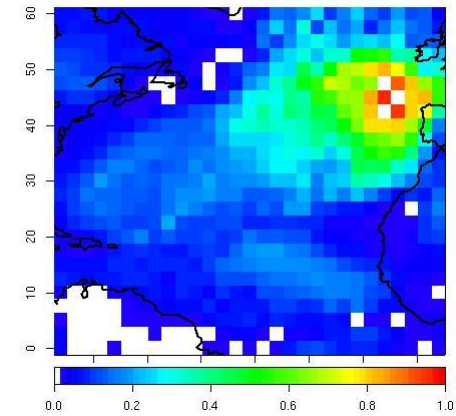
Volcanic SO₂



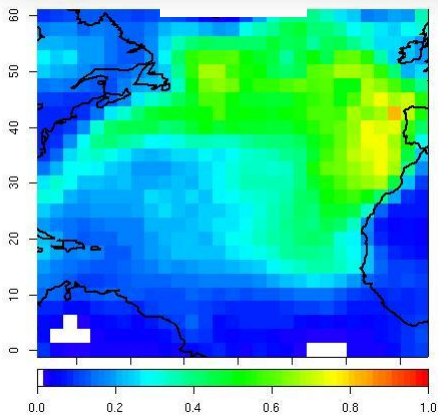
Anthrop SO₂



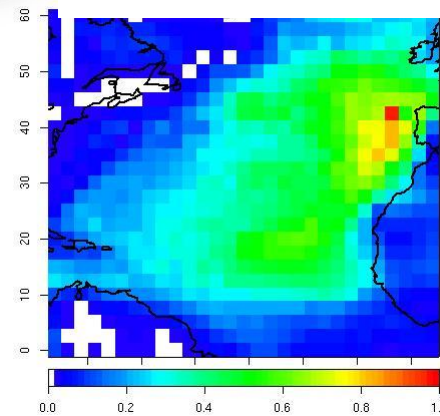
Subgrid SO₄



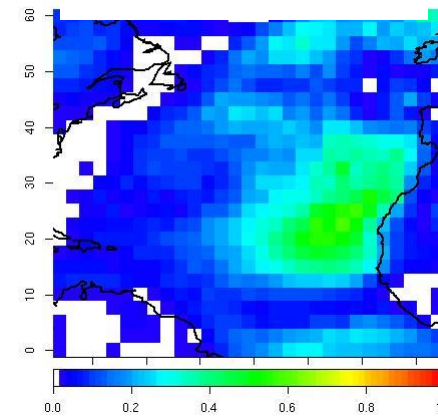
JUL DMS %24



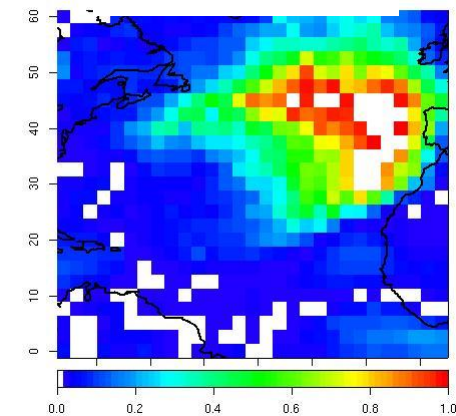
Acc mode width



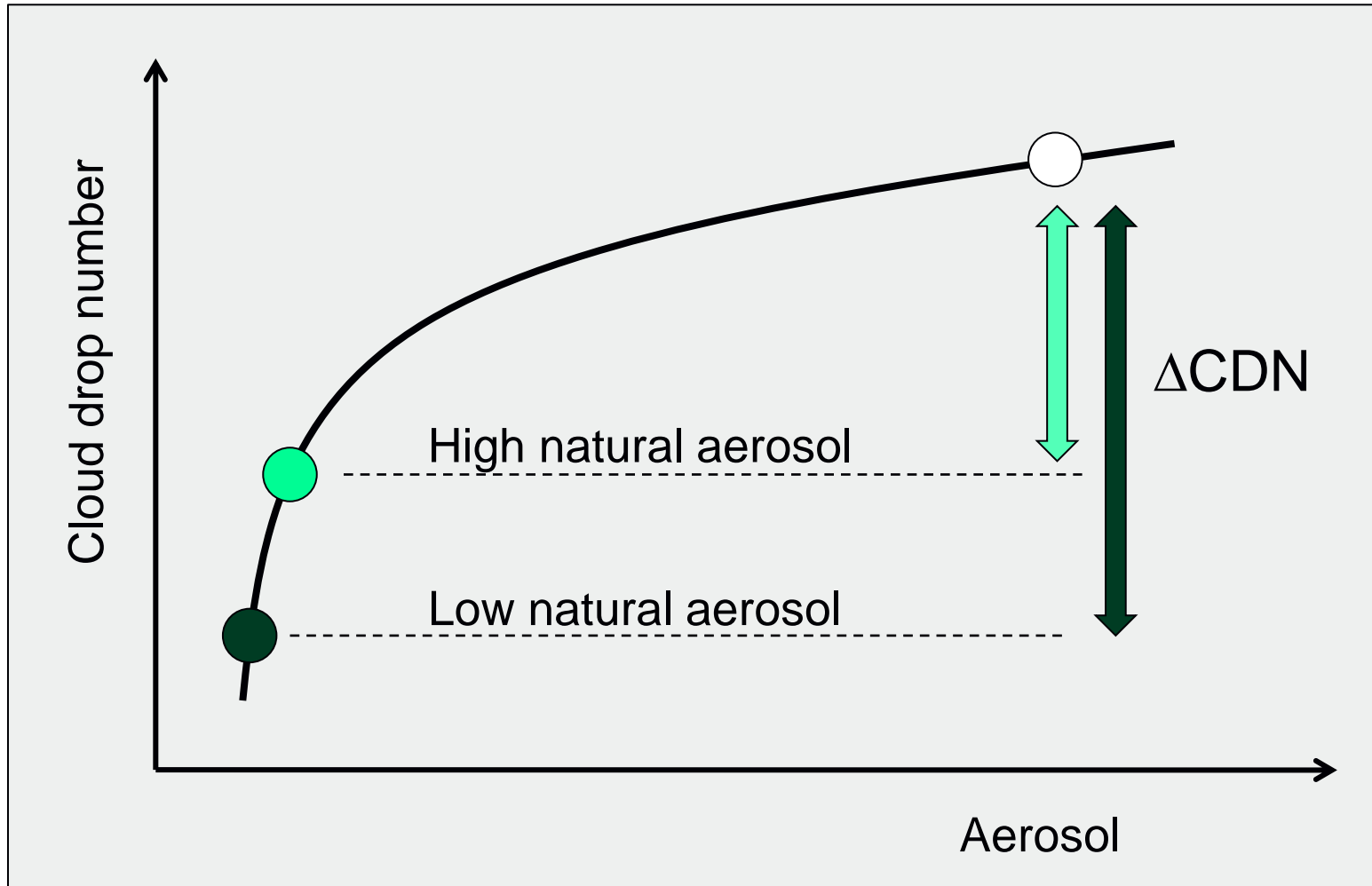
Activation diam



BB diameter



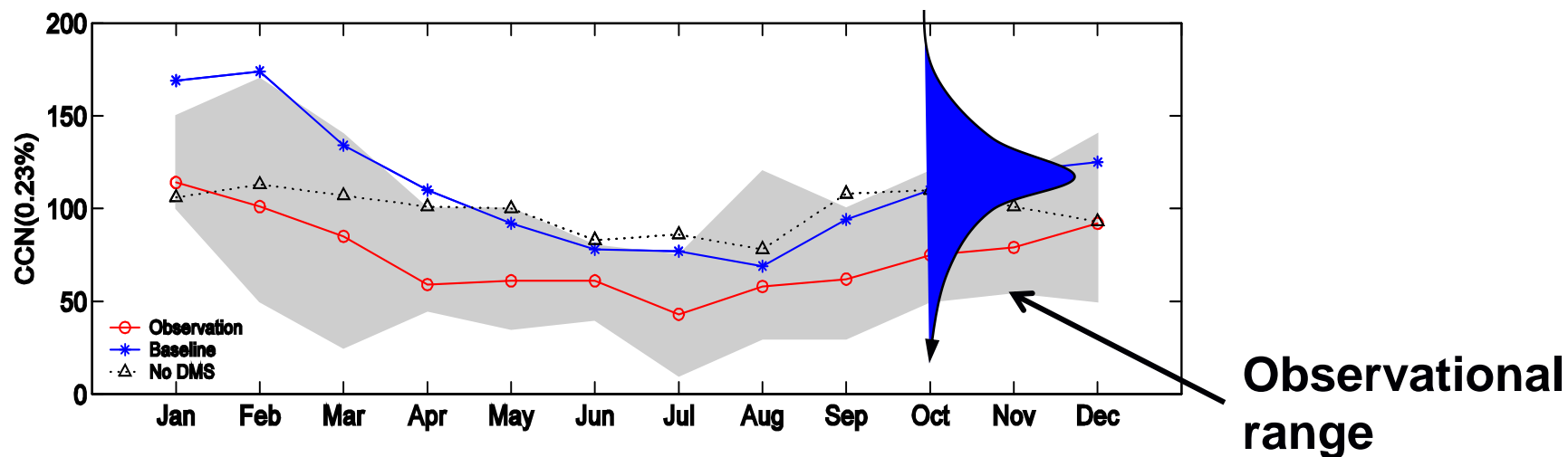
Why do natural emissions contribute to uncertainty in forcing?



Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate, A. Schmidt et al., Atmos. Chem. Phys. Discuss., 12, 8009-8051, 2012

Sensitivity and uncertainty “one at a time”

GLOMAP predictions of CCN at Cape Grim using “one-at-a-time” sensitivity tests



- One-at-a-time tests sample only a tiny fraction of parameter space
- They don't provide any statistical information