Sensitivity of Climate Forcings to Aerosol Physics and Chemistry

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Climate Forcing Uncertainties

- AEROCOM✈: Large diversities among models: Improved representation of processes necessary, especially regarding the fine aerosol composition
- This study: Range of uncertainties involving microphysical processes within one model

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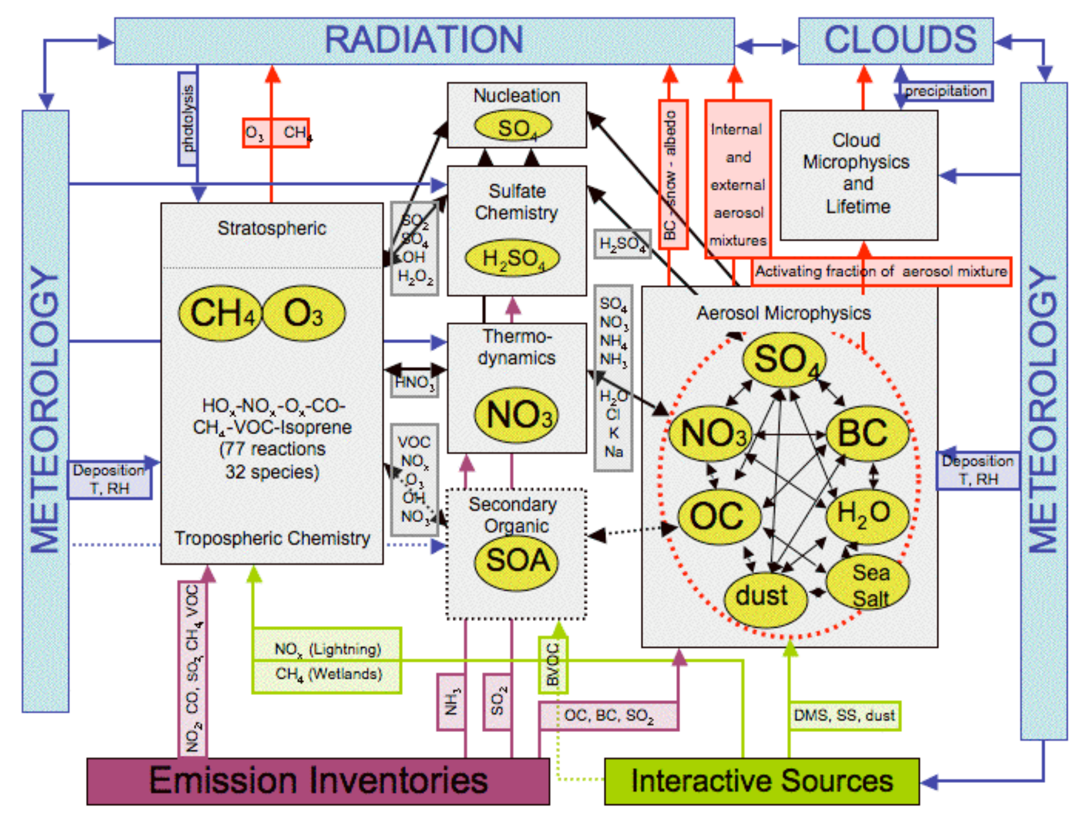
✈ Textor et al, ACP 2006, Kinne et al. ACP 2006, Schulz et al. ACP 2006

GISS GCM

GISS climate model (modelE) *Schmidt et al. 2006*
Fully coupled ocean-atmosphere climate model
Including gas and aerosol phase chemistry

Experimental set up:
4° x 5° horizontal res.
23 vertical layers

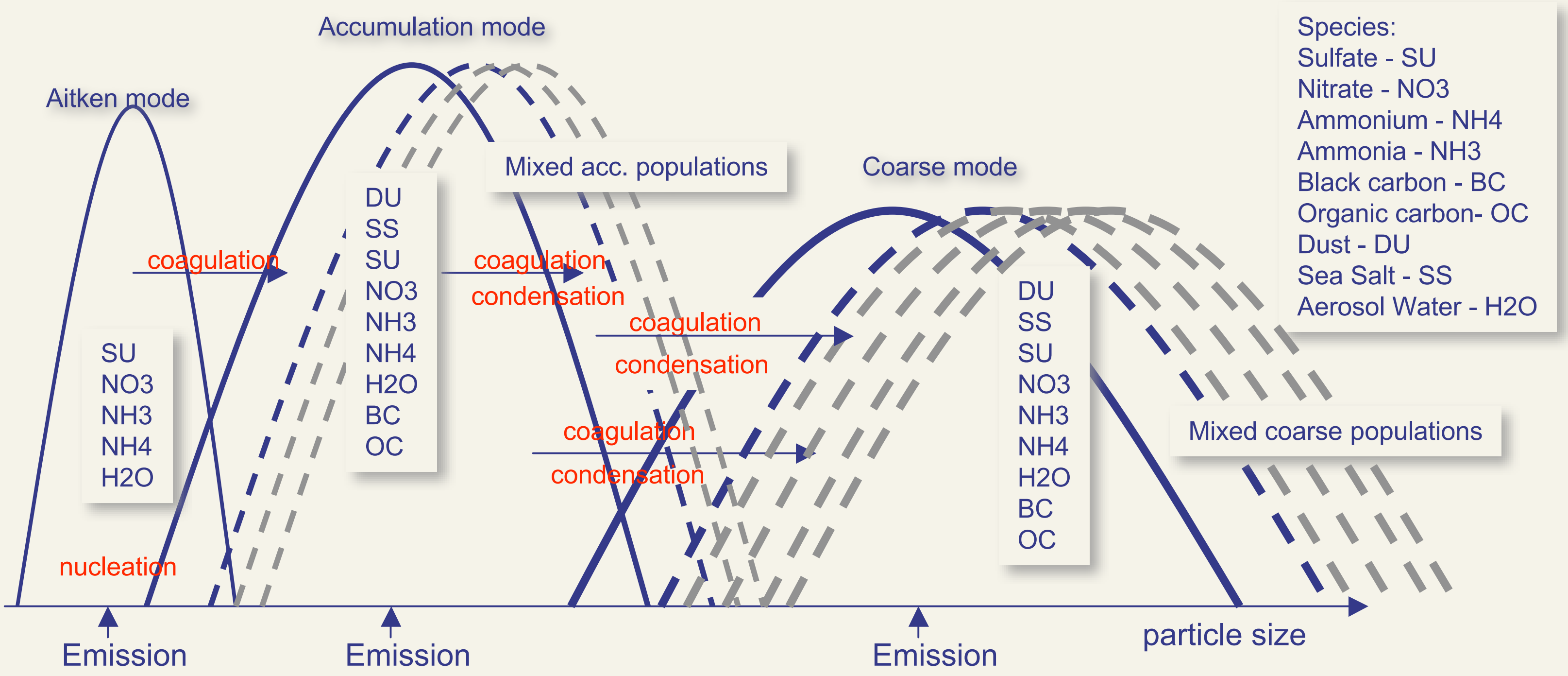
Model results:
5 year average



MATRIX

Aerosol Microphysical Model based on the Methods of Moments

Bauer et al. ACP 2008



Aitken mode

Accumulation mode

Mixed acc. populations

Coarse mode

Mixed coarse populations

SU
NO3
NH3
NH4
H2O

DU
SS
SU
NO3
NH3
NH4
H2O
BC
OC

DU
SS
SU
NO3
NH3
NH4
H2O
BC
OC

nucleation

coagulation

coagulation
condensation

coagulation
condensation

coagulation
condensation

Emission

Emission

Emission

particle size

Species:
Sulfate - SU
Nitrate - NO3
Ammonium - NH4
Ammonia - NH3
Black carbon - BC
Organic carbon- OC
Dust - DU
Sea Salt - SS
Aerosol Water - H2O

- Partitioning of semi volatile species: EQSAM 3 (Metzger et al. 2006)
- Droplet activation: Abdul Razzak and Ghan(1998, 2000)

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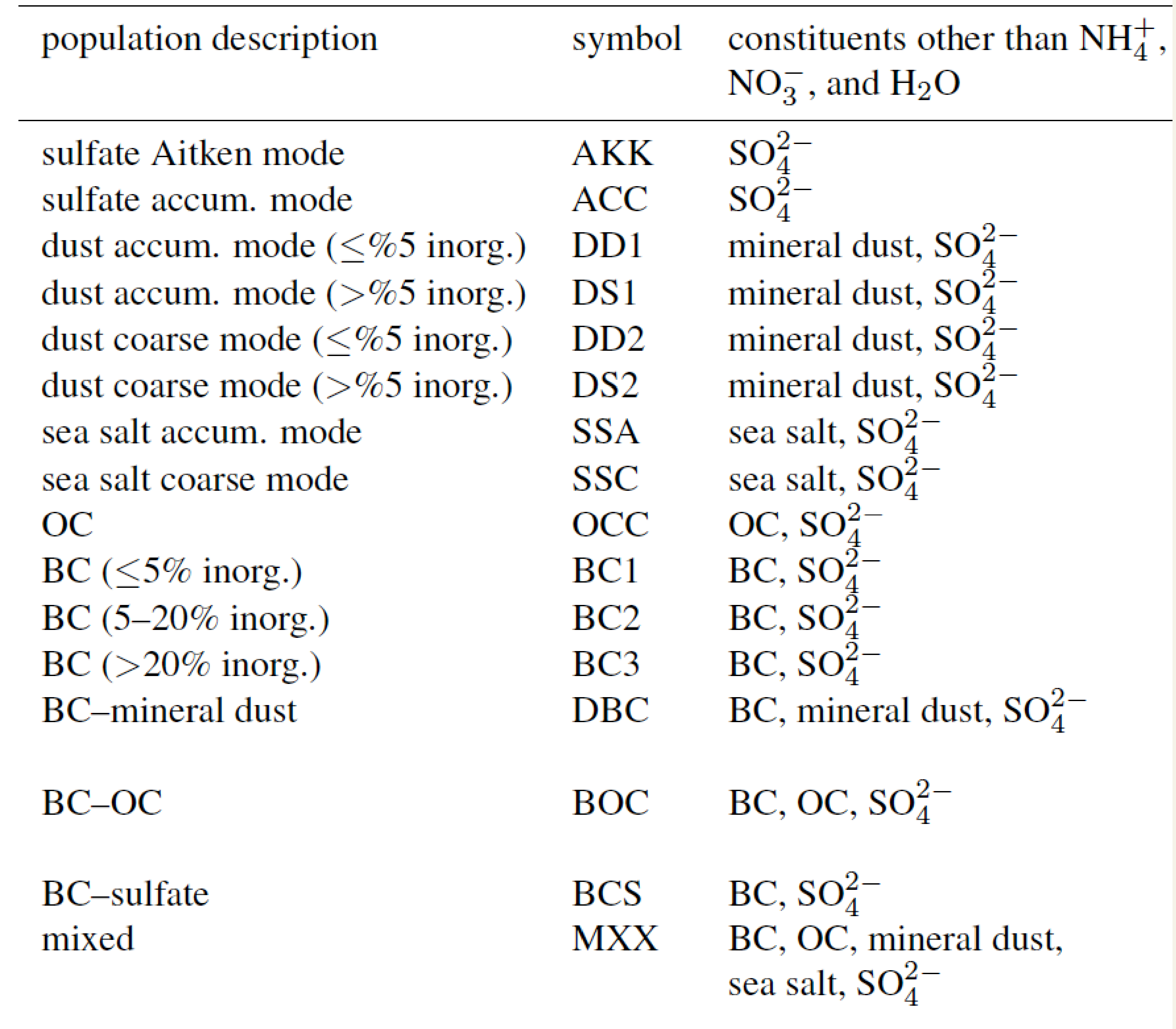

population description	symbol	constituents other than NH_4^+, NO_3^-, and H_2O
sulfate Aitken mode	AKK	SO_4^{2-}
sulfate accum. mode	ACC	SO_4^{2-}
dust accum. mode ($\leq$%5 inorg.)	DD1	mineral dust, SO_4^{2-}
dust accum. mode ($>$%5 inorg.)	DS1	mineral dust, SO_4^{2-}
dust coarse mode ($\leq$%5 inorg.)	DD2	mineral dust, SO_4^{2-}
dust coarse mode ($>$%5 inorg.)	DS2	mineral dust, SO_4^{2-}
sea salt accum. mode	SSA	sea salt, SO_4^{2-}
sea salt coarse mode	SSC	sea salt, SO_4^{2-}
OC	OCC	OC, SO_4^{2-}
BC ($\leq$5% inorg.)	BC1	BC, SO_4^{2-}
BC (5–20% inorg.)	BC2	BC, SO_4^{2-}
BC ($>$20% inorg.)	BC3	BC, SO_4^{2-}
BC–mineral dust	DBC	BC, mineral dust, SO_4^{2-}
BC–OC	BOC	BC, OC, SO_4^{2-}
BC–sulfate	BCS	BC, SO_4^{2-}
mixed	MXX	BC, OC, mineral dust, sea salt, SO_4^{2-}

Nucleation

Kulmala et al 2004 :'The formation rate of 3 nm particles during regional nucleation events lies typically in the range 0.01–10 particles cm^{-3} s^{-1} in the boundary layer. In coastal environments and industrial plumes, however, formation rates as high as $10e^4$–$10e^5$ particles cm^{-3} s^{-1} have been reported.'

	Scheme	Author	Nucleation rate [s-1]	New Particle Formation Rate * at 3 nm [cm-2 s-1]
1	Ternary H2SO4- NH3 -H20	Napari et al (2002)	1.08e+4	6590
2	Binary H2SO4 - H2O	Jaecker-Voirol and Mirabel (1989)	4.88e+3	2263
3	Binary H2SO4 - H2O	Vehkamaki et al (2002)	1.21e+3	713
4	Fit to observation	Eisele and McMurry (1997)	1.5e-4	4
5	No Nucleation		0	0

* Transformation of nucleation rate into new particle formation rate after Kerminen et al. 2004 and Bauer et al. 2008 ACD

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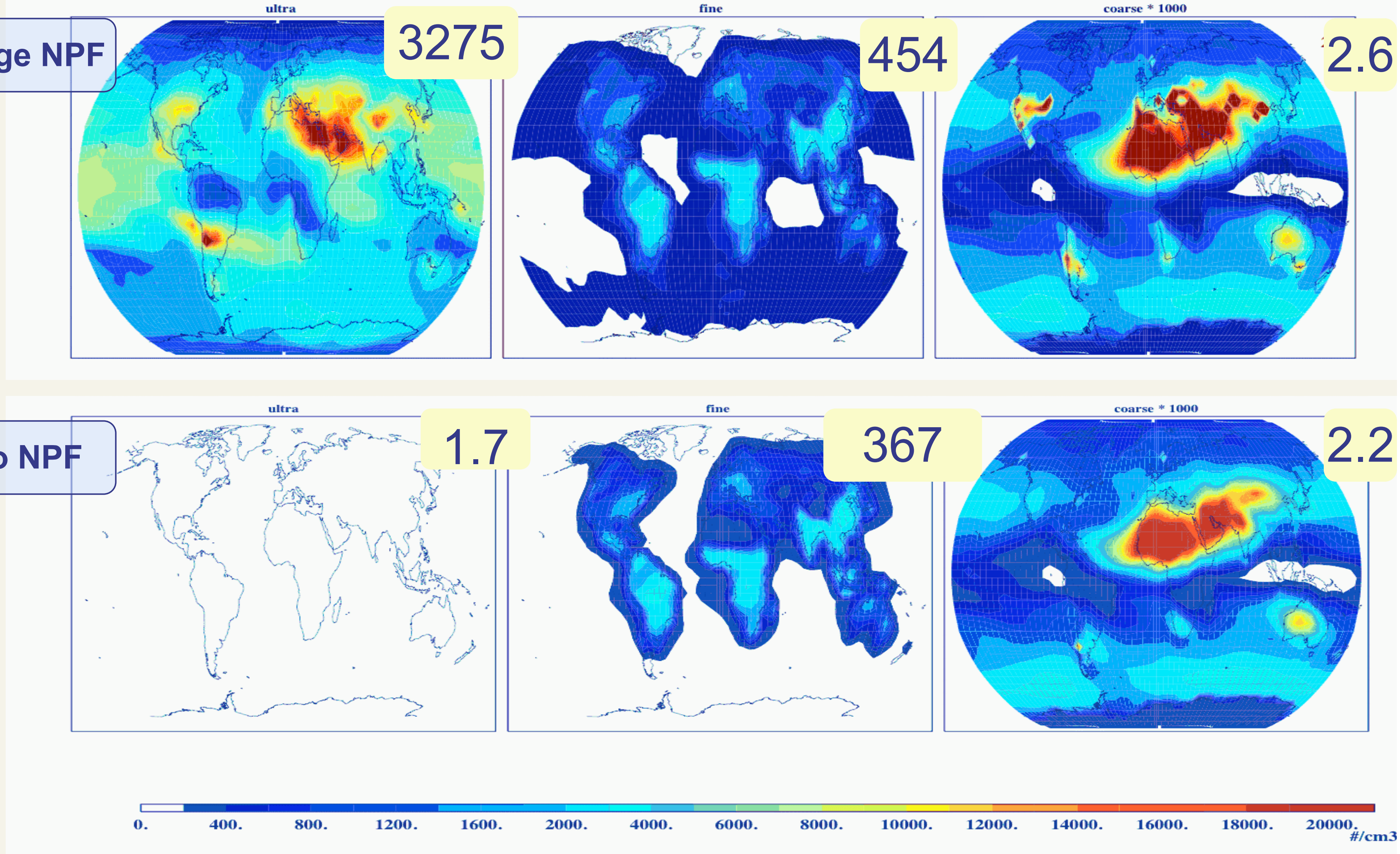
Number Concentrations [#/cm^3]:

Ultra [> 0.05 um] **Fine [0.05 - 1 um]** **Coarse x 1000 [< 1 um]**



Large NPF

3275

454

2.6

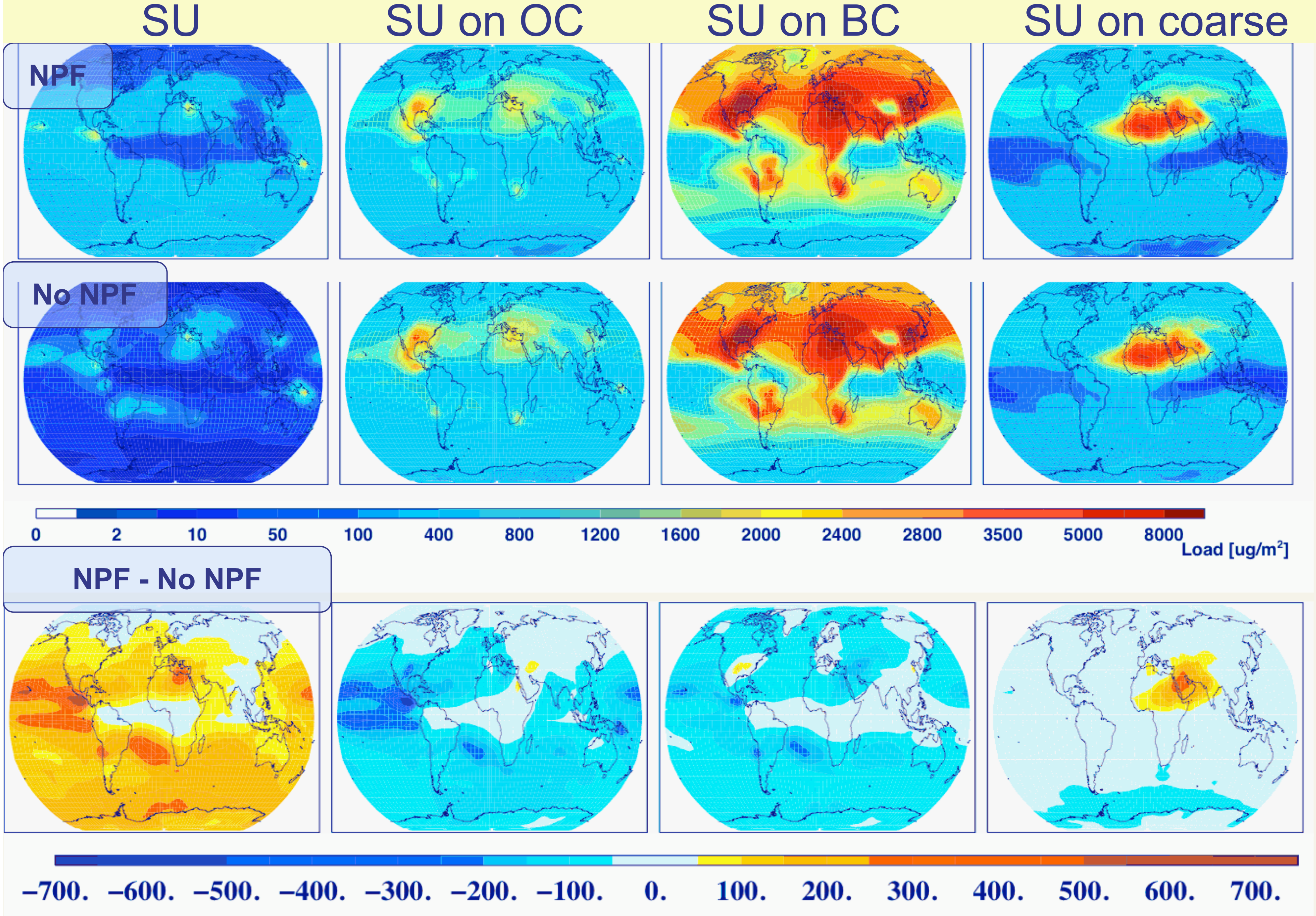
No NPF

1.7

367

2.2

Sulfate Mixing State



SU

SU on OC

SU on BC

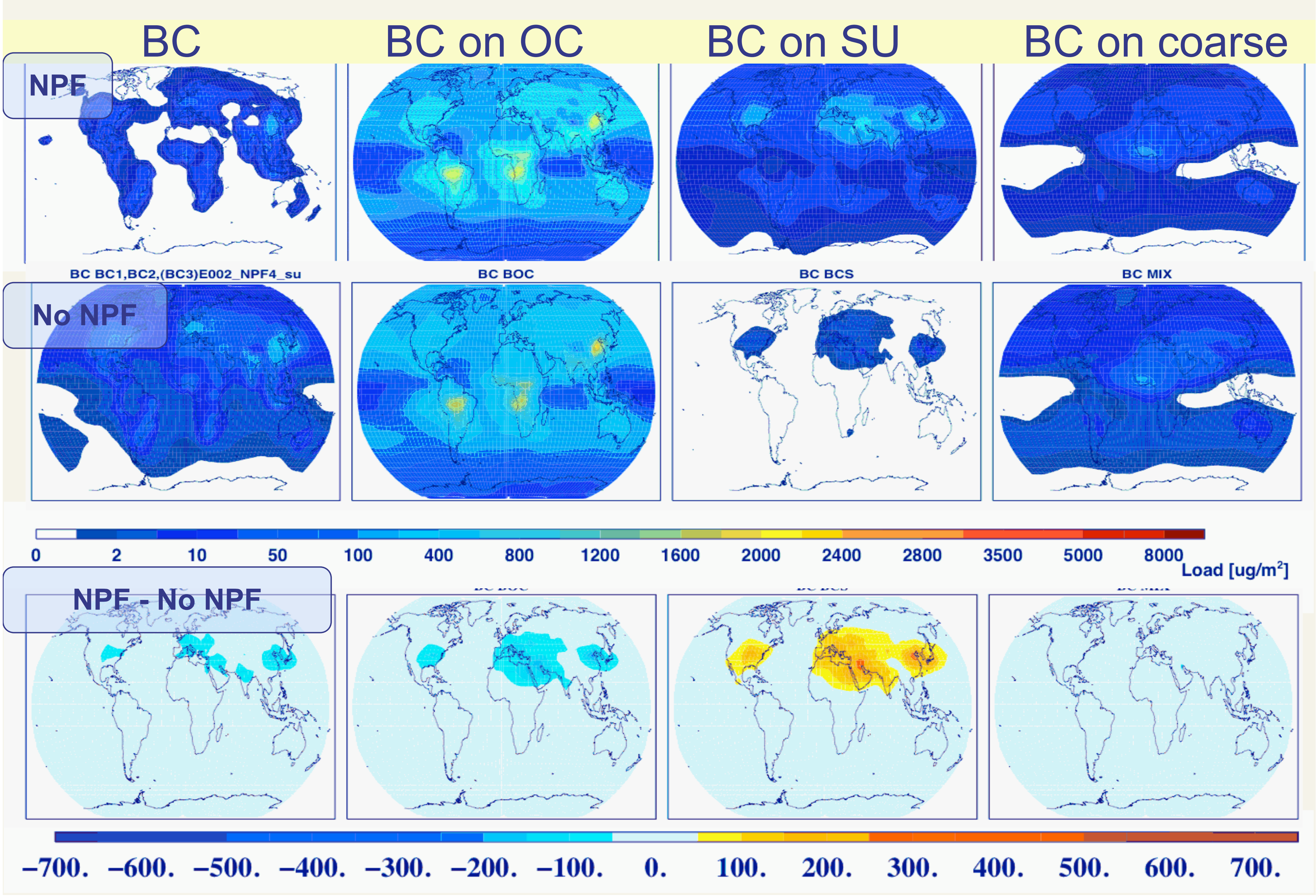
SU on coarse

NPF

No NPF

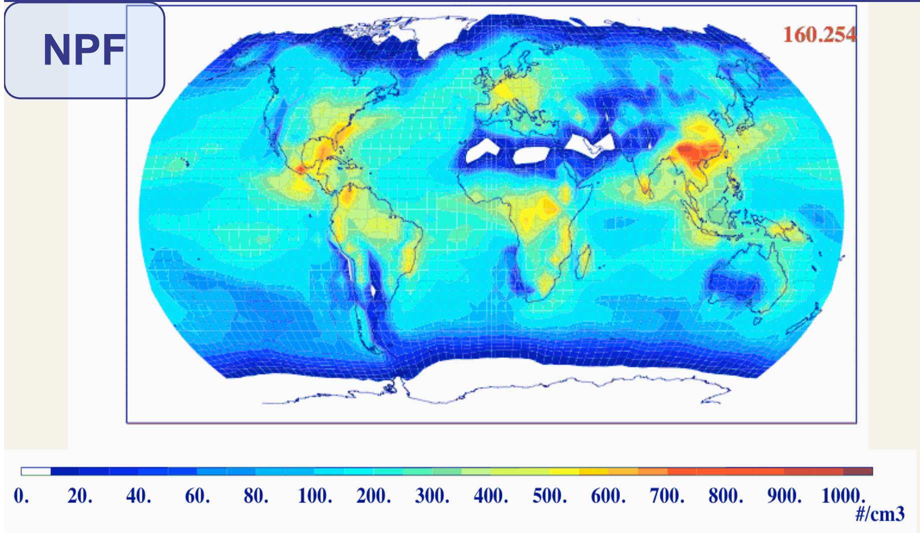
NPF - No NPF

Black Carbon Mixing State

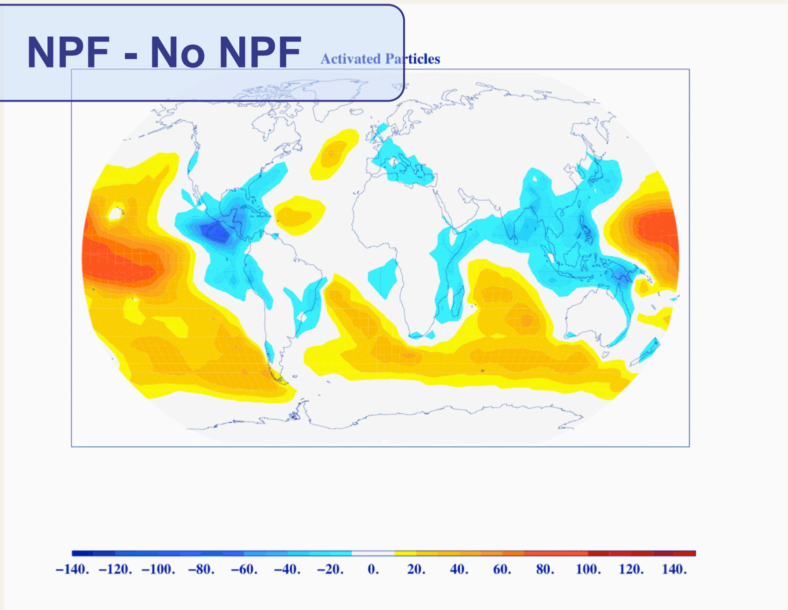


CCN Number concentrations [column cm^{-3}]

NPF



NPF - No NPF



- **5.6 % more activated particles through nucleation**

- 5.3 % more CCN through binary nucleation, *Wang and Penner 2008 ACPD*

Why not larger?
Competition for sulfuric acid gas and ammonia between nucleation and primary particles.

CCN → activated particle → CDNC

Emission Size Uncertainty Experiments

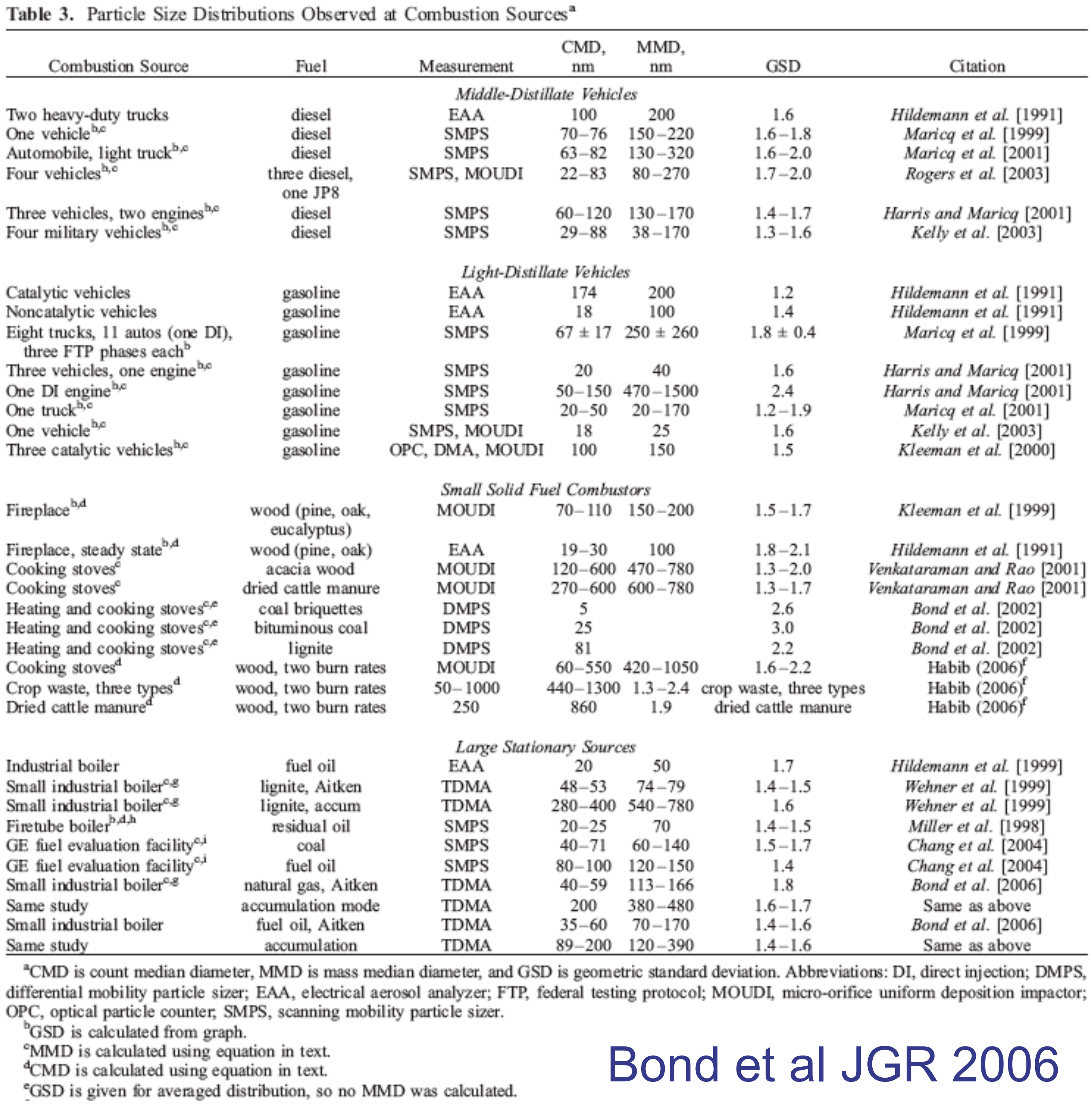


Table 3. Particle Size Distributions Observed at Combustion Sources[a]

Combustion Source	Fuel	Measurement	CMD, nm	MMD, nm	GSD	Citation
		Middle-Distillate Vehicles				
Two heavy-duty trucks	diesel	EAA	100	200	1.6	*Hildemann et al.* [1991]
One vehicle[b,c]	diesel	SMPS	70–76	150–220	1.6–1.8	*Maricq et al.* [1999]
Automobile, light truck[b,c]	diesel	SMPS	63–82	130–320	1.6–2.0	*Maricq et al.* [2001]
Four vehicles[b,c]	three diesel, one JP8	SMPS, MOUDI	22–83	80–270	1.7–2.0	*Rogers et al.* [2003]
Three vehicles, two engines[b,c]	diesel	SMPS	60–120	130–170	1.4–1.7	*Harris and Maricq* [2001]
Four military vehicles[b,c]	diesel	SMPS	29–88	38–170	1.3–1.6	*Kelly et al.* [2003]
		Light-Distillate Vehicles				
Catalytic vehicles	gasoline	EAA	174	200	1.2	*Hildemann et al.* [1991]
Noncatalytic vehicles	gasoline	EAA	18	100	1.4	*Hildemann et al.* [1991]
Eight trucks, 11 autos (one DI), three FTP phases each[b]	gasoline	SMPS	67 ± 17	250 ± 260	1.8 ± 0.4	*Maricq et al.* [1999]
Three vehicles, one engine[b,c]	gasoline	SMPS	20	40	1.6	*Harris and Maricq* [2001]
One DI engine[b,c]	gasoline	SMPS	50–150	470–1500	2.4	*Harris and Maricq* [2001]
One truck[b,c]	gasoline	SMPS	20–50	20–170	1.2–1.9	*Maricq et al.* [2001]
One vehicle[b,c]	gasoline	SMPS, MOUDI	18	25	1.6	*Kelly et al.* [2003]
Three catalytic vehicles[b,c]	gasoline	OPC, DMA, MOUDI	100	150	1.5	*Kleeman et al.* [2000]
		Small Solid Fuel Combustors				
Fireplace[b,d]	wood (pine, oak, eucalyptus)	MOUDI	70–110	150–200	1.5–1.7	*Kleeman et al.* [1999]
Fireplace, steady state[b,d]	wood (pine, oak)	EAA	19–30	100	1.8–2.1	*Hildemann et al.* [1991]
Cooking stoves[c]	acacia wood	MOUDI	120–600	470–780	1.3–2.0	*Venkataraman and Rao* [2001]
Cooking stoves[c]	dried cattle manure	MOUDI	270–600	600–780	1.3–1.7	*Venkataraman and Rao* [2001]
Heating and cooking stoves[c,e]	coal briquettes	DMPS	5		2.6	*Bond et al.* [2002]
Heating and cooking stoves[c,e]	bituminous coal	DMPS	25		3.0	*Bond et al.* [2002]
Heating and cooking stoves[c,e]	lignite	DMPS	81		2.2	*Bond et al.* [2002]
Cooking stoves[d]	wood, two burn rates	MOUDI	60–550	420–1050	1.6–2.2	Habib (2006)[f]
Crop waste, three types[d]	wood, two burn rates	50–1000	440–1300	1.3–2.4	crop waste, three types	Habib (2006)[f]
Dried cattle manure[d]	wood, two burn rates	250	860	1.9	dried cattle manure	Habib (2006)[f]
		Large Stationary Sources				
Industrial boiler	fuel oil	EAA	20	50	1.7	*Hildemann et al.* [1999]
Small industrial boiler[c,g]	lignite, Aitken	TDMA	48–53	74–79	1.4–1.5	*Wehner et al.* [1999]
Small industrial boiler[c,g]	lignite, accum	TDMA	280–400	540–780	1.6	*Wehner et al.* [1999]
Firetube boiler[b,d,h]	residual oil	SMPS	20–25	70	1.4–1.5	*Miller et al.* [1998]
GE fuel evaluation facility[c,i]	coal	SMPS	40–71	60–140	1.5–1.7	*Chang et al.* [2004]
GE fuel evaluation facility[c,i]	fuel oil	SMPS	80–100	120–150	1.4	*Chang et al.* [2004]
Small industrial boiler[c,g]	natural gas, Aitken	TDMA	40–59	113–166	1.8	*Bond et al.* [2006]
Same study	accumulation mode	TDMA	200	380–480	1.6–1.7	Same as above
Small industrial boiler	fuel oil, Aitken	TDMA	35–60	70–170	1.4–1.6	*Bond et al.* [2006]
Same study	accumulation	TDMA	89–200	120–390	1.4–1.6	Same as above

[a] CMD is count median diameter, MMD is mass median diameter, and GSD is geometric standard deviation. Abbreviations: DI, direct injection; DMPS, differential mobility particle sizer; EAA, electrical aerosol analyzer; FTP, federal testing protocol; MOUDI, micro-orifice uniform deposition impactor; OPC, optical particle counter; SMPS, scanning mobility particle sizer.

[b] GSD is calculated from graph.

[c] MMD is calculated using equation in text.

[d] CMD is calculated using equation in text.

[e] GSD is given for averaged distribution, so no MMD was calculated.

Bond et al JGR 2006

Emission Size Uncertainty Experiments

AEROCOM emission size range* [*Textor et al. ACP 2006*] :

SU: sulfate: 0.13 - 0.6 µm

BC: black carbon 0.025 - 0.6 µm

OC: organic carbon 0.03 - 0.8 µm

*emissions may be distributed over several size-bins / modes or differ between source sectors

Sizes of the emitted particles [µm]

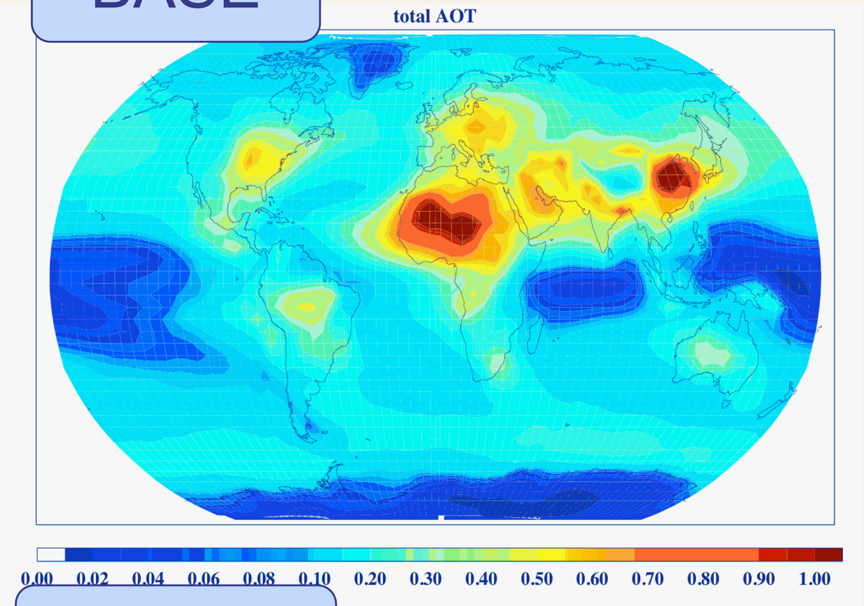
Mass median diameters of log normal distributions

	Accumulation mode emissions			Several modes	
	SU	BC	OC	SS	DU
BASE	0.068	0.03 fossil 0.037 bio	0.03 0.037	0.2 - 4.	0.3 - 6
C1	BASE	BASE	BASE	2xBASE	2x BASE
F1	0.07	0.07	0.1	BASE	BASE
F2	0.3	0.3	0.3	BASE	BASE
F3	0.6	0.6	0.6	BASE	BASE

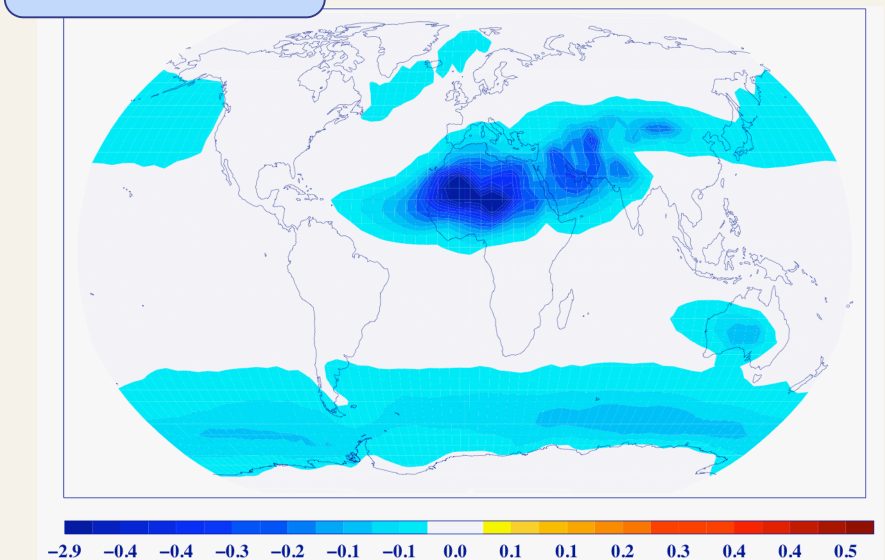
Note!
Just emission sizes.
Ambient particle sizes determined by microphysics

Aerosol Optical Thickness

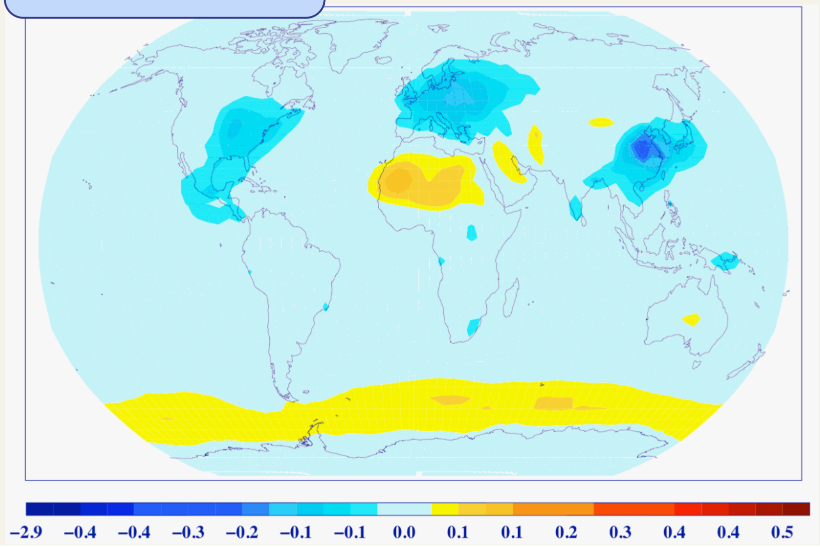
BASE



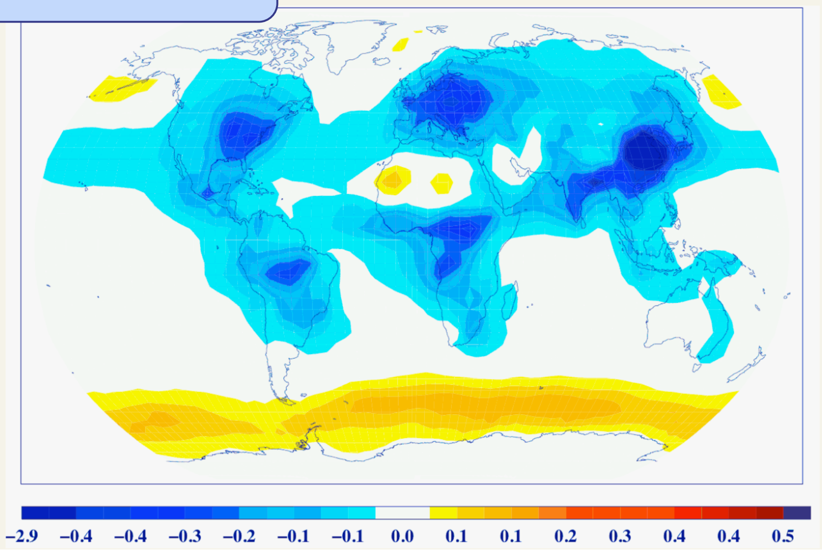
C1 - BASE



F1 - BASE

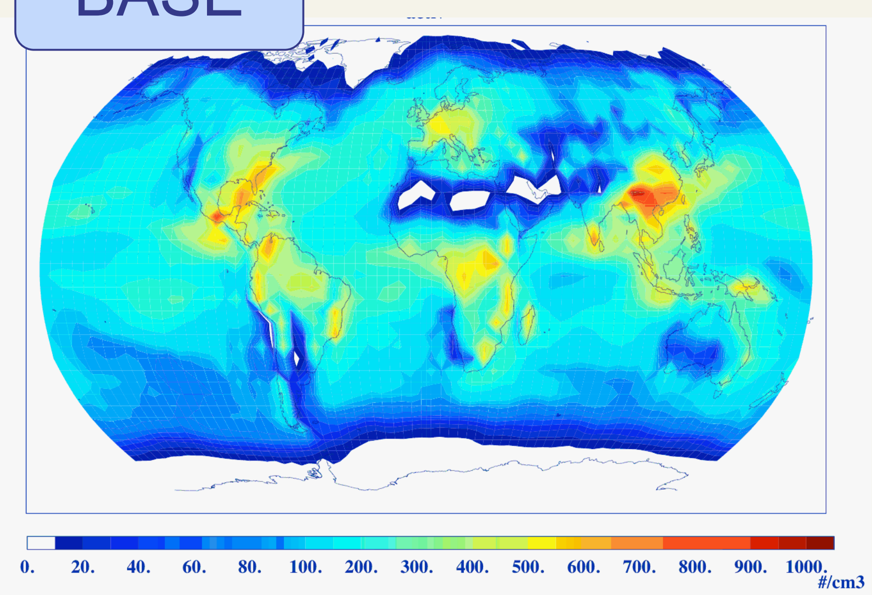


F2 - BASE



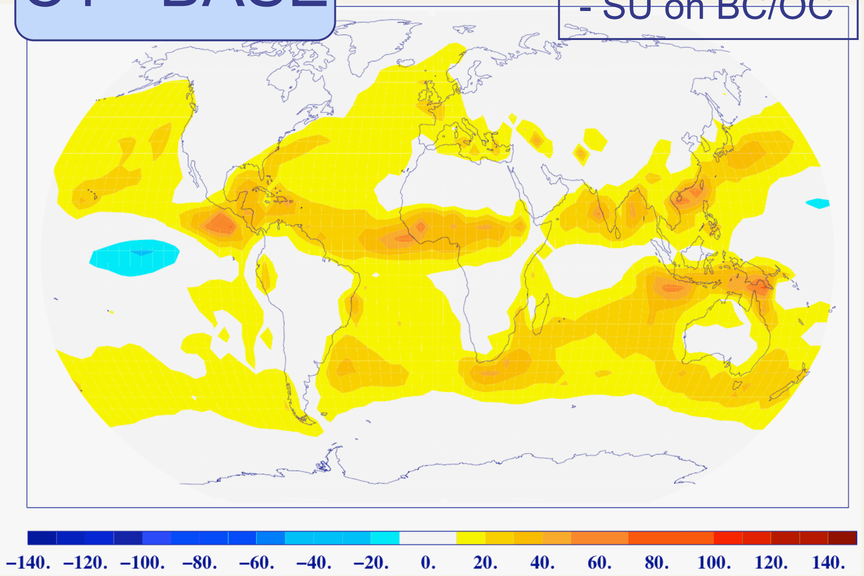
CCN Number concentrations [column cm^{-3}]

BASE



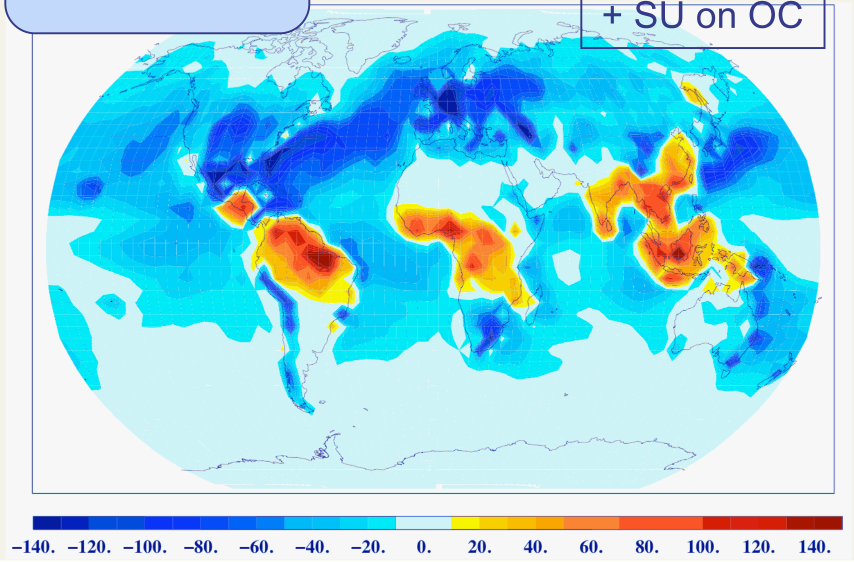
C1 - BASE

+ SU on coarse
- SU on BC/OC



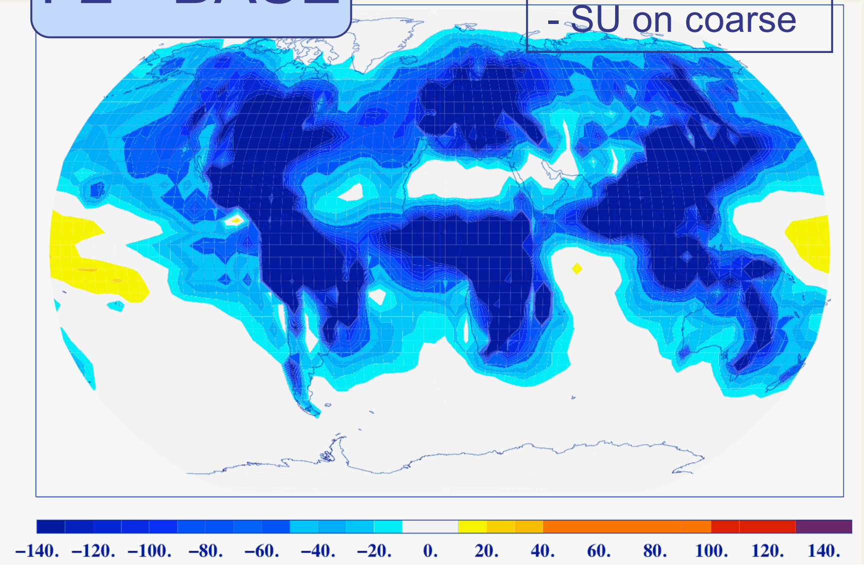
F1 - BASE

- SU on BC
+ SU on OC



F2 - BASE

+ SU on BC/OC
- SU on coarse



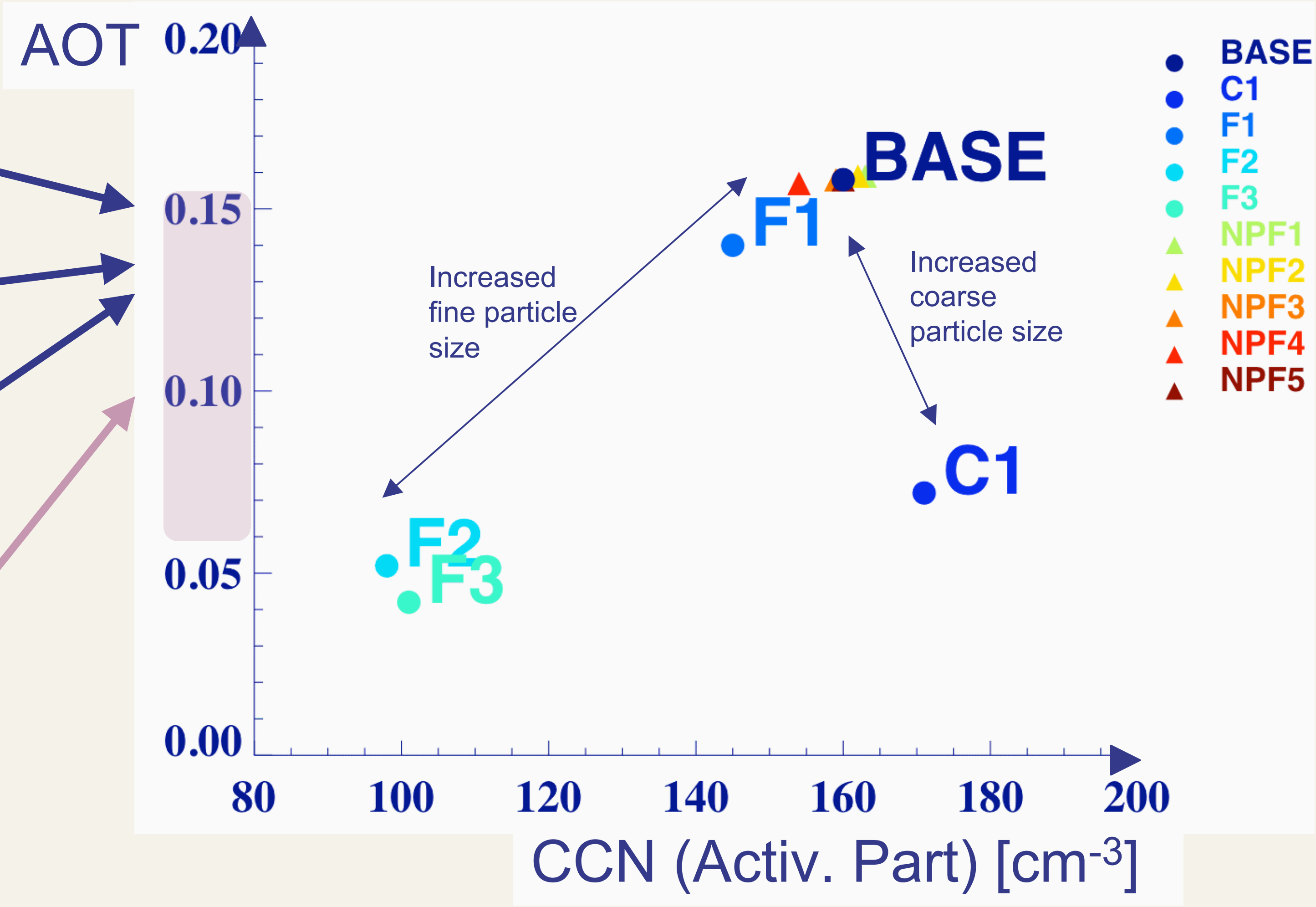
Aerosol Optical Thickness
versus
ACTIVATED aerosol numbers

Satellite✢
0.15

AERONET✢
0.135

AEROCOM✢
Model median
0.127

AEROCOM✢
Max 0.151
Min 0.065



✢ Kinne et al. 2006 ACP

Summary

Nucleation:

- The quantification of new particle formation through nucleation events is still an open research question. Nucleation events greatly impact aerosol size distributions and mixing state.
- The impact of nucleation on activated particle number concentrations is estimated here (comparing two extreme cases) to be about 5%.

Emission size information:

- Our experiments demonstrate that the entire AOT diversity of the AeroCom models can be reproduced by our model simply by changing the sizes of emitted particles.
- AOT and activated particle number concentrations may vary by up to 100%, comparable to the range of uncertainty of the direct and the indirect aerosol forcing.

- The knowledge of emission size per chemical species and emission sector is crucial for aerosol modeling → emission inventories
- Observations of aerosol size distributions and mixing state needed!

Acknowledgements

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