Sensitivity of Aerosol Indirect Forcing to Organic Aerosol Hygroscopicity

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Introduction

- Organic aerosol is one of the largest uncertainties in aerosol treatment
  - Emissions and processes:

    Multiple sources (primary & secondary; fossil fuel, bio fuel, biomass burning, biogenic, etc.)
  - Numerous organic species with different properties:

    Complicated chemistry involved with secondary organic aerosol (SOA)

- Hygroscopicity of organics $\rightarrow$ CCN $\rightarrow$ cloud droplet number $\rightarrow$ aerosol indirect forcing

Organics hygroscopicity (κ) from environmental chamber & field studies

Organics Class	Range of κ	References
Biogenic SOA	0.06-0.23	Prenni et al., 2007; King et al., 2007, Duplissy et al., 2008; Engelhart et al., 2008; Gunthe et al., 2009; Juranyi et al., 2009; King et al., 2009; King et al., 2010.
Anthropogenic SOA	0.06-0.14	Prenni et al., 2007
Biomass burning organic matter	0.06-0.33	Vestin et al., 2007; Asa-Awuku et al., 2008; Carrico et al., 2008; Gunthe et al., 2009; Petters et al., 2009

Primary organic aerosols from fossil fuel consumption are generally considered as non-hygroscopic (i.e., with κ=0)

Benchmark 7-Mode Modal Aerosol Model (MAM)

Aitken
number
sulfate
ammonium
secondary OA
sea salt

Accumulation
number
sulfate
ammonium
secondary OA
Primary OA
BC
sea salt

Fine Soil Dust
number
soil dust
sulfate
ammonium

Fine Sea Salt
number
sea salt
sulfate
ammonium

coagulation
condensation

Primary Carbon
number
Primary OA
BC

Coarse Soil Dust
number
soil dust
sulfate
ammonium

Coarse Sea Salt
number
sea salt
sulfate
ammonium

All modes log-normal with prescribed width.

Total transported aerosol tracers: 31

Cloud-borne aerosol and aerosol water predicted but not transported.

CAM5 Simulations

- 7-mode MAM
- 5 years present-day (PD) and preindustrial (PI) simulations at 1.9°x2.5° resolution

- IPCC AR5 emissions for anthropogenic OA, BC, SO2, SO4 (Lamarque et al.)
- AEROCOM emissions for natural DMS, SO2, SO4, injection heights and primary particle sizes
- SOA (g) emission: apply yields on MOZART VOCs emissions

Simulated cases

Case Name	Description
CTL	Standard CAM, κ= 0.0 (POA), κ=0.14 (SOA)
POA-hiK	κ = 0.1 (POA), κ=0.14 (SOA)
SOA-loK	κ = 0.0 (POA), κ=0.07 (SOA)
SOA-hiK	κ = 0.0 (POA), κ=0.21 (SOA)

Sources of POA:

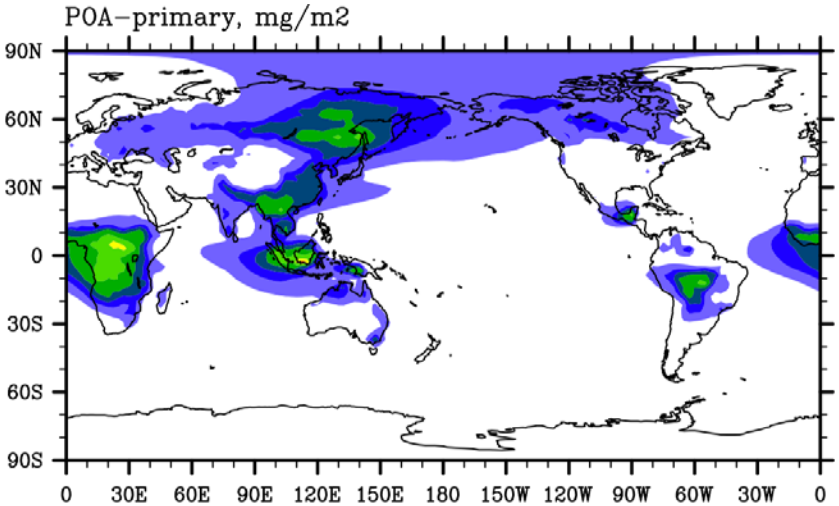
- Fossil fuel burning (κ~0)
- Biofuel burning
- Biomass burning (0.06-0.33)

Sources of SOA:

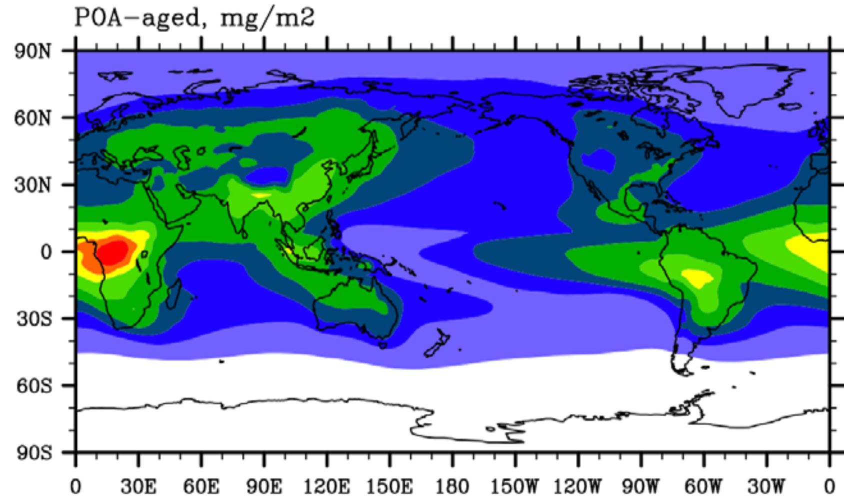
- Anthropogenic precursors (0.06-0.14)
- Biogenic precursors (0.06-0.23)

OA Column Burden at PD

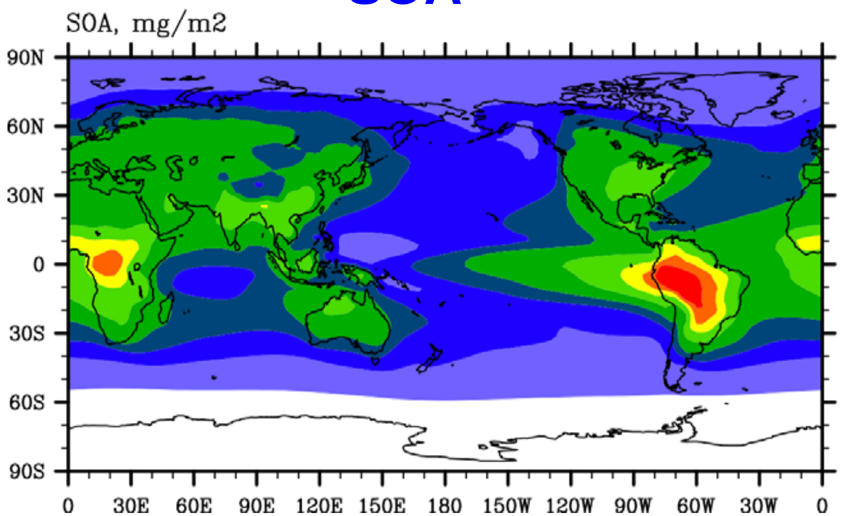
POA in primary mode

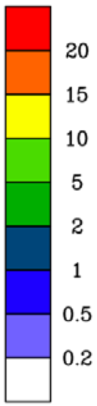


POA in aged mode



SOA

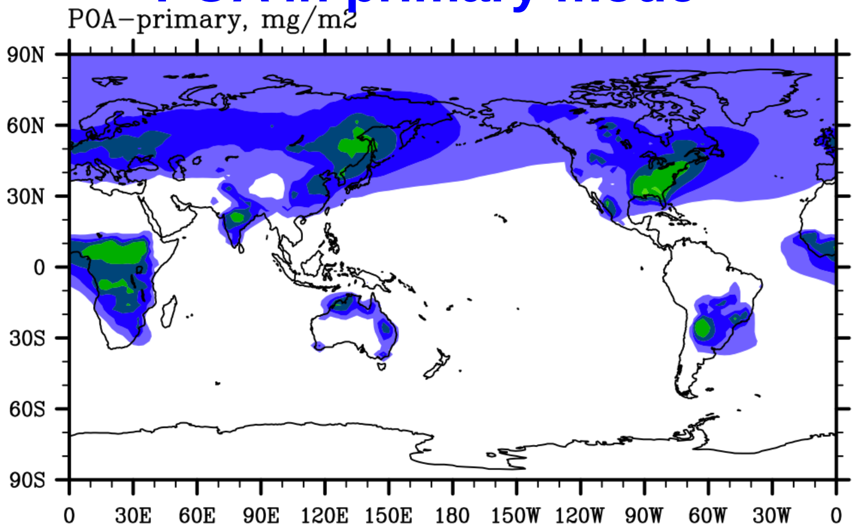




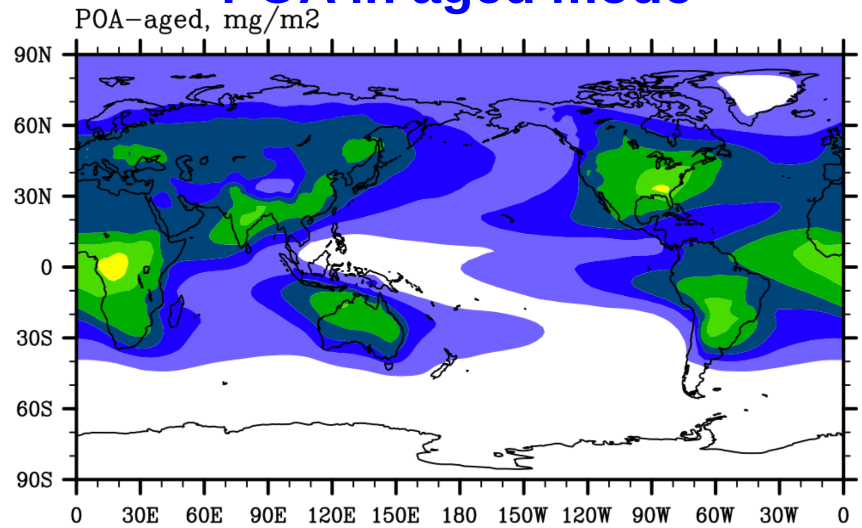
Control case
κ=0.0 (POA), κ=0.14 (SOA)

OA Column Burden at PI

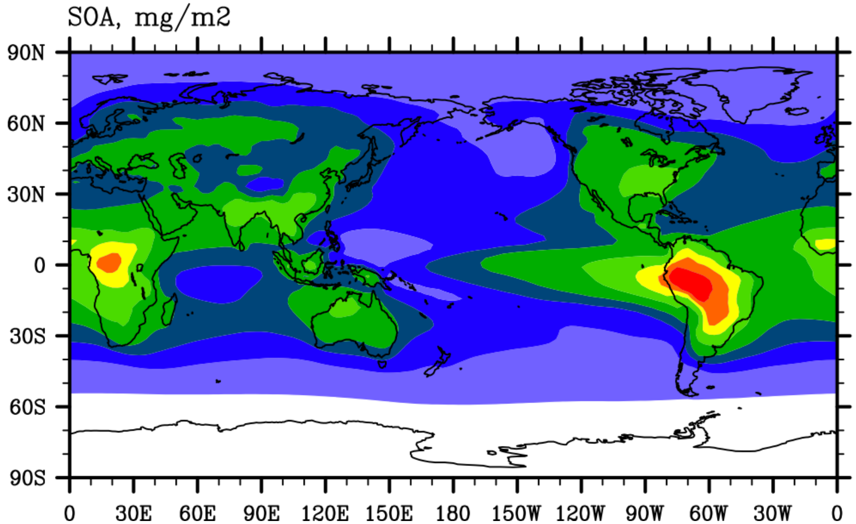
POA in primary mode

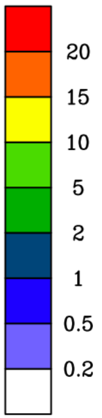


POA in aged mode



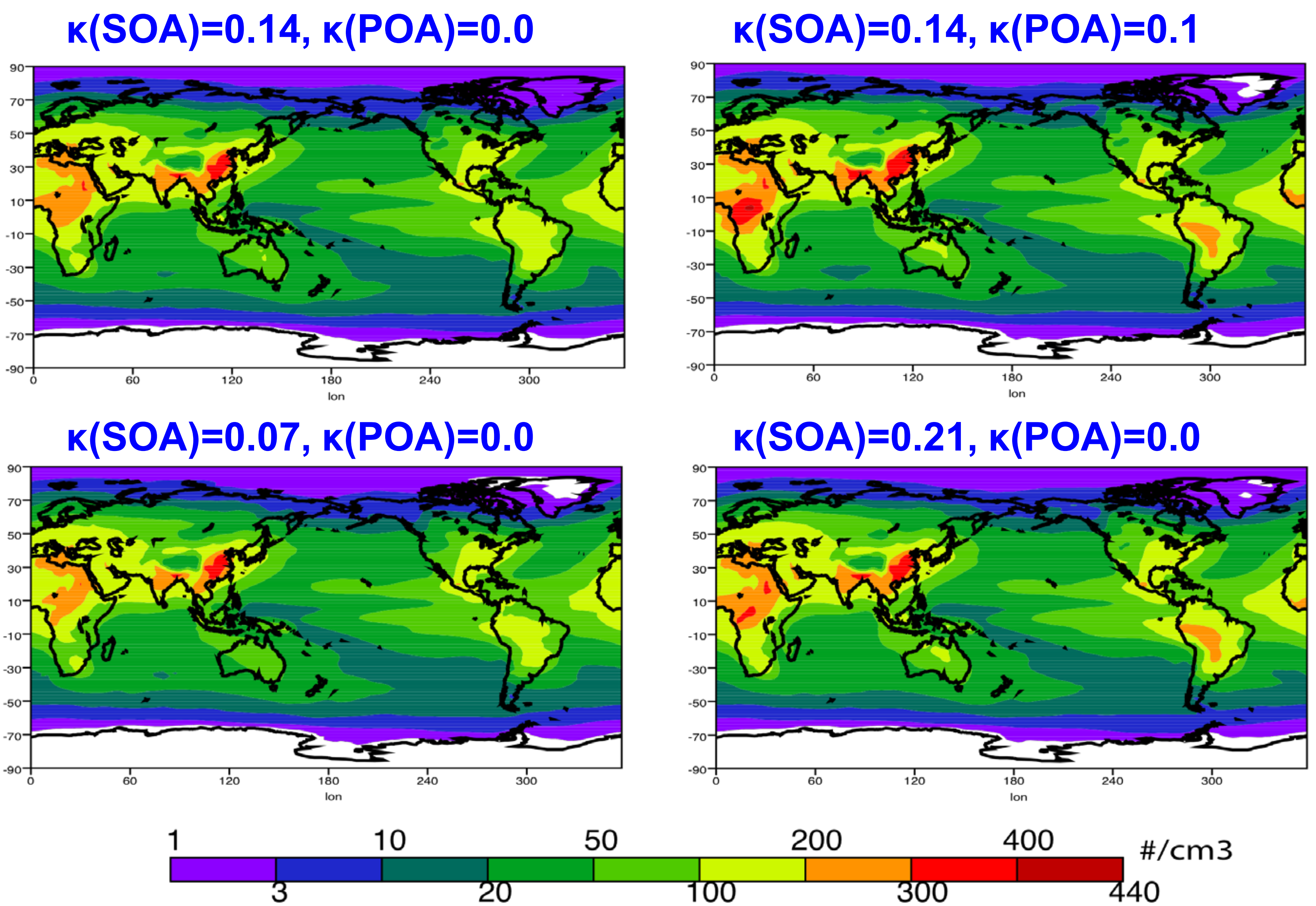
SOA





Control case
κ=0.0 (POA), κ=0.14 (SOA)

CCN (S=0.1%) at 859 hPa at PD

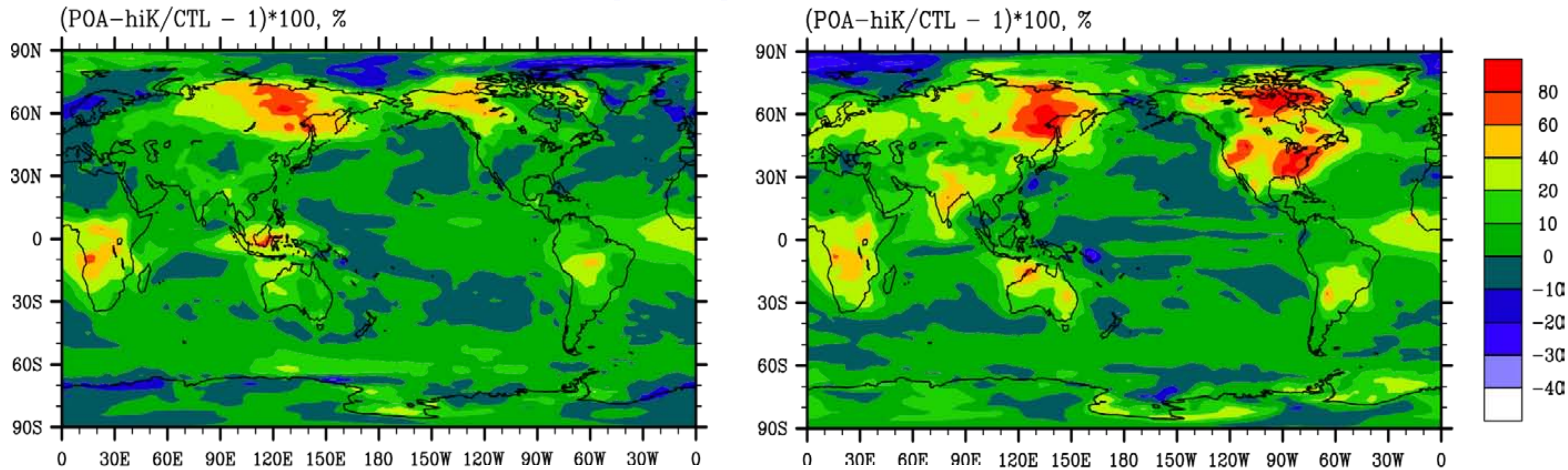


Percentage Change in CCN (S=0.1%) at 859 hPa

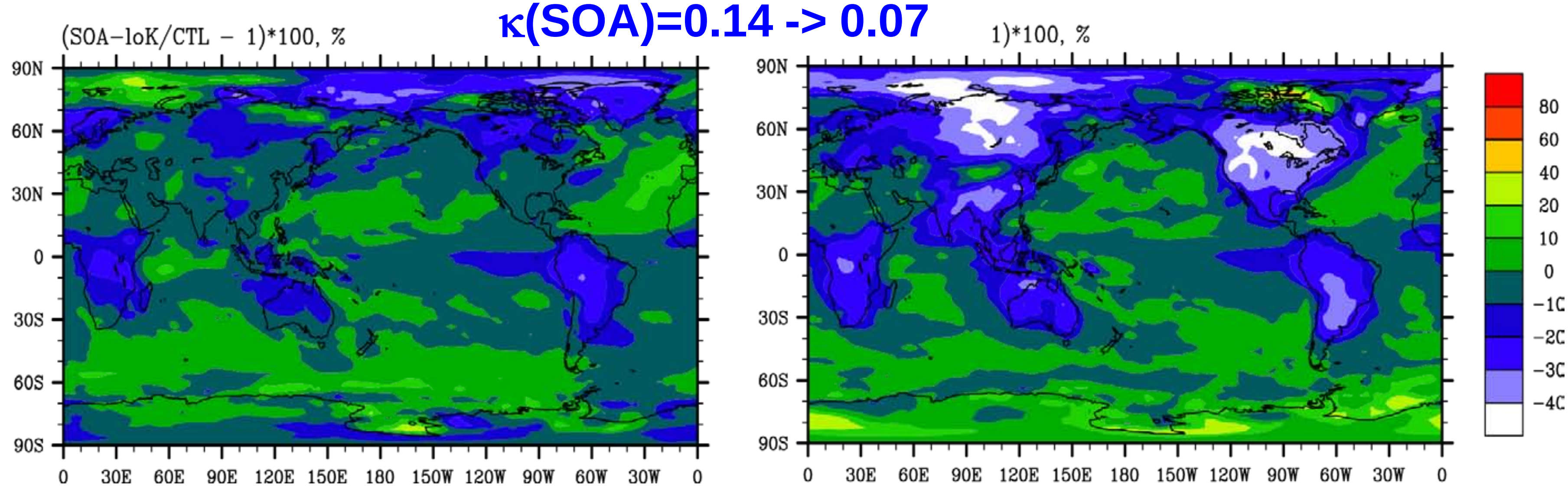
PD

κ(POA)=0.0 -> 0.1

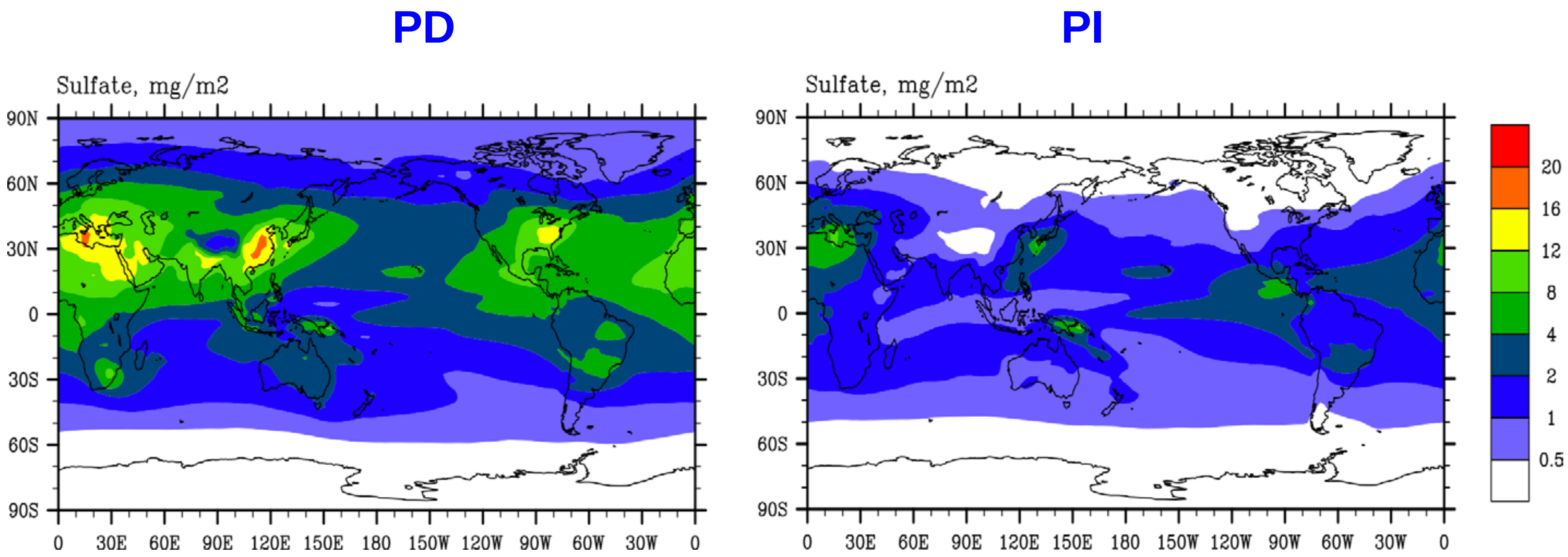
PI



κ(SOA)=0.14 -> 0.07



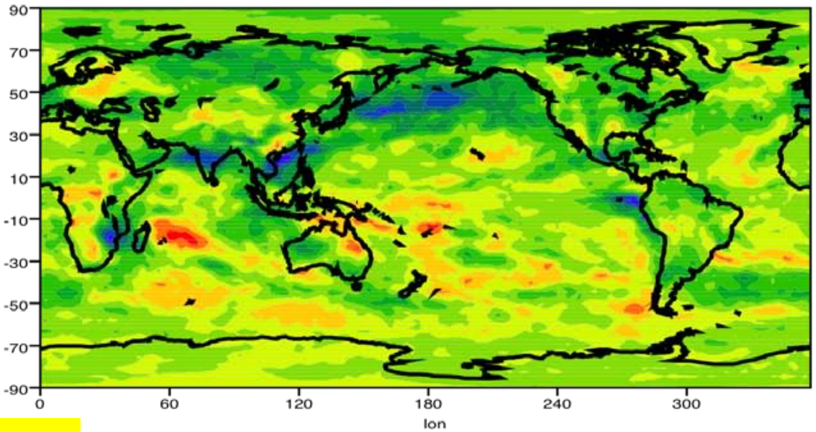
SO4 Column Burden



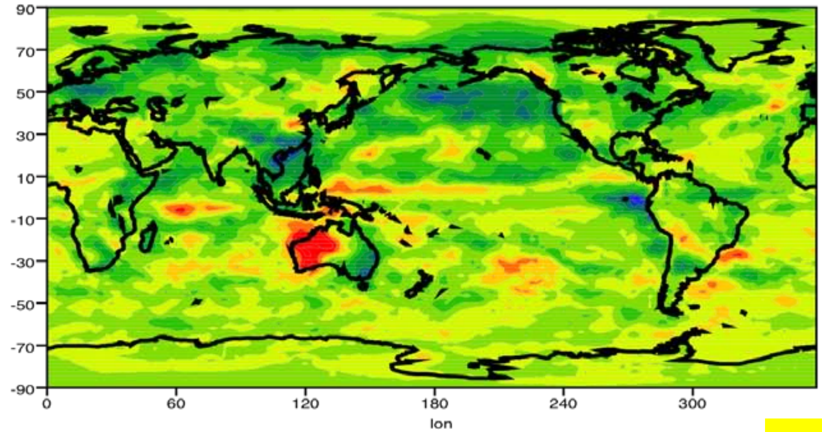
At PD, after organics are internally mixed with substantial amount of sulfate, particle hygroscopicity (κ) is dominated by sulfate, and is relatively insensitive to the change in organics hygroscopicity (κ).

Indirect Forcing (ΔSWCF, PD-PI, W/m2)

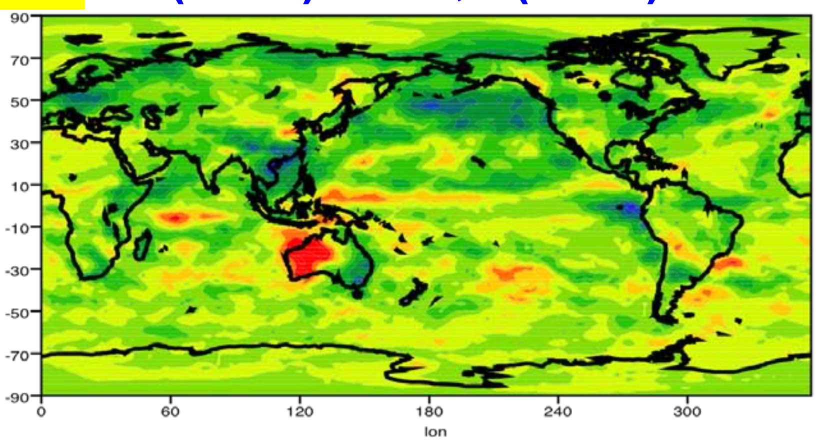
-1.26 κ(SOA)=0.14, κ(POA)=0.0



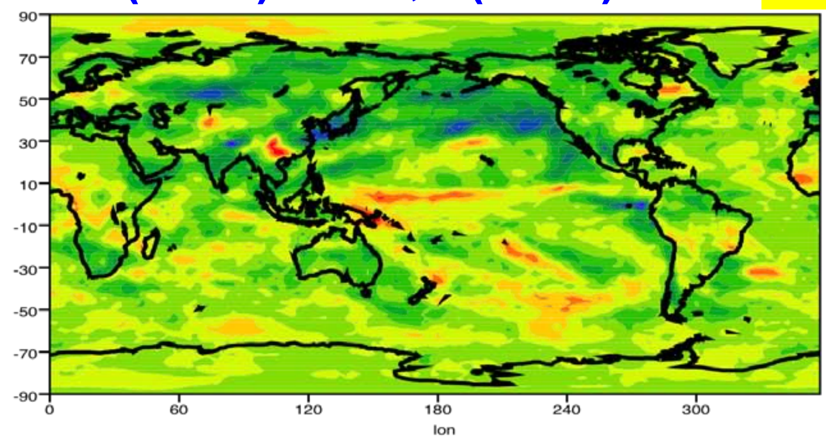
κ(SOA)=0.14, κ(POA)=0.1 -1.10

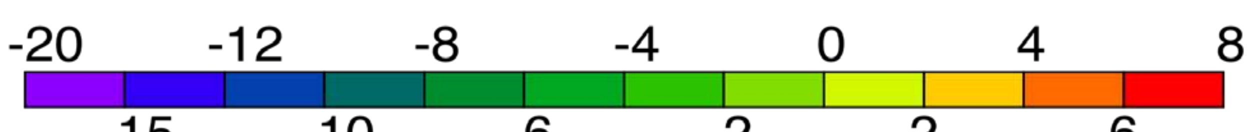


-1.47 κ(SOA)=0.07, κ(POA)=0.0



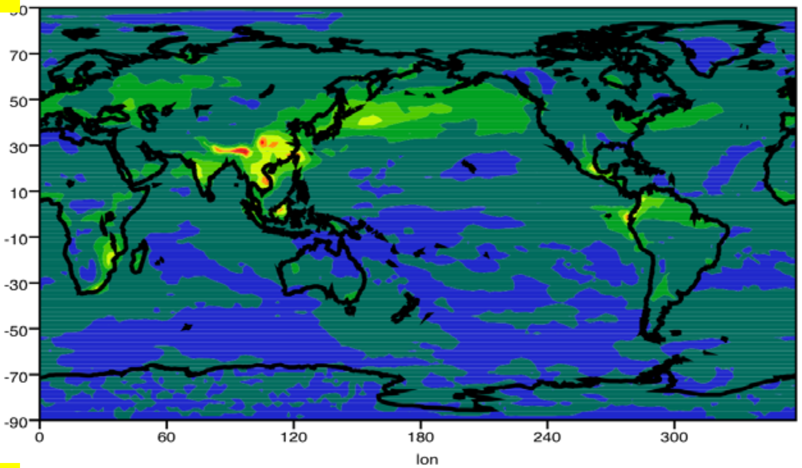
κ(SOA)=0.21, κ(POA)=0.0 -1.25



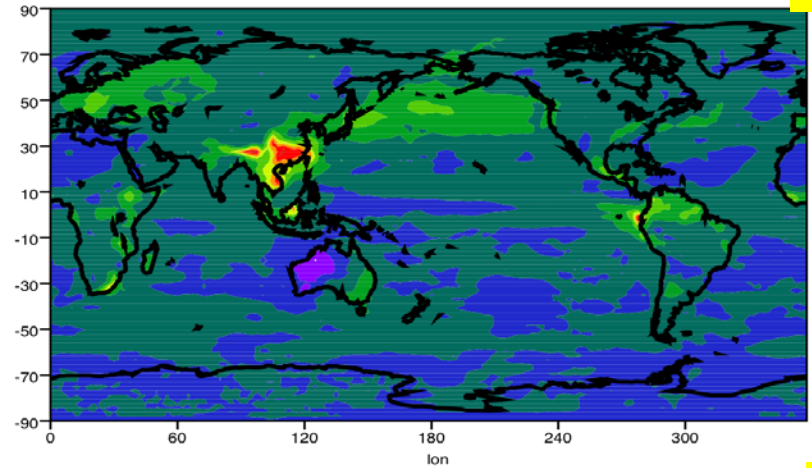


Indirect Forcing (ΔLWP, PD-PI, g/m2)

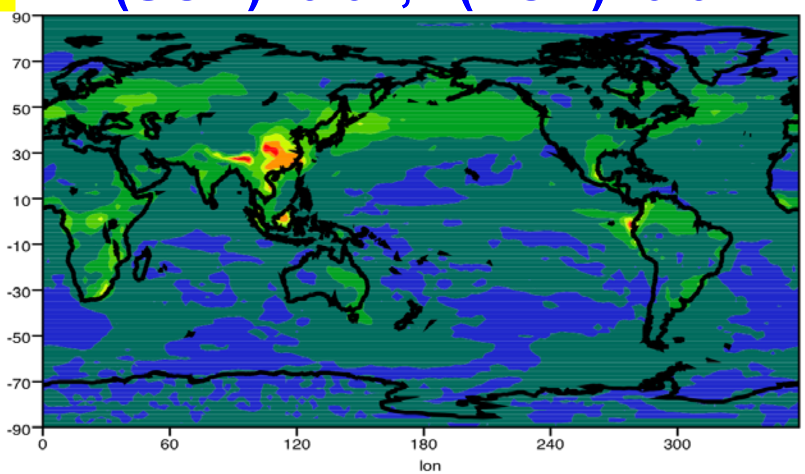
+3.9 κ(SOA)=0.14, κ(POA)=0.0



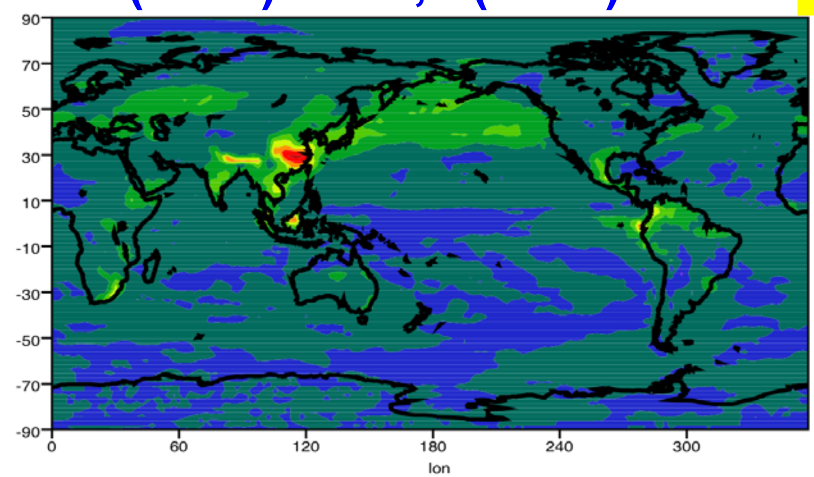
κ(SOA)=0.14, κ(POA)=0.1 +3.6

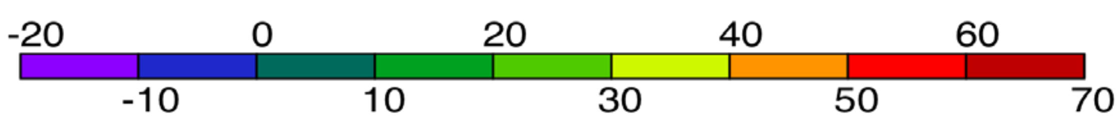


+4.6 κ(SOA)=0.07, κ(POA)=0.0

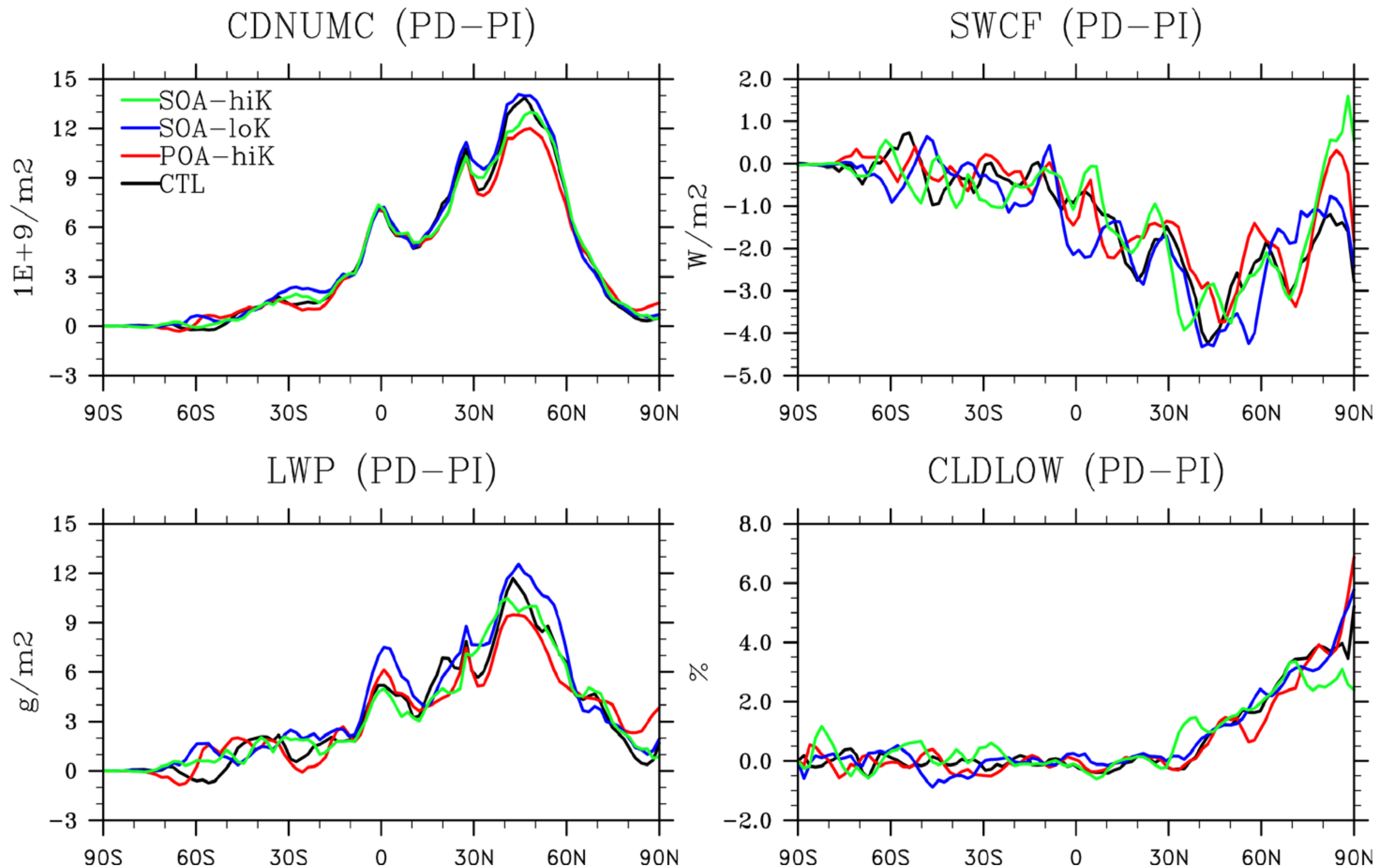


κ(SOA)=0.21, κ(POA)=0.0 +3.8





Changes between PD and PI



Summary

- Change in organic hygroscopicity causes 0.4 W/m2 (~30%) change in AIE. This uncertainty is comparable or even larger than those due to autoconversion parameterization and tuning parameters related to entrainment, drizzle and snow formation (Liu et al., 2008; Lohmann and Ferrachat, 2010).
- Higher hygroscopicity of organics (POA, SOA) reduces AIE
  - Disproportional larger increase of CCN at PI reduces droplet number increase from PI to PD, thus reduces AIE
- Future improvements:
  - Better characterization and representation of organics hygroscopicity, especially under PI conditions (Amazon).
  - More organic classes in models- separate biomass burning OA from fossil fuel OA
  - Representation of the variation of organic hygroscopicity during aerosol aging.

Acknowledgements

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