

Chemistry Climate Interactions

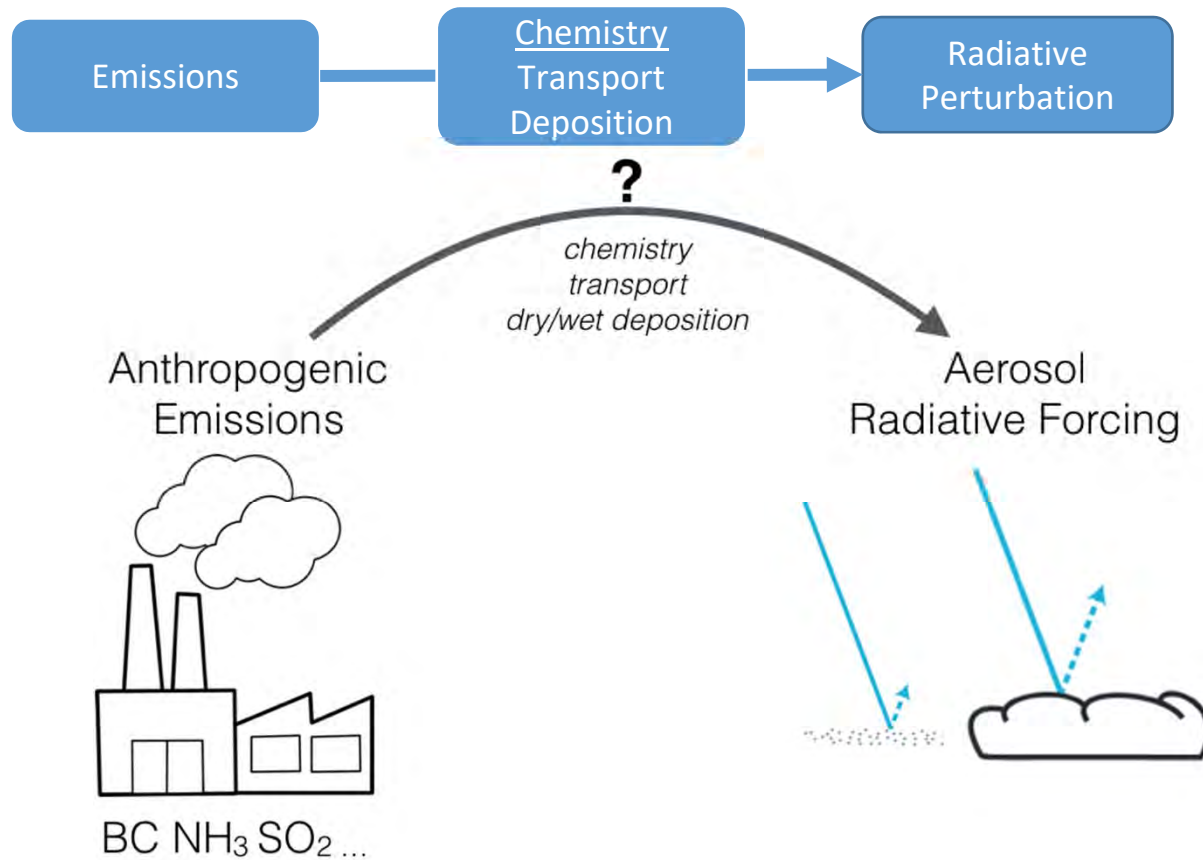
Fabrice Paulot

Laboratory Review

2019



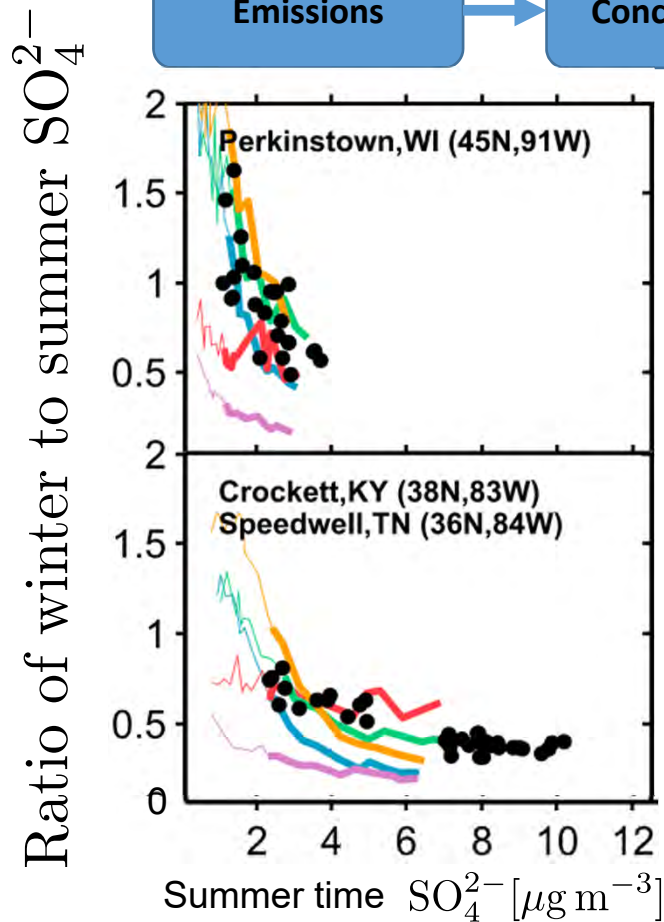
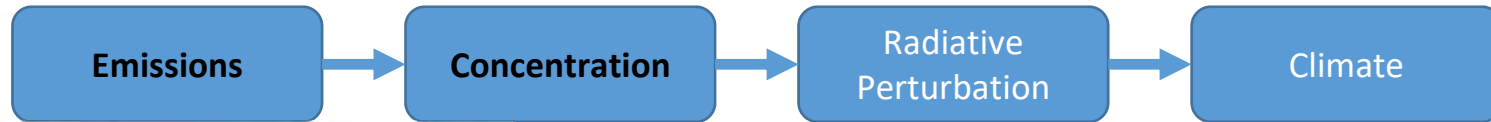
Going from emissions to climate impact: a chemical detour



- Leverage long-term observations to constrain relationship between emissions and concentration/radiative effects

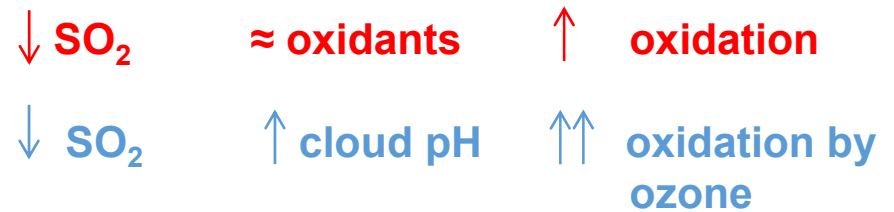
- Develop chemical mechanisms that can provide robust estimates under a wide range of emission conditions

Observed non-linearity in the response of sulfate



- SO_2 emissions have decreased by $\sim 70\%$ in the last 30 years. Long-term observations of sulfate in the US show **faster decrease in summer than in winter**

Mechanism

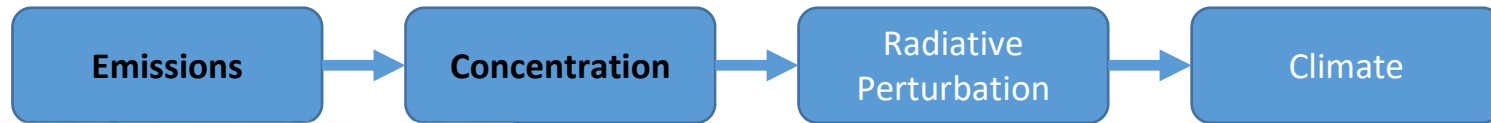


most important under low oxidant levels (winter)

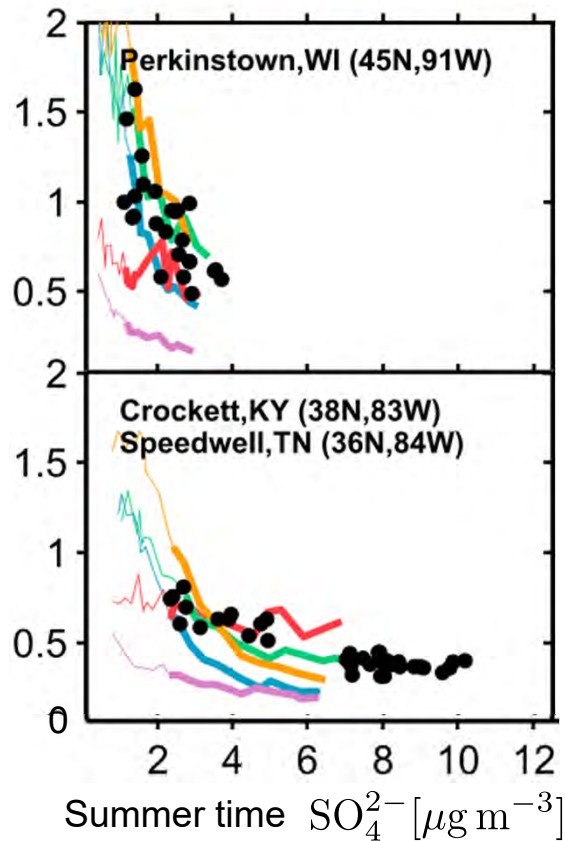
Observations	<u>Dynamic cloud pH</u>	<u>Constant cloud pH</u>
Base + Transition Metals	Base model - seasonal NH_3 emissions	Base model with PI NH_3

Paulot et al. (GRL, 2017)

Observed non-linearity in the response of sulfate



Ratio of winter to summer SO_4^{2-}



- SO_2 emissions have decreased by $\sim 70\%$ in the last 30 years. Long-term observations of sulfate in the US show **faster decrease in summer than in winter**

Mechanism



most important under low oxidant levels (winter)

Implications

- high sensitivity to NH_3 emissions
- SO_4 forcing does not follow SO_2 emissions

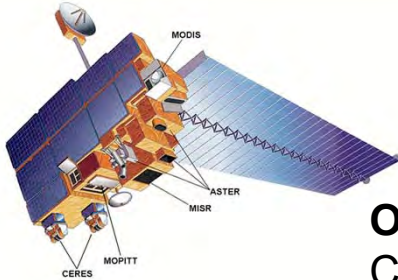
Observations Dynamic cloud pH Constant cloud pH

Base + Transition Base model - Base model with
Metals seasonal NH_3 PI NH_3
 emissions

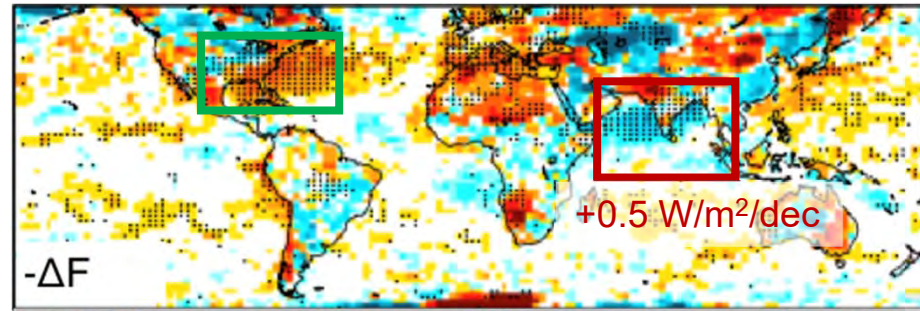
Paulot et al. (GRL, 2017)



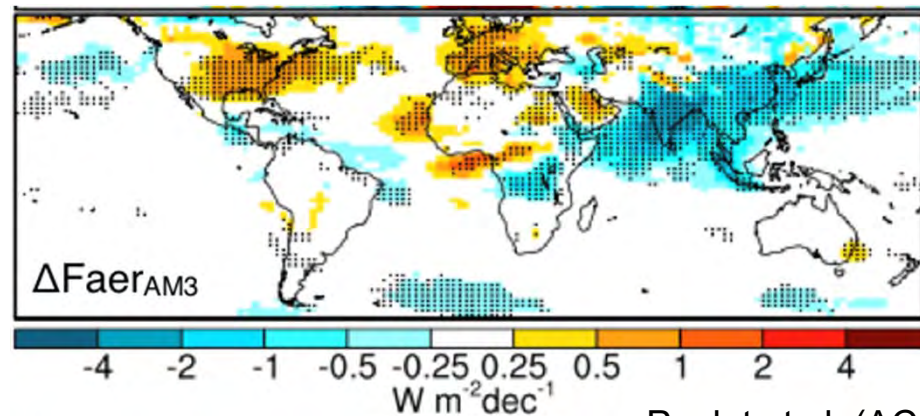
Extracting changes in aerosol effect from CERES



Observation (CERES)
Change in outgoing
clear-sky SW radiation
(2001-2015)

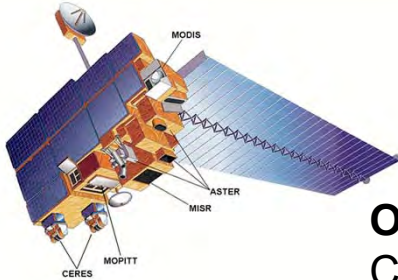


**Simulated change in
aerosol effect (AM3)**



Paulot et al. (ACP, 2018)

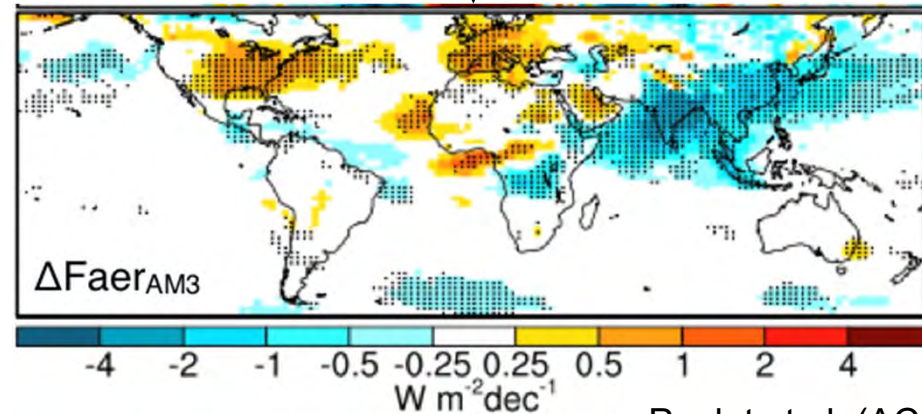
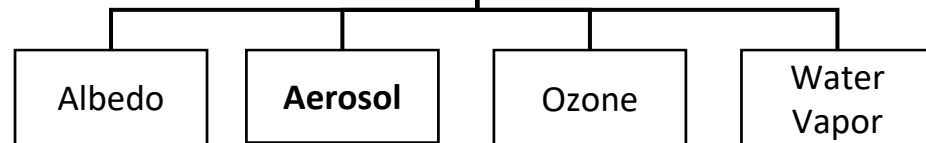
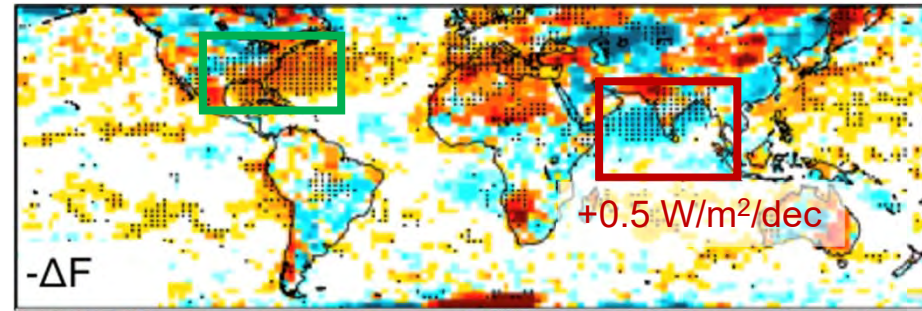
Extracting changes in aerosol effect from CERES



Observation (CERES)
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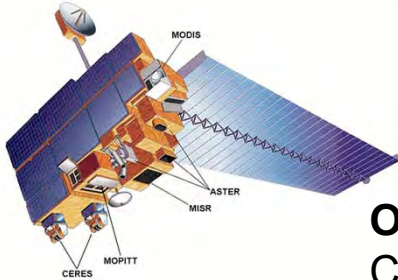
Use **RTM** to correct for
changes in albedo, water
vapor, and ozone

**Simulated change in
aerosol effect (AM3)**



Paulot et al. (ACP, 2018)

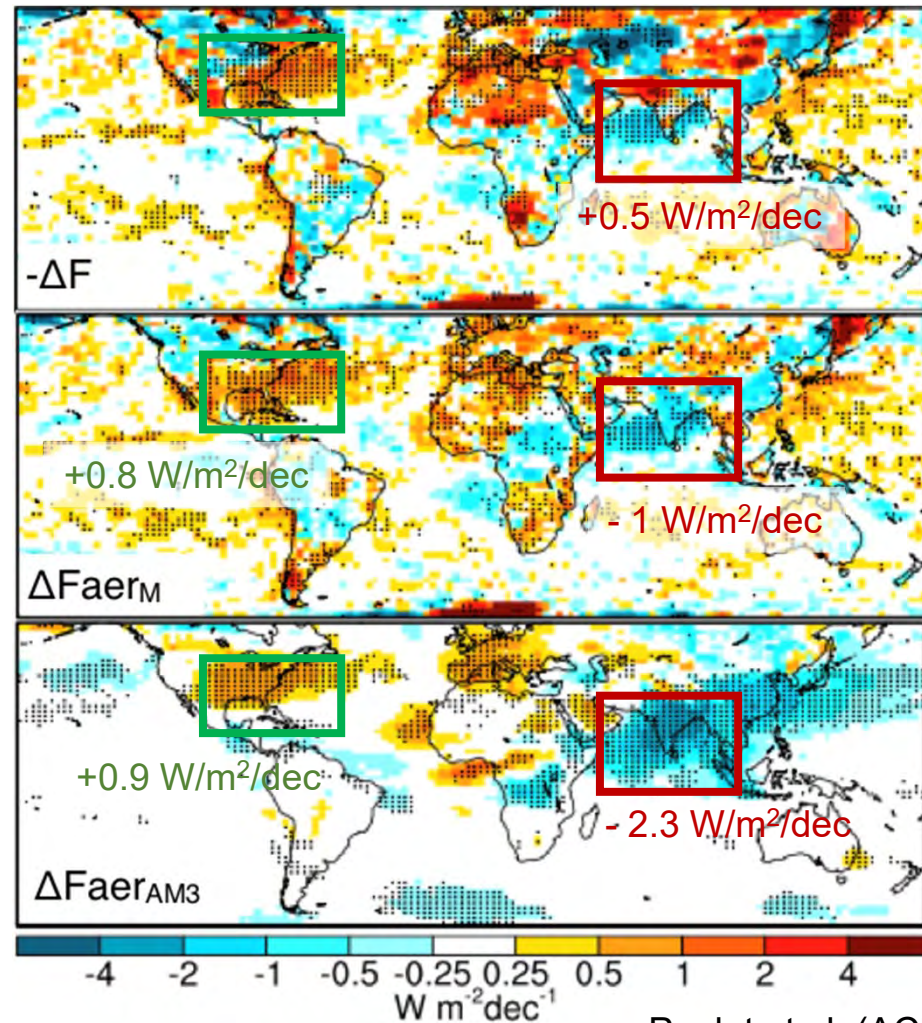
Regional constraints on changes in the aerosol effect



Observation (CERES)
Change in outgoing clear-sky SW radiation (2001-2015)

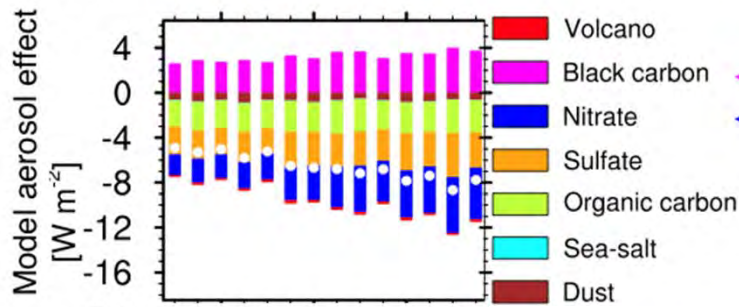
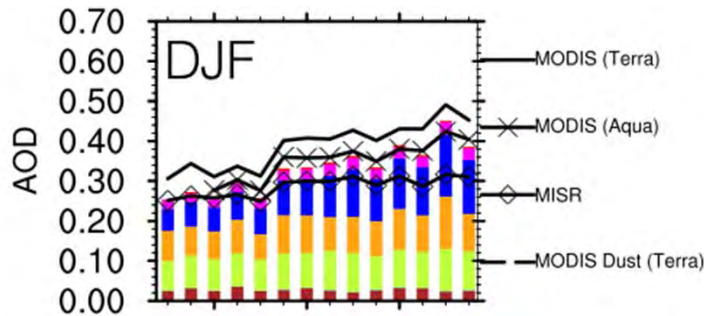
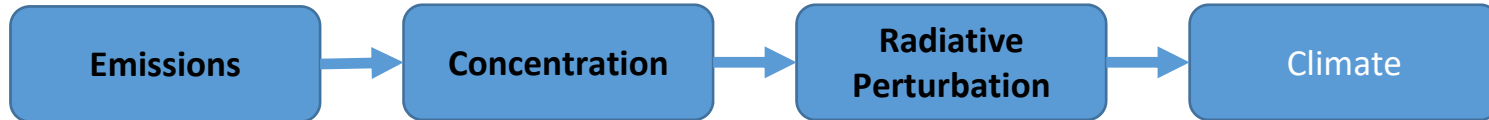
Estimated change in aerosol effect
observation corrected for albedo, water vapor, and ozone using RTM

Simulated change in aerosol effect

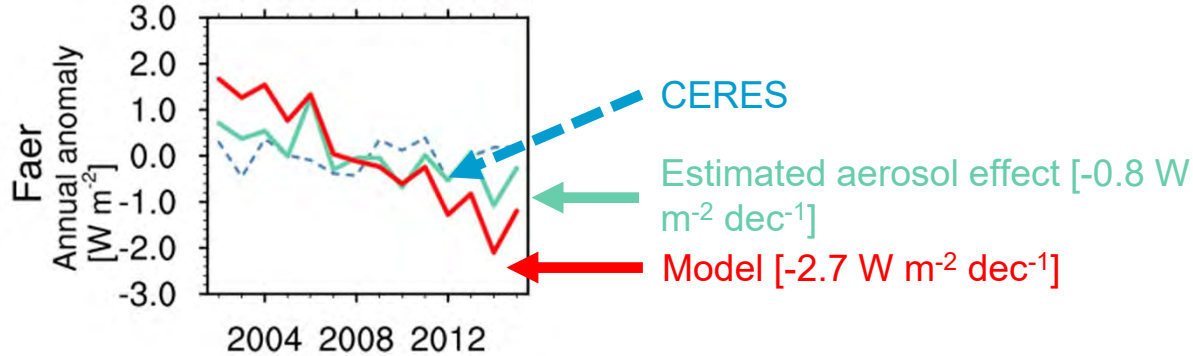
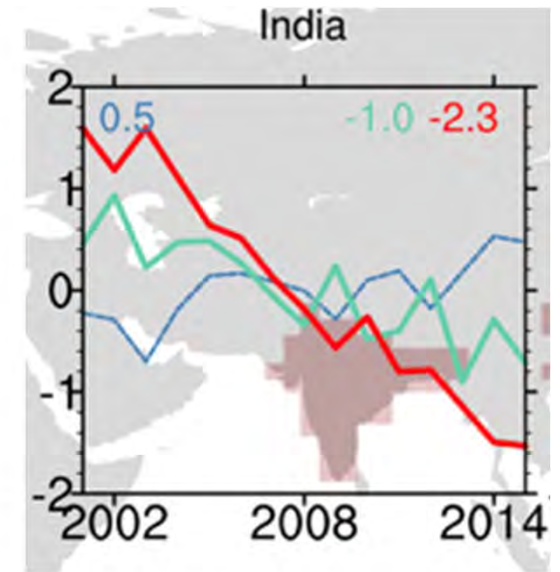


Paulot et al. (ACP, 2018)

Leverage seasonal trends to understand source of biases over India: nitrate and BC



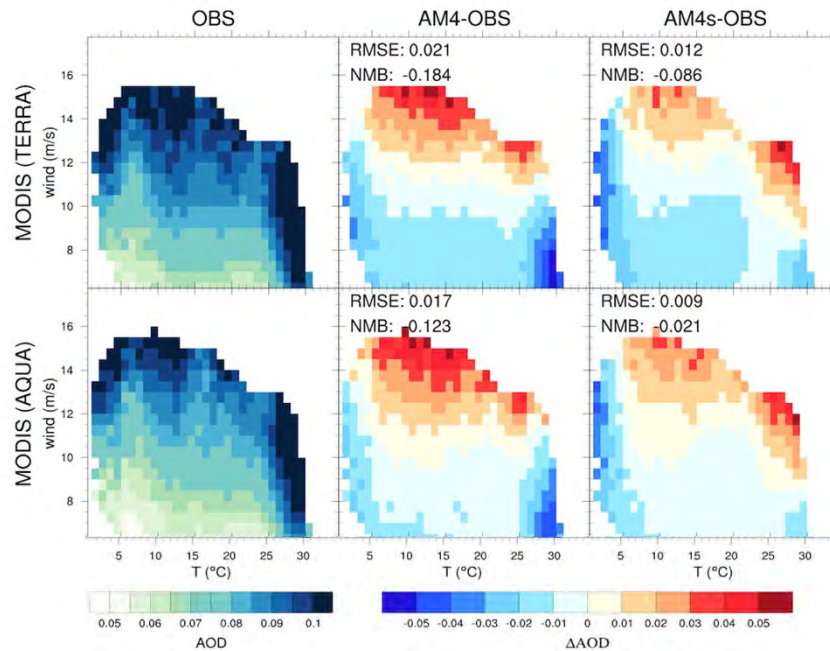
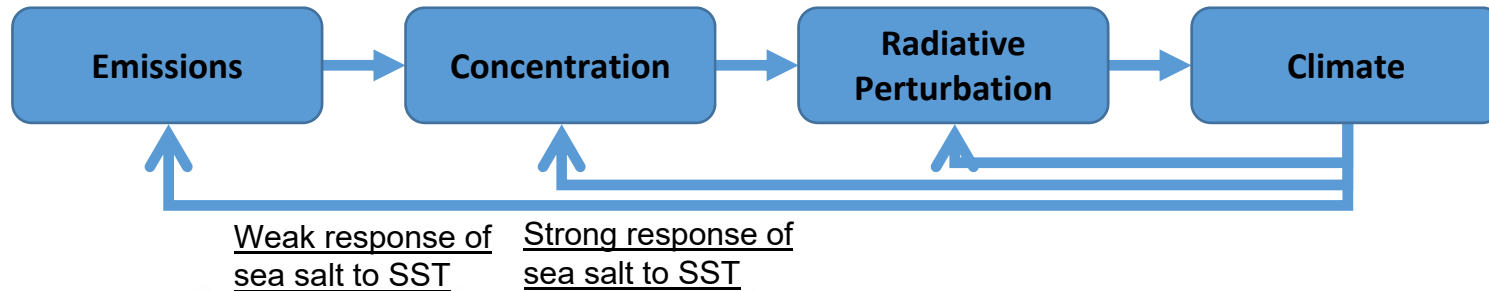
$0.9 \text{ W m}^{-2} \text{ dec}^{-1}$
 $-2.4 \text{ W m}^{-2} \text{ dec}^{-1}$
 => included in ESM4



- Additional information helps constraints sources of biases
- Changes are not dominated by sulfate

Paulot et al. (ACP, 2018)

Closing the loop: sea salt radiative feedback



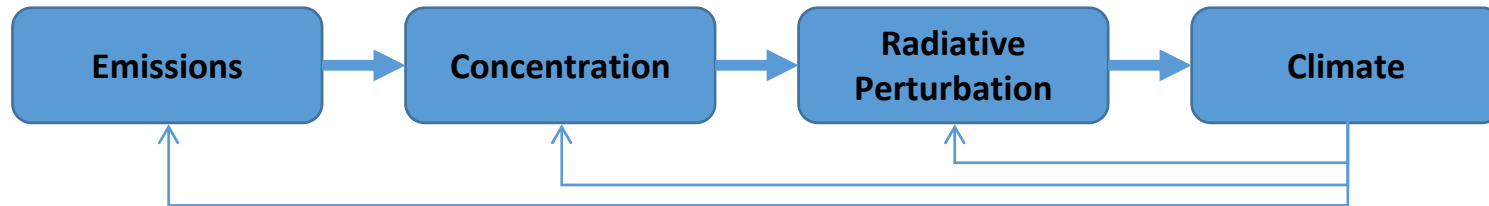
Recent field/lab experiments point to a **sensitivity of sea salt emissions to SST**

Sea salt aerosols – **most important scattering aerosol** in the atmosphere.

Use nudged configuration to compare simulated and observed relationship between **AOD, SST, and wind speed** over “pristine” marine regions

Strong T dependence reduces model bias at high temperature (equatorial/subtropical Pacific)

Closing the loop: natural aerosols feedback



	ECS [°C]
CM4 Weak response of sea salt to SST	5
ESM4 Strong response of sea salt to SST	3.1
CM4s Strong response of sea salt to SST	4.5

- Change in sea salt parameterization contribute ~25% of the difference in climate sensitivity between CM4 and ESM4.
- Other factors include: stratospheric ozone, biogenics, land, ...
- **Negative feedback $\sim -0.08 \text{ W m}^{-2} \text{ K}^{-1}$**
AerChemMIP Considerable uncertainty on the size distribution, temperature sensitivity, and radiative effect of sea salt emissions.
- Highlights uncertainty in the representation of marine chemistry

Summary & Future plans

- High-frequency/long-term observations can provide emerging constraints for the sensitivity of climate forcers to changes in anthropogenic emissions/climate. [Paulot et al. (2017ab GRL), Paulot et al. (2018, ACP), He et al. (2019, ACP)]
- Comprehensive coupling between land/ocean/atmosphere to improve and advance the scientific understanding of the role of biogeochemical feedbacks on climate and air quality
 - Interactive emissions
 - Interactive fire emission (Ginoux et al., in preparation)
 - Agricultural activities (-> Minjin Lee's talk)
 - Natural aerosols (dust (in ESM4), marine aerosols)
 - Interactive deposition (-> Meiyun Lin's talk)