

Exploring natural aerosol formation from DMS oxidation and implications for aerosol forcing

Exploring natural aerosol formation from DMS oxidation and implications for aerosol forcing
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Goal: Better Estimation of Aerosol Forcing

Key Results: Sulfate Aerosol and Its Radiative Effects

1) Diversified Products of DMS Oxidation

Figure 6. Branching ratios (%) of the multistage DMS oxidation pathways in our modified GCM-Chem simulation considering HPHMT, SO₂, and MSAs as the branching products. The key intermediate and diverse fates of SO₂ are presented in parallel reaction rates while their color indicates phases of the corresponding reactions. The remaining 2% of DMS will be further oxidized to SO₂, and then SO₂ & MSAs will convert into sulfate.

SO ₂	35%
MSA	45%
HPHMT	22%

1) DMS

Next: Indirect Effects and Forcing

The results shown before are preliminary and will await further analyses.

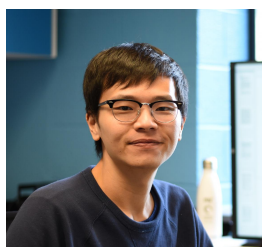
1) Indirect Radiative Effect

Figure 10. Parameters related to indirect radiative effect (IRE) in this study. IRE is defined as change in direct radiative forcing of a subsequent generation. For each of a 100-year time slice, the model simulates the atmosphere and the oceans at the beginning and the end of the time slice. The difference in the CO₂ concentration is 100 ppm. The IRE is defined as the difference in the radiative forcing at the end of the time slice minus the radiative forcing at the beginning of the time slice, divided by the change in CO₂ concentration.

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GOAL: BETTER ESTIMATION OF AEROSOL FORCING

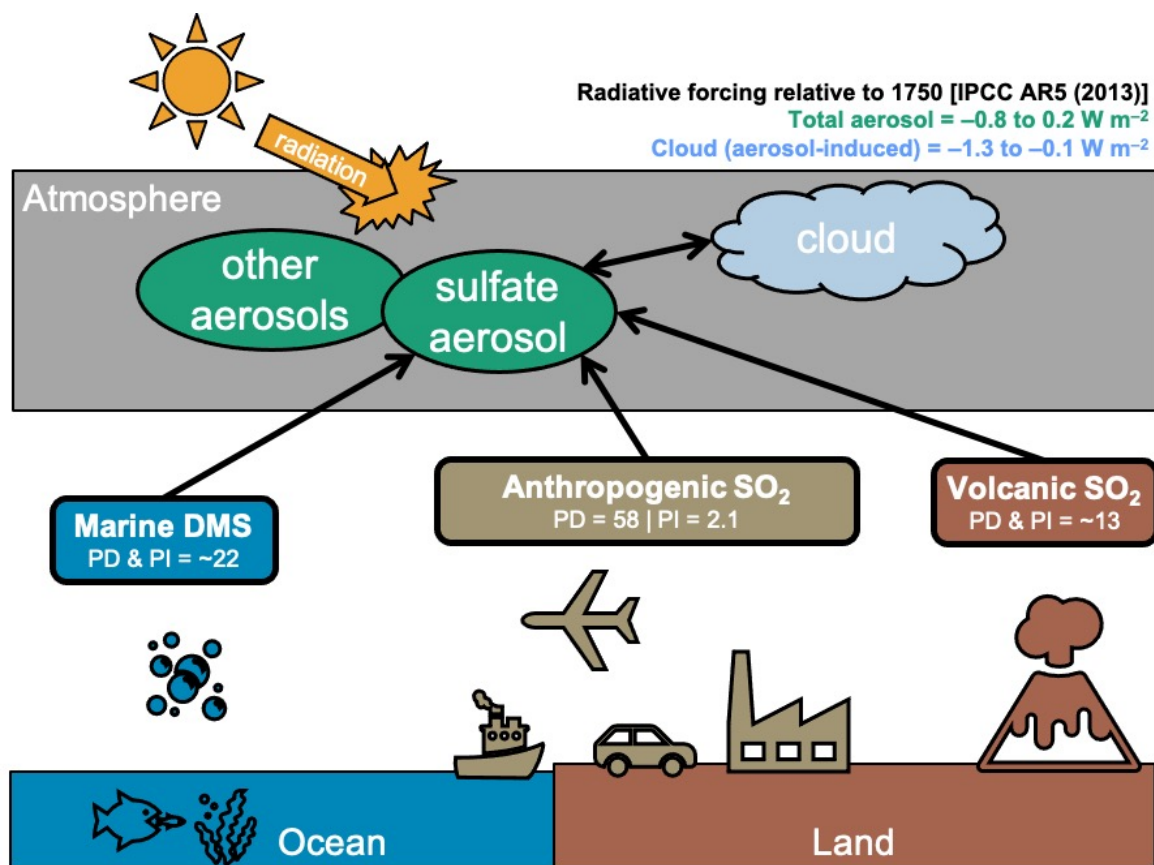
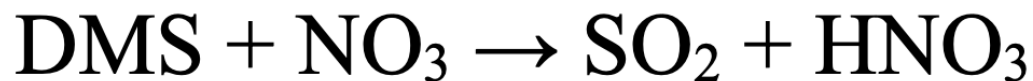
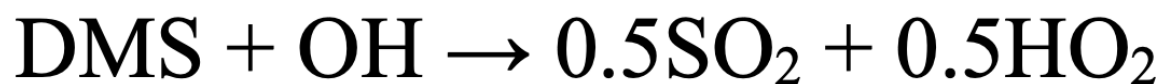


Figure 1. Primary sources of sulfur and their annual global fluxes in Tg-S yr^{-1} at present-day (PD) and pre-industrial (PI) levels [CEDS, Hoesly *et al.* (2017)]. Some of the sulfur results in sulfate aerosol production. Sulfate aerosol alters the Earth's radiative budget both directly (scattering) and indirectly (facilitating cloud formation). Large uncertainties remain in recent aerosol radiative forcing estimations.

- Atmospheric aerosol particles interact with the incoming and outgoing radiation directly (via scattering and absorption) and indirectly (via changing cloud properties and lifetime).
- According to IPCC AR5 (2013), aerosols are the dominant contributor to uncertainty in global climate forcing.
- To reduce such uncertainty, it's important to accurately determine the formation of aerosols and their radiative effects in both the present-day (PD) and pre-industrial (PI) atmospheres.
- Marine dimethyl sulfide (DMS) is the primary natural source and has been the largest contributor to sulfate aerosols before anthropogenic SO_2 took up the place since the industrial revolution.
- Over the pristine marine atmosphere, the direct radiative effect (DRE) of sulfate yielded from DMS oxidation is considerably greater than its direct radiative forcing (DRF), highlighting the crucial role os natural aerosols play in evaluating the radiative budget of the Earth.

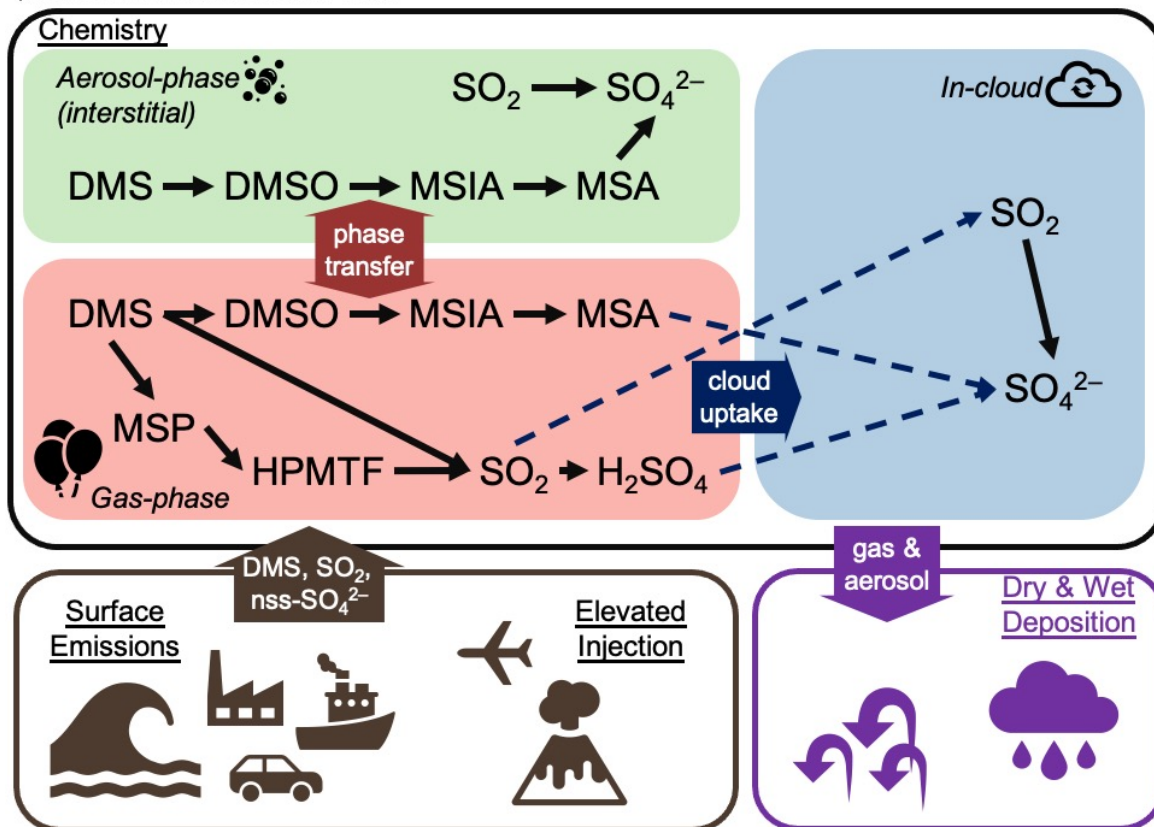
- Similar to most global circulation models (GCMs), in consideration of computation cost, the default version of the Community Earth System Model (CESM) uses three single-step reactions to represent DMS oxidation:



- Oversimplifying DMS oxidation chemistry may undercut the ability of the model to estimate sulfate formation and its climate effects.
- Thus, this study focuses on evaluating how a comprehensive DMS oxidation scheme in CESM can better quantify natural sulfate aerosol formation, its radiative effect, and its climate forcing.

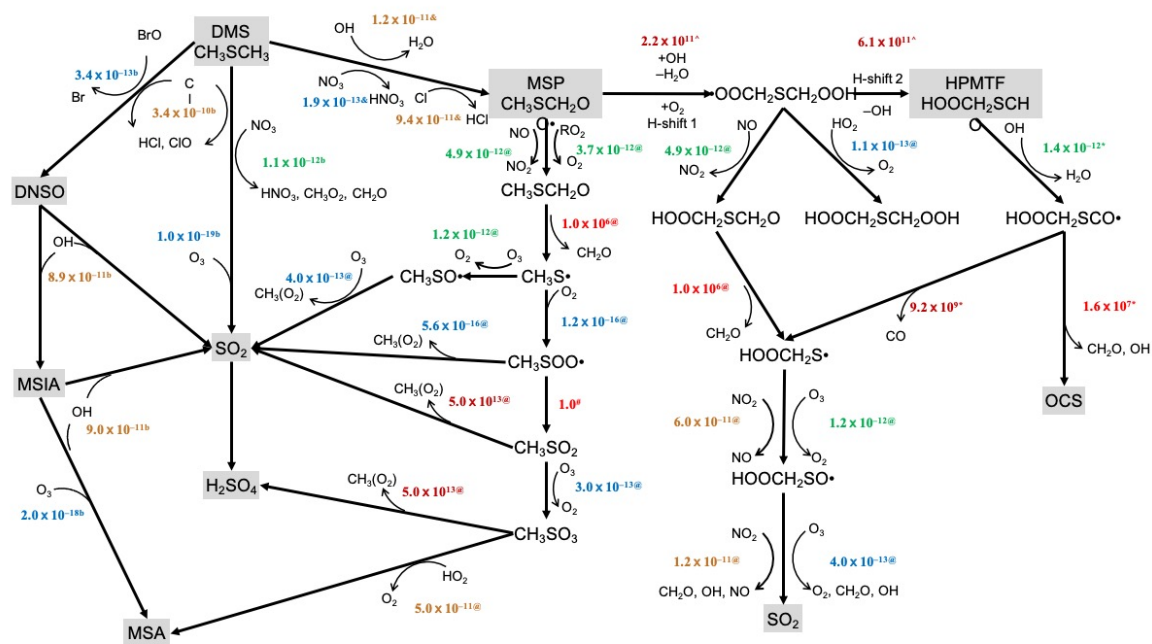
EXPANDING THE DMS OXIDATION CHEMISTRY IN CESM2

Figure 2. Sulfur cycle in this study's modified CAM6-chem. We expand the gas- and aerosol-phase chemistry to include important intermediates such as MSA, and HPMTF. Sea-to-air DMS fluxes are calculated online while emissions of SO₂ and non-sea-salt sulfate are prescribed [Hoesly *et al.* (2017)]. Cloud uptake and deposition processes follow standard CAM6-chem.



- Our study based on the Community Earth System Model (CESM) version 2.1.
- The land component is the Community Land Model (CLM) version 5 with Satellite Phenology, with 2000-year land use.
- The atmosphere component is driven by the Community Atmosphere Model version 6 with chemistry (CAM6-chem). SO₂ emissions are prescribed by emission inventories at the 2000-level. DMS emission is based on OASISS which calculates online sea-to-atmosphere DMS flux based on Lana *et al.* (2011) sea-surface DMS climatology. Sea-surface temperature also remains constant at the 2000 level. Aerosol simulation is based on the MOSAIC model for CAM-chem.
- We expand the DMS oxidation chemistry in CAM6-chem by adding two new pathways based on recent literature, which are
 1. A multiphase addition pathway produces DMSO and MSIA as intermediates and yielding MSA, SO₂, and sulfate as final products.
 2. A gas-phase H-abstraction pathway that produces a series of short-lived organ-sulfur chemicals and yields MSA and SO₂.
- The detailed gas-phase and aqueous-phase DMS oxidation chemistry scheme are shown below:

Figure 3. The expanded gas-phase DMS oxidation scheme in this study.

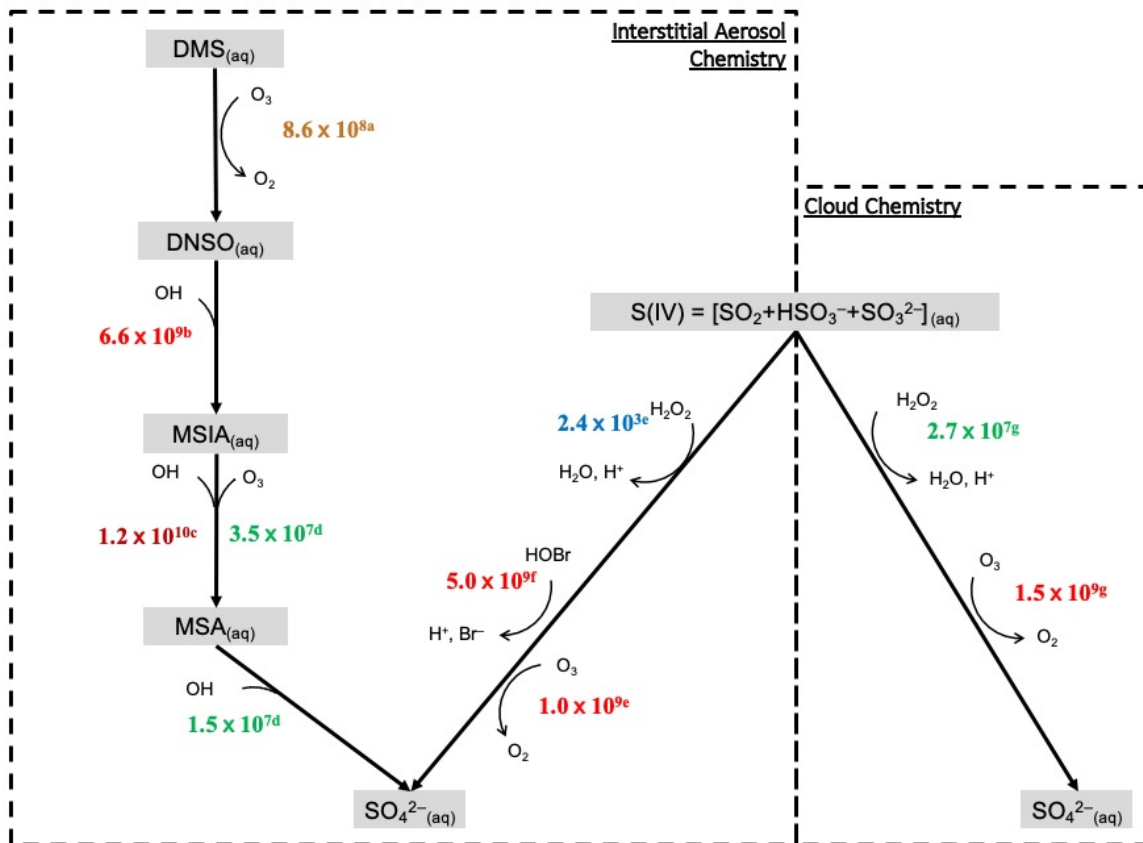
**Rate Constants (s⁻¹ or cm³ molecule⁻¹ s⁻¹)**

very fast
fast
medium
slow
very slow

Reference:

& Cuevas *et al.* (2018)
^ Veres *et al.* (2020)
* Wu *et al.* (2015)
@MCM, Saunders *et al.* (2003)
Campolongo *et al.* (1999)
b Bukrholder *et al.* (2015)

Figure 4. The expanded aqueous-phase DMS oxidation scheme in this study.

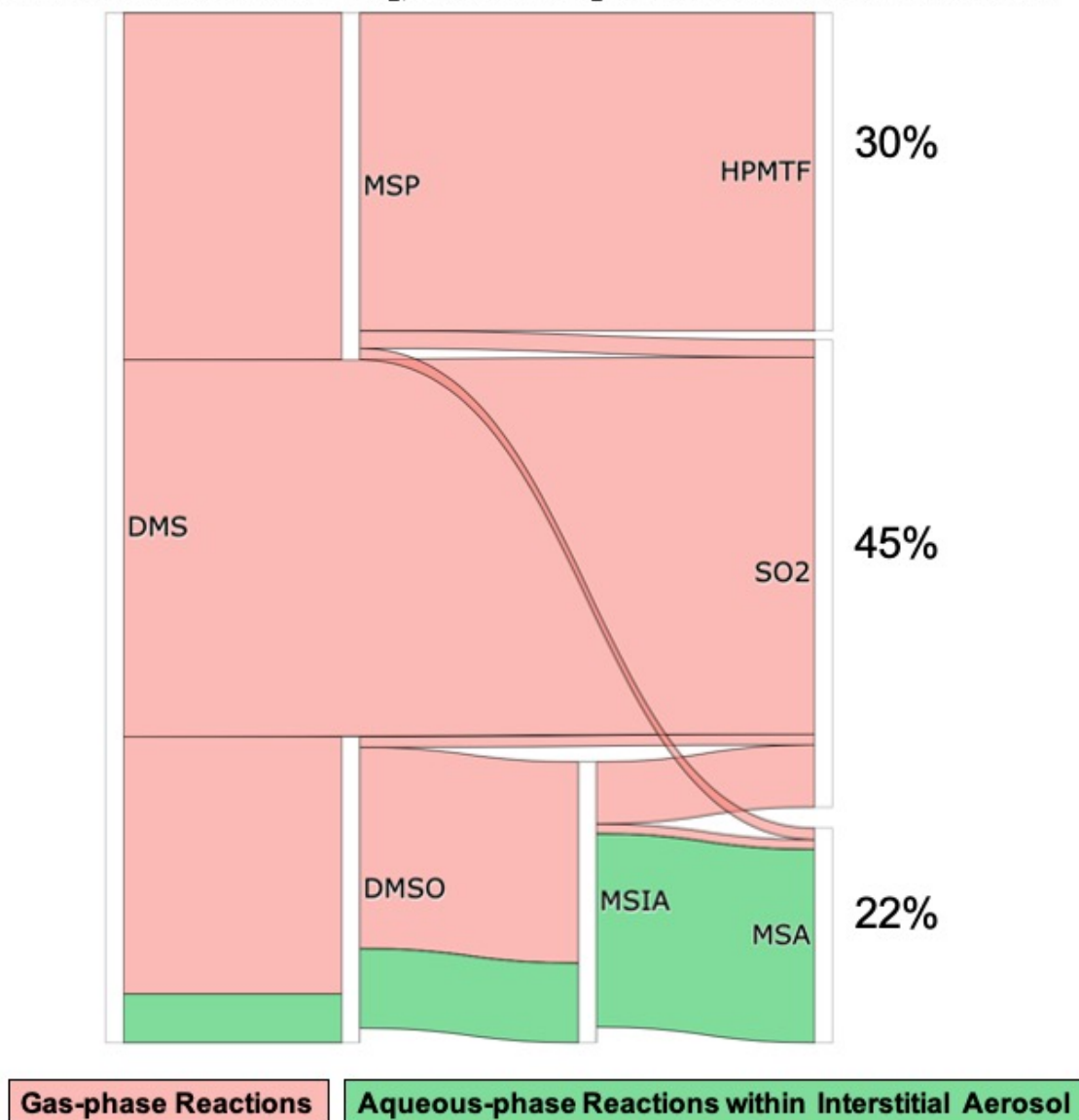
**Rate Constants (M⁻¹ s⁻¹)****very fast****fast****medium****slow****very slow****Reference:**

- a Gershenzon *et al.* (2001)
 b Zhu *et al.* (2003)
 c Bardouki *et al.* (2003)
 d Hoffmann *et al.* (2016)
 e Jacob (1986)
 f Troy & Margerum (1991)
 g Hoffmann & Calvert (1985)

KEY RESULTS: SULFATE AEROSOL AND ITS RADIATIVE EFFECTS

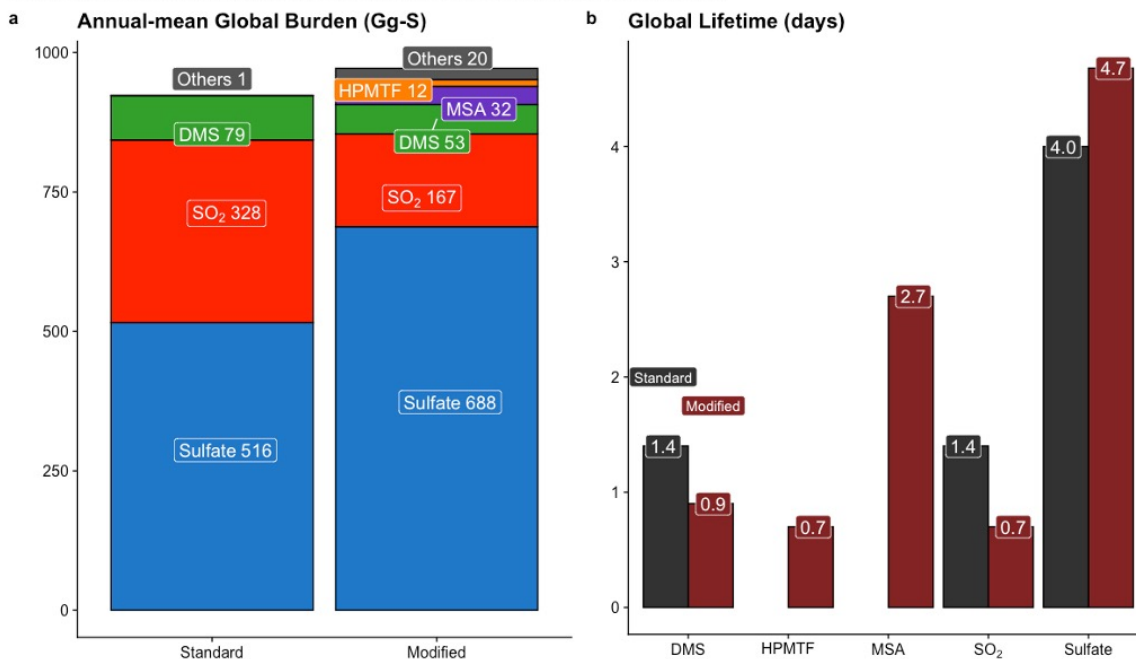
1) Diversified products of DMS oxidation

Figure 5. Branching ratios (%) of the multi-phase DMS oxidation pathways in our modified CAM6-chem simulation (considering HPMTF, SO₂, and MSA as the terminating products). Only key intermediates are shown. Widths of ribbons are proportionate to global-total reaction rates while their colors indicate phases of the corresponding reactions. The remaining 3% of DMS-sulfur losses to dry/wet deposition. Our model assumes that HPMTF will be further oxidized to SO₂, and that SO₂ & MSA will convert into sulfate.



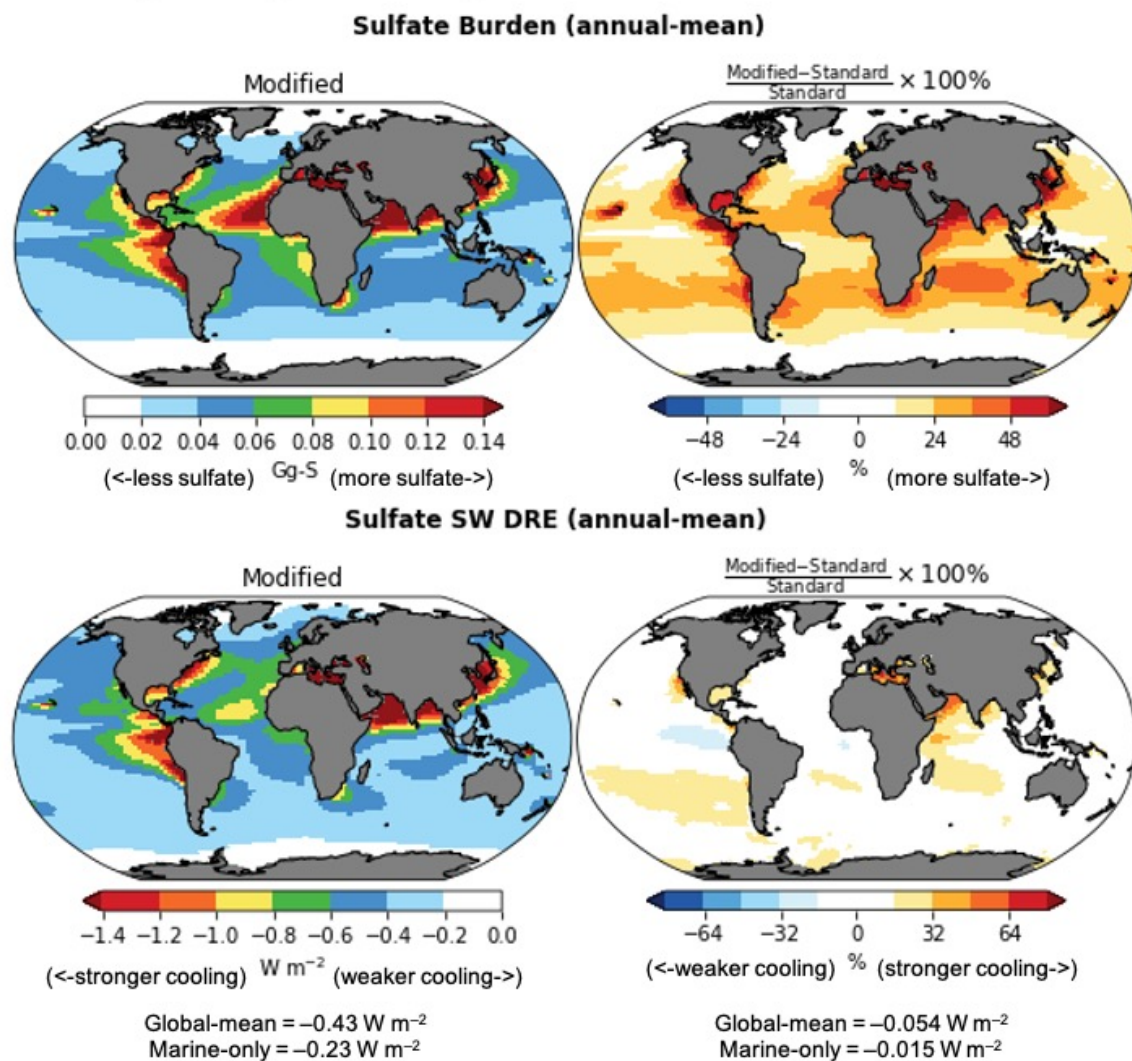
2) Longer lifetime of sulfate and other atmospheric sulfur species

Figure 6. Burden and lifetime of atmospheric sulfur compounds in the model runs using our modified chemistry ("Modified") and CAM6-chem's default chemistry ("Standard"). Our expanded chemistry diverts some DMS-derived SO₂ to MSA and HPMTF, increasing the total global sulfur burden. MSA and HPMTF, which have longer lifetimes than SO₂, delay the production of sulfate, allowing sulfate to form further downwind from its sources.



3) Increased marine sulfate burden and enhanced direct cooling effects

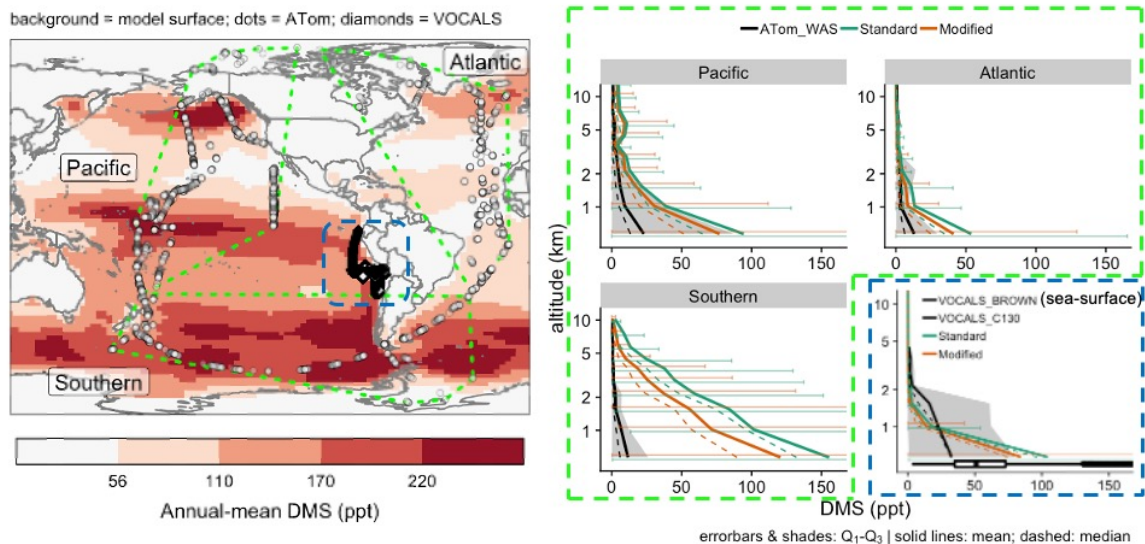
Figure 7. Sulfate burden (Gg-S) and shortwave direct radiative effect (SW DRE; Wm^{-2}) from our model runs. Only marine values are shown. Yet sulfate burden is higher along coastlines near anthropogenic sources, our expanded chemistry has resulted in substantial increases in sulfate burden over Pacific, allowing for more SW scattering and leading to stronger local cooling effects.



MODEL VS. OBSERVATIONS

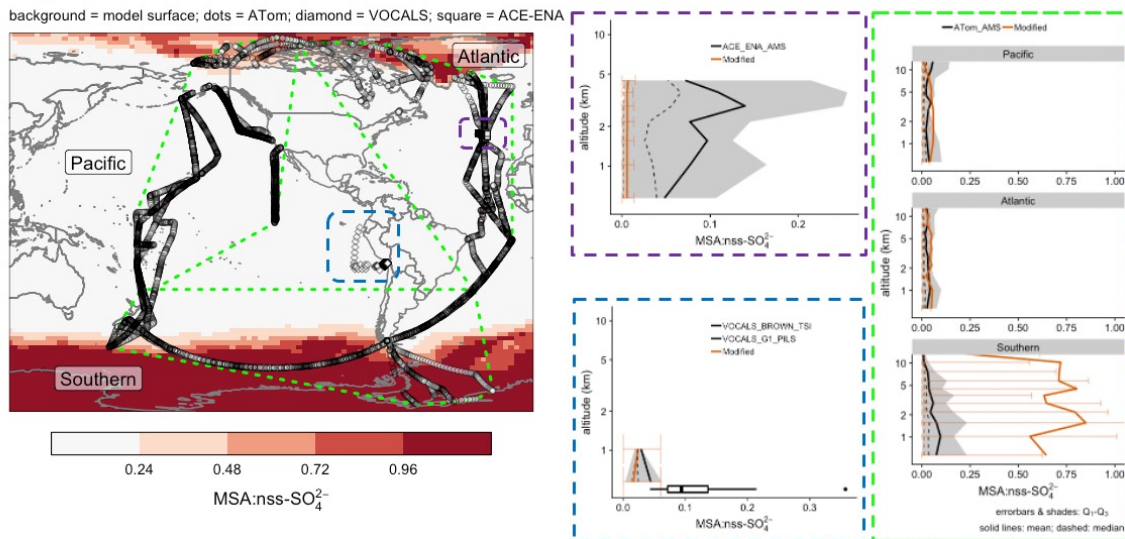
1) DMS

Figure 8. DMS (ppt) comparison between our simulation and ATom and VOCALS-Rex measurements. Only marine data are shown. Observations are resampled and vertically binned to match on model grids while model values shown are spatial-temporal matching with re-gridded observations. Compared to ATom, CAM6-chem biases high (esp. over the Southern Oceans) while our expanded DMS scheme help reduce the biases in all regions. Compared to VOCALS-Rex, both runs fairly agree with measurement, except the near-surface overestimation.



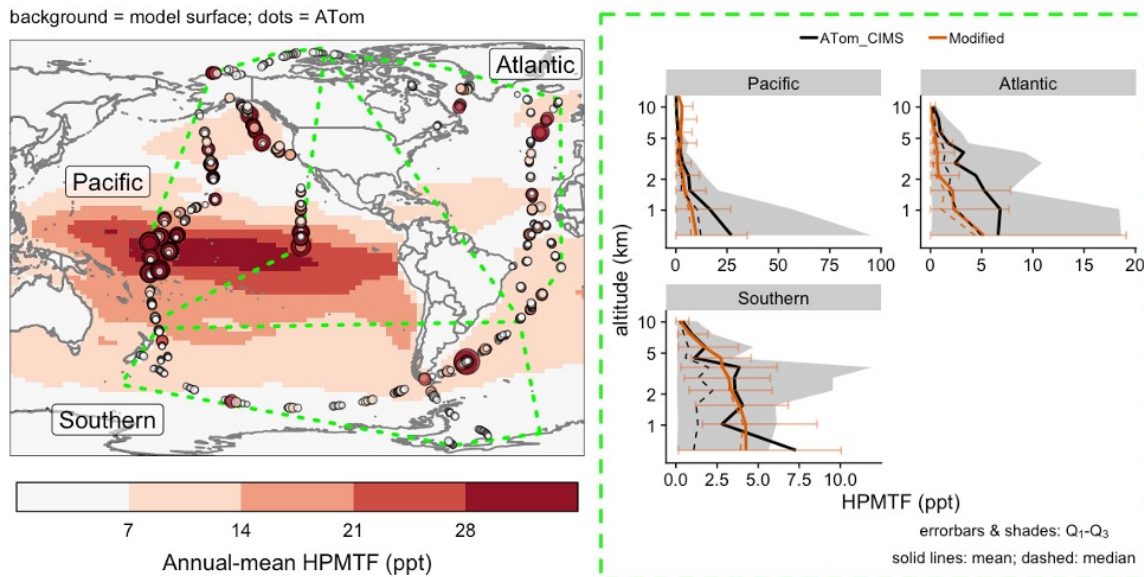
2) MSA:nss-SO₄²⁻ ratio

Figure 9. Aerosol MSA to non-sea-salt sulfate ratio (MSA:nss-SO₄²⁻) comparison between our simulation and ATom, ACE-ENA, and VOCALS-Rex campaigns. Only marine data are shown. Observations are resampled and vertically binned to match on model grids while model values shown are spatial-temporal matching with re-gridded observations. Our modified CAM6-chem fairly agrees with VOCALS-Rex and ATom over Pacific and Atlantic. and VOCALS-Rex. Model high-biases in Southern is likely due to overestimated MSA and underestimated sulfate compared to ATom's Southern measurements. Model low-biases compared with ACE-ENA can be explained by much higher observed-MSA and sulfate.



3) HPMTF

Figure 10. HPMTF comparison between our simulation and ATom measurements. Only marine data are shown. Observations are resampled and vertically binned to match with model grids while model values shown are spatial-temporal matching with re-gridded observations. Our modified CAM6-chem fairly agrees with ATom over Pacific and Southern Oceans but biased low over Atlantic.

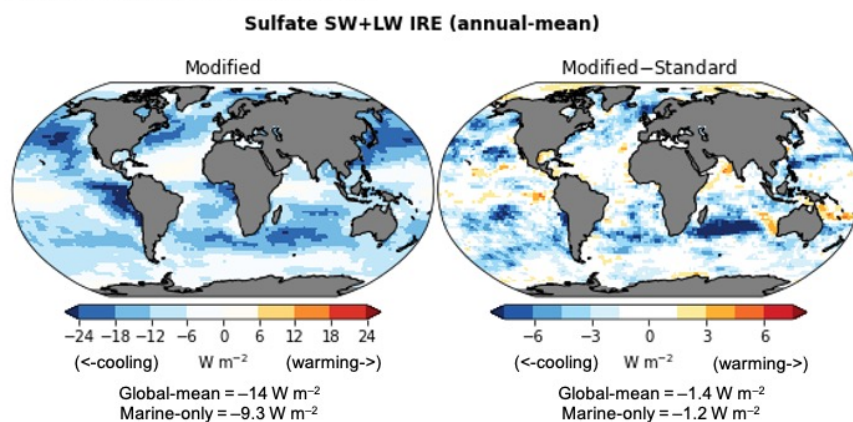


NEXT: INDIRECT EFFECTS AND FORCING

The results shown below are preliminary and still await further analysis.

1) Indirect Radiative Effect

Figure 11. Preliminary estimates of present-day sulfate indirect radiative effect (IRE; W m^{-2}) from our model runs. IRE is defined as difference of the cloud radiative forcing of a full-aerosol atmosphere from that of a sulfate-free one. Only marine values are shown. The difference between the "Modified" and the "Standard" runs (-1.2 W m^{-2}) is comparable to the IPCC's reported cloud forcing (relative to 1750) of -1.3 W m^{-2} to -0.2 W m^{-2} , hinting the importance of the DMS oxidation chemistry in accurately determining aerosol radiative forcing.



2) Sulfate Radiative Forcing

Three long-term runs using the "standard" and "modified" DMS chemistry and a sulfate-free under the preindustrial emissions (1750) will be performed to determine sulfate radiative forcing. It will allow us to compare the present-day and preindustrial DRE and IRE (i.e., the forcings), and hence evaluate how our extended DMS oxidation scheme can reduce the uncertainties in forcing estimation.

DISCLOSURES

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We would like to thank Jesse Kroll, Qing Ye, and Matthew Goss for consultation on DMS oxidation chemistry.

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ATom DMS mixing ratio is collected and analyzed by Donald R. Blake using whole air sampling (WAS). ATom MSA and sulfate concentrations are measured by Jose-Luis Jimenez using High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR ToF AMS). ATom HPMTF mixing ratio is collected by Patrick Veres and Andy Neuma.

VOCALS-REx DMS in-flight measurement (on NCAR's C-130) is conducted by Alan Bandy while the ship measurement (on NOAA R/V Ron Brown) is done by Barry J. Huebert. VOCALS-REx ship measurement of MSA and sulfate (on NOAA R/V Ron Brown) is done by Timothy Bates, while the in-flight measurement (on G-1) is done by Stephen R. Springston.

ACE-ENA measurements of MSA and sulfate are collected and analyzed by John E. Shilling and Maria A. Zawadowicz.

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ABSTRACT

Atmospheric aerosol influences the Earth's overall radiative balance, both directly via scattering and absorbing insolation or outgoing radiation reflected by the Earth's surface, and indirectly through promoting cloud formation and changing cloud properties. To quantify the radiative forcing requires the comparison of the net radiative budgets of a "polluted" atmosphere versus a "cleaner" preindustrial one. The accurate determination of aerosol radiative forcing thus partly depends on our ability to model the poorly understood preindustrial atmosphere. Here, we focus on modeling the oxidation of dimethyl sulfide (DMS) – a primary natural precursor of non-sea-salt sulfate.

Based on previous laboratory studies, we extend the simple DMS oxidation scheme used in the Community Atmospheric Model with chemistry (CAM-chem) version 6 (and similarly in many global ESMs) by adding new gas- and aqueous-phase reactions and including intermediate compounds, e.g., methanesulfonic acid (MSA). This implementation delays the formation of the sulfate, changes the spatial distribution of sulfate aerosol, allows more time for the intermediates to undergo dry or wet deposition, and eventually reduces the effective sulfate yield from DMS. We explore whether a more comprehensive DMS oxidation scheme improves the ability of the model to capture observations of DMS, sulfate, and other relevant species from a series of airborne and in situ measurements. Finally, we perform a series of simulations under present-day and preindustrial climate and emission scenarios to characterize the impact of a more comprehensive DMS scheme on the estimation of aerosol indirect effect.

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