# Aerosol and Dimethyl Sulfide Sensitivity to Sulfate Chemistry Schemes

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#### Key Points:

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- Seven DMS and sulfate chemistry schemes are tested in a single global climate model.
- The simulated spread in AOD and CDNC is more than twice as large as the change from pre-industrial to present-day.
- Constraining the chemistry of atmospheric sulfur is critical to constrain aerosolcloud interactions.

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#### 20 Abstract

Dimethyl sulfide (DMS) is the largest source of natural sulfur in the atmosphere and undergoes oxidation reactions resulting in gas-to-particle conversion to form sulfate aerosol.

<sup>23</sup> Climate models typically use independent chemical schemes to simulate these processes,

however, the sensitivity of sulfate aerosol to the schemes used by CMIP6 models has not

been evaluated. Here, we implemented seven DMS and sulfate chemistry schemes in an

atmosphere-only Earth system model. A large spread in aerosol optical depth (AOD)
 is simulated (0.077), almost twice the magnitude of the pre-industrial to present-day in-

is simulated (0.077), almost twice the magnitude of the pre-industrial to present-day increase in AOD. Differences are largely driven by the inclusion of the nighttime DMS ox-

 $_{29}$  idation reaction with NO<sub>3</sub>, and in the number of aqueous phase sulfate reactions. Our

<sup>30</sup> analysis identifies the importance of DMS-sulfate chemistry for simulating aerosols. We

<sup>31</sup> suggest that optimizing DMS/sulfur chemistry schemes is crucial for the accurate sim-

32 ulation of sulfate aerosols.

#### <sup>33</sup> Plain Language Summary

Dimethyl sulfide (DMS) is a sulfur-bearing gas predominantly emitted from ma-34 rine biological activity. DMS is the largest natural contributor to the global sulfur cy-35 cle, but its contribution is highly uncertain. Representing the complex chemical conver-36 sion of DMS to form natural sulfur atmospheric particles accurately in Earth System Mod-37 els is difficult. Complex atmospheric chemistry is expensive to implement, therefore sim-38 plistic approaches to represent the chemistry are used. Here we examine the variability 39 between different chemistry schemes. To achieve this, we employ a state-of-the-art Earth 40 System Model to compare seven simulations with differing sulfur-related chemical reac-41 tions. We show that sulfate chemistry contributes to large uncertainties in aerosol and 42 cloud formation. This work underscores the need to improve sulfur chemistry to improve 43 the accuracy of cloud and aerosol projections in a warming world. 44

#### 45 1 Introduction

Dimethyl sulfide (DMS; CH<sub>3</sub>SCH<sub>3</sub>) is the primary natural source of atmospheric
sulfur-containing species (Breider et al., 2010; Boucher et al., 2003). DMS is produced
from the biogeochemical activity of marine biota (Charlson et al., 1987; Keller et al., 1989;
Bates et al., 1987), and when emitted into the atmosphere, undergoes numerous chemical reactions, some of which lead to the formation of sulfate aerosols (Hoffmann et al.,
2021; Chen et al., 2018). Aerosols play an important role in cloud formation and influence Earth's energy balance (Carslaw et al., 2013; Novak et al., 2021).

Both natural (biogenic) and anthropogenic emissions contribute to the global sul-53 fur cycle. In the Northern Hemisphere (NH), atmospheric sulfur originates primarily from 54 anthropogenic sources such as power stations and ship emissions (e.g. Smith et al., 2011). 55 In contrast, natural sources dominate atmospheric sulfur loading in the Southern Hemi-56 sphere (SH), with anthropogenic sources contributing only 30% - 50% (Kloster et al., 57 2006; Korhonen et al., 2008). Emissions of anthropogenic sulfur-containing gases are well-58 represented in climate models (Hoesly et al., 2018; Hardacre et al., 2021; Turnock et al., 59 2020). In contrast, there are significant uncertainties regarding natural sulfur emissions, 60 especially over the remote Southern Ocean where DMS emissions are large and obser-61 vations are sparse (Bhatti et al., 2023; Bock et al., 2021; Hulswar et al., 2022). 62

The Southern Ocean region has a vital role in the global sulfur cycle but is predominately of natural origin, which is one of the largest sources of uncertainty for the sulfur cycle (Hoesly et al., 2018; Fung et al., 2022). This region is where global DMS production maximizes but is poorly constrained in models (Belviso et al., 2004; Bock et al., 2021; Revell et al., 2019), and is closely examined in this work.

Previously we examined the sensitivity of atmospheric DMS to oceanic DMS con-68 centrations and sea-to-atmosphere transfer velocities in a global climate model (Bhatti 69 et al., 2023). Here, we examine the sensitivity of sulfate aerosol formation to the model's 70 DMS and sulfate chemistry scheme. Whilst there is active work in the improvement of 71 DMS mechanisms used for modelling (e.g. Cala et al., 2023), current generation climate 72 models use relatively similar DMS and sulfate chemistry schemes, but with slight dif-73 ferences (e.g. Archibald et al., 2020; Horowitz et al., 2020; Sheng et al., 2015). We im-74 plemented seven such chemistry schemes taken from other climate models into a single 75 model, and assessed uncertainties in aerosol and cloud properties associated with sul-76 fate chemistry. Model configurations and simulation descriptions are described in Sec-77 tion 2, and results are shown in Section 3. 78

#### 79 2 Methods

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#### 2.1 Model Configuration and Simulations Performed

Simulations were performed with the atmosphere-only configuration of the UK Earth System Model (UKESM1-AMIP), which operates on a grid with a resolution of  $1.25^{\circ}$ latitude  $\times 1.85^{\circ}$  longitude (Sellar et al., 2019). All simulations were performed with an oceanic DMS data set calculated from satellite chlorophyll *a* observations, which is described and evaluated by Bhatti et al. (2023). DMS emissions are calculated using the transfer velocity from Blomquist et al. (2017). Atmospheric oxidation of DMS is handled via the StratTrop chemistry scheme (labelled here as 'REF'; Archibald et al., 2020; Mulcahy et al., 2020), which is modified for the sensitivity simulations.

Aerosol microphysics is determined using the Global Model of Aerosol Processes 89 (GLOMAP-mode) – a two-moment modal aerosol microphysics scheme. This scheme sim-90 ulates various aerosol species across five lognormal size modes: nucleation mode, solu-91 ble Aitken mode, accumulation mode, coarse mode, and insoluble Aitken mode (Mulc-92 aby et al., 2020). Typically, aerosols with a radius of  $\geq 25$  nm (Aitken mode) are acti-93 vated into cloud condensation nuclei (CCN) and cloud droplets (Walters et al., 2019; Abdul-94 Razzak & Ghan, 2000). A constant cloud water pH of 5.0 is used in the UKESM1, which 95 is important for aqueous-phase chemistry (Turnock et al., 2019). Greenhouse gas con-96 centrations and anthropogenic aerosol emissions follow Coupled Model Intercomparison 97 Project phase 6 (CMIP6) recommendations. 98

Simulations were run for three years, from January 2016 to December 2018, with
 the first year discarded as spin-up. Wind and temperature are nudged to values from
 the ERA-5 reanalysis at 6-hourly intervals (Hersbach et al., 2020; Dee et al., 2011).

Six sensitivity simulations were performed using DMS and sulfate chemistry schemes 102 from other Earth system models, many of which participated in CMIP6 (Table 1). For 103 detailed DMS and non-DMS sulfur reactions, refer to Tables S1 and S2. In terms of gas-104 phase chemistry, all schemes feature an OH addition and abstraction pathway. All schemes 105 also include a NO<sub>3</sub> oxidation reaction for DMS, except MIROC. None include the newly-106 identified hydroperoxymethyl thioformate, a DMS oxidation product (HPMTF; Veres 107 et al., 2020), although its role is actively researched (Cala et al., 2023; Fung et al., 2022). 108 All schemes involve at least two aqueous-phase in-cloud reactions. StratTrop, CHEM3, 109 and GEOS-CHEM have a third aqueous-phase reaction with  $O_3$ . CHEM3 and GEOS-110 CHEM have the largest number of aqueous-phase reactions (Revell et al., 2019; Chen 111 et al., 2017, 2016). Methanesulfonic acid (MSA), which is produced by DMS reacting 112 with OH, is treated as a sink for DMS in UKESM1-AMIP and is not transported or ad-113 vected (Mulcahy et al., 2020). Differences between each model tuning mean that the spread 114 between chemistry schemes will be smaller if doing an intercomparison using their own 115 model. Here we go further than just testing expansions of sulfate chemistry schemes, but 116

quantifying how much variability in DMS and aerosol can result just from using CMIP6
 or well-established chemistry schemes.

We quantify the spread between the simulations in the values derived from the various chemistry schemes using the relative range in percentage. This is calculated by the difference between the largest and smallest values, divided by the smallest value, and then multiplied by 100.

**Table 1.** Chemical reactions used in each simulation. Light gray shading: DMS oxidation reactions. Medium gray shading: gas-phase reactions involving DMS oxidation products. Dark gray shading: aqueous phase reactions involving sulfur-containing species. All reactions are gas-phase unless otherwise indicated. All models contain two DMS + OH reactions; abstraction and addition. For reaction rates and references see Tables S1 and S2.

Chemical Reaction	REF	SEN-SOCOL <sup>a</sup>	SEN-MIROC <sup>b</sup>	SEN-GFDL <sup>c</sup>	SEN-GEOS-CHEM <sup><math>d</math></sup>	SEN-CHEM3	$\mathbf{SEN}\text{-}\mathbf{Nor}\mathbf{ESM}^{e}$
DMS + OH (abs)							
DMS + OH (add)							
$DMS + NO_3$							
DMS + ClO						[	
DMS + Br							
DMS + BrO							
$DMS + O_3$							
DMS + Cl							
$DMS(aq) + O_3(aq)$							
$SO_2 + OH$							
$SO_2 + O$							
$SO_2 + O_3$							
$SO_3 + H_2O$							
DMSO + OH		[					
$MSIA + O_3$		[					
MSIA + OH		[					
$S(IV) + H_2O_2(aq)$							
$S(IV) + O_3(aq)$							
$S(IV) + O_3(aq)$			[				
S(IV) + HOBr(aq)		[	[				
S(IV) + HOBr(aq)		[	[				
$O_{3(aq)} + MSI^{-c}_{(aq)}$							
$O_{3(aq)} + MSIA_{(aq)}$							
$S(IV) + HO_2NO_2(aq)$							

<sup>a</sup> Solar-climate Ozone Links (SOCOL)

<sup>b</sup> Model for Interdisciplinary Research on Climate (MIROC)

<sup>c</sup> Geophysical Fluid Dynamics Laboratory (GFDL)

<sup>d</sup> Goddard Earth Observing System (GEOS)

<sup>e</sup> Norwegian Earth System Model (NorESM)

#### 123 2.2 Observational Data Sets

Satellite, ground, and ship-based observations were used for model evaluation (Table S3). Data from the Southern Ocean, representing a region largely untouched by anthropogenic aerosol emissions, are limited. To evaluate atmospheric DMS in this region
we merge ground and ship-based data into one data set, each weighted equally across
each month. Only two sources offer DMS data for the Austral winter, but the summer
and autumn are represented by seven datasets (Table S3).



Figure 1. Southern Ocean atmospheric DMS surface concentrations showing climatological monthly-means for the simulations comparing differing sulfate chemistry schemes with observations. Lines show the observational means (in grey) and their standard deviation (error bars) compared with the simulations for the same grid cells. The Southern Ocean measurements are compiled from 3 ground-based stations and 4 voyages, all weighted equally. The average  $\mathbb{R}^2$  value represents the seasonal correlation coefficient between the simulation and each respective observation.

#### <sup>130</sup> 3 Results and Discussion

The Southern Ocean (40 °S to 60 °S) is a focus of this study to investigate the different sulfate chemistry schemes used in the model simulations, and to compare with observations of atmospheric DMS. This region's global importance for DMS is highlighted by the high proportion of atmospheric DMS from the Southern Ocean contributing towards the global atmospheric DMS burden with 49% to 70% shown by our simulations.

Average DMS concentrations are relatively well constrained between the simula-136 tions, which is unsurprising given that all simulations used the same oceanic DMS source 137 and sea-to-air transfer velocity. Previous work has evaluated DMS and other aerosol prop-138 erties in UKESM1-AMIP (Bhatti et al., 2023; Mulcahy et al., 2020). Bhatti et al. (2023) 139 identified a 171% spread in DJF atmospheric DMS from Southern Ocean DMS concen-140 trations and emissions, whereas the spread identified from the simulations performed here 141 during the same period and region is 48%. Although the emissions and concentrations 142 drive much of the spatial and seasonal variability of atmospheric DMS, we demonstrate 143 that differences in CMIP6 chemistry also have a profound influence on Southern Ocean 144 atmospheric DMS. Here we examine the seasonal cycle in DMS from available observa-145 tions (Figure 1). 146

As shown in Figure 1, all simulations overestimate austral wintertime (JJA) atmospheric DMS but generally are closer to observations in summer months, except at Amsterdam Island. The year-long observational stations display a clear seasonal cycle for Southern Ocean DMS; however, none of the simulations successfully capture the DMS depletion during winter. The SOCOL chemistry scheme enables UKESM1-AMIP to best represent the seasonal cycle in atmospheric DMS ( $R^2 = 0.357$  compared to observations;  $R^2 < 0.2$  for all other simulations).

The current representation of the UKESM1-AMIP Southern Ocean DMS may be 154 flawed during the wintertime, as demonstrated by increases in DMS concentrations which 155 are not observed in any observations (Figure 1). DMS is mostly oxidized via  $NO_3$  dur-156 ing austral winter, however, most simulations do not oxidize DMS quickly enough, re-157 sulting in an accumulation of DMS during winter from the less efficient wintertime loss 158 pathway, which SEN-SOCOL shows. The additional Cl and Br chemistry is therefore 159 an important source for DMS oxidation during winter, in agreement with Chen et al. (2018). 160 Although the distribution of atmospheric DMS is mostly controlled by the oceanic DMS 161 and DMS emissions (Bhatti et al., 2023), we demonstrate the importance of choosing a 162 sulfate chemical reaction scheme appropriately over the Southern Ocean. As a result, 163 we investigate the global differences in chemical oxidation of DMS between each simu-164 lation. 165

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#### 3.1 Chemical oxidation of DMS

Globally, DMS + OH reactions account for 56% to 65% of total DMS loss in our simulations (Figure 2), in agreement with Fung et al. (2022). DMS oxidation via the OH addition and abstraction pathways dominates other oxidation reactions in the Southern Hemisphere, while DMS + NO<sub>3</sub> is largest in the Northern Hemisphere where there are large anthropogenic nitrate emissions. The hemispheric distribution of the widely used DMS reactions (DMS + OH and DMS + NO<sub>3</sub> is consistent with Chen et al. (2018).

Global DMS lifetimes of 1.2 to 1.4 days are consistent with the literature estimates of 0.72 to 2.34 days (Breider et al., 2010; Mulcahy et al., 2020). Figure S1 shows the spatial distribution of DMS lifetimes for each simulation. SEN-SOCOL (Figures 2b and S1b) has more DMS oxidized by NO<sub>3</sub> at the high SH latitudes than the other chemistry schemes, therefore reducing the lifetime of DMS, especially during winter. This reduces the relative importance of DMS oxidation via OH.

In SEN-GEOS-CHEM, there is more DMS oxidation by  $NO_3$  over continental re-179 gions than in the other simulations due to the high availability of NO<sub>3</sub> (Figures 2e and S1e). 180 Reducing the reaction rate constant for DMS oxidation with  $NO_3$  tends to extend the 181 DMS lifetime for the simulations, particularly during the night when  $NO_3$  is often the 182 only DMS loss pathway for many chemistry schemes. SEN-MIROC, without a DMS + 183  $NO_3$  reaction, lengthens the DMS lifetime over high northern latitudes (Figure S1), in 184 contrast to the other simulations. However, SEN-MIROC contains an alternative DMS 185 loss pathway via DMSO, and essentially becomes a night loss mechanism for DMS (not 186 shown), compensating for the lack of  $DMS + NO_3$  reaction. 187

The inclusion of additional DMS oxidation pathways (e.g. ozone, reactive chlorine, and bromine) as in SOCOL and CHEM3 has a relatively minor impact on DMS oxidation. UKESM1-AMIP currently only has a stratospheric source of inorganic chlorine or bromine, which explains why the contribution of these pathways is close to zero in Figure 2b,f. Future work will investigate the impacts on sulfur chemistry by implementing tropospheric inorganic chlorine and bromine sources.

Of all the chemistry schemes used here, only SEN-GFDL has up-to-date rate con-194 stant parameters for DMS oxidation as recommended by the latest JPL report (Table S1; 195 Burkholder et al. (2020)). Our results show some, but minor differences in DMS lifetime 196 and concentrations when comparing simulations with up-to-date rate constants to those 197 without (Figures 2 and 1) We therefore suggest that other modeling groups regularly en-198 sure rate constant parameters are up-to-date to have more accurate DMS reactions. Ex-199 panding the analysis to other sulfur species, including DMS, which are instrumental to-200 wards aerosol formation is further evaluated below. 201



Figure 2. Zonal annual means of the relative proportions of all chemical reactions (%) involved in DMS oxidation. The sum of all reactions for each simulation equals 100%. The dashed line indicates the equator.  $Br_x = Br$  and/or BrO and  $Cl_x = Cl$  or ClO. See Table 1 for more details.

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#### 3.2 Aerosol Response to Sulfate Chemistry Schemes

To better understand the differences in aerosol between the simulations with differing sulfate chemistry, we examine differences in the aerosol modes and CCN and H<sub>2</sub>SO<sub>4</sub> concentrations. Number concentrations of aerosol in different modes from our simulations are shown in Figure 3 (a-c for NH, g-i for SH). We exclude coarse mode aerosols which are dominated by sea spray in UKESM1-AMIP.

The nucleation mode over both hemispheres has a much higher sensitivity to changes in the chemistry scheme than the other modes, from the higher number of nucleation mode particles from anthropogenic sulfur sources. Simulations over the NH have an aerosol number concentration around three times higher than the SH with the NH having 129% more nucleating particles (Figures 3a, g and 3f, l). Similarly,  $H_2SO_4$  has much higher concentrations over the NH (Figures 3d,j). This is due to a much higher anthropogenic contribution.

In both the NH and SH SEN-NorESM has lower Aitken and accumulation mode 215 concentrations compared to most of the other simulations which leads to lower AOD and 216 CCN. This can be attributed to the scheme using only one  $SO_2$  oxidation pathway, re-217 ducing atmospheric sulfate available for conversion to aerosol. As aerosols grow from the 218 nucleation mode, they have a greater influence on cloud formation (Figure 3e, k). SEN-219 GEOS-CHEM and SEN-CHEM3 have the highest CCN concentrations due to a larger 220 number of in-cloud reactions which enhances aerosol growth into the accumulation mode. 221 Simulations with more aqueous-phase chemistry may allow more sulfate aerosol to trans-222 fer into larger aerosol modes. For example, shown in Figure 3a, the SEN-GEOS-CHEM 223



Figure 3. Global averaged aerosol number concentrations from the (a,g) nucleation-mode, (b,h) Aitken-mode, (c,i) accumulation-mode. (d,j) The  $H_2SO_4$  abundance is shown in parts per trillon (ppt). (e,k) Cloud condensation nuclei and (f,l) Global aerosol number size distributions are also shown. (a - f): Northern Hemisphere average; (g - l): Southern Hemisphere average.

and SEN-CHEM3 simulations contain fewer nucleation mode aerosol concentrations than the REF simulation. However, SEN-GEOS-CHEM and SEN-CHEM3 contain the highest concentrations within the larger modes (Figure 3c,i).

Accurately representing cloud-water pH in climate models is crucial for aqueous-227 phase chemistry and cloud formation (Turnock et al., 2019). Global cloud water pH varies 228 between 3-8, however, UKESM1 uses a uniform value of 5 (Shah et al., 2020). A small 229 increase in pH could reduce the number of aerosols serving as CCN (Turnock et al., 2019). 230 Chemistry schemes used in their native models are effectively tailored to that models spe-231 cific configurations. For example, GEOS-CHEM has interactive cloud-water pH affect-232 ing aerosol modes differently to the UKESM1 (Alexander et al., 2012). Other models, 233 like GFDL, assume cloud water pH of 4.5, leading to differences in their aqueous-phase 234 reactions (Krasting et al., 2018; Turnock et al., 2016). DMS oxidation into  $SO_4^{-2}$  can also 235 impact pH (Shah et al., 2020), particularly in DMS-rich areas like the Southern Ocean. 236 Excessive oxidation can lower the pH, impacting cloud formation. Thus, assuming a uni-237

form cloud-water pH for the Southern Ocean will lead to model spread, given the significant oxidation variations across simulations. Further work in updating the UKESM1
chemistry sources is therefore needed to better represent aerosols and DMS. For instance,
the inclusion of a BrO inorganic source from sea-spray would provide a much greater avenue for DMS oxidation during the winter, which has been shown to have a substantial
impact (Breider et al., 2010; Fung et al., 2022).

3.3 Sensitivity to Sulfate Chemistry Schemes

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To assess the overall sensitivity of sulfate formation to atmospheric chemistry, we analyze simulated DMS, AOD, cloud droplet number concentration (CDNC), and allsky shortwave radiation at the top of the atmosphere relative to the SO<sub>4</sub> mass fraction (which is integrated between the surface and top-of-atmosphere; Figure 4).

The spread between all the simulations annual mean is 20% for DMS globally. The 249 DMS burden is reasonably well constrained (ranging between 35-44 Gg S globally; (Fig-250 ure 4a). The vertical error bars are larger in the SH than in the NH because of the large 251 seasonality in marine biogenic activity at southern high latitudes (Figure 4e, i; Jarníková 252 & Tortell (2016); Curran & Jones (2000)). With the exception of the MIROC scheme, 253 the spread is 12% in DMS burden across the models. The MIROC chemistry scheme omits 254 the  $DMS + NO_3$  oxidation reaction, which is important in the removal of DMS over the 255 NH due to anthropogenic nitrate emissions (Archer-Nicholls et al., 2023; Chen et al., 2018). 256

The global contribution of atmospheric DMS to the overall sulfur burden  $(DMS_{sulfur})$ 257 across all simulations, varies between 13.6% to 25.4% (Table S4). Four simulations (SEN-258 SOCOL, REF, SEN-GFDL, SEN-CHEM3) have the lowest DMS<sub>sulfur</sub>, shown in Fig-259 ure 4a and Tables S4 and S5. Although SEN-NorESM has the lowest sulfur burden, it 260 has the highest proportion of sulfur from DMS (Table S4). Existing estimates of global 261  $DMS_{sulfur}$  are between 10% to 32% (Gondwe et al., 2003; Kloster et al., 2006; Fung et 262 al., 2022). SEN-MIROC has almost double the average DMS burden over the NH likely 263 due to the lack of  $NO_3$  reactions with DMS as discussed above. Around 80% of the global 264 annual average DMS<sub>sulfur</sub> is sourced entirely from SH annual average DMS (Table S4). 265

Figure 4a,e,i demonstrates that simulations with very similar DMS burdens have 266 very different  $SO_4$  burdens driven by the different sulfate mechanisms/oxidation path-267 ways. The spread in the global annual mean  $SO_4$  mass fraction across the simulations is 308%. SO<sub>4</sub> is crucial for the formation of clouds and aerosol (Figure 4) and consequently 269 global annual mean AOD and CDNC have a spread of 79% (0.077) and 70% (44 cm<sup>-3</sup>) 270 across our simulations (Figure 4b,c). The spread in AOD between our simulations is much 271 greater than the AOD of 0.031 from CMIP6 models (Vogel et al., 2022). Additionally, 272 global annual mean AOD is suggested to have only increased by 0.04 to 0.046 since pre-273 industrial times, with CDNC also increasing by 10 to 20  $\rm cm^{-3}$ , highlighting the large vari-274 ation between the chemistry schemes (Seo et al., 2020; Tsigaridis et al., 2006; Bhatti et 275 al., 2022; Bauer et al., 2020; Kirkevåg et al., 2018). The spread of the simulations be-276 tween both hemispheres is the same for CDNC but is two times greater over the NH than 277 the SH for AOD. For all simulations, there is a linear relationship between AOD and CDNC 278 vs  $SO_4$  mass fraction: the chemistry schemes that oxidize DMS and sulfate more efficiently 279 (such as SEN-GEOS-CHEM and SEN-CHEM3) also produce more AOD and CDNC. 280 All simulations showing AOD and CDNC over the SH are closer to the observed AOD 281 and CDNC averages. Future work will quantify what fraction of the spread in aerosol 282 is driven by DMS and from anthropogenic sulfur. 283

SEN-NorESM has the lowest AOD, CDNC, and SO<sub>4</sub> mass fraction, but an average DMS burden. This is likely due to inefficient sulfur-to-aerosol conversion over the NH demonstrated by a higher sulfur burden and fewer reactions involving SO<sub>4</sub> products than other schemes (Table S2). More specifically, the lower aerosol likely results from



Figure 4. Annual-mean atmospheric DMS sulfur burden, AOD, CDNC, and shortwave radiation as a function of the vertically integrated  $SO_4$  mass fraction. Top row: global average; middle row: Northern Hemisphere; bottom row: Southern Hemisphere. The dashed horizontal line represents the average value from observations. First column: atmospheric DMS (no global observations are available); second column: MODIS AOD (Platnick et al., 2017); third column: CDNC satellite measurements at cloud top between 2017 to 2018 from Grosvenor et al. (2018); fourth column: TOA all-sky shortwave radiation from CERES (Loeb et al., 2018). Error bars show the standard deviation on spatially averaged quantities calculated over the two-year simulations.

decreased  $SO_4$  mass fraction (Figure 4) and less efficient  $SO_2$  oxidation into  $H_2SO_4$ , as discussed in Section 3.2.

The CHEM3 scheme is a modified version of the SEN-GEOS-CHEM DMS and sulfate chemistry scheme, with both simulations simulating the largest global CDNC (Chen et al., 2018; Revell et al., 2019). Revell et al. (2019) showed that SEN-CHEM3 leads to increased CDNC over the Southern Ocean due to the inclusion of additional aqueousphase sulfate reactions. Despite pronounced differences in global mean CDNC (63-107 cm<sup>-3</sup>, Figure 4c), differences in top-of-atmosphere all-sky shortwave radiation are relatively small between the simulations (Figure 4d,h,l).

#### <sup>297</sup> 4 Summary and Conclusions

This study compares the differences between sulfate chemistry schemes using identical base configurations. The sensitivity of DMS and its oxidation products to changes in sulfate chemistry was investigated using a nudged configuration of the UKESM1-AMIP model. We show that testing 7 sulfate chemistry schemes in one model causes large variations in SO<sub>4</sub>, CNDC, and AOD across simulations; twice the change in AOD and CDNC between the pre-industrial and the present-day when simulated in UKESM1 (Seo et al., 2020; Bauer et al., 2020). Additional aqueous-phase chemistry increases the inter-model

variance through an increased number of larger aerosols. Our results need further inves-305 tigation to determine if they would be universally robust. For example, because of dif-306 ferences in model formulation (grid box sizes, oxidant levels and many other processes), 307 we don't expect the same sensitivities we have calculated here across other "base mod-308 els" – and we encourage work that would address this issue as a priority. As each model 309 treats DMS and sulfur chemistry and aerosol microphysical processes differently from 310 UKESM1, such as differences in cloud water pH or aerosol size distributions, the spread 311 in AOD (0.077) is more than double that compared between CMIP6 simulations (0.031;312 Vogel et al. (2022)). Therefore, careful consideration is necessary when modifying sul-313 fate chemistry schemes in climate models as aerosol response may vary significantly. We 314 highlight the associated uncertainty to aerosol and CDNC from sulfate chemistry seems 315 to be significantly large enough that it may alter the pre-industrial baseline and there-316 fore be an important source of uncertainty in aerosol ERF estimates. Therefore our study 317 builds on previous perturbed parameter ensemble studies of aerosol parameters which 318 did not consider these parameters (Carslaw et al., 2013) 319

We demonstrate that differences between well-established DMS and sulfate aerosol 320 chemistry schemes can strongly impact the global spread of DMS concentrations by as 321 much as 20% between the simulations, with larger fluctuations over the NH. These global 322 differences arise from differences in DMS oxidation pathways. Large seasonal differences 323 are also present between the simulations over the Southern Ocean, with the closest sim-324 ulation to observations coming from the SOCOL chemistry scheme ( $\mathbb{R}^2$  of 0.36). The UKESM1-325 AMIP currently lacks an inorganic BrO source from sea-spray which may provide im-326 provements to comparisons with observations, especially during the winter. The spread 327 in Southern Ocean DJF atmospheric DMS associated with chemistry (48%) is less than 328 the spread from oceanic concentration and emissions (171%). 329

This work demonstrates the importance of DMS and sulfate chemistry in future model intercomparison projects for future aerosol modeling. Overall, we find that testing different sulfate chemistry schemes in a single model can strongly affect aerosols and cloud formation.

#### <sup>334</sup> 5 Open Research

The MODIS-aqua satellite data from AOD and chlorophyll a are available in https:// 335 giovanni.gsfc.nasa.gov/giovanni/. CDNC observational data are available from https:// 336 doi.org/10.5285/864a46cc65054008857ee5bb772a2a2b. The CERES data was obtained 337 in https://ceres.larc.nasa.gov/data/. DMS measurements are available at https:// 338 ebas-data.nilu.no/Default.aspx. Model simulation data are archived at New Zealand 339 eScience Infrastructure (NeSI; https://www.nesi.org.nz/). As all simulation data is 340 over 1 Terabyte, they will be managed and made available for at least 5 years by con-341 tacting the corresponding author. 342

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