Authors’ Response to Referee Comments for

An advanced modelling study on the role of dimethyl sulfide in new particle formation in the pristine marine boundary layer

Wollesen de Jonge et al.
Atmospheric Chemistry and Physics,

RC: ***Referee Comment***, AR: Authors’ Response, ☐ Manuscript Text

1. Referee #1

1.1. Overview

RC: ***The present work provides a comprehensive and systematic analysis of how DMS-derived new particle formation (NPF) and subsequent growth respond to varying meteorological conditions within the marine boundary layer. The results clearly demonstrate that DMS-derived NPF and growth occur only under a limited set of conditions, specifically, cloud-free periods following moderate to heavy precipitation, despite a wide range of meteorological and environmental parameters. In addition, the study identifies priority areas for future research on gas-phase DMS oxidation chemistry, based on detailed quantum chemical calculations and sensitivity analyses. This represents an important step toward addressing the role of DMS in clouds and climate. I recommend publication after the authors address the minor concerns outlined below.***

AR: We thank referee #1 for the positive feedback and refer the reader to the general and technical comments outlined below.

1.2. General comments

RC: ***Throughout Section 3.2: My primary concern is the lack of model evaluation against field observations. For example, the modeled DMS concentrations, ranging from 200 ppt (Line 448) to 2 ppb (Line 559), are substantially higher than typical observed values, which are mostly below 300 ppt and rarely exceed 500 ppt in the summertime Southern Ocean (e.g., Figure 8 of Kang et al., 2025). Similarly, the modeled SO2 concentrations (100–500 ppt; Lines 386–387) are considerably higher than observations, which are generally below 50 ppt (Figure S3 of Kang et al., 2025). I recognize that this is a modeling study aimed at qualitatively demonstrating how DMS-derived NPF responds to different environmental conditions. However, these large discrepancies between model results and observations raise concerns about the reliability of the conclusions. I suggest that the authors discuss the possible reasons for the overestimation of atmospheric sulfur species and assess its potential impact on their conclusions.***

AR: We agree with the referee that some model evaluation against field measurements is warranted, despite the fact that the present study focuses its efforts on modelling. Firstly, however, we must emphasize that the sea-to-air flux of DMS in the model is calculated based on a transfer velocity specifically calculated for DMS (Blomquist et al., 2017). This stands in contrast to other transfer velocities that have been calculated for different gases (Liss and Merlivat, 1986; Wanninkhof, 2014; Nightingale et al., 2000), which are often used in modelling studies to calculate the sea-to-air flux of DMS. The CAMS-GLOB-OCE dataset (v3.1), for example, uses the transfer velocity from Nightingale et al. (2000) to calculate global DMS emissions. The wind-speed dependency in the transfer velocity from Nightingale et al. (2000) is quadratic, as opposed to the wind-speed dependency in the transfer velocity from Blomquist et al. (2017), which is super-linear. This means that the DMS flux presented in this study is significantly lower than the fluxes presented in modelling studies that calculate their transfer velocity based on the method from Nightingale et al. (2000), especially at high wind speeds. Secondly, all surface-ocean concentrations of DMS in the model have been based on realistic concentrations presented in Lana et al. (2011) and Hulswar et al. (2022).

As shown in this study, the gas-phase concentration of DMS in the marine boundary layer (MBL) is highly dependent on the surface-ocean concentration of DMS, the wind speed, the sea surface temperature (SST), and the oxidation capacity of the atmosphere. The relatively short lifetime of DMS also means that concentrations are likely to vary over the ocean in accordance with changes in these external factors. The gas-phase DMS concentrations simulated in this study are generally representative of an environment with moderate to high DMS concentrations in the surface ocean and relatively high wind speeds, but marine regions with little to no surface-ocean DMS and/or low wind speeds would experience much lower concentrations. It is therefore correct that DMS concentrations are typically in the range of 50–600 ppt over the ocean (Novak et al., 2022), but observations of gas-phase DMS in regions with high surface-ocean DMS concentrations have also shown that concentrations can range from one to several ppb (Marandino et al., 2008; Lawson et al., 2020; Owen et al., 2021). As for SO_2, the concentration in the model is affected by the fact that we simulate cloud-free conditions from day three to day six of the simulation. If sporadic cloud cover is introduced into the model, then SO_2 is efficiently removed from the gas phase, and its concentration becomes significantly lower (Fig. S73). We have added a segment in Section 3.1 that compares the simulated DMS concentration in the BaseCase simulation with observations from the MBL.

> The formation and growth of particles is driven by emissions of DMS, which enters the model at 8.3×10^{9} molecules cm^{-2} s^{-1} (6 m/s wind speed) and gradually builds up to reach a concentration of 0.92 ppb at the end of the simulation (Fig. 2b). Atmospheric DMS concentrations in the MBL have been shown to range from tens of ppt in regions with low surface-ocean DMS concentrations to a couple of ppb over waters with high surface-ocean DMS concentrations (Marandino et al., 2008; Novak et al., 2022; Lawson et al., 2020). The scenario prescribed in the BaseCase simulation is representative of a marine environment with moderate to high surface-ocean DMS concentrations, and the simulated gas-phase concentrations of DMS are therefore in line with observations from the MBL.

RC: ***Section 3.2.1 and 3.2.4, and Figure 3: Is it correct that the SST sensitivity tests were conducted with a fixed air temperature of 283 K? It is somewhat difficult to envision scenarios in which air temperature and sea surface temperature vary independently, although I appreciate the value of isolating the sensitivity to individual parameters. If feasible, I suggest performing additional sensitivity tests in which both air temperature and SST are varied simultaneously by the same amount. Would their effects on PN concentrations offset each other, or does one parameter exert a stronger control on PN concentrations?***

AR: It is correct that the sea surface temperature (SST) sensitivity tests were conducted at a fixed air temperature, with the SST as the only variable factor. This was done to test the impact of changes in the SST on the formation and growth of DMS-derived aerosol particles. We also acknowledge that the SST and the air temperature close to the sea surface can be similar, especially along the equator. However, in the Southern Ocean and in the North Atlantic Ocean (where phytoplankton blooms and moderate to high DMS sea surface concentrations tend to be most prevalent), the SST and air temperature can be very different. During spring and summer in the Southern Ocean, air masses from Antarctica are significantly colder than the surrounding ocean. This creates a scenario in which NPF is favoured, both because of the high sea-to-air flux of DMS and

NH_3 from the relatively warm ocean and because of the increased cluster stability in the cold air. During spring and summer in the North Atlantic Ocean, air masses from the continents are often significantly warmer than the ocean. Here, NPF is likely to be inhibited due to the decreased cluster stability in the warm air. In other scenarios, warm ocean currents or cold-water upwelling zones can also create situations in which the ocean temperature and the air temperature differ. Consequently, the SST can differ from the air temperature, which warrants these specific sensitivity runs. To keep things simple, we will therefore not perform sensitivity runs in which multiple parameters are varied.

RC: ***Line 148-161 and Line 379-391: The rapid cloud uptake of HPMTF (Novak et al., 2021) is not considered in this model. Given the importance of this process in suppressing gas-phase SO_2 formation, I recommend that the authors conduct an additional sensitivity test including this process to ensure that it does not affect their conclusions. References such as Fung et al. (2022) and Tashmim et al. (2024) may be useful for constraining the rate constants. Alternatively, the authors should justify why this process is not included.***

AR: On line 72–79 in 'Methods - Model Description', we state that the uptake of key DMS-derived water-soluble compounds (including HPMTF), is calculated using Henry's law solubilities and pKa values obtained with COSMO*therm* calculations as presented in Wollesen de Jonge et al. (2024). This means that we do consider the uptake of HPMTF to the aqueous-phase of both deliquesced particles and cloud droplets. In accordance with these calculations, however, the uptake of HPMTF to the cloud droplets remains fairly limited, and the compound is unlikely to stay in the particle-phase upon cloud evaporation. At the same time, the chemistry for the aqueous-phase oxidation of HPMTF still remains unknown. This means that the approach used in Novak et al. (2021) where all HPMTF taken up by the clouds is converted to SO_4^{2-} PM is quite uncertain, as it does not consider alternative reaction pathways that could potentially produce volatile species that will not remain in the aqueous-phase of the cloud droplets upon evaporation. We agree, however, that a pathway for the aqueous-phase oxidation of HPMTF most likely exists. The problem is just that we do not know it yet.

RC: ***Line 763-766 (and 20-26): This recommendation for future work would benefit from rephrasing in light of both the results of the present study and existing observational evidence. As summarized in Kerminen et al. (2018) (their Section 3.2.4) and Zheng et al. (2021), the prevailing view that "NPF is rare in the marine boundary layer" is based on the limited number of observed NPF events relative to the substantial body of field observations across various regions and seasons, particularly at the surface level. The present study, which shows that DMS-derived NPF occurs only under a limited set of conditions, is consistent with these observational findings from a theoretical perspective. I am not convinced that simply deploying long-term, station-based observations would be sufficient to demonstrate the influence of MBL NPF on CCN concentrations. A more critical question is where within the MBL such conditions are met. At a minimum, new observational efforts should target locations with favorable conditions (e.g., frequent cloud-free periods) and be combined with detailed air-mass history analyses to determine whether such events occur near the surface or aloft.***

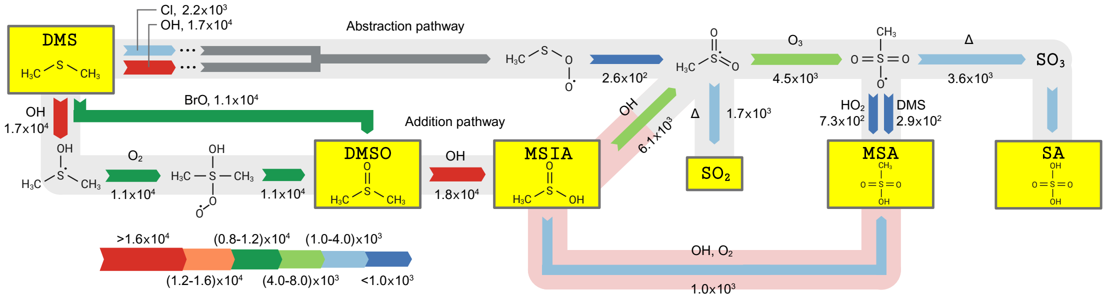
AR: We agree with the reviewer that simply deploying long-term observations in the pristine marine environment with no thought of the region in which they would measure would not help us to understand NPF in the MBL better. Still, studies like Zheng et al. (2021) do tell us that NPF takes place within the MBL with *"... regular and frequent occurrence following cold front passages"*. As these events require specific circumstances, such as moderate to high concentrations of surface ocean DMS, moderate wind speeds, and cloud-free conditions, it becomes harder for cruise campaigns and other short-term measurement efforts to capture them. We therefore propose long-term measurements as a means to increase the chances of capturing these events. As for the location, current research stations in the regions impacted by DMS emissions tend to be situated relatively far away from the areas with high concentrations of surface ocean DMS. Examples of such stations include Villum (81°36' N, 16°40' W), Pallas (67°58'N, 24°07'E), Neumayer (70°40'S, 08°16'W) and Aboa (73°03'S, 13°25'W). All these stations are relatively unaffected by anthropogenic emissions, but still measure regular NPF events. Lagrangian modelling studies using air-mass back-trajectories generated with HYSPLIT and FLEXPART have linked some of these NPF events to emissions of DMS and NH_3 from the ocean (Xavier et al., 2024; Wollesen de Jonge et al., 2024), but without measurements from the actual MBL these results remain speculative. One suggestion could be to place a research station in a location such as the Faroe Islands, where surface ocean DMS concentrations in the North Atlantic Ocean tend to reach their peak in May–July. The island also experiences high variations in cloud cover, precipitation, and wind speeds, potentially providing optimal conditions for new particles to form and grow. We have rephrased the section on future work to accommodate these considerations.

> As observations of NPF over the oceans remain scarce, we also encourage the scientific community to expand measurement efforts in the marine environment. As demonstrated in this study, DMS-driven NPF in the MBL takes place under specific conditions, and while NPF has been observed in flight and ship campaigns (Baccarini et al., 2021; Zheng et al., 2021), long-term measurements over the pristine ocean would increase the likelihood of meeting these conditions and thereby capturing such events. Locations such as the Faroe Islands may comply with these requirements, as the islands are surrounded by high concentrations of surface ocean DMS during spring and summer and are subject to significant variations in cloud cover, precipitation, and wind speeds. At the same time, research groups conducting measurements in open-ocean or coastal environments should seek to collaborate with modellers capable of reproducing the emissions, chemistry, and aerosol processes in the air masses that affect the measurements made at research stations and during campaigns. Such efforts may help to determine the source of NPF in the marine environment and quantify the extent to which natural emissions of DMS and NH_3 impact NPF and ultimately CCN concentrations in the MBL.

RC: ***Figure 1: The placement of the DMS + BrO pathway is potentially misleading. In the current scheme, this pathway appears to be categorized under the abstraction pathway, which is not consistent with chemical terminology. In an abstraction pathway, oxidants remove an H atom from a methyl group of DMS, whereas in an addition pathway, oxidants form a bond with the central sulfur atom of DMS in the initial reaction step. Please see Barnes et al. (2006, their Section 2.3.8) for a detailed discussion of this mechanism. The initial product of the DMS + BrO reaction is generally assumed to be the (CH3)2S–OBr adduct. In addition, the DMS + Cl reaction can proceed via both abstraction and addition pathways (see also Barnes et al., 2006, Section 2.3.2). These pathways are treated separately in the present model (Nr G4 and G5 in Table S1). Please check whether the flux value reported for DMS + Cl in Figure 1 (2.2 × 103) corresponds solely to the abstraction pathway.***

AR: We agree that Figure 1 could be read in the wrong way. The intention of the figure was to show the two main pathways for the oxidation of DMS in the addition channel (OH and BrO) in addition to the two main pathways in the abstraction channel (OH and Cl). The grey arrows originating from the arrows showing the pathways for OH/Cl H-abstraction have been marked as the abstraction pathway in the figure, whereas the OH/BrO addition have been marked as the addition pathway. The mechanism used in this study also considers Cl addition to DMS (reaction G5, Table S1), but as the CH_3SCH_3Cl radical easily decomposes back into DMS and Cl (reaction G21 Table S1) this pathway becomes negligible compared to the addition of BrO to DMS and the H-abstraction from DMS by Cl. The flux denoted in the figure for the reaction of DMS with Cl therefore only relates to the abstraction pathway. We have added a description of the initial reactions with DMS in the figure caption to avoid these misconceptions. We have also changed the figure to visually separate the two pathways.

The fluxes are shown next to the arrow for each reaction together the reactant for said reaction. Key compounds, all of which are able to partition to the aqueous-phase, are highlighted in yellow. New and modified pathways are marked in red. The initial addition of OH and BrO to DMS marks the addition pathway in the DMS oxidation mechanism, whereas the H-abstraction from DMS by OH and Cl marks the abstraction pathway.



1.3. Technical Comments

RC: ***Line 48, 327 and more: "metrological" may be a typo of "meteorological". Please check throughout the manuscript. Line 268, 352, 356, and more: "continues" may be a typo of "continued" or "continuous". Please check throughout the manuscript. Line 464: "As a results, . . . " -> "As a result, . . . ". Line 759: "formation SA" -> "formation of SA". Line 763: Delete one "to" from "the scientific community to to provide. . . ".***

AR: All technical comments have been corrected in the manuscript.

2. Referee #2

2.1. Overview

RC: ***This manuscript presents a detailed box modeling study of dimethylsulfide (DMS) oxidation in the box model ADCHAM, which resolves gas-phase and multiphase chemistry in aerosol and cloud water. The study presents an extensive array of sensitivity simulations beyond a base case that is meant to represent NPF observations made from aircraft over the Eastern North Atlantic. The authors implement detailed DMS oxidation chemistry in both gas and liquid phases, and find that with moderate sea-air exchange of NH3(g), DMS emitted from the surface ocean is sufficient to drive nucleation and particle growth following precipitation. This result is significant because, as the authors describe, the prevailing view is that the condensation sink in the marine boundary layer is too high for new particle formation and growth to occur. This work is suitable for publication in Atmospheric Chemistry and Physics, once the following largely minor comments are addressed.***

AR: We thank reviewer #2 for the positive and constructive comments. It is always a pleasure to receive feedback that is clearly based on an extensive knowledge on the oxidation chemistry of DMS. Our response is found under the general and specific comments outlined below.

2.2. General comments

RC: ***Further evidence and context is required to establish the range of surface ocean NHx concentrations applied in this study, as these choices appear critical for the modelled nucleation and growth by DMS-derived sulfuric acid. L216-218 describes justification for the chosen low, moderate, and high NHx concentrations, but no citations are given. Section 3.2.6 does not describe whether the chosen NHx(aq) concentrations and flux parameterization produce reasonable NH3(g) concentrations relative to what has been observed in the marine boundary layer. The low value of 0.1 mmol/m3 appears somewhat high; for example, approximately half of surface ocean samples from Wentworth 2016 contained NHx < 0.02 mmol/m3. Further on the flux parameterization, it is not immediately clear how this was parameterized: NH3(g) fluxes arise through bidirectional exchange, and so depend on the balance between NH3(g) and NHx(aq) concentrations, see Wentworth et al., 2016.***

AR: We agree that the description on the NH_x concentration from section 2.3 ('Sensitivity Runs') can be misleading, as it assumes that the reader remembers the reference to the study by Paulot et al. (2020) in section 2.1, L119. This study provides global surface ocean concentrations on NH_x from the Earth System Model 4 (ESM4.1) and evaluates the model performance against observations from different regions around the world. On L115-L120, we describe how the concentrations of NH_x were determined in accordance with the study by Paulot et al. (2020). It is correct therefore, that the concentration of NH_x may be lower than 0.1 mmol/m^3 in many regions of the world, especially in tropical and mid-latitude oceans (with the exception of certain coastal regions). However, if you look at the concentration of NH_x in the higher latitude regions where the phytoplankton blooms form (and where high concentrations of surface ocean DMS are often seen), said concentrations will generally be higher than 0.1 mmol/m^3. The 0.1-0.4 mmol/m^3 range is therefore meant to constitute a realistic representation of NH_x concentrations in regions with high concentrations of surface ocean DMS.

We also agree that the description of the flux parametrization for NH_3 could be better. On L120-121, however, we actually mention that the flux of NH_3 was calculated using the method by Wentworth et al. (2016). This means that we consider the bidirectional exchange of NH_3 in the model, e.g. meaning that the sea-air flux of NH_3 is regulated based on the concentration of NH_3 in the air. This also means that the sea-air flux of NH_3

can become negative if the concentration of NH_3 in the air becomes too high. As for the actual concentration of NH_3 in the model, the 'BaseCase' run experiences a maximum concentration of 50 ppt with a mean concentration of 17 ppt throughout the simulation. The concentration is highly pH dependant and decreases after the precipitation event as the majority of sea-spray particles are removed and the remaining particles acidify (Fig. S3). These concentrations fall within the range of 40-870 ppt observed in the Arctic MBL by Wentworth et al. (2016) and should therefore represent realistic concentrations of NH_3 in the MBL with minor impact from anthropogenic sources.

We have modified the following sections with a more detailed description on the surface ocean concentration of NH_x, the flux parametrization for NH_3, and the concentration of NH_3 in the model.

> $[NH_x]_{0.1}$, $[NH_x]_{0.2}$ and $[NH_x]_{0.4}$ demonstrate the effect of the sea surface concentration of total dissolved ammonium, which in turn impacts the emissions of NH_3 to the marine atmosphere. NH_x concentrations of 0.2 mmol/m^3 are representative of conditions in the North Atlantic Ocean between May and July, where phytoplankton blooms are at their peak in the region (Paulot et al., 2020). The same is true in the Southern Ocean during December to February, where NH_x concentration of 0.4 mmol/m^3 are common. Both higher and lower concentrations of NH_x can be found in different regions of the Worlds oceans, or in different periods of the year. The range of NH_x concentrations presented here are nevertheless representative of pristine ocean regions during periods with active phytoplankton blooms.

> The transfer velocity of NH_3 was calculated in accordance with the method by Wentworth et al. (2016). This approach employs a bidirectional exchange of NH_3, meaning that the air-sea flux of NH_3 in the model is regulated by the gas-phase concentration of NH_3. In the case that the concentration of NH_3 in the gas-phase exceeds the so-called 'compensation-point', the sea-air flux of NH_3 can also become negative.

> At the conditions given in the BaseCase model setup, a distinct NPF event takes place on the third day of the simulation following the precipitation event (Fig. 2a). The formation and growth of particles is driven by emissions of DMS, which enters the model at 8.3×10^9 molecules cm^{-2} s^{-1} (6 m/s wind-speed) and gradually builds up to reach a concentration of 0.92 ppb at the end of the simulation (Fig. 2b). The flux parametrization for the sea-air exchange of NH_3 results in a mean NH_3 gas-phase concentration of 17 ppt throughout the simulation with a maximum concentration of 50 ppt. These concentrations fall within the range observed in the Arctic MBL (Wentworth et al., 2016), and constitute a realistic representation of NH_3 in the MBL without impact from anthropogenic emissions.

RC: ***The discussion of controls on surface ocean DMS concentration is somewhat cursory and should be improved. See specific comments below.***

AR: We refer the reader to the specific comments.

RC: ***The authors may consider adjusting the article's title to better reflect the major findings of the work, rather than what was done.***

AR: As the study is based solely on modelling (with no support from measurements), we choose to present the results in a more neutral and descriptive manor so as not to sound over-confident in the results from our model. We are of course quite confident in the results presented in this paper, and hope that future measurements will help to confirm the role of DMS on NPF in the pristine MBL. However, as the topic remains controversial, we have chosen to go with the descriptive title.

RC: ***The manuscript contains a number of typos (e.g., 'to' instead of 'too'; 'continues' instead of 'continued'; 'summery' instead of 'summary'; 'bellow' instead of 'below').***

AR: All these typos have been corrected in the revised manuscript.

2.3. Specific Comments

RC: ***L12-14: Please adjust the description of the new rate coefficients for MSIA gas-phase reactions to better reflect what was done and what was found (i.e., as written, it is unclear).***

AR: We acknowledge that the description in the abstract could be clearer. However, to stay within the 250 word limit of the abstract, the details of the quantum chemical calculations and the chemistry sensitivity runs are saved for the main text. Still, the revised section now includes a better description of the method and the reason for the quantum chemical calculations.

> Finally, we employ quantum chemical calculations to provide temperature dependant rate coefficients for the OH-initiated oxidation of methane sulphinic acid (MSIA). We also assess other key uncertainties in the DMS oxidation mechanism to illustrate their impact on the formation and growth of DMS-derived aerosol particles in the MBL.

RC: ***L20-22: While these statements are true, the authors neglect to include a number of high-latitude ship-borne and aircraft studies that also show this. For example: https://doi.org/10.5194/acp-17-13119-2017; https://doi.org/10.1002/2017GL075671; https://doi.org/10.5194/acp-17-5515-2017; https://doi.org/10.5194/acp-16-7663-2016; https://doi.org/10.1038/s41467-020-18551-0; https://doi.org/10.1038/s41561-021-00751-y***

AR: The suggested studies on marine NPF observed during cruise (Collins et al., 2017; Baccarini et al., 2020; Burkart et al., 2017a; Brean et al., 2021) and flight campaigns (Burkart et al., 2017b; Willis et al., 2016) have been included.

> Observations on NPF in the MBL nevertheless remains scarce, and while certain studies report NPF events during cruise (Collins et al., 2017; Burkart et al., 2017a; Baccarini et al., 2020; Brean et al., 2021; Baccarini et al., 2021) and flight campaigns (Willis et al., 2016; Burkart et al., 2017b; Zheng et al., 2021), it is commonly believed that such events are rare (Pirjola et al., 2000; Quinn and Bates, 2011).

RC: ***L31 (and elsewhere): The authors refer to the major DMS source as phytoplankton, which is a true but incomplete description of DMS production in the surface ocean. A number of reviews exist that describe the role of bacteria in converting DMSP to DMS (and methanethiol, MeSH, which is not mentioned in this manuscript), and the authors should provide a more accurate description in their introduction and elsewhere.***

AR: It is true that DMS is not simply released directly from macro- and microalgae (including phytoplankton), but forms through different fates of the precursor dimethylsulfoniopropionate (DMSP) which is released when the algae species experience physiological stress (Carpenter et al., 2012; Hulswar et al., 2022). However, as this manuscript focuses on the fate of DMS after its formation in the surface ocean and following its emission into the atmosphere, any detailed description of these processes seems to us to be outside the scope of the study. We have however included some additional details in the initial description of DMS in the introduction.

> Over the oceans, dimethyl sulfide (DMS: $(CH_3)_2S$), formed through the breakdown of dimethylsulfoniopropionate (DMSP: $(CH_3)_2S^+CH_2CH_2COO^-$) released from macro- and microalgea (including phytoplankton) during physiological stress (Carpenter et al., 2012; Hulswar et al., 2021), comprises the largest source of natural sulfur and thus SA to the atmosphere (Lovelock et al., 1972; Bates et al., 1992).

RC: ***L40: Include Veres et al., 2020?***

AR: We have added a reference to the study by Veres et al. (2020). Note that this study is also credited on L149-152 for being the first study to observe HPMTF in the atmosphere and for revising the theoretical reaction rates for the production of HPTMF originally calculated by Wu et al. (2014).

> DMS has also been found to undergo autoxidation to form hydroperoxymethyl thioformate (HPMTF: $HOOCH_2SCHO$) (Wu et al., 2014; Berndt et al., 2019; Veres et al., 2020; Ye et al., 2022).

RC: ***L75-77: What is meant by "decrease in the aerosol particle deliquescence"? A decrease in particle water content with a change in humidity? Or a change in particle composition that leads to a change in the particle deliquescence point?***

AR: We agree that the phrasing is misleading. We meant to describe that certain low-volatile compounds might form through oxidation in the particle phase and stay there upon a decrease in the water content of deliquesced aerosol particles or through the evaporation of cloud droplets. The sentence has been changed.

> Some of these species contribute to the growth of the aerosol particles by undergoing aqueous-phase oxidation to form low-volatile compounds that remain in the particle-phase, even upon a decrease in the water-content of deliquesced aerosol particles or through the evaporation of cloud droplets.

RC: ***L82: "bellow cloud-scattering of particles" is below-cloud scavenging meant here?***

AR: We meant to write below-cloud scavenging. This has been corrected.

RC: ***L84: How does this dry deposition parameterization compare to more recent parameterizations, such as Emerson et al., 2020 (https://doi.org/10.1073/pnas.2014761117)? Does this have significant impact on the results of this work?***

AR: It is correct that newer parametrizations such as Emerson et al. (2020) has revised the treatment of aerosol particle dry deposition from some of the first parametrizations, e.g. Slinn and Slinn (1980) in the case of dry deposition over the ocean. The parametrization from Emerson et al. (2020) finds that the deposition of particles over the ocean should be lower than that presented in the older parametrizations, but admits that their treatment of ocean deposition remains uncertain. For one, the approach considers the ocean to be a flat surface and therefore relies solely on Brownian diffusion and impaction without considering the interception of particles by waves. In the simulations performed in this study, a 6 m/s wind-speed was considered in the BaseCase and throughout the sensitivity runs (except those focusing explicitly on the impact of wind-speed). These conditions do not form the basis for a flat ocean surface, and while the waves formed might not be big, they do create a surface roughness that would increase the deposition of particles as described in the parametrization from Emerson et al. (2020).

We agree though, that future development of the ADCHAM model should focus on testing alternative parametrizations for dry deposition from the one utilized in the original development of the model (Roldin et al., 2014). If the Slinn and Slinn (1980) parametrization does in fact over-predict dry deposition in the

500-2000 nm size range (Emerson et al., 2020), sea-spray PM concentrations may be under-estimated in the current model setup. It should be noted though, that the considerable uncertainty related to the emission parametrizations of sea-spray particles (Sofiev et al., 2011; Salter et al., 2015; Barthel et al., 2019) already makes the concentration of sea-spray in the marine atmosphere very uncertain.

RC: ***L94-96: It may be useful to lead with this justification. This reader persistently wondered how this simulation scheme was decided upon, until the end of the description.***

AR: This is a good point. We have moved the reference to the study by Zheng et al. (2021) to the start of the section.

> To investigate the influence of DMS on NPF in the pristine MBL, a model scenario reproducing conditions over the remote open ocean was constructed. The simulation was made to imitate the conditions during the flight-campaign described in Zheng et al. (2021), where frequent NPF within the MBL was observed following the passage of cold fronts. In the BaseCase scenario, the movement of an air-parcel ...

RC: ***L111: Sea-ice melt or break-up?***

AR: It is true that sea-ice melting also incentivise phytoplankton growth and production of DMS in the surface ocean. The rapid exposure of the ocean to sunlight during sea-ice breakout has nevertheless been shown to give rise to some of the highest concentrations of surface ocean DMS (Lana et al., 2011). Since concentrations also increase during sea-ice melting, we have added this statement to the sentence.

> It should be noted that even higher concentrations can be found in the ocean surrounding Antarctica during sea-ice breakout and melting.

RC: ***Figure 1: It appears visually in Figure 1 that DMS+BrO is an H-abstraction pathway. Suggest to revise the figure to separate the addition and abstraction pathways more clearly.***

AR: A similar question was raised by Referee #1. We refer the reader to the answer given in section 1.2.

RC: ***Section 2.2 (Table S1 & S2): Table S1 & S2 should show units of all rate coefficients used.***

AR: In modelling studies similar to this one (Hoffmann et al., 2016; Jacob et al., 2024), it is generally implicitly understood that second order rate coefficients are provided in units of cm^3 molecules^{-1} s^{-1} in the gas-phase and in L mol^{-1} s^{-1} in the aqueous-phase. First order rate coefficients are always provided in units of s^{-1}. To make this clear, we have added a description of the units in the caption of Table S1 and S2.

> Table S1. Gas-phase DMS reactions. Second order rate coefficients are given in units of cm^3 molecules^{-1} s^{-1} and first order rate coefficients in units of s^{-1}.

> Table S2. Aqueous-phase DMS reactions. Second order rate coefficients are given in units of L mol^{-1} s^{-1} and first order rate coefficients in units of s^{-1}.

RC: ***L142-145 (Table S2): Why do the authors implement on the reaction of MSIA + O3 and not MSI- + O3? Is the pH of cloud droplets and aerosol always below the pKa of MSIA in the model?***

AR: We do actually consider the oxidation of MSI-, just under name the $CH_3SO_2^-$ (e.g. the deprotonated form of MSIA). Reaction A25-30 in Table S2 highlights how this compounds is oxidized by OH, SO_4^{2-}, Cl_2^-, H_2O_2 and O_3. However, with a pKa of 2.56 (at 300K), MSIA does not dissolve efficiently into the deliquesced aerosol particles after the precipitation event were the pH value was found to be below two (Fig. S3). Before the precipitation event, however, the high concentration of sea-spray and associated water-content allowed for a higher pH (above three in the deliquesced particles and higher than five during cloud periods). During this period, $CH_3SO_2^-$ does in fact contribute to the production of MSA in the aqueous-phase (Fig. S7).

RC: ***L155-156: This is an indirect estimation of the rate coefficient. Why aren't the authors using the temperature dependent measurement of Assaf 2023 https://doi.org/10.1021/acs.jpca.2c09095, which suggests a somewhat lower value at room temperature?***

AR: We were reluctant to use the temperature dependant rate-coefficient from Assaf et al. (2023), as the temperature range of 314-433 K which was used in the experiment does not represent the conditions in the actual atmosphere. Extrapolating outside this temperature range, which is needed to perform the simulations in our study, would therefore add unnecessary uncertainty to the results. At the same time, other experimental studies (Berndt et al., 2019; Ye et al., 2021, 2022; Jernigan et al., 2022) performed at relevant temperatures on this reaction all present reaction rates that are higher than the one presented in Assaf et al. (2023). As the rates presented in these studies are also reasonable similar, we choose to go with the rate associated with the lowest uncertainty (Ye et al., 2022) and use it as a benchmark to scale the temperature dependant rate presented in Veres et al. (2020).

RC: ***L167-171: Do the authors use the rate coefficients calculated by Chen et al., 2023? https://doi.org/10.1021/acs.est.3c07120.***

AR: Throughout this study we sought to prioritize reaction rate coefficients that have been obtained both through theoretical calculations and through measurements. Given that the theoretical reaction coefficients presented in Chen et al. (2023), e.g. for the addition of O_2 to CH_3S and CH_3SO_2, have already been validated from experimental data in MCMv3.3.1, we choose to give priority to the experimental results. The same goes for the thermal decomposition of CH_3SO_2, which also appears to be orders of magnitude higher in Chen et al. (2023) compared to the experimental rate from MCMv3.3.1 and the theoretical estimates from Jacob et al. (2024) and Berndt et al. (2023).

RC: ***L177-181: The MSIA+OH addition pathway is also proposed by Chen et al., 2023 (https://doi.org/10.1021/acs.est.3c07120) as a source of MSA. Also, why do the authors computations provide a quite different result than Lv 2019 that suggests this reaction proceeds to H2SO3 and CH3 radical?***

AR: As stated on L177-181, this study does not claim to have discovered the reaction pathway leading to the production of MSA. This was done as early as in the study by Lucas and Prinn (2002), and later in Shen et al. (2022) and Ye et al. (2022). In these papers, however, the rate was never determined through experiments or quantum chemical calculations, but merely by qualified guesswork. Here, we use quantum chemical calculations to provide a better alternative for the reaction coefficient for this rate, which may be used until an experimental rate is determined. It is true though that this pathway is also discussed in Chen et al. (2023), and we have added the reference to this sentence. As for the study by Lv et al. (2019), they only focused on the pathway leading to the production on H_2SO_3 and not MSA as was done in this study. Here, we also consider the reaction pathway presented in Lv et al. (2019). However, in accordance with our calculations, this pathway is irrelevant compared to the production of MSA from the addition of OH to MSIA.

> This pathway has been proposed theoretically in previous publications (Lucas and Prinn, 2002; Shen et al., 2022; Ye et al., 2022; Chen et al., 2023), but the rate of reaction has so far only been based on estimates or taken from similar reactions.

RC: ***L186: It is known whether this isomerization actually occurs? Current literature seems to assume this is the case.***

AR: I agree that it is hard to trace the origins of this reaction. Hoffmann et al. (2016) references some of the earlier model studies like Lucas and Prinn (2002) for the reaction rate, which in turn references Kukui et al. (2000) which in turn references Turnipseed et al. (1992) which does not seem to mention it. There is however an estimate in the experimental study by Turnipseed et al. (1993) that mention that the reaction should have an upper limit of 20-25 s^{-1}. The rate used in this study was adapted from Hoffmann et al. (2016) as it was described to be a key component in the direct production of H_2SO_4. As described in this study, however, this pathway is negligible compared to the production of CH_3SO_2 from the OH-initiated oxidation of MSIA (a pathway that what not included by Hoffmann et al. (2016)).

RC: ***L192: Not only NO, but also HO2 or RO2 (See Chen et al., 2023) Do the authors use the equilibrium constant for CH3SO2 + O2 from Chen et al., 2023? In Table S1, the reaction of CH3SO2 (+O2) –> CH3SO2O2 is attributed to MCM.***

AR: This is true. $CH_3S(O)_2OO$ can also react with HO_2 or RO_2 to form CH_3SO_3 (see reactions G113 and G119 in Table S1). These reactions, in addition to the CH_3SO_2/$CH_3S(O)_2OO$ equilibrium reactions (see reactions G99 and G118 in Table S1), were taken from MCMv3.3.1 in accordance with the previously mentioned principle of giving priority to reaction coefficients obtained experimentally. L191-192 has been modified to include a description of these pathways.

> CH_3SO_2 either undergoes thermal decomposition to form SO_2 or reacts with O_3, NO_2, or O_2 followed by NO, HO_2, or RO_2 to form CH_3SO_3.

RC: ***Figure 2: Consider showing the concentration of DMSO and NH3 in this figure.***

AR: As DMSO was not vital to the results discussed in the paper, we have decided to keep it in the supplementary overview figure of specific DMS-derived oxidation products (e.g. Fig. S2, S13, S23, S33, etc.). It also proved difficult to include NH_3 without overcrowding Figure 2. NH_3 concentrations were therefore added to the same supplementary overview figures as DMSO.

RC: ***Does the model capture the aerosol pH effects on this partitioning? e.g., https://www.nature.com/articles/s43247-025-03041-2.***

AR: ADCHEM is able to capture the release of compounds with semi-volatile characteristics such as MSIA and MSA in accordance with the pH of the aerosols particles. In the aqueous-phase chemistry, we treat the $CH_3SO_2H/CH_3SO_2^-$ and $CH_3SO_3H/CH_3SO_3^-$ equilibrium in accordance with the pKa obtained from Wollesen de Jonge et al. (2024) and the pH of the particles. The evaporation of these species are then treated in accordance with their Henry's law solubilities.

RC: ***L316-317: How does this depend on pH? Do the authors assume that the reaction always proceeds through MSIA+O3 (rather than MSI- + O3)? L320: What is the pH of these particles?***

The production of MSA from dissolved and dissociated MSIA ($CH_3SO_2H/CH_3SO_2^-$) is highly dependant on pH. Before the precipitation event, the pH in the aerosols is higher (approximately three in the deliquesced aerosols particle and five in the cloud droplets) than after the event (lower than two in the deliquesced particles) due to the significant amount of sea-spray particles and associated water-content (see Fig. S3). This impacts the $CH_3SO_2H/CH_3SO_2^-$ equilibrium, meaning that a higher fraction of MSIA dissociates into $CH_3SO_2^-$ before the precipitation event when the pH is higher. Consequently, the $CH_3SO_2^-$ + O_3 reaction forming MSA is more important before the precipitation event than after (see Fig. S7). Note that the pH of the aerosol particles is displayed in the supplementary overview figures on PM concentrations (Fig. S3, S15, S25, S35, etc.). For the difference in the $CH_3SO_2H/CH_3SO_2^-$ oxidation by O_3 in the aqueous-phase, we have added a reference in the text to Figure S7.

> This happens as 85% of MSA is formed in the aqueous-phase of the deliquesced aerosol particles and cloud droplets from the oxidation of dissolved and dissociated MSIA by O_3 (Fig. S7).

RC: ***L324: Does the model not capture aerosol liquid water reactions of S(IV), or is the pH and liquid water content simply not sufficient for rapid conversion? What might be the effect enhanced reaction rates due to ionic strength in aerosol (eg. https://pubs.acs.org/doi/10.1021/acs.est.3c00212; https://doi.org/10.1021/acs.est.0c06496)?***

The impact of SO_2 on the SO_4^{2-} PM concentration in deliquesced aerosol particles and cloud droplets, respectively, depends both on the uptake and dissolution of SO_2. The Henry's law solubility of SO_2 is approximately six orders of magnitude lower than that of e.g. MSIA, and while the SO_2 concentration in the gas-phase is generally higher than MSIA it is not enough to drive a significant uptake of SO_2 to the deliquesced particles. During cloud cover, the increase in water content helps to increase the uptake of SO_2 to the aqueous-phase, and the increase in pH helps the compound to dissolve into HSO_3^- and SO_3^{2-}. It should also be noted that H_2O_2, which drives the oxidation of SO_2 in the aqueous-phase across the pH spectrum, dissolves more efficiently into the cloud droplets compared to the deliquesced particles. This means that we do consider the liquid water reactions of S(IV) in the model at all times. They are simply more effective during cloud periods in the model.

As for the impact of ionic strength, the aerosol pH in our BaseCase simulation is strongly acidic (mean bulk particle-phase pH of approximately 1.4–3 across the sensitivity runs). At this pH, virtually all dissolved S(IV) exists as $SO_2{\cdot}H_2O$ rather than HSO_3^- or SO_3^{2-}. The ionic strength enhancements discussed in the cited literature are most pronounced for the O_3 + S(IV) pathway, which proceeds primarily through SO_3^{2-} and is therefore strongly suppressed at low pH. The H_2O_2 + S(IV) pathway is also limited by H_2O_2 availability, as it partitions more efficiently into cloud droplets than into deliquesced particles. In the experiments by Yu et al. (2023) the estimated aerosol pH was $\sim$4.0, based on the pH in the bulk solution used to generate the aerosol particles. Yu et al. (2023) also illustrates that at pH<3 the S(IV) oxidation by H_2O_2 will still be more than 2 orders of magnitude faster than the S(IV) oxidation by O_3, in the remote marine condition case investigated in their figure 4, even if you consider their observed ion strength effect on the O_3 oxidation of S(IV). We agree with the conclusions from Yu et al. (2023) that "*atmospheric models should consider the ionic strength effects on the multiphase oxidation of SO_2 by O_3 in sea salt aerosols to improve the predictions of the sulfate formation rate and the sulfate aerosol budget in the marine atmosphere.*" ADCHAM use the AIOMFAC to calculate the activity coefficients of ions in the aqueous solutions, including H^+, Na^+, K^+, $NH0^+$, Mg^{2+}, Ca^{2+}, Cl^-, Br^-, I^-, NO_3^-, IO_3^-, OH^-, HSO_4^-, SO_4^{2-},HCO_3^-, CO_3^{2-} and $CH_3SO_3^-$ but we currently lack the theoretical framework to calculate activity coefficients for $SO_2{\cdot}H_2O$, HSO_3^- and SO_3^{2-} that could account for the non-ideal mixture conditions in concentrated electrolyte solutions. We identify this as a priority for future development, particularly for scenarios involving higher aerosol pH.

RC: ***L357-358: At what NOx concentrations?***

AR: NO_x concentrations remain low in the simulations so as to reproduce the conditions in the pristine MBL. We consider a small flux of natural marine NO in accordance with the study by Braeuer et al. (2013), which yields a mean NO_x concentration of $\sim$ 6 ppt in the BaseCase simulations. This information has been included in the sentence on L357-358.

> However, due to the low NO_x conditions in the simulation (mean concentration of ~ 6 ppt in BaseCase, representative of the pristine MBL), the NO_x driven oxidation of CH_3SO and CH_3SOO_2 in the abstraction pathway make up < 1% of the CH_3SO_2 source flux throughout all temperature sensitivity runs.

RC: ***L454-455: What is the particulate methansulfonate/methansulfonic acid lifetime against OH oxidation in the model?***

AR: At the moment, the model only considers the loss of MSA PM (e.g. $CH_3SO_3H/CH_3SO_3^-$) via the oxidation by dissolved OH. In the model, OH does not partition efficiently to the deliquesced particles and consequently MSA PM is not lost during non-cloud periods. In the cloud droplets, however, the lifetime of CH_3SO_3H and $CH_3SO_3^-$ against dissolved OH oxidation is 3.8 and 4.4 hours, respectively, assuming a liquid water content of $3.3 \cdot 10^{-10}$ kg/cm^3. We acknowledge that MSA PM in the deliquesced particles might also be lost due to heterogenous oxidation by OH, but at the moment we are not able to reproduce these reactions in the model.

RC: ***L511-513: This doesn't seem correct? The Henry's Law constant will decrease with increasing temperature, but this would not change the concentration gradient at some given Cw and Ca? Perhaps this statement can be clarified.***

AR: We acknowledge that this phrase was unprecise. We meant to say that an increase in temperature would enhance the (C_w-C_aH^{-1}) term in the flux calculation through a change in the dimensionless Henry's law constant (H). The sentence has been modified. We also noticed that H was wrongly referenced in the text as the Henry's law solubility, while the H presented in the equation (and in Ziska et al. (2013)) denotes the Henrys law volatility. As these terms are simply related via $H_v = H_s^{-1}$, the flux expression in equation (1) has been changed to: $F = K(C_w\text{-}C_aH)$. This allows us to keep referring to H as the Henry's law solubility, which is consistent with the nomenclature used throughout the rest of the manuscript.

> In addition, the Henrys law solubility dictates that gases are less soluble in warm as opposed to cold water, thereby increasing the sea-air flux in warmer waters.

RC: ***L542-543: This statement needs to be clarified and referenced. While high latitude oceans are often regions of elevated DMS production compared to the global average, this is not necessarily because of nutrients, but rather because of biological species distribution (e.g., prevalence of strong DMS producers in polar regions). Polar regions often have nutrient limitation, which varies depending on region and season.***

AR: We acknowledge that this statement is imprecise. What we meant by this is that the high latitude oceans (> 60°) generally experience more phytoplankton growth through a combination of nutrient availability (from strong upwelling) and from long daylight hours during spring and summer. We understand that this does not transfer directly to the production of DMS, as the release of DMSP and subsequent transformation to DMS is generally stress-induced and therefore driven by external factors. To keep the argument simple, we refer to the studies by Lana et al. (2011) and Hulswar et al. (2022) which describe how high DMS concentrations are generally found at high latitudes in colder waters.

> It should be noted that elevated concentrations of DMS and NH_x in the surface ocean are generally found at high latitudes in colder waters, whereas middle and low latitudes with warmer waters on average experience lower concentrations (Lana et al., 2011; Hulswar et al., 2022; Paulot et al.,2015).

RC: ***L546: Similar to the above comment, this is an overly simplified statement. DMS does not scale simply***

with total biomass abundance, but is more related to the abundance of DMS producing algal functional groups (see e.g. https://link.springer.com/article/10.1007/s10533-007-9091-5), as well as mixed layer depth (see e.g. https://doi.org/10.1038/46516 and https://doi.org/10.1029/2001GB001829).

AR: Again, we acknowledge that the statement lacks detail. To account for this, we have rephrased the section and included additional information that gives a better understanding for the spatial and temporal distribution of DMS in the surface ocean throughout the worlds oceans.

> The concentration of DMS in the surface ocean varies globally in accordance with various physical and chemical ecosystem parameters. These parameters include the availability of nutrients and sunlight that helps to form and sustain the growth of phytoplankton blooms, in addition to variations in temperature, salinity, UV-light intensity, and certain algal and bacterial enzymes that either drive the stress-induced released of DMSP or contribute to the conversion DMSP to DMS (Stefels et al. (2007); Carpenter et al. (2012)). In $[\mathrm{DMS}]_5$, BaseCase, $[\mathrm{DMS}]_{15}$ and $[\mathrm{DMS}]_{20}$, we demonstrate the impact that low, moderate, and high concentrations of DMS may have on the DMS-derived NPF and growth within the MBL. It should be noted that DMS may reach even higher concentrations in certain regions of the worlds oceans (Lana et al., 2011; Hulswar et al., 2022). The concentrations applied here are nevertheless representative of surface ocean concentrations of DMS in the pristine marine environment during spring and summer in both the northern and southern hemisphere.

RC: ***L554: What is "natural nutrient availability"?***

AR: Here we meant to describe regions of the world where man-made fertilizers spill into the ocean and contribute to so-called "unnatural" algae growth. In this study, we wished to describe regions of the world where the plankton blooms formed naturally under so-called "natural" nutrient availability. We have however rephrased this entire paragraph based on the question stated above, and therefore refer the reader to this question.

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