



Oxidation mechanisms for volatile methylated sulfur compounds and the major contribution of methanesulfonic acid to Southern Ocean aerosol particles

Samuel Ruhl¹, Matthias Kohl¹, Rima Baalbaki², Christos Xenofontos², Ryan Vella^{1,a}, Sergey Gromov¹, Xu-Cheng He³, Jiali Shen^{3,4}, Dina Alfaouri³, Samira Atabakhsh⁵, Lubna Dada⁶, Jenna DeVivo⁷, Jonathan Duplissy³, Imad El Haddad⁶, Hartwig Harder¹, Tuija Jokinen², Heikki Junninen⁸, Vijay P. Kanawade^{2,9}, Milin K. Sebastian¹⁰, Hannah Klebach¹¹, Markku Kulmala³, Felix Kunkler², Jos Lelieveld^{1,2}, Katrianne Lehtipalo^{3,12}, Clara J. Lietzke⁷, Lu Liu⁶, Roy Mauldin⁷, Bernhard Mentler¹³, Ottmar Möhler⁹, Tuukka Petäjä³, Douglas M. Russell¹¹, Mario Simon¹¹, Roseline C. Thakur³, Wenjuan Yu³, Jiangyi Zhang³, Zhensen Zheng^{13,14}, Jasper Kirkby^{11,15}, Holger Tost¹⁶, Theodoros Christoudias², Rolf Sander¹, and Andrea Pozzer^{1,2}

¹Department of Atmospheric Chemistry, Max Planck Institute for Chemistry, Mainz, Germany.

²Climate and Atmosphere Research Center (CARE-C), The Cyprus Institute, Nicosia, Cyprus.

³Institute for Atmospheric and Earth System Research/Physics, Faculty of Science, University of Helsinki, Helsinki, Finland.

⁴Helsinki Institute of Physics, University of Helsinki, Helsinki, Finland

⁵Leibniz Institute for Tropospheric Research (TROPOS), Leipzig, Germany.

⁶PSI Center for Energy and Environmental Sciences, Paul Scherrer Institute, Villigen, Switzerland.

⁷Department of Chemistry, Carnegie Mellon University, Pittsburgh, PA, USA.

⁸Institute of Physics, University of Tartu, Tartu, Estonia.

⁹Global Center for Clean Air Research (GCARE), School of Engineering, Civil and Environmental Engineering, University of Surrey, Guildford, Surrey, United Kingdom.

¹⁰Karlsruhe Institute of Technology, Institute of Meteorology and Climate Research Atmospheric Aerosol Research, Karlsruhe, Germany.

¹¹Goethe University Frankfurt, Institute for Atmospheric and Environmental Sciences, Frankfurt am Main, Germany.

¹²Atmospheric composition research, Finnish Meteorological Institute, Helsinki, Finland

¹³Ion Molecule Reactions and Environmental Physics Group, Institute of Ion Physics and Applied Physics, Leopold-Franzens University Innsbruck, Innsbruck, Austria.

¹⁴IONICON Analytik GmbH, 6020 Innsbruck, Austria.

¹⁵CERN, European Organization for Nuclear Research, CH-1211 Geneva, Switzerland

¹⁶Institute for Atmospheric Physics, Johannes Gutenberg University Mainz, Mainz, Germany.

^anow at: Institute for Atmospheric and Climate Science, ETH Zurich, Zurich, Switzerland

Correspondence: Samuel Ruhl (s.ruhl@mpic.de)

Abstract. Volatile methylated sulfur compounds (VMS), particularly dimethyl sulfide (DMS) and methanethiol (MeSH), are important natural sources of atmospheric sulfur. Their oxidation pathways and contribution to aerosols and cloud condensation nuclei (CCN) remain uncertain. Here, we investigate four gas-phase chemical mechanisms of increasing complexity for VMS oxidation using the global chemistry-climate model EMAC, and evaluate the results against shipborne and ground-based observations of DMS, sulfuric acid (SA), and methanesulfonic acid (MSA) between 2016 and 2019. In the marine boundary layer, DMS mixing ratios are largely insensitive to the choice of mechanism and agree well with observations, whereas simulated



SA and MSA differ markedly between mechanisms. Notably, oxidation by bromine monoxide (BrO) is the dominant process controlling the DMS loss rates and concentrations in the Southern Ocean. We also evaluate the contribution of MSA to global new particle formation in the marine boundary layer, based on recent measurements of (SA+MSA)-NH₃-H₂O nucleation at the CERN CLOUD chamber. Our simulations show that, under the cold and humid conditions of the Southern Ocean and Antarctic, MSA-induced nucleation rates become comparable to those of SA, with MSA accounting for around 25% of CCN_{0.4} over the Southern Ocean and up to 40% over the Antarctic. MSA is therefore a key trace gas in the sulfur budget of these regions and a substantial source of CCN. This is particularly relevant for the Southern Ocean, where climate models exhibit a large positive shortwave radiation bias that has been linked to underestimated CCN concentrations.

1 Introduction

Sulfuric acid concentrations in the atmosphere are dominated by anthropogenic emissions, particularly from coal and oil combustion, metal smelting, vehicular and maritime traffic, and oil refining. These activities release sulfur dioxide (SO₂), which is subsequently oxidized by hydroxyl radicals (OH) to form sulfuric acid (SA, H₂SO₄). In recent decades, stringent air quality regulations have substantially reduced anthropogenic SO₂ emissions (Vestreng et al., 2007; Sun et al., 2018; Yuan et al., 2024), increasing the relative importance of natural sulfur sources.

Volatile methylated sulfur (VMS) compounds are an important natural source of sulfur in the atmosphere. Estimates of total VMS emissions range from 16 to 37 Tg(S)yr⁻¹ (Wohl et al., 2024; Cala et al., 2023; Chen et al., 2018; Hezel et al., 2011; Cosme et al., 2002), while volcanoes account for an additional 21 – 25 Tg(S)yr⁻¹ (Carn et al., 2017). The most prominent VMS is dimethyl sulfide (DMS, CH₃SCH₃) which is produced in the ocean surface layer through enzymatic and bacterial degradation of dimethylsulfoniopropionate (DMSP). It is subsequently oxidized in the atmosphere to low-volatility compounds such as methanesulfonic acid (MSA, CH₃SO₃H) and SA that can drive new particle formation.

While DMS has long been the focal point of marine sulfur research, growing evidence suggests that other VMS species also play an important role. In particular, methanethiol (MeSH, CH₃SH), which is also produced from DMSP degradation, may contribute up to one-third of the total oceanic sulfur flux (Wohl et al., 2024). In the atmosphere, MeSH competes with DMS for oxidants such as the hydroxyl radical (OH), nitrate (NO₃), bromine monoxide (BrO), bromine (Br) and chlorine (Cl), thereby modifying the oxidative fate of DMS, extending its atmospheric lifetime, and influencing the partitioning between MSA and SA (Tashmim et al., 2025). Despite these implications, MeSH is still largely neglected in global model studies, which may result in a systematic underestimation of total biogenic marine sulfur input to the atmosphere.

Observations indicate similar abundances of MSA as SA in the marine boundary layer (Eisele and Tanner, 1993; Berresheim et al., 2002; Mauldin III et al., 2003; Brean et al., 2021; Boyer et al., 2024; Jokinen et al., 2018; Thakur et al., 2022; Schmale and Baccarini, 2021; Beck et al., 2021), indicating a likely important role of this gas in the atmospheric sulfur budget. However, substantial uncertainties remain in representing MSA and its precursors in global models. Depending on the ambient temperature, humidity and available gas phase bases, MSA exhibits highly variable gas-aerosol partitioning (Berresheim et al., 2002; Hodshire et al., 2019), complicating its representation in chemical transport models.



40 Another major source of uncertainty in the MSA budget stems from the oxidation chemistry of DMS itself (Cala et al., 2023; Fung et al., 2022b; Chen et al., 2018; Hezel et al., 2011; Cosme et al., 2002; Jacob et al., 2026), particularly the oxidation by halogen radicals. Several studies have shown that including reactions with BrO is essential to capture the observed seasonality and latitudinal variability of DMS and its oxidation products (Jongebloed et al., 2025; von Glasow and Crutzen, 2004). Most
45 instance, none of the CMIP6 models accounts for DMS oxidation explicitly, treating DMS as a prescribed, non-reactive tracer (Thornhill et al., 2021). These limitations are particularly problematic over the Southern Ocean, where VMS derived sulfur is the main driver of particle formation and growth (He et al., 2026).

Whereas it is well established that SA drives atmospheric particle nucleation (Dunne et al., 2016; Jokinen et al., 2018), it has only recently been found that MSA nucleates together with ammonia (NH_3) below -10°C , at rates comparable to SA-NH_3
50 (Baalbaki et al., 2026). This underscores the need for model studies to evaluate the impact of MSA on global aerosol formation and growth (Hodshire et al., 2019; Ning et al., 2026).

The climate relevance of these sulfur-derived particles is twofold: they exert, firstly, a direct radiative forcing by scattering incoming solar radiation (Myhre et al., 2013) and, secondly, an indirect effect by modifying cloud properties, which is particularly important in remote oceanic regions where CCN sources are limited (Charlson et al., 1987; Niu et al., 2025). Marine
55 sulfur emissions occur primarily over low-albedo ocean surfaces, enhancing the net cooling (Charlson et al., 1987).

Here, we use the global chemistry-climate model EMAC to evaluate four gas-phase sulfur oxidation mechanisms, each with varying levels of chemical complexity, and assess their ability to reproduce observed DMS mixing ratios, SA and MSA concentrations. In addition, we perform targeted sensitivity simulations to (i) test heterogeneous sulfur chemistry on deliquescent aerosols, (ii) evaluate the role of MeSH emissions and chemistry on MSA abundances, (iii) assess the importance of halogens
60 on VMS oxidation, and (iv) evaluate the contribution of MSA to new particle formation (NPF). Our study aims to identify the key processes for marine VMS chemistry and to quantify the impact of VMS oxidation products on aerosol particle formation and CCN concentrations.

2 Model Configuration

The ECHAM/MESSy Atmospheric Chemistry (EMAC) model is a numerical chemistry and climate simulation system that
65 includes sub-models describing processes in the troposphere and middle atmosphere, and their interaction with oceans, land and human influences (Jöckel et al., 2006). It uses the second version of the Modular Earth Submodel System (MESSy2, Jöckel et al., 2010) to link multi-institutional computer codes. The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al., 2003).

In this study we used a set-up based on Kohl et al. (2023), and here we summarize the most important aspects for this work.
70 The model integrations are performed on a T63 spectral decomposition which corresponds to a Gaussian grid resolution of $1.875^\circ \times 1.875^\circ$ (approx. $200\text{ km} \times 200\text{ km}$ at the equator) with 31 vertical pressure levels up to 10 hPa (about 32 km) altitude and a corresponding time-step of 10 minutes. For this study, we operate EMAC in the quasi chemistry-transport model mode,



i.e., without feedbacks between chemistry and dynamics, as described in Deckert et al. (2011), with the dynamics nudged (Jeuken et al., 1996; Jöckel et al., 2006) towards the fifth-generation European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis (ERA5, Hersbach et al. (2023)).

In the model, terrestrial DMS emissions are prescribed using a climatological dataset from Spiro et al. (1992), contributing $0.66 \text{ Tg(S)yr}^{-1}$, or approximately 2% of the total annual DMS flux. The remaining 98% of atmospheric DMS originates from oceanic emissions. For the oceanic emissions, the AIRSEA submodel (Pozzer et al., 2006) has been applied using an ocean surface climatology and Wanninkhof (1992) for the water-side transfer velocity parametrization. For the ocean surface concentration climatology we used the climatologies compiled by Hulswar et al. (2022) and Wohl et al. (2024) for DMS and MeSH, respectively.

Bromine and chlorine emissions from sea ice and snow-covered land are calculated online as described in Toyota et al. (2011). Bromocarbon emissions are included using the annually averaged emission fields from Warwick et al. (2006), processed by the submodel OFFEMIS (Kerkweg et al., 2006). BrO is a major sink for DMS and plays a crucial role in the resulting oxidized sulfur compounds. Bromine enters the atmosphere from the ocean either by the direct emission of bromocarbons (CHBr_3 , CH_2Br_2 , CH_2ClBr , CHClBr_2 and CHCl_2Br) from the sea surface or by debromination from sea-salt aerosols. In addition, bromine and chlorine are emitted from snow pack and sea ice. Sander et al. (1999), Zhang et al. (2024) and Schill et al. (2025) pointed out that bromine is also recycled on non-sea-salt aerosols (non-SSA). We account for that within the AERChem sub-submodel which includes heterogeneous bromine chemistry on deliquescent aerosols.

2.1 VMS gas-phase chemical mechanisms

The gas-phase chemistry is calculated by the submodel MECCA (Module Efficiently Calculating the Chemistry of the Atmosphere, Sander et al., 2019). The four mechanisms are based on the same underlying mechanism, solely the reactions related to VMS have been modified. In addition, all mechanisms have been extended or updated, so as to have the same reactions between DMS and halogens, as well as MeSH chemistry, taken from Burkholder et al. (2019). Table 1 gives an overview of the four VMS chemical schemes, which are described in more detail below, the full lists of reactions are provided as supplement material.

MECCA: The simplest VMS chemistry scheme evaluated in this study contains 5 gas-phase species and 11 reactions associated with VMS (Jöckel et al., 2016). Reaction with the hydroxyl radical (OH) proceeds via H abstraction or addition to the DMS molecule. Another addition pathway is the reaction with BrO. Hydrogen abstraction is also initiated by the nitrate radical (NO_3), chlorine and bromine. Products and major intermediates are sulfur dioxide (SO_2) and MSA. In this mechanism, MSA is almost exclusively produced from the oxidation of CH_3SO_2 by ozone and the hydroperoxyl radical (HO_2), where CH_3SO_2 is an intermediate product of the DMS abstraction pathway. The intermediate CH_3S is not included, thus MeSH is neglected.

Fung22: Adapted from Fung et al. (2022a), this mechanism covers a broad range of VMS-derived reactions and intermediates. In total, the mechanism contains 37 species and 45 reactions related to VMS. The H-abstraction pathway is mainly based



on Hoffmann et al. (2016). The OH-addition pathway was adapted from Fung et al. (2022a), although the temperature-dependent reaction rates are different in this study than those in Table 6 of Fung et al. (2022a).

MCM: The Master Chemical Mechanism (MCM) v3.3.1 (Saunders et al., 2003) uses DMS-halogen reactions from Burkholder et al. (2019). In total, 34 gas-phase species and 77 gas-phase reactions in the *MCM* are associated with VMS.

- 110 **Shen:** The Shen et al. (2022) scheme introduces an improved DMS oxidation mechanism for the production of gas-phase MSA, based on experimental results from the Cosmics Leaving Outdoor Droplets (CLOUD) chamber at CERN. The proposed reactions represent an add-on/correction to the *MCM* scheme. The VMS chemistry in this scheme involves 47 species and 109 reactions.

Table 1. Overview of VMS chemical mechanisms: name, number of species and reactions associated with VMS. Further details are provided in the Supplement.

Name	Complexity	Reference
<i>MECCA</i>	5 species, 11 reactions	Sander et al. (2019)
<i>Fung22</i>	37 species, 45 reactions	Fung et al. (2022a)
<i>MCM</i>	34 species, 77 reactions	Saunders et al. (2003)
<i>Shen</i>	47 species, 109 reactions	Shen et al. (2022)

2.2 Multiphase sulfur chemistry

- 115 Aerosol dynamics and microphysics are simulated by the submodel GMXe (Pringle et al., 2010), simulating aerosols in a modal distribution, with the aerosol-thermodynamic equilibrium estimated by ISORROPIA Fountoukis and Nenes (2007). Additional non-equilibrium water-phase chemistry in the aerosol phase is further calculated with the AERCHEM sub-submodel (Rosanka et al., 2024). The uptake and release of gases by cloud and rain droplets, as well as their aqueous-phase processing, are represented by the SCAV submodel (Tost et al., 2006). Atmospheric sulfur compounds DMS, dimethyl sulfoxide (DMSO,
- 120 CH_3SOCH_3), hydroperoxymethyl thioformate (HPMTF, HOCH_2SCHO), methanesulfinic acid (MSIA, $\text{CH}_3\text{SO}_2\text{H}$), MSA, and SA can partition to cloud and rain droplets, and to deliquescent aerosols, where they may undergo chemical transformation. In addition, MSIA, MSA, and SA can dissociate or hydrate in the aqueous phase, thereby influencing acidity levels. The aqueous-phase DMS oxidation schemes implemented in SCAV and AERCHEM build on von Glasow and Crutzen (2004); Fung et al. (2022b); Hoffmann et al. (2016). Henry's law constants are taken from Sander (2023); Brimblecombe and Clegg
- 125 (1988), while dissociation constants follow Clarke and Woodward (1966); Wudl et al. (1967); Seinfeld and Pandis (1998). The phase transfer and the aqueous-phase oxidation scheme, adopted in all the simulations, is presented in the electronic supplement.



2.3 MSA condensation and new particle formation

Following the approach of Vignati et al. (2004): the change in gas-phase MSA concentration by condensation MSA_g^c over time depends on the number of particles N_i and the condensation coefficient C_i per mode i

$$\frac{d[\text{MSA}_g^c]}{dt} = - \sum_{i=1, \text{n modes}} N_i C_i [\text{MSA}_g^c] \quad (1)$$

where the condensation coefficient C_i for mode i is calculated as described in Fuchs (1964)

$$C_i = \frac{4\pi D \bar{r}_{gi}}{\frac{4D}{\alpha \nu \bar{r}_{gi}} + \frac{\bar{r}_{gi}}{\bar{r}_{gi} + \Delta}} \quad (2)$$

with $\bar{r}_{gi}$ geometric mean radius of mode i , D diffusion coefficient, ν thermal velocity, Δ mean free path length and α accommodation coefficient. For the insoluble modes, α is 0.3.

For the soluble modes, the accommodation coefficient α is varied depending on MSA volatility regimes. The transition temperature T_{trans} has been estimated along the equilibrium vapor pressure line $C_{eq} = 10^{-2} \mu \text{ gm}^{-3}$, following the work of Hodshire et al. (2019), based on the Extended Aerosol Inorganic Model (E-AIM) (Clegg et al., 1992) and dependent on temperature (Donahue et al., 2006) and relative humidity (RH) (Chen et al., 2018):

$$T_{\text{trans}}(\text{RH}) = a - b \cdot \text{RH} + c \cdot \text{RH}^2 - d \cdot \text{RH}^3 + e \cdot \text{RH}^4, \quad (3)$$

where the coefficients are $a = 2.52 \cdot 10^2$, $b = 6.19 \cdot 10^{-1}$, $c = 3.49 \cdot 10^{-2}$, $d = 5.6 \cdot 10^{-4}$ and $e = 3.32 \cdot 10^{-6}$. Three MSA volatility regimes and corresponding α values have been defined, based on ambient temperature, RH, and ammonia (NH_3) concentration: 1. *Volatile* ($\alpha = 0.0$, no condensation), when RH is below 90% and the ambient NH_3 concentration is lower than three times the ambient MSA concentration; 2. *Semi-volatile* ($\alpha = 0.1$, De Bruyn et al. (1994)), when either the RH is less than or equal to 90%, or the ambient NH_3 concentration is at least three times greater than the ambient MSA concentration, and the temperature T is higher than the transition temperature T_{trans} ; 3. *Negligible-volatility* ($\alpha = 1.0$), when the ambient NH_3 concentration is greater than or equal to three times the ambient MSA concentration, and the temperature T is lower than T_{trans} . We note that these estimates are likely to understate the role of MSA, i.e., MSA may condense to the particle phase and contribute to NPF already at lower RH and/or higher temperatures than we assume for this study (Hanson, 2005; Yu et al., 2026).

Nucleation of MSA has been implemented based on parameterization of recent CLOUD results (Baalbaki et al., 2026). MSA is allowed to nucleate when (i) the gas-phase ammonia mixing ratio in the ambient air is at least three times larger than the MSA mixing ratio, (ii) the ambient temperature is lower than T_{trans} (see equation 3), so that MSA is in the negligible-volatility regime, and (iii) the temperature is below -10°C , consistent with the experimental basis of Baalbaki et al. (2026). If these conditions are fulfilled, the sum of SA and MSA is taken as input for ternary nucleation as described in Dunne et al. (2016), otherwise only SA is considered. An overview of the NPF channels used in this study is given in Table 2.



Table 2. Summary of the NPF channels used in this study, with their molecular composition and respective publication. Dunne et al. (2016) fitting parameters are taken from Gordon et al. (2017). The brackets indicate that MSA only contributes under certain atmospheric conditions to NPF, described in the text. HOMs are highly oxygenated molecules from monoterpenes.

New Particle Formation Channels	Molecular Composition	Reference
SA neutral & ion-induced binary	H_2SO_4	Dunne et al. (2016)
SA (MSA) neutral & ion-induced ternary	$[\text{H}_2\text{SO}_4 + (\text{CH}_3\text{SO}_3\text{H})] - \text{NH}_3$	Dunne et al. (2016); Baalbaki et al. (2026)
Neutral & ion-induced pure HOMs	HOMs	Kirkby et al. (2016)
Synergistic	$\text{H}_2\text{SO}_4 - \text{NH}_3 - \text{HNO}_3$	Wang et al. (2022)

2.4 Model simulations

To investigate the impact of DMS oxidation pathways, MeSH emissions, halogen reactions, and (SA+MSA)- NH_3 - H_2O NPF on the fate of marine sulfur emissions, we conducted a series of simulations spanning the years 2016-2019 to match available observations. Eight numerical integrations were performed, with the model setups summarized in Table 3.

The **MECCA**, **Fung22**, **MCM** and **Shen** simulations represent the EMAC model integrations based on the four corresponding chemical mechanisms (see Sect. 2.1). Among the four, the **Shen** mechanism offers the most balanced performance, combining the lowest spread for both SA and MSA with the highest correlation for MSA (for SA it is within 0.02 of the best value, obtained by **MECCA**, which however strongly underestimates SA), which motivates its use as the baseline configuration for the sensitivity experiments: 1. **Shen no ACM**, with heterogeneous chemistry on deliquescent aerosols switched off, enabling to study the effect of aerosol multi-phase processes, 2. **Shen no MeSH**, in which we excluded MeSH emissions and chemistry, allowing us to quantify the impact of MeSH on oxidant partitioning and 3. **Shen no halog** switching off DMS reactions with halogens, assessing the sensitivity of VMS oxidation to halogens, 4. **Shen no NPF**, in which (SA + MSA)- NH_3 - H_2O nucleation is disabled, isolating the role of MSA in new particle formation. The ensemble of simulations allows a quantification of the individual contributions of chemical complexity, heterogeneous aerosol chemistry, precursor diversity, and oxidant availability to the global distribution of sulfur species and their role in aerosol formation. The corresponding model-observation comparisons for the four sensitivity experiments are provided in the Appendix (Figs. A1, A2 and A3 for DMS, SA and MSA, respectively).

3 Observations

To evaluate the model results, we compare simulated surface DMS mixing ratios as well as SA and MSA concentrations with ambient observations. These include measurements from five shipborne campaigns and nine ground-based monitoring stations, all located in regions influenced by marine boundary layer conditions. Fig. 1 shows the locations of the stations and the ship



Table 3. VMS chemical mechanisms and sensitivity tests for the Shen mechanism. The setups are identical except for the differences indicated here.

Simulation	Name	Description
1. Baseline chemical mechanism	MECCA	<i>MECCA</i> chemical mechanism Sander et al. (2019).
	Fung22	<i>Fung22</i> chemical mechanism Fung et al. (2022a).
	MCM	<i>MCM</i> chemical mechanism Saunders et al. (2003).
	Shen	<i>Shen</i> chemical mechanism Shen et al. (2022).
2. Sensitivity test	Shen no ACM	As Shen but without heterogeneous chemistry on deliquescent aerosols (AERCHEM (ACM) off).
	Shen no MeSH	As Shen but MeSH emissions and chemistry excluded.
	Shen no halog	As Shen but without DMS halogen reactions.
	Shen no NPF	As Shen but with no MSA NPF.

tracks. Further information about the stations such as location, altitude, measured species, timespan of the measurement and the respective reference are given in Table 4. Equivalent information for the ship campaigns are given in Table 5.

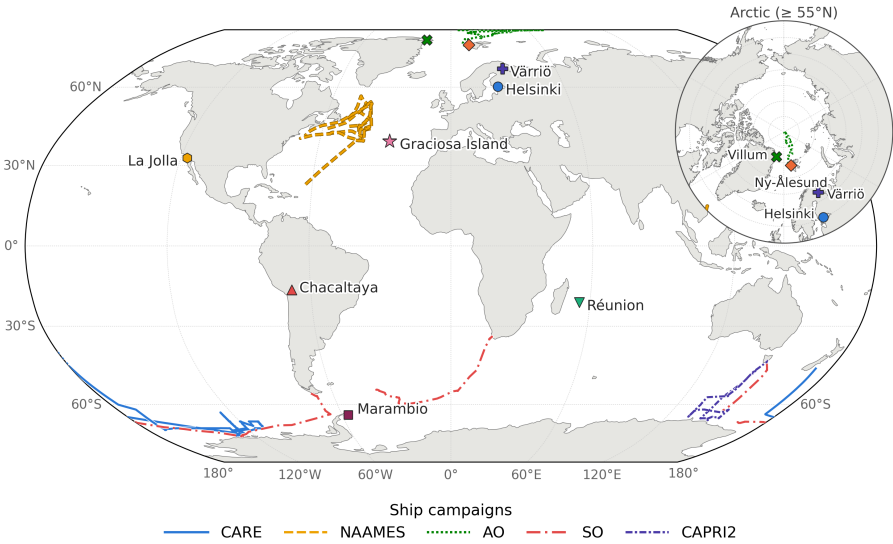


Figure 1. Ship tracks and locations of land-based stations that provide ambient measurements for this study (see Tables 4 and 5 for further details).

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For the model-observation comparison, simulated fields are sampled in the grid cell containing each measurement location and at the lowest model level, consistent with the near-surface altitude of the observations (Tables 4 and 5). For the moving ship campaigns, the model is sampled along the cruise track using the reported ship position, advancing the sampling location with



the ship. Model output is co-sampled in time with the observations: only time steps for which a concurrent observation exists
185 enter the comparison, and both model and observations are aggregated to a common temporal resolution before the statistical
metrics are computed. Because the observations span different years from the nudged simulation period (2016-2019), records
from outside this window (Villum, Graciosa Island, NAAMES1) are compared against the climatologically nearest simulated
year, as indicated in the footnotes of Tables 4 and 5; this introduces an unavoidable meteorological mismatch for those two
sites.

Table 4. Name, location, altitude and reference of land-based ground observations used in this work. DMS measurements have been taken from Graciosa Island and La Jolla. MSA and SA observations are taken from all other stations listed.

Station	Location (°N,°E)	Altitude a.s.l (m)	Species	Timespan	Ref.
Chacaltaya	(−16.35,−68.13)	5240	SA,MSA	05.05.2018 - 28.05.2018	Scholz et al. (2023)
Helsinki	(60.20,24.96)	25	SA,MSA	26.06.2019 - 17.08.2019	Thakur et al. (2022)
Marambio	(−64.24,−56.63)	198	SA,MSA	29.01.2018 - 21.02.2018	Quéléver et al. (2022)
Ny-Alesund	(78.93,11.93)	45	SA,MSA	25.03.2017 - 25.08.2017	Beck et al. (2021)
Reunion Island	(−21.08,55.35)	2150	SA,MSA	11.04.2018 - 17.04.2018	Salignat et al. (2023)
Varrio	(67.75,29.61)	370	SA,MSA	04.04.2019 - 24.10.2019	Jokinen et al. (2022)
Villum ¹	(81.60,−16.66)	24	SA,MSA	27.02.2015 - 24.08.2015	Beck et al. (2021)
Graciosa Island ²	(39.09,−28.03)	5	DMS	01.06.2022 - 15.07.2022	Bertram et al. (2023)
La Jolla	(32.87,−117.26)	5	DMS	03.09.2019 - 30.09.2019	Novak et al. (2022)

1. Cast to the year 2016 for model comparison

2. Cast to the year 2019 for model comparison

Table 5. Name, region, species, time-span and respective reference of the ship based observations used in this work.

Campaign	Region	Species	Time-span	Ref.
MOCCHA (AO-ship)	Arctic Ocean	SA, MSA	01.08.2018 - 19.09.2018	Baccarini et al. (2020)
NAAMES1 ³	North Atlantic	DMS	05.11.2015 - 02.12.2015	Bell et al. (2021)
NAAMES2	North Atlantic	DMS	11.05.2016 - 05.06.2016	Bell et al. (2021)
NAAMES3	North Atlantic	DMS	30.08.2017 - 24.09.2017	Bell et al. (2021)
NAAMES4	North Atlantic	DMS	20.03.2018 - 13.04.2018	Bell et al. (2021)
ACE (SO-ship)	Southern Ocean	SA, MSA	20.12.2016 - 19.03.2017	Baccarini et al. (2021)
CHINARE 34 (CARE)	Southern Ocean	DMS	24.02.2018 - 31.03.2018	Zhang et al. (2020)
CAPRICORN-2 (CAPRI2)	Southern Ocean	SA,MSA	10.01.2018 - 22.02.2018	Miljevic et al. (2025)

3. For model comparison the first leg was cast to the year 2016



190 4 Results

In this section we evaluate the eight simulations shown in Table 3 with boundary layer observations of DMS, SA and MSA, and analyse the modeled global distributions, emissions and oxidation pathways of the marine sulfur species. A consistent picture emerges across the four chemical mechanisms. While surface DMS is reproduced well and is largely insensitive to the choice of mechanism, both SA and MSA are overestimated by most of the mechanisms (the exceptions being SA in **MECCA** and MSA in **MCM**). This points to errors beyond the gas-phase oxidation scheme, which we will isolate with the sensitivity simulations, as discussed below.

4.1 Dimethyl sulfide (DMS)

Observations from two shipborne campaigns (CARE and NAAMES) and two ground-based monitoring sites (Graciosa Island, Azores and La Jolla, California) were used to evaluate the model results at the surface for DMS. A summary of the comparison is given in Table 6. The comparisons for the four chemical mechanisms are also shown in Figure 2, which reveals largely

Table 6. Statistical metrics of DMS model simulations compared with observations: Mean Pearson correlation coefficients r , mean standard deviation of the logarithmic model-to-observation difference $\sigma_{\log}$ and the mean over all sites of the median model-to-observation ratio (Median (M/O)). The latter is obtained by first computing the median model-to-observation ratio at each site and then averaging these per-site medians with equal weight per site. This site-averaged metric is a different aggregation from the value annotated in the corresponding violin figures (Figs. 2, 6 and 7), which is the median of the model-to-observation ratios taken over all individual measuring points pooled across sites. The two values therefore need not coincide, the site-averaged metric being more sensitive to individual sites with extreme model bias.

Simulation	r	$\sigma_{\log}$	Median (M/O)
MECCA	0.07	0.49	0.90
Fung22	0.10	0.48	0.97
MCM	0.11	0.47	1.00
Shen	0.10	0.47	1.01
Shen no ACM	0.11	0.49	1.00
Shen no MeSH	0.10	0.46	0.93
Shen no halog	0.13	0.46	1.28
Shen no NPF	0.10	0.47	1.00

200

consistent behavior across the different mechanisms. The model results reproduce the NAAMES campaign measurements over the North Atlantic with reasonable accuracy, and inter-mechanism variability is generally small. An exception occurs during the NAAMES-3 leg, where the numerical integrations with the different chemical schemes consistently overestimate DMS mixing ratios relative to observations. On the contrary, model results from all simulations tend to underestimate DMS levels



205 observed during the CARE campaign in the Southern Ocean and on Graciosa Island. Across the ship campaigns, model results–observation agreement is somewhat similar between the integration runs, with correlation coefficients r ranging from 0.07 to 0.13, log-transformed standard deviations ($\sigma_{\log}$) between 0.46 and 0.49, and median model-to-observation ratios (Median (M/O)) from 0.90 to 1.28. Station comparisons (Figure 2) at Graciosa Island (39°N, 28°W) and La Jolla (32°N, 117°W) show similarly robust agreement with observations, with no substantial sensitivity to the choice of chemical mechanism. The

210 overall low correlation reflects the time resolution of the monthly DMS sea surface climatological concentration on which flux calculations are based. While still timestep resolved physical parameters are taken into account (e.g. wind speed) to calculate the fluxes, a monthly sea-surface DMS climatology is too coarse to capture the variability in time observed by ambient measurements. Except for the model results from the **MECCA** simulation, all other integrations yield a median (M/O) of approximately 1. The **MECCA** simulation does not include MeSH chemistry, resulting in a shorter DMS lifetime because of

215 the missing competition for oxidants. This effect can also be seen in the **Shen no MeSH** sensitivity run, in which the Median (M/O) decreases by 0.08 relative to the **Shen** baseline run, solely due to the absence of oxidant competition between DMS and MeSH. This is in agreement with the results of Wohl et al. (2024), which observed an extended DMS lifetime due to MeSH presence.

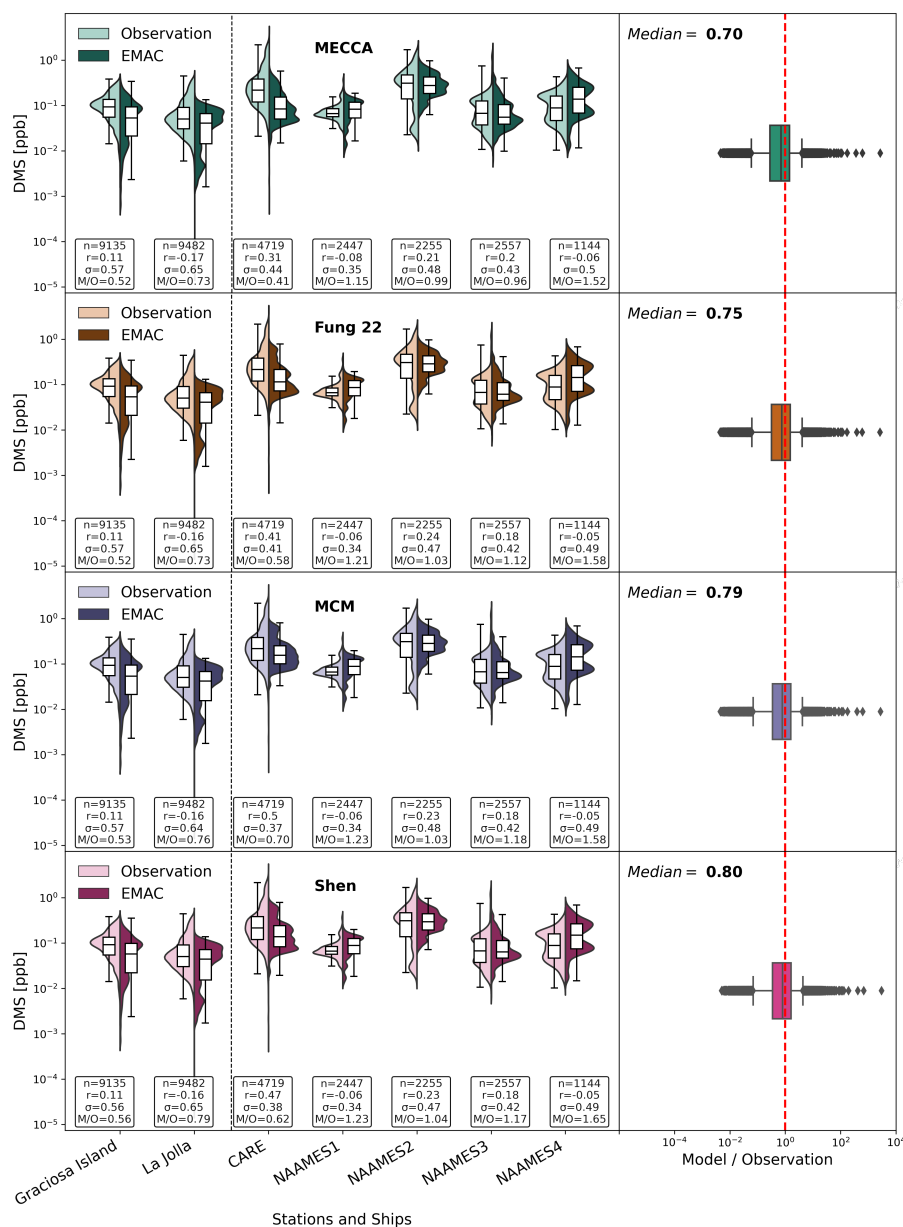


Figure 2. Comparison of modelled DMS mixing ratios with ambient observations for four mechanism integrations (**MECCA**, **Fung22**, **MCM**, and **Shen**), evaluated against two ship campaigns (NAAMES and CARE) and two stationary sites (Graciosa Island and La Jolla). Left: split violin plots with overlaid box-and-whisker elements, where the whiskers span the full data range. Right: box-and-whisker plots of the model-to-observation ratio, where the whiskers indicate the 5th and 95th percentiles. Boxes give the interquartile range. Annotated metrics are the number of sampling points n , the Pearson correlation coefficient r , the standard deviation of the logarithmic model-to-observation difference $\sigma_{\log}$, and the median model-to-observation ratio pooled over all measurement points. This pooled median differs from the site-averaged M/O in Table 6.



Global DMS fluxes, the modeled surface mixing ratio distribution, and the dominant chemical loss pathways, including their fractional contributions, are presented in Figure 3.

In Figure 3b, we see elevated DMS mixing ratios over the Southern Ocean despite the relatively modest emission fluxes in

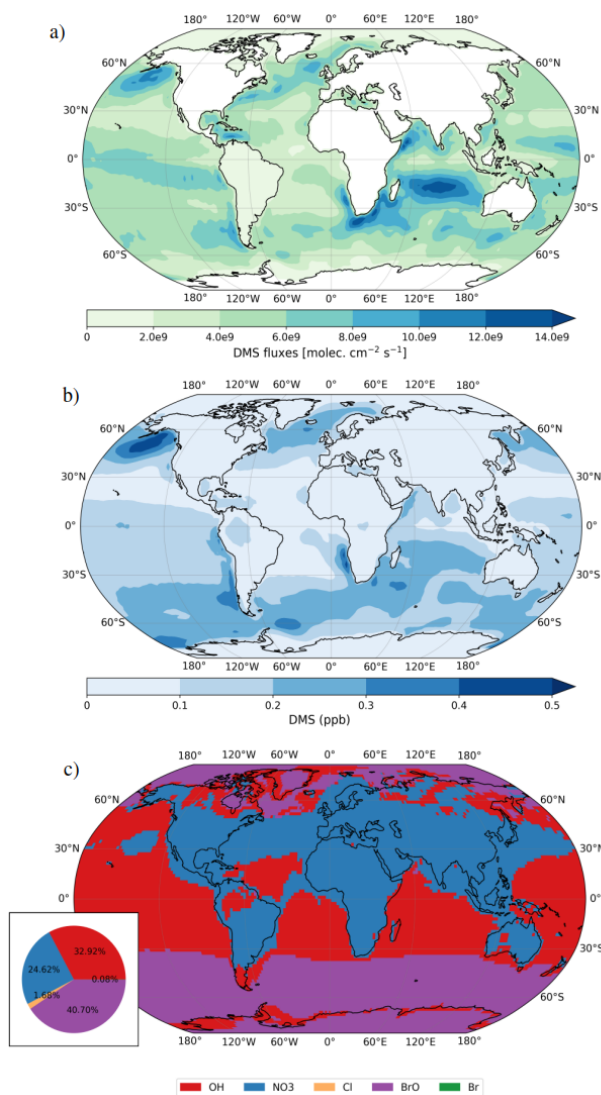


Figure 3. Global maps from the **Shen** baseline simulation showing (a) DMS emission fluxes averaged over the simulation period, (b) annual mean surface DMS mixing ratios, and (c) dominant DMS oxidation pathways at the surface. Panel (c) includes a pie chart indicating the fractional contributions of individual oxidation pathways (in percent).

that region. This reflects a weak total oxidation: OH is limited by the low photochemical activity at high latitudes, and NO₃ is suppressed in clean, low-NO_x environments. With both fast oxidants scarce, BrO becomes the leading DMS oxidant over the



Southern Ocean (Figure 3c), despite the relatively slow BrO+DMS reaction rate (which is slower than those of OH and NO₃),
225 because BrO is abundant. Since all oxidation pathways are slow in absolute terms, the overall DMS loss rate remains slow,
extending the DMS lifetime and allowing mixing ratios to build up in the marine boundary layer over the Southern Ocean.

During daytime at mid-latitudes, OH dominates DMS oxidation over the high-emission ocean regions. Over the continents,
where there is almost no local DMS source, the DMS has been advected from marine regions. In the more polluted, NO_x-rich
continental air it is oxidised predominantly by NO₃ at night. As a result, NO₃ is the dominant DMS oxidant above low-emission
230 regions such as the continents (Figure 3c).

Table 7. Annual DMS atmospheric emissions, lifetime and burden.

	Emis. Tg(S)yr ⁻¹	Burden Gg(S)	Lifetime	Reference	
Previous studies	16.0	63-66	1.5 d		Cala et al. (2023)
	22.5	74	1.2 d		Chen et al. (2018)
	23.6	110	1.8 d		Hezel et al. (2011)
	19.4	65	1.2 d		Cosme et al. (2002)
	23.5	135	2.1 d		Wohl et al. (2024)
This work	31.0	56	0.7 d	MECCA	Sander et al. (2019)
	30.8	67	0.8 d	Fung22	Fung et al. (2022a)
	30.8	70	0.8 d	MCM	Saunders et al. (2003)
	30.8	70	0.8 d	Shen	Shen et al. (2022)
	30.7	82	0.9 d	Shen no ACM	
	30.6	58	0.7 d	Shen no MeSH	
	30.0	129	1.6 d	Shen no halog	
	30.8	70	0.8 d	Shen no NPF	

A compilation of global DMS emissions, atmospheric burden, and lifetimes derived in this study, alongside values found
in the literature, is provided in Table 7. Nearly all atmospheric DMS is removed by oxidation, with deposition accounting for
only a negligible part ($2 \cdot 10^{-5}$ Tg(S)yr⁻¹).

In our simulations, the DMS lifetime (0.8 d) is shorter than previous values reported in the literature. The DMS lifetime is
235 substantially increased if reactions with halogens are omitted (**Shen no halog**), highlighting their importance. Since bromine
emissions are often underestimated or neglected in global models, this can lead to systematic overestimation of DMS lifetimes
in many studies. When MeSH chemistry is excluded, the increased availability of oxidants leads to an even shorter DMS
lifetime.



Interestingly, the model run **Shen no ACM** shows an increase in the atmospheric lifetime of DMS by 0.1 d compared with the **Shen** baseline run. This can be explained by the recycling of bromine on deliquescent aerosols, which is calculated by AERCHEM. Switching it off leads to a reduction in reactive BrO and, consequently, an increase in DMS lifetime.

We note that the DMS emissions derived here ($\sim 31 \text{ Tg(S)yr}^{-1}$) exceed the range of the earlier estimates compiled in Table 7 ($16\text{--}23.6 \text{ Tg(S)yr}^{-1}$). This mainly reflects the AIRSEA flux formulation, which dynamically accounts for the atmospheric DMS saturation just above the ocean surface and therefore yields higher sea-to-air fluxes; combined with the short simulated lifetime (0.8 d), it results in an atmospheric burden ($56\text{--}70 \text{ Gg(S)}$) that remains within the range of previous studies. The remaining differences in DMS emission estimates across models can arise from several sources, including differences in sea-ice coverage, land-sea masks, or air-sea exchange parameterizations. Differences in annual DMS emission fluxes between the model integrations within EMAC arise primarily from variations in the atmospheric DMS saturation just above the ocean surface, as the fluxes are computed based on the air-sea concentration gradient and governed by Henry's law solubility (Pozzer et al., 2006).

4.2 Methanethiol (MeSH)

In this study, annual MeSH emissions from the ocean to the atmosphere are estimated at 8.1 Tg(S)yr^{-1} , corresponding to about 20% of the total VMS flux. The MeSH atmospheric burden in the **Shen** simulation is 16 Gg(S) , giving an estimated lifetime of 18 h, shorter than the value previously reported by Wohl et al. (2024). Although the burdens remain within the range of comparable literature values, the annual sulfur emissions derived here are about 40% higher compared with Wohl et al. (2024), despite using the same surface ocean MeSH climatology. Both this study and Wohl et al. (2024) adopt the classical two-layer model for air-sea gas exchange; however, they diverge substantially in implementation. The method of Wohl et al. (2024) is compound-specific, relying on empirically tailored solubility and transfer velocity parameterizations for MeSH and DMS. Here, the AIRSEA submodel is designed as a generalized framework within the MESSy infrastructure, calculating sea-to-air fluxes based on a standardized set of species-dependent parameters. One notable distinction is that AIRSEA dynamically incorporates atmospheric gas concentrations into the flux calculation, allowing for elevated emissions depending on local atmospheric saturation. Furthermore, in Wohl et al. (2024) the MeSH/DMS approach imposes a flux cutoff above 85% sea-ice cover, whereas AIRSEA handles fractional ice coverage, potentially contributing to discrepancies in polar emission estimates. In addition, the studies differ in their choice of transfer velocity parameterization: Wohl et al. (2024) adopt the formulation of Johnson (2010), whereas the present work applies that of Wanninkhof (1992), which likely accounts for the largest portion of divergence in emission estimates.

MeSH emission fluxes, surface mixing ratios, and dominant atmospheric reaction pathways are presented in Figure 4. In tropical regions, the spatial emission patterns of MeSH closely follow those of DMS. At higher latitudes, distinct MeSH emission maxima are observed, particularly over the Southern Ocean, the North Pacific, and the North Atlantic.

Figure 4c presents the dominant and total fractional oxidation pathways of MeSH at the surface. Overall, 66 % of atmospheric MeSH is oxidized by OH, forming CH_3SO , which dominates the chemical sink across the Southern Hemisphere, over both land and ocean. Regionally, NO_3 becomes more important, particularly under nighttime or low-OH conditions. In the Northern



Hemisphere, NO_3 is the dominant oxidant over land surfaces. Over the ocean in the Northern Hemisphere, OH remains the primary oxidant, consistent with expectations based on daytime photochemistry.

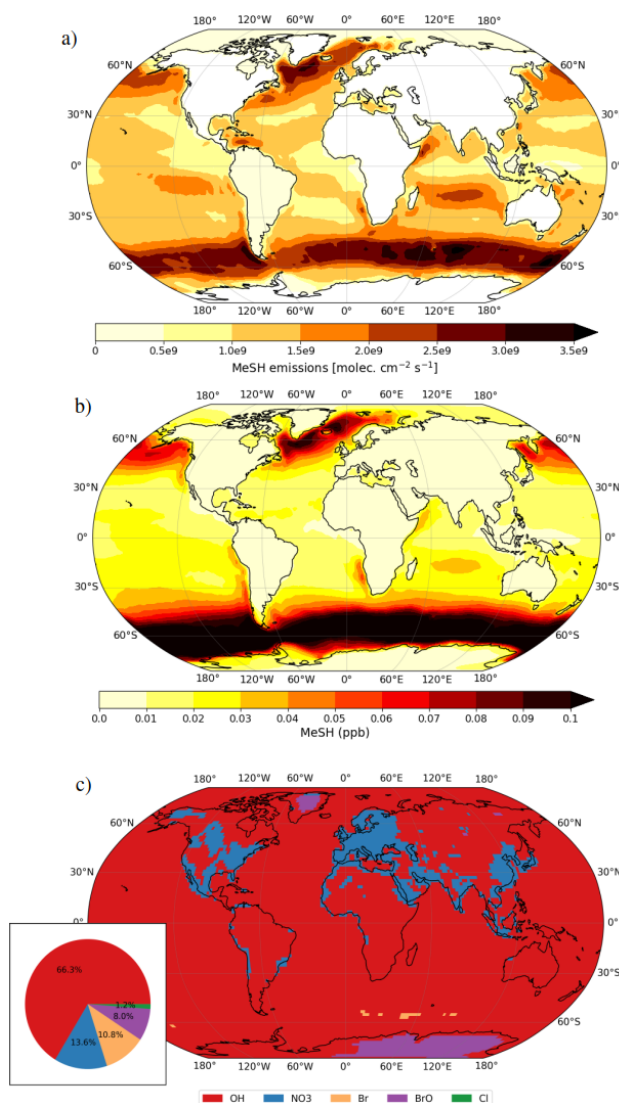


Figure 4. As Figure 3 but for MeSH.

275 The chemical loss pathways of MeSH are largely consistent between this study and Wohl et al. (2024), as both adopt identical rate coefficients for key gas-phase reactions. However, MeSH oxidation by atomic bromine (Br) is included in this work but was omitted in the earlier study. The inclusion of halogen chemistry is important for MeSH oxidation, especially in polar and marine boundary layer environments where bromine activation is relatively strong. Omitting it would lead to a lengthening of the lifetime by 12 h, partly reconciling our shorter lifetime with that of Wohl et al. (2024).

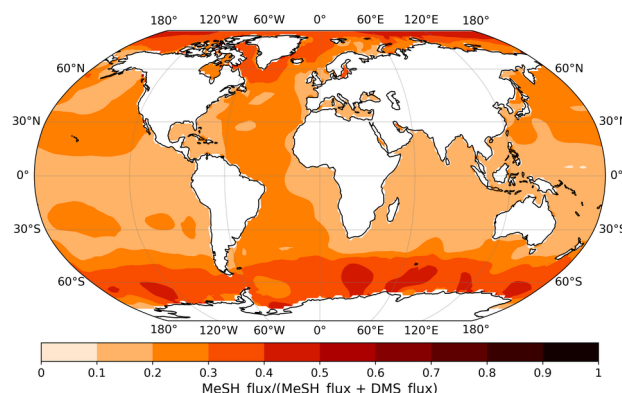


Figure 5. Global map of the fractional MeSH flux from the **Shen** baseline run.

280 The variability in the MeSH-to-DMS emission flux ratio in our model arises primarily from the sea surface concentration climatologies employed in this study. The disparity in the climatology reflects differences in the biogeochemical pathways of DMSP degradation, where MeSH and DMS are produced via competing enzymatic processes (Moran et al., 2012; Moran and Durham, 2019). These processes are modulated by regional oceanic conditions, including microbial community structure, nutrient availability, and water temperature, which likely contribute to the observed emission contrasts across latitude bands.

285 Lawson et al. (2020) reported that MeSH accounts for approximately 20% of the combined DMS - MeSH flux over the southwest Pacific Ocean. Other studies have found that MeSH concentrations can comprise up to 50% of the total volatile sulfur pool in surface waters of the Atlantic Ocean, with a pronounced increase toward higher latitudes (Gros et al., 2023).

The findings in Gros et al. (2023) are consistent with our model results, exhibiting similar latitudinal trends in the MeSH-to-total sulfur emission fraction, particularly in the Southern Ocean and North Atlantic regions (see Figure 5). VMS fluxes

290 are highest during summer, driven by increased production of DMSP at the ocean surface, which is primarily produced by phytoplankton, with MeSH exhibiting a stronger seasonal variability than DMS, likely due to differences in its production pathways from DMSP. As higher solar radiation intensifies ocean stratification, limiting the nutrient supply, MeSH production is favored over DMS (Chen et al., 2021). Given the projected increase in ocean stratification due to climate change (Li et al., 2020), we suggest that the role of MeSH in the marine sulfur cycle may become increasingly significant in the future.

295 4.3 Sulfuric acid (SA)

The comparison of numerical results for SA with observations from shipborne campaigns and coastal or island-based monitoring stations, primarily situated in remote marine environments, is presented in Figure 6 and summarized in Table 8. In the **MECCA** simulation, SA concentrations are generally underestimated. The model performs reasonably well at continental or near-continental stations with relatively stronger anthropogenic influence, such as Helsinki, Chacaltaya, and Varrio, yet

300 largely underestimates SA concentrations in the Southern Ocean. This suggests that the default mechanism is more suitable for simulating SA concentrations over land or in regions with anthropogenic influence, but fails to capture SA abundances



Table 8. SA model simulations compared with observations (see Table 6 for an explanation of the quantities).

Simulation	r	$\sigma_{\log}$	Median (M/O)
MECCA	0.40	0.97	0.66
Fung22	0.37	0.71	17.74
MCM	0.32	0.74	3.50
Shen	0.38	0.68	6.21
Shen no ACM	0.39	0.66	7.13
Shen no MeSH	0.38	0.73	2.55
Shen no halog	0.38	0.70	4.72
Shen no NPF	0.38	0.67	6.33

in remote marine environments. Previous EMAC studies comparing simulated SO_2 (Makroum, 2024) and particulate SO_4^{2-} (Pozzer et al., 2022) with observations in polluted regions of the United States, Europe, and East Asia actually have shown that the model tends to overestimate sulfur species in such environments.

305 The model results from simulation **Fung22** generally overestimates SA concentrations compared with both station and ship-based observations.

Model results from **MCM** compare relatively well in terms of Median (M/O) agreement, yielding a ratio of 3.5. However, its Pearson correlation coefficient is $r = 0.32$, indicating limited temporal or spatial correspondence with observations. The results from **MCM** exhibit a moderate logarithmic standard deviation (0.74), suggesting a reasonable spread. While **MCM** 310 overestimates SA at Helsinki and Varrio (two closely located sites) it substantially underestimates the SO-ship campaign, which plays a crucial role in constraining VMS-derived SA.

The **Shen** simulation generally overestimates SA concentrations. It obtains a Pearson correlation ($r = 0.38$) with a $\sigma_{\log}$ of 0.68. While most stations, as well as AO- and CAPRI2 ship campaigns are strongly overestimated, good agreement is present with the SO-ship campaign (Median (M/O)= 1.15).

315 Except for **MECCA** simulations, all other numerical integrations overestimate SA concentrations compared with the observational datasets. The ship campaigns show the largest spread between the mechanisms, indicating substantial uncertainties arising from the choice of DMS oxidation mechanism. While the modeled DMS mixing ratios remain similar between mechanisms, this suggests that high uncertainties are caused by second- or higher-generation effects within the chemical schemes.

The temporal correlations for the acids are moderate for SA ($r = 0.32$ -0.40) and low for MSA ($r = 0.12$ -0.26), and vary 320 little (Table 8 and 9) between mechanisms. Unlike DMS, whose weak correlations mainly reflects the monthly emission climatology, gas-phase SA and MSA have lifetimes of only minutes to hours and are governed by a fast, local balance between photochemical production and the condensation sink. Reproducing their short-term variability is therefore limited by the model resolution.

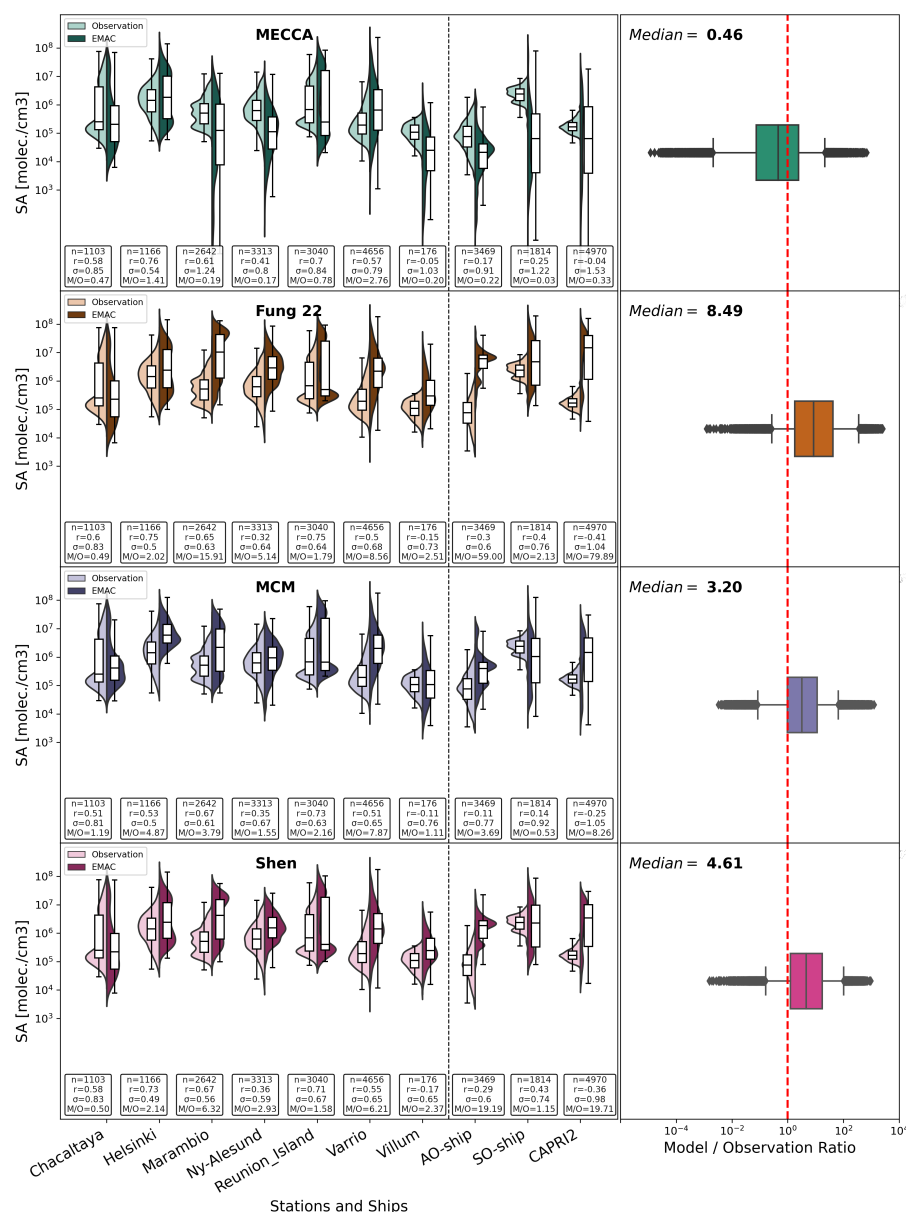


Figure 6. Comparison of modeled SA concentrations with ambient observations for the four mechanism model integrations (**MECCA**, **Fung22**, **MCM**, and **Shen**). In the left column SA concentrations are shown as split-violin-plots with Box-Whisker overlays. The Pearson correlation coefficient (r) and $\sigma_{\log}$ the standard deviation of the log difference between model and observation for each run are given. The right column shows the Box-Whisker of model over observation ratio for the four scenarios. Boxes represent the interquartile range (25th–75th percentiles), where the whiskers in the left column panels encompassing the full range of data points while the whiskers in the right column panels indicating the 5th and 95th percentile bounds. The value annotated in the right column is the median of the model-to-observation ratios over all individual measuring points, which is a different aggregation from the site-averaged Median (M/O) of Table 8 (the mean of the per-site median ratios) and need not coincide with it.



The model overestimates SA most strongly at high latitudes, over the Southern Ocean and the Arctic, where simulated concentrations generally exceed the observations (Fig. 6). We attribute the general tendency to overestimate SA at high latitudes to an underestimated condensation sink. Sea-salt emissions, and hence the aerosol surface area available for condensation, are likely underestimated over the Southern Ocean and the Arctic. A too-weak condensation sink allows gas-phase SA to accumulate rather than condense onto pre-existing particles, biasing simulated concentrations high. The same mechanism would explain the concurrent overestimation of MSA in these regions (Sect. 4.4), since MSA partitions onto deliquescent sea-salt and sulfate particles in an analogous manner. The **MECCA** simulation is the exception: it underestimates SA almost everywhere (avg. Median (M/O)= 0.66) and does not show the high-latitude bias. This is consistent with **MECCA** omitting MeSH emissions and chemistry, and thus lacking an important precursor source for both MSA and SA.

Within the Southern Ocean, however, the magnitude of this bias depends strongly on the campaign used for evaluation. Against CAPRI2 the overestimation is severe (**Fung22** Median (M/O)= 78.89, **Shen** Median (M/O)= 19.71), whereas against SO-ship the same simulations overestimate only mildly (Median (M/O)= 2.13 and 1.15, respectively). The **MCM** mechanism even reverses sign between the two campaigns, underestimating SA against SO-ship (Median (M/O)= 0.53) while overestimating it against CAPRI2 (Median (M/O)= 8.26). This campaign-to-campaign spread is driven in part by the observational datasets themselves rather than by the oxidation mechanism, consistent with the bias being similar across all chemical schemes.

4.4 Methanesulfonic acid (MSA)

The same set of observations used for SA is also used for the MSA comparison. An overview of the results of the comparison for the eight model simulations is given in Table 9. Figure 7 compares modeled MSA concentrations from the four different

Table 9. MSA model simulations compared with observations (see Table 6 for an explanation of the quantities).

Simulation	r	$\sigma_{\log}$	Median (M/O)
MECCA	0.12	0.89	6.19
Fung22	0.21	0.86	3.91
MCM	0.14	0.98	0.29
Shen	0.25	0.70	3.37
Shen no ACM	0.26	0.70	3.41
Shen no MeSH	0.24	0.71	2.56
Shen no halog	0.25	0.70	2.59
Shen no NPF	0.25	0.71	3.48

chemical mechanisms with observations from station sites and ship campaigns. The **MECCA** simulation overestimates MSA concentrations at all sites and ship routes, except CAPRI2, yielding the highest overall model-to-observation median ratio of 6.19, with a correlation of $r = 0.12$ and a logarithmic standard deviation of $\sigma_{\log} = 0.89$.



345 The **Fung22** simulation underestimates MSA concentrations at Chacaltaya, Helsinki and Reunion-Island. However, it overestimates MSA in general, in particular at Marambio station and the AO-ship campaign. It exhibits an overall spread of $\sigma_{\log} = 0.86$, with particularly large scatter at Chacaltaya, Helsinki, and Varrio.

MCM is the only simulation which underestimates MSA concentrations across all sites. It yields Pearson correlation coefficient of $r = 0.14$ and a spread of $\sigma_{\log} = 0.98$, yielding consistent underprediction without substantial variability. The underestimation of DMS oxidation products in MCM was already reported in Rosati et al. (2021).

350 The **Shen** simulation performs similarly to **Fung22** in terms of overall concentration levels, with a slightly lower Median (M/O) ratio. However, it provides improved correlation ($r = 0.25$) and a lower spread ($\sigma_{\log} = 0.70$) compared with **Fung22**. The **Shen** simulation shows good agreement with Ny-Alesund and Varrio sites, SO-ship and CAPRI2 campaigns, while underestimating Chacaltaya, Helsinki and Reunion Island station and overestimating MSA elsewhere.

355 As for SA, the low MSA correlations are consistent across mechanisms and reflect the difficulty of phase-matching a short-lived, locally controlled species at the model and emissions spatio-temporal resolution rather than the choice of oxidation scheme.

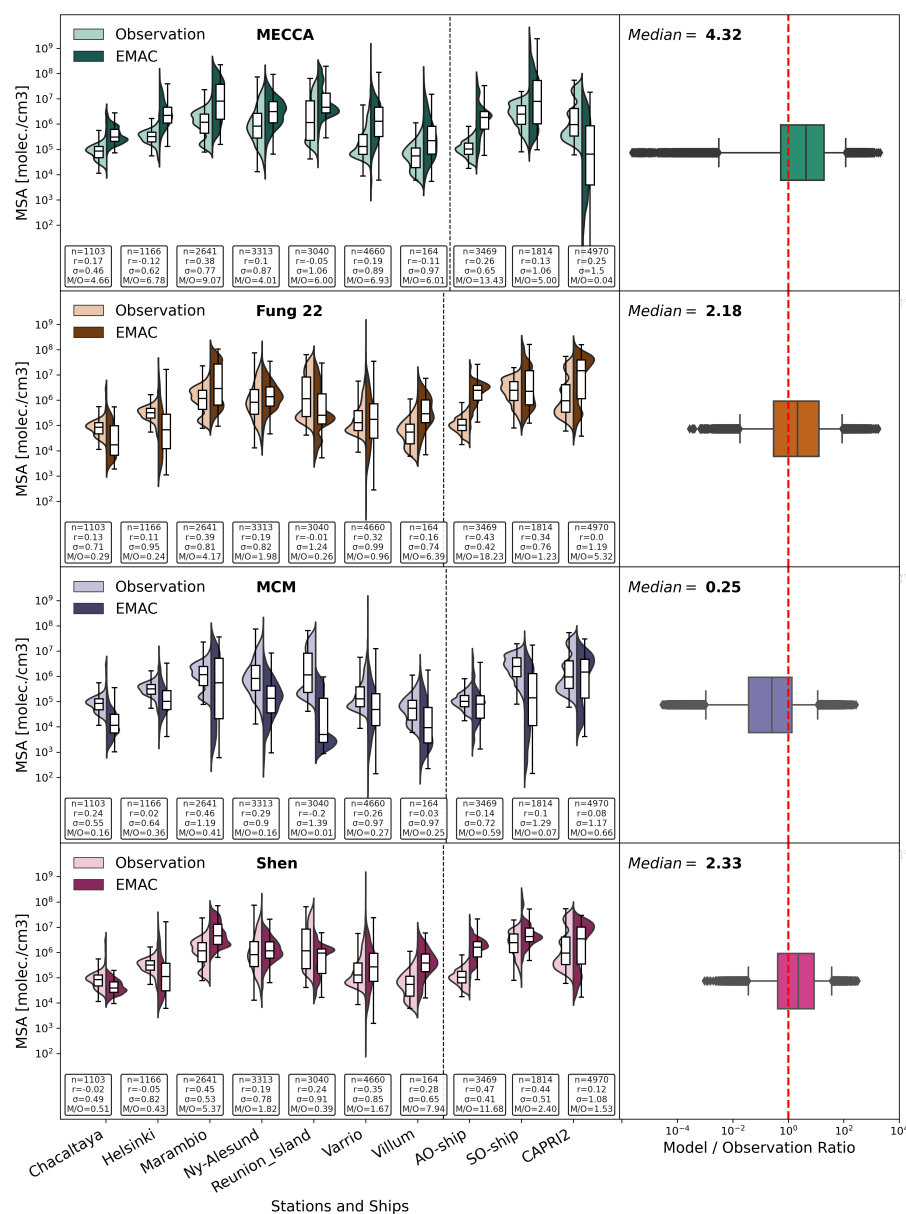


Figure 7. Comparison of modeled MSA concentrations ratios with ambient observations for the four mechanism model integrations (MECCA, Fung22, MCM, and Shen). The left column shows MSA concentrations as split-violin-plots with Box-Whisker overlays. The Pearson correlation coefficient (r) and $\sigma_{\log}$ the standard deviation of the log difference between model and observation for each run are given. The right column shows the Box-Whisker of model over observation ratio for the four scenarios. Boxes represent the interquartile range (25th–75th percentiles), where the whiskers in the left column panels encompassing the full range of data points while the whiskers in the right column panels indicating the 5th and 95th percentile bounds. The value annotated in the right column is the median of the model-to-observation ratios over all individual measuring points, which is a different aggregation from the site-averaged Median (M/O) of Table 9 (the mean of the per-site median ratios) and need not coincide with it.



In remote marine environments, MSA is similarly abundant to SA, despite being uniquely derived from the oxidation of VMS. A recent global modeling study using several DMS oxidation schemes reported divergent results across mechanisms (Jongebloed et al., 2025). To date, a robust and consistent model representation of MSA is lacking, underscoring the need for further investigation into the controlling processes of MSA formation, particularly under varying oxidant conditions.

In Figure 8, the global distribution of MSA surface mixing ratios and chemical production rates are shown. The highest chemical production occurs in coastal areas in the Northern Hemisphere between 30° N and 60° N, with additional regional maxima in the Southern Ocean, the west coast of South America, and southern Africa. In contrast, the largest MSA mixing ratios are found in the Southern Hemisphere south of 60° S and along the coast of Antarctica indicating that MSA tends to accumulate in clean, remote environments. Cold, dry, and acidic conditions in these regions favor elevated gas-phase MSA through enhanced chemical production and reduced condensation sink.

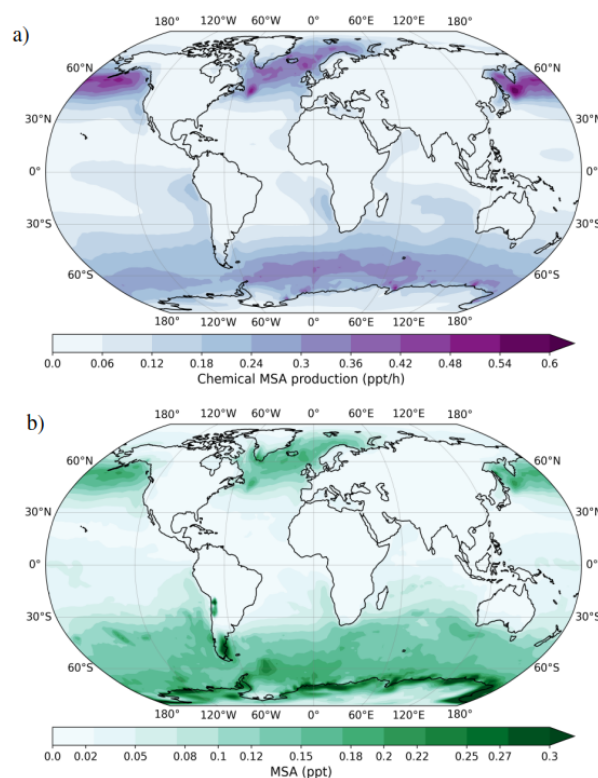


Figure 8. Simulations of the Shen mechanism showing annual mean values for a) MSA production rates and b) MSA mixing ratios in the surface layer.



4.5 Aqueous-phase methanesulfonic acid

Significant amounts of MSA are present in the liquid phase when aqueous-phase VMS chemistry is calculated in deliquescent aerosols, reflecting the dynamic partitioning of MSA between the gas and aerosol phases. The sensitivity simulation **Shen** **no ACM** excludes this aerosol-phase chemistry, yet the effect on gas-phase MSA concentrations is small. The high effective Henry's law solubility of $6.5 \cdot 10^{13} \text{ mol}^2 \text{ kg}^{-2} \text{ atm}^{-1}$ (Brimblecombe and Clegg, 1988) keeps MSA in the aqueous phase, suppressing evaporation while enhancing the condensation sink. The small difference between the two simulations indicates that the condensation scheme in GMXe already removes most of the available gas-phase MSA before the gas–aqueous equilibrium is computed by AERCHEM. The effective Henry's law solubility constant for MSA is, however, subject to large uncertainties (Wang et al., 2017; Campolongo et al., 1999). An underestimation can drive excessive MSA degassing from both aerosols and cloud droplets, leading to an overprediction of gas-phase MSA. Additional uncertainties arise from the aqueous-phase oxidation pathways of reduced sulfur species (Hoffmann et al., 2021), which may overproduce particulate MSA, oversaturate the dissolved fraction, and thereby strongly affect the modeled gas–particle equilibrium.

Because MSA is less acidic than sulfuric acid, its partitioning in the aqueous phase is also strongly influenced by the degree of aerosol neutralization. Highly acidic solutions suppress MSA dissociation, thereby shifting the equilibrium toward higher vapor pressures and enhancing gas-phase MSA. This coupling between aerosol composition and MSA volatility suggests that accurately representing aqueous-phase processes and acid–base equilibria is essential for reproducing observed MSA abundances (Miljevic et al., 2025).

4.6 Methanesulfonic acid induced nucleation

Gas-phase MSA in the atmosphere condenses onto existing aerosols and nucleates together with SA, ammonia and water vapour. We have implemented the SA(-MSA)-NH₃ nucleation and growth rates from Baalbaki et al. (2026). Figure 9 shows the fraction of the nucleation rate at 1.7 nm that is induced by MSA, defined as $(J_{1.7\text{-base}} - J_{1.7\text{-nNPF}})/J_{1.7\text{-base}}$. Here, $J_{1.7\text{-base}}$ is the nucleation rate of the baseline **Shen** simulation, and $J_{1.7\text{-nNPF}}$ that of the sensitivity simulation **Shen no NPF**, in which only MSA nucleation was switched off. MSA-enhanced nucleation dominates at high latitudes (poleward of 60°), reaching 90% above Antarctica. MSA is therefore a major driver of NPF above the Southern Ocean and the Antarctic.

A potential concern is that the overestimation of SA and MSA biases the absolute nucleation rates and hence the inferred CCN contributions (presented in the next section). Three considerations indicate that the relative MSA contribution reported here is robust. First, the contributions are reported as ratios $((J_{1.7\text{-base}} - J_{1.7\text{-nNPF}})/J_{1.7\text{-base}})$, and the analogous CCN ratio), in which a common bias in the precursor concentrations largely cancels. Second, the central result refers to the Southern Ocean, where the model reproduces both SA and MSA reasonably well. Third, SA is overestimated roughly twice as strongly as MSA (global Median (M/O) of 6.21 versus 3.37), so reducing both biases toward the observations would increase the contribution of MSA relative to SA. Our results are therefore conservative with respect to the conclusion that MSA-induced nucleation rivals that of SA.



400 Nevertheless, nucleation from iodine oxoacids (IOA), namely iodic acid (HIO_3) and iodous acid (HIO_2), as described by He et al. (2021); Sipilä et al. (2016); Thakur et al. (2022), were not included in the present study. Neglecting them may alter the relative dominance of different NPF channels in the Southern Ocean and other marine environments.

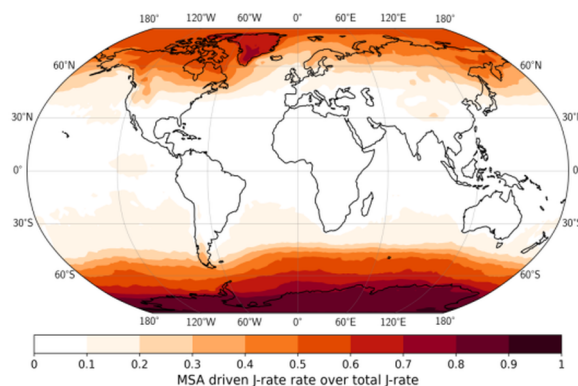


Figure 9. Relative contribution of MSA to the nucleation rate at 1.7 nm, $(J_{1.7\text{-base}} - J_{1.7\text{-nNPF}})/J_{1.7\text{-base}}$, integrated from the surface to 4 km altitude. $J_{1.7\text{-base}}$ is the baseline (**Shen**) nucleation rate, which includes MSA nucleation, and $J_{1.7\text{-nNPF}}$ that of the sensitivity run (**Shen no NPF**) with MSA nucleation switched off.

4.7 MSA nucleation and MeSH emissions contributing to CCN

The climatic importance of stable cluster formation is particularly pronounced in pristine regions with a slow condensation
405 sink, where newly formed clusters can evade scavenging and grow to CCN sizes (Carslaw et al., 2013). During the CAPRI2 campaign a correlation between SA and MSA and increased CCN Miljevic et al. (2025) was observed. Other studies found an increase in CCN over the Antarctic to nucleation events connected to high phytoplankton activity (Kim et al., 2019; Park et al., 2023). In Figure 10 a) we show that MSA-induced nucleation is responsible for up to around 40 % of the CCN concentration in the high-latitude Southern Hemisphere, peaking above the Antarctic and gradually decreasing towards the equator.

410 In Figure 10 a) we show the relative contribution of MSA nucleation to CCN at 0.4 % supersaturation, defined as $(\text{CCN}_{0.4\text{-base}} - \text{CCN}_{0.4\text{-nNPF}})/\text{CCN}_{0.4\text{-base}}$, averaged from the surface to 4 km altitude and over the full simulation period. Here, $\text{CCN}_{0.4\text{-base}}$ is the CCN concentration of the baseline **Shen** simulation and $\text{CCN}_{0.4\text{-nNPF}}$ that of the sensitivity simulation **Shen no NPF**, in which MSA nucleation was switched off. The relative increase is large, around 25 % over the Southern Ocean and up to 40 % over Antarctica. The shortwave cloud albedo (Twomey) response scales approximately with the logarithm of cloud droplet
415 number concentration N_d (Twomey, 1977), which in marine boundary layers is largely controlled by CCN abundance. Because background CCN (and N_d) are very low in these regions, a doubling of CCN produces a relatively large increase in negative radiative forcing from the first indirect cloud effect.

The presence of MeSH emissions and chemistry alters the SA and MSA concentrations, affecting the nucleation and growth of marine aerosol particles and changing CCN abundances. In Figure 10 b) we show the corresponding contribution of MeSH



emissions, defined as $(CCN_{0.4-base} - CCN_{0.4-noMeSH})/CCN_{0.4-base}$, where $CCN_{0.4-noMeSH}$ is the CCN concentration of the sensitivity simulation **Shen no MeSH**, in which MeSH emissions were switched off. In our simulation, MeSH emissions are responsible for 30-50 % of the $CCN_{0.4}$ in the Southern Ocean, indicating an effect on clouds that is comparable to that of DMS in this region. Similarly strong contributions from MeSH are observed in the equatorial Pacific and the Indian Ocean. In the Arctic, around 30 % of the $CCN_{0.4}$ originates from MeSH emissions.

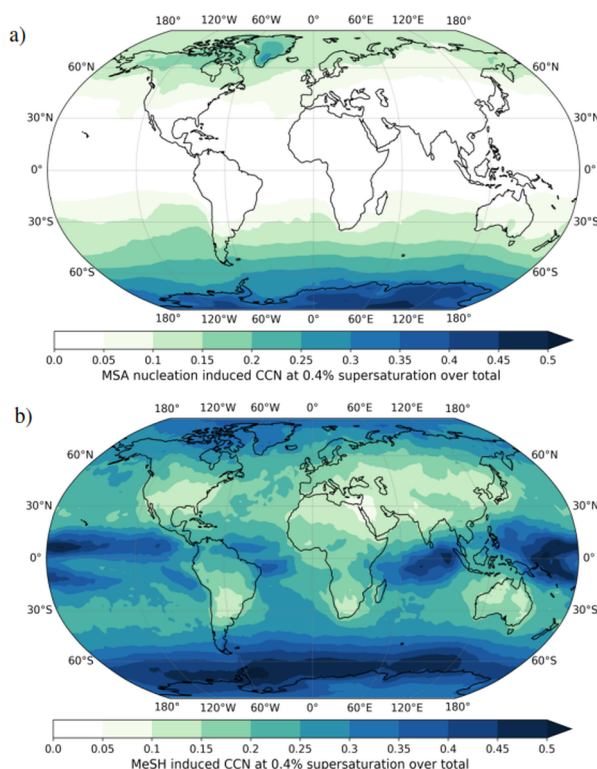


Figure 10. CCN at 0.4 % supersaturation, averaged from the surface to 4 km altitude and over the full simulation period. **(a)** Relative contribution of MSA nucleation to CCN, $(CCN_{0.4-base} - CCN_{0.4-nNPF})/CCN_{0.4-base}$. **(b)** Relative contribution of MeSH emissions to CCN, $(CCN_{0.4-base} - CCN_{0.4-noMeSH})/CCN_{0.4-base}$. $CCN_{0.4-base}$ is the baseline (**Shen**) value, which includes MSA nucleation and MeSH emissions, $CCN_{0.4-nNPF}$ that of the sensitivity run (**Shen no NPF**) with MSA nucleation switched off, and $CCN_{0.4-noMeSH}$ that of the sensitivity run (**Shen no MeSH**) with MeSH emissions switched off.

5 Conclusions

In this work we have simulated with the EMAC model four different chemical schemes representing VMS degradation in the atmosphere. Model results were compared with observational datasets covering different regions in the world. The four mechanisms reproduce surface DMS almost equally well, with median model-to-observation ratios of 0.90–1.01 and near-



identical correlation and spread (Table 6). This insensitivity confirms that the simulated DMS field is governed primarily by emissions and oxidant availability. The choice of mechanism instead becomes decisive for the higher-generation products: despite the convergence in DMS, the median SA bias ranges over more than an order of magnitude, from an underestimate in **MECCA** (Median (M/O)= 0.66) to a more than fifteen-fold overestimate in **Fung22** (Median (M/O)= 17.74, Table 8), while the MSA bias spans from a factor-of-three underestimate in **MCM** (Median (M/O)= 0.29) to a six-fold overestimate in **MECCA** (Median (M/O)= 6.19, Table 9).

No single mechanism reproduces all three species (i.e. DMS, MSA and SA) simultaneously in a satisfactory way. **MECCA** is the only scheme that does not overestimate SA, yet it lacks MeSH chemistry, yields the poorest DMS agreement and has the largest MSA overestimate. **MCM** is the only scheme to underestimate MSA while still overestimating SA, pointing to an imbalanced partitioning between the two acids. Notably, increasing mechanistic complexity does not translate into improved realism, as the intermediate-complexity **Fung22** scheme produces by far the largest SA overestimate.

Overall, the inter-comparison shows that VMS oxidation schemes remain subject to large uncertainties, but that these uncertainties do not arise from the gas-phase mechanism alone. The overestimation of both acids by nearly all schemes, concurrent with a well-captured DMS, indicates additional and largely mechanism-independent factors, namely the magnitude and wind-speed dependence of VMS emissions, the abundance of oxidants (in particular halogens), the strength of the aerosol condensation sink, and the treatment of heterogeneous and multiphase sulfur chemistry. Because these processes affect the modeled acids, no single dominant source of uncertainty can be isolated, and further progress will require observational constraints on emissions, oxidants, and the condensation sink to be improved alongside the refinement of the oxidation chemistry itself. The sensitivity experiments identify halogen and MeSH chemistry as the two dominant controls on the marine VMS budget, both acting largely independently of the choice of gas-phase scheme. Disabling the DMS-halogen reactions (**Shen no halog**) doubles the global DMS lifetime from 0.8 to 1.6 d and roughly doubles its burden, confirming that BrO is the leading DMS sink over the Southern Ocean and that the common neglect or underestimation of bromine in global models likely biases simulated DMS lifetimes high. MeSH exerts a strong indirect control through competition for shared oxidants: removing it (**Shen no MeSH**) shortens the DMS lifetime and, notably, nearly halves the SA overestimate (Median (M/O) from 6.21 to 2.55) while also reducing the MSA bias. By contrast, multiphase chemistry (**Shen no ACM**) influences gas-phase DMS mainly through bromine recycling on deliquescent aerosols, while leaving gas-phase MSA essentially unchanged, as its high effective Henry's law solubility retains MSA in the condensed phase.

Beyond its role as an acid, MSA is a major driver of new particle formation and growth at high latitudes (poleward of 60°), in the climate-sensitive Southern Ocean and Antarctic regions. Under the cold conditions found there, MSA-induced nucleation rivals that of SA and accounts for around 25% of CCN_{0.4} over the Southern Ocean and up to 40% over Antarctica. Climate models strongly underestimate CCN in this climatically important region, producing a large positive radiation bias (Fiddes et al., 2025). Although the absolute CCN enhancement is modest, the large relative increase against the low background CCN of these pristine regions translates into a large first indirect (Twomey) forcing. MeSH leaves a comparable impact on CCN_{0.4}: its omission reduces CCN_{0.4} by up to 50% over the Southern Ocean, the equatorial Pacific and the Indian Ocean, and by about 30% in the Arctic, an effect comparable to that of DMS in these regions. Because climate-driven ocean stratification is projected



to favour MeSH production over DMS, the contribution of MeSH to marine CCN may grow in the future, underscoring the need to better constrain its emissions and oxidation chemistry.

Several limitations must be taken into account. As discussed above, nearly all schemes overestimate both acids while reproducing DMS, and the sensitivity model integrations point to too-rapid (halogen-mediated) oxidation, too-high VMS emissions, and an underestimated condensation sink as the principal contributors. VMS emissions are most uncertain at the high sea-surface wind speeds characteristic of the North Atlantic storm track and the Southern Ocean, amplifying the bias in those regions.

A further limitation is the omission of iodine emissions and iodine-related chemistry. Neglecting iodine likely overestimates the oxidant pool otherwise consumed by iodine reactions and excludes iodine-driven NPF, thereby overstating the relative role of sulfur-driven nucleation in remote marine regions (Prados-Roman et al., 2015; Baccarini et al., 2020; Xavier et al., 2024; He et al., 2021).

This study represents a first step toward a comprehensive and observationally evaluated representation of oceanic sulfur chemistry within EMAC, focused here on gas-phase VMS species at the surface and their role in NPF and CCN. Owing to the still-large uncertainties in the treatment of MSA gas–aerosol partitioning, which controls the availability of gas-phase MSA for NPF, we restrict the present evaluation to the lower troposphere. Future work will aim for a more general representation of MSA partitioning that also captures its behaviour in the upper troposphere. Recent and upcoming airborne campaigns are beginning to provide the vertical-profile constraints needed to extend this evaluation throughout the troposphere (Klebach et al., 2026). The recent finding that MSA contributes to NPF in the upper troposphere, thereby altering the net top-of-atmosphere radiative forcing (Bhatti et al., 2023), illustrates why a well-constrained vertical distribution of sulfur species is essential for robust statements on the role of biogenic sulfur as a climate driver and for assessing its sensitivity to anthropogenic perturbations, such as changing oxidant levels and ocean-surface nutrient availability.

In summary, this study quantifies the influence of MeSH on VMS-derived acid abundances, explicitly represents MSA-NH₃-H₂O nucleation, and evaluates four VMS gas-phase mechanisms against observations, demonstrating that not only SA-driven but also MSA-induced new particle formation is important over the Antarctic and the Southern Ocean. Given that oceanic VMS emissions are the largest natural sulfur source to the atmosphere, and increasingly so under future low-anthropogenic-emission scenarios, constraining VMS oxidation together with the MSA and MeSH contributions to aerosol formation is essential for reducing the persistent uncertainty in aerosol radiative forcing and aerosol-cloud interactions over the remote ocean.

Code and data availability. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code are licensed to all affiliates of institutions that are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium website (<http://www.messy-interface.org>). The code presented here is available as git commit #f070b968 in the MESSy repository, and all changes will be included in the main repository.

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Appendix A: Comparison of sensitivity test model simulations to observations

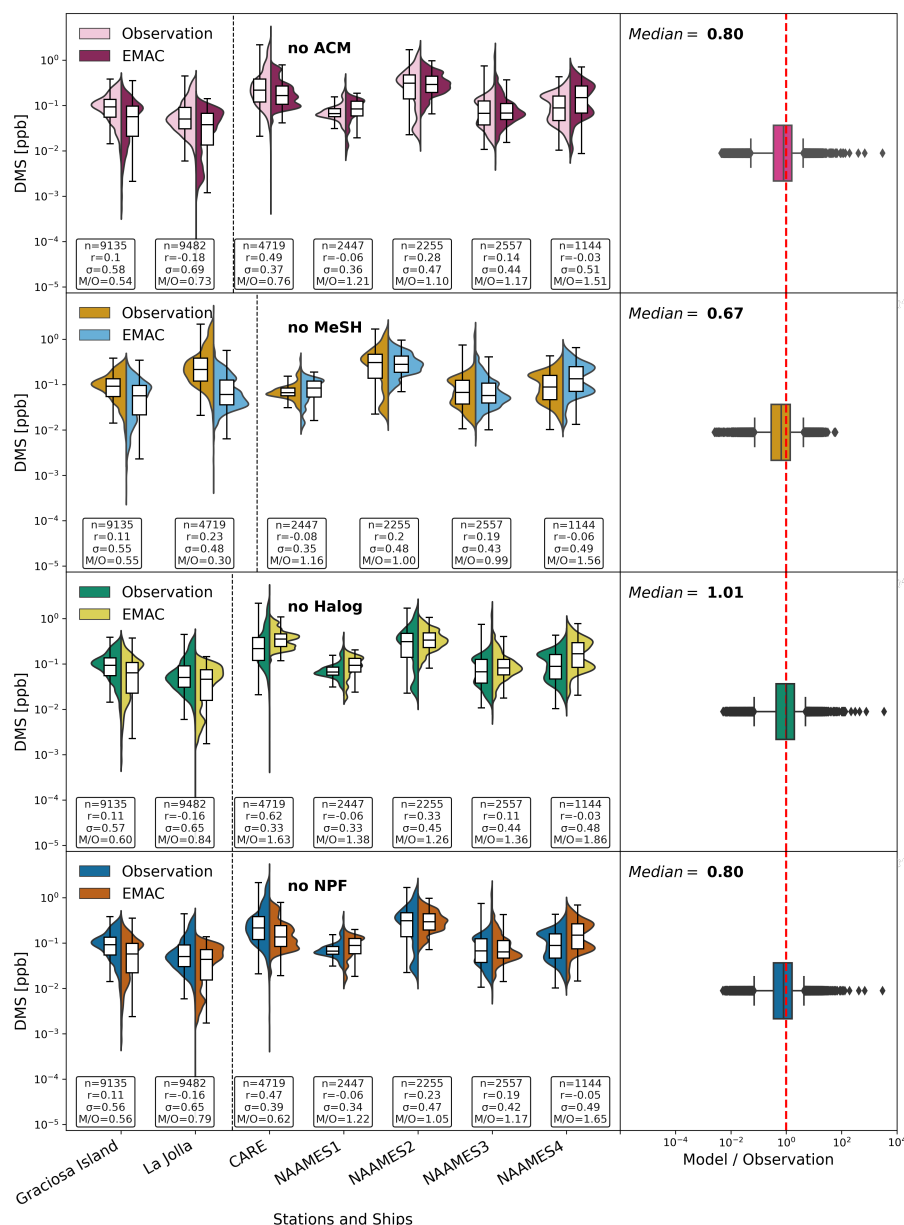


Figure A1. Comparison of modelled DMS mixing ratios with ambient observations for four sensitivity integrations (**Shen no ACM**, **Shen no MeSH**, **Shen no halog**, and **Shen no NPF**), evaluated against two ship campaigns (NAAMES and CARE) and two stationary sites (Graciosa Island and La Jolla). Left: split violin plots with overlaid box-and-whisker elements, where the whiskers span the full data range. Right: box-and-whisker plots of the model-to-observation ratio, where the whiskers indicate the 5th and 95th percentiles. Boxes give the interquartile range. Annotated metrics are the number of sampling points n , the Pearson correlation coefficient r , the standard deviation of the logarithmic model-to-observation difference $\sigma_{\log}$, and the median model-to-observation ratio pooled over all measurement points. This pooled median differs from the site-averaged M/O in Table 6.

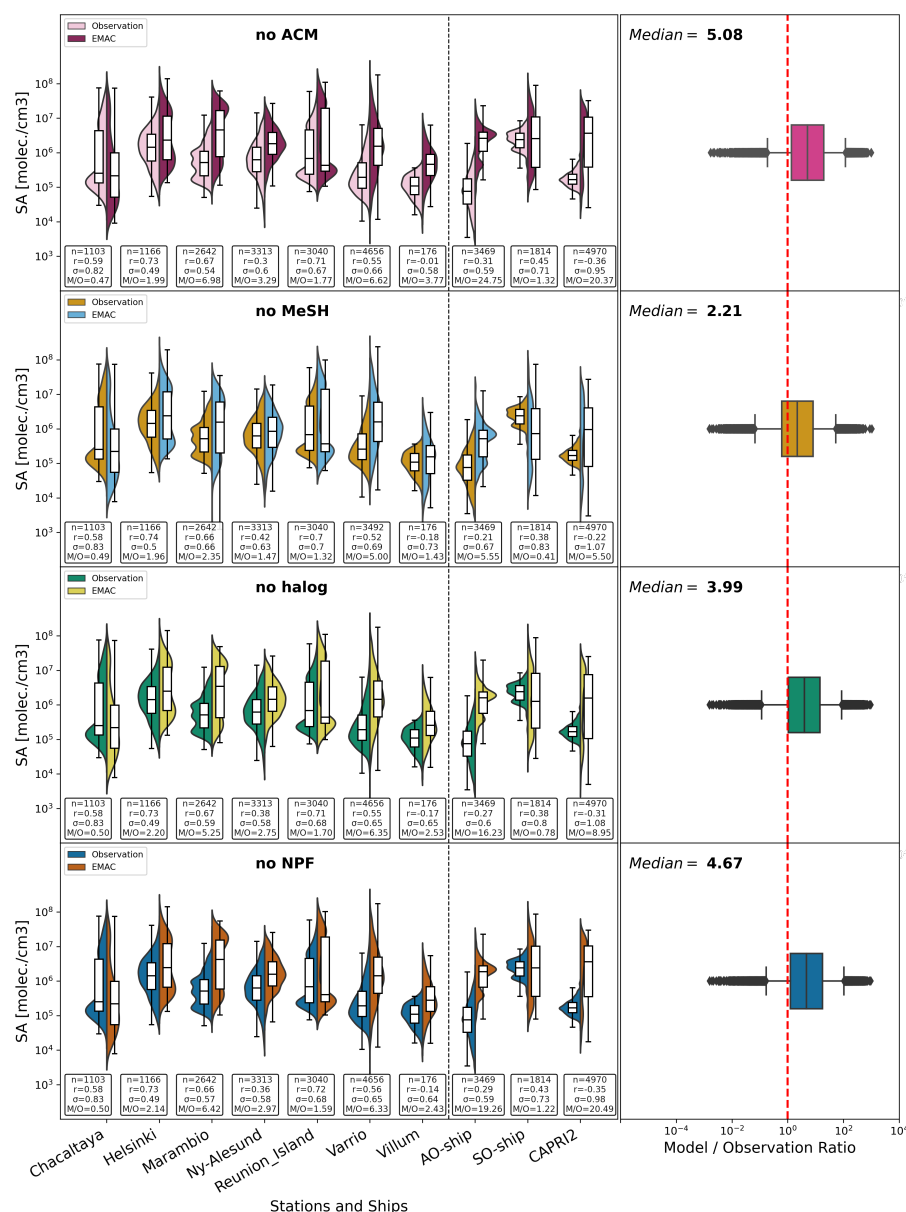


Figure A2. Comparison of modeled SA concentrations with ambient observations for four sensitivity model integrations (**Shen no ACM**, **Shen no MeSH**, **Shen no halog**, and **Shen no NPF**). In the left column SA concentrations are shown as split-violin-plots with Box-Whisker overlays. The Pearson correlation coefficient (r) and $\sigma_{\log}$ the standard deviation of the log difference between model and observation for each run are given. The right column shows the Box-Whisker of model over observation ratio for the four scenarios. Boxes represent the interquartile range (25th–75th percentiles), where the whiskers in the left column panels encompassing the full range of data points while the whiskers in the right column panels indicating the 5th and 95th percentile bounds. The value annotated in the right column is the median of the model-to-observation ratios over all individual measuring points, which is a different aggregation from the site-averaged Median (M/O) of Table 8 (the mean of the per-site median ratios) and need not coincide with it.

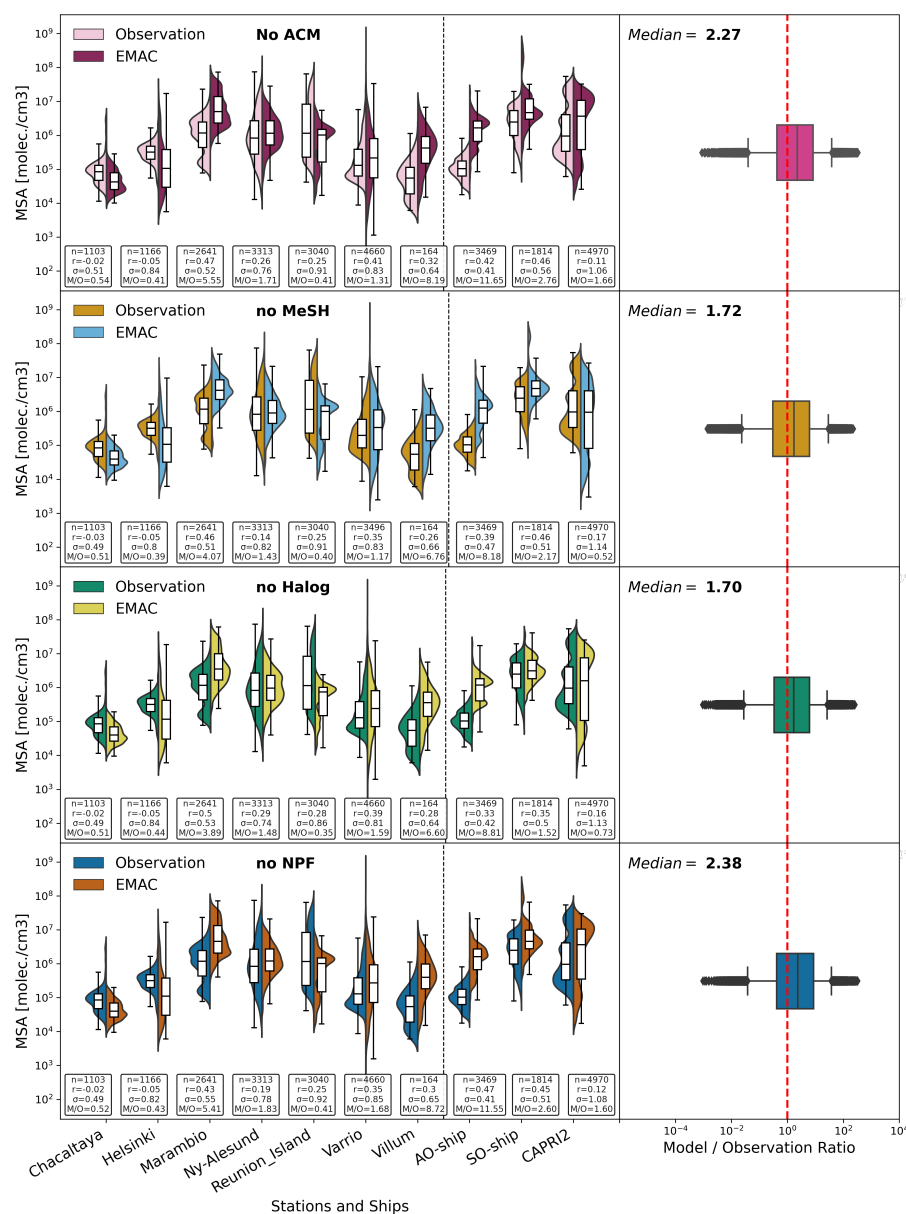


Figure A3. Comparison of modeled MSA concentrations with ambient observations for four sensitivity model integrations (**Shen no ACM**, **Shen no MeSH**, **Shen no halog**, and **Shen no NPF**). The left column shows MSA concentrations as split-violin-plots with Box-Whisker overlays. The Pearson correlation coefficient (r) and $\sigma_{\log}$ the standard deviation of the log difference between model and observation for each run are given. The right column shows the Box-Whisker of model over observation ratio for the four scenarios. Boxes represent the interquartile range (25th–75th percentiles), where the whiskers in the left column panels encompassing the full range of data points while the whiskers in the right column panels indicating the 5th and 95th percentile bounds. The value annotated in the right column is the median of the model-to-observation ratios over all individual measuring points, which is a different aggregation from the site-averaged Median (M/O) of Table 9 (the mean of the per-site median ratios) and need not coincide with it.



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