Answers to reviewer comments for "Methanesulfonic and sulfuric acids are major contributors to tropical Indo-Pacific aerosol"

Hannah Klebach et al.

We thank the reviewers for the constructive feedback and have tried to improve our manuscript and answer all open questions. The reviewer comments are printed in black, our replies in blue and changes to the manuscript are indicated in **bold**.

Reviewer 1

This paper measures methanesulfonic acid (MSA) and sulfuric acid (SA) in the tropical Indo-Pacific from 0-14 km in altitude using the HALO aircraft. MSA and SA were measured in both gas-phase and particle phase, however, due to the instrument limitations (evaporation of the species in the inlet), some of the data have been excluded, especially in the mid-troposphere. Additionally, zero-dimensional box modelling of the oxidation of DMS in the upper troposphere has been performed to compare with the observations.

Overall, I think this is a useful contribution to the field; due to the importance of MSA and SA for new particle formation (NPF) and contribution to cloud condensation nuclei (CCN), more fieldwork measurements, particularly in areas that have been underexplored, are important. The observations of MSA and SA in the upper troposphere that are likely due to deep convection were interesting, and the comparison of MSA and SA concentrations with time from convection (Figure 9) provides a nice insight into the possible chemistry and lifetime of these species in the upper troposphere.

The paper is well written and the figures are nice, however, the discussion on the formation of MSA and SA from DMS oxidation is lacking in depth, and the limitations of the model are understated. Further details on this are included in the following comments. In addition, some smaller technical comments are included.

Major comments:

1. Due to the complicated nature of MSA formation, especially in the gas phase, the discussion of the formation of MSA from the gas phase should be expanded. This includes how MSA formation occurs from the complicated equilibrium chemistry of CH_3SO_x species. Whether CH_3SO_x species form MSA, SA or SO_2 (which could eventually contribute to SA) depends on RO_2, H-donors, NO_x and temperature. This discussion, and how it impacts the results, is missing from this work. In the particle phase, MSA is primarily formed from aqueous reactions of MSIA, with MSIA formed due to the DMS addition pathway (OH or BrO initiated), and reaction of DMS and O_3 in cloud droplets. Additionally, there seems to be a direct sulfate formation pathway from the uptake of HPMTF in aerosol/cloud droplets Jernigan et al. [24]. Understanding these pathways is important for the interpretation of the results, and should be discussed in more detail.

We thank the reviewer for pointing out the lack of discussion on MSA formation in the manuscript. We have added the following paragraphs to the introduction of the paper:

Main oxidation products of DMS are SO_2, sulfuric acid (SA), methanesulfonic acid (MSA) and hydroperoxymethyl thioformate (HPMTF). The initial oxidation step occurs mostly by OH, via H-abstraction or OH-addition. Regionally NO_3 and halogen species, predominantly BrO, can contribute significantly to the oxidation [44, 27]. The initial DMS oxidation is followed by multiple steps, including reactions with O_3, NO_x, HO_2, RO_2 and photochemical reactions. The exact pathways and reaction rates are still an active area of research with multiple schemes differing in complexity proposed (e.g. Chen et al. [9], Shen et al. [43], Fung et al. [15], Tashmim et al. [44], Jacob et al. [22]).

One important group of intermediates are the CH_3SO_x radicals (CH_3S, CH_3SO, CH_3SOO,CH_3SO_2O). Their relative abundance and further reactions depend on temperature, NO_x, HO_x, O_3 and RO_2 concentrations [22]. The key competition is between pathways that keep the carbon-sulfur skeleton intact long enough to form MSA, and pathways that break it down toward SO_2 or SA (via SO_3).

Temperature plays an important role in the MSA/SA ratio from DMS oxidation. At cold temperatures the addition pathway is favoured over the abstraction pathway [7, 44]. Additionally, decreased thermal decomposition reduces fragmentation towards SO_3 and SO_2. Overall, this leads to an enhanced MSA production at cold temperatures resulting in higher yields at high latitudes [43, 7, 44].

In addition to the gas-phase mechanism, DMS is also oxidised in the aqueous phase. This pathway contributes substantially to total atmospheric MSA (predominantly present as methanesulfonate, MS^-, in cloud and aerosol water) and DMS-derived sulfate, especially in cloud-processed marine air [9, 20, 27]. HPMTF is efficiently scavenged by cloud droplets, which leads to enhanced sulfate formation and a decrease in gas-phase SO_2 [38].

2. Although some modelling of DMS has been included, I think there has been a lack of commentary on the DMS oxidation mechanisms in the literature, and the differences between them. The DMS mechanism from Shen et al. [43] has been used in this modelling work, however there is no discussion on how that mechanism compares/differs from other DMS mechanisms in the literature, such as those from Jernigan et al. [24], Ye et al. [50], Jacob et al. [22, 23]. Specifically, Jacob et al. [22] found that the mechanism from Shen et al. [43] tended to underestimate SO_2 concentrations when compared to other chamber studies and mechanisms. A mechanism comparison is particularly lacking in the discussion of model results from line 440 onwards, and the conclusion.

In light of the many different DMS oxidation schemes that have been proposed over the past decades, we agree that a deeper discussion of the Shen et al. mechanism is required. For the box modelling in the upper troposphere, we additionally performed the simulation with the chemical mechanism proposed in Jacob et al. [22] to show the variability between mechanisms. The results can be seen in the updated figure 10 of the main manuscript and Fig. 1a.

The methods section was adapted accordingly:

To adequately represent marine sulfur chemistry, the default DMS scheme was extended following the recent mechanistic work of Shen et al. [43] and Jacob et al. [22]. The Shen mechanism is based on MCMv3.3.1 as well as reactions from Hoffmann

et al. [20] and other recent publications. It was validated against CLOUD chamber measurements. Similarly, the Jacob mechanism is based on the MCM but added 73 reactions and changed 21. It performed best in comparison to several other chemical schemes [22]. Both schemes include HPMTF reactions but no aqueous-phase or halogen reactions. For a sensitivity test, halogen reactions for Br_2 and Cl_2 were added from Burkholder et al. [6].

In the gas phase section, we have extended the discussion about the discrepancy between measurements and model due to the uncertainties and limitations of the Shen et al. scheme:

Figure 3c compares the ratios measured during the CAFE-Pacific campaign with the kinetic model simulation by Shen et al. [43], which has been verified by chamber experiments. Both in the model and the measurements a clear shift towards MSA at colder temperatures can be confirmed. However, the measured SA/MSA ratios lie consistently under the model prediction with the strongest discrepancy in the BL. A reason for this could be uncertainties in the chemical mechanism. Shen et al. [43] is only one of multiple DMS oxidation schemes proposed. However, in a comparison it has been shown to underpredict SO_2 rather than MSA [22]. The effect of NO_x on the chemistry should be negligible due to the low mixing ratios above the ocean (mostly below 100 pptv, see Fig. A10). Even higher mixing ratios closer to the coast or shipping routes would only slightly enhance the SA/MSA ratio according to experimental results in Shen et al. [43], although the literature on the effect of NO_x on DMS oxidation is not conclusive [32, 33, 50].

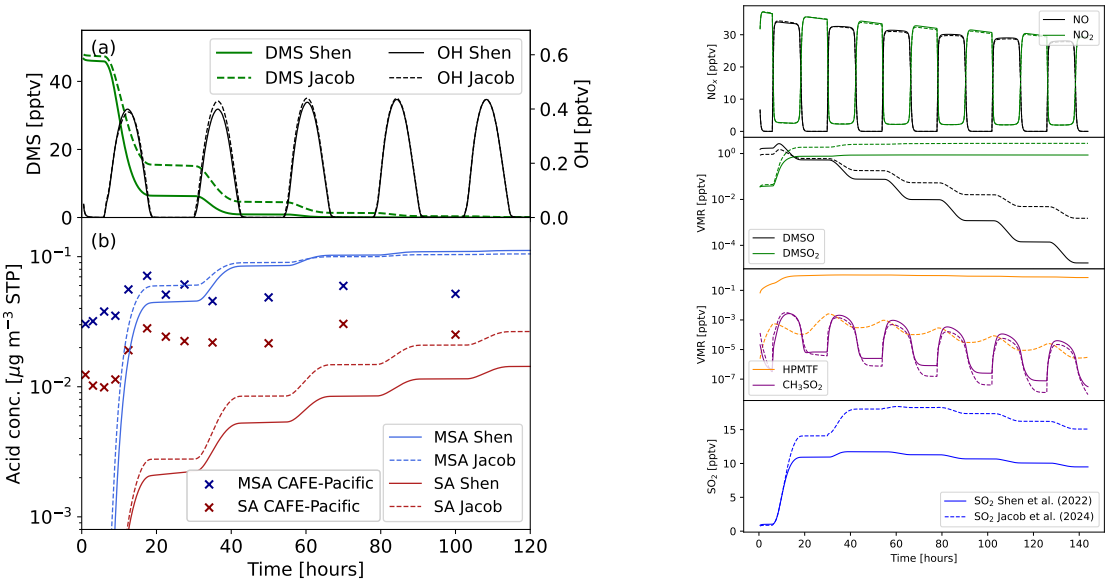
Processes that are not considered in the kinetic model but can impact the atmospheric SA/MSA ratio include, for example, the oxidation of DMS by halogen compounds. These reactions promote the addition pathway and, consequently, enhance MSA formation. However, the effect of Cl is expected to be minor and BrO has a significant impact mainly at higher latitudes [29, 9, 44]. Another missing mechanism are aqueous-phase reactions. For example, HPMTF is efficiently taken up into cloud droplets eventually reducing gas-phase SO_2 concentrations and subsequent SA formation [38, 30]. This is a likely reason for the low SA/MSA ratios in the BL measurements. Another important source could be evaporation of MSA from particles at low ambient RH values. While the RH in the BL is mostly high (∼80%, Fig. A9), the range of values in the free troposphere is broad. This might also explain the high MSA values measured for low marine fractions in Fig. 3b. The higher values cannot be caused by evaporation in the instrument since only data points with a very low ΔT and sufficiently high RH in the inlet are considered.

In the box modelling section we added a comparison between both schemes:

The DMS mixing ratio decreases more slowly in the Jacob mechanism than in the Shen mechanism. However, more MSA and SA are produced using the Jacob scheme, which indicates a more efficient conversion of DMS to acids. The DMS in the Shen mechanism is largely converted to HPMTF, which has mixing ratios 3–4 orders of magnitude higher than in the Jacob scheme (Fig. A20). This could be caused by the low OH reaction rate in the Shen scheme or the additional HPMTF photolysis included by Jacob et al. The higher SA formation in the Jacob scheme likely results from the enhanced production of SO_2 (Fig. A20), while the Shen scheme has been shown to underpredict SO_2 formation [22]. Overall, acid production is comparable between the two schemes, with the Jacob mechanism yielding 10–60% more SA than the Shen mechanism. MSA concentrations are initially up

to 20% higher using the Jacob scheme but later remain up to 20% lower than using the Shen scheme. [...]

The branching ratio between MSA and SA can also be influenced by the O_3 or NO_x concentrations. However, NO_x is expected to have a minor influence (Fig. A10, [43, 39]) and the approximate agreement in MSA concentrations rules this out as an explanation for the discrepancy unless the initial DMS concentration is significantly higher. Chemical processes that could increase the SA yield are a more efficient conversion of SO_2 to SA, of $DMSO_2$ to SO_2 or of HPMTF to SO_2. It should be considered that most DMS chemical schemes and experiments have been focused on marine BL conditions potentially missing pathways or accurate reaction rates for the cold conditions of the upper troposphere.



(a) Revised Fig. 10

(b) Revised Fig. A20

Figure 1: Box modelling of upper tropospheric DMS oxidation. (a) shows the revised Fig. 10 from the main manuscript with the addition of a second chemical mechanism based in Jacob et al. [22]. (b) shows the additional species for both mechanisms, it is show in Fig. A20 in the appendix of the original manuscript.

3. Line 432: 'Since we measure the combined gas and particle phase in the upper troposphere, we can directly compare our data to the model results.' I disagree with this. As mentioned previously in this paper, the major source of MSA in particle phase/cloud droplets is from aqueous phase reactions (Hoffmann et al, 2016). I understand that the observed results are a combination of both gas and aqueous phase, however, as this modelling does not include aqueous chemistry, it cannot be directly compared to the combined gas and particle phase. This should be made much clearer, and included in the discussion of the model results and conclusions.

This sentence was included in the manuscript simply to emphasise that no loss processes such as condensation or nucleation are included in the box model, hence the results are representative of the total production of SA and MSA in the gas phase. However, we agree that the statement requires refinement.

Aqueous-phase reactions can significantly alter the yield of MSA and SA as suggested by multiple publications [20, 9, 38]. In the proposed process of DMS transport by convective clouds, aqueous-phase processes can occur at two points: first, within the convective cloud during uplift. DMS

uptake can lead to the formation of MSA and sulfate in the cloud droplets. As the cloud evaporates, they may be retained in the particle phase. This is likely the origin of the MSA and SA concentrations measured in RF14 (Figure 6 original manuscript). However, generally cloud outflows show low concentrations of aerosols [49], and also in RF14, we observed very low acid and particle number concentrations. Aqueous-phase reactions are likely the reason why the acid concentrations in Figure 9 (original manuscript) have enhanced values even very shortly after the convection. However, the comparison of RF14 and RF18 and the increase in concentrations in Figure 9 10-20 hours after convection shows that the aqueous-phase reactions only account for a fraction of the observed acids.

The second point where aqueous-phase reactions could occur is after the DMS is injected into the upper troposphere. If the air mass encounters clouds within the first approximately 48 hours after convection, the gas-phase DMS could be taken up and undergo aqueous phase reactions. If the cloud encounter occurs after that time the ratio of SA and MSA can still be impacted for example by the uptake of DMSO, $DMSO_2$ or HPMTF.

To estimate this effect, we traced back the trajectories to the last encounter of very low, low, medium or high clouds using the Himawari satellite cloud type. Cirrus clouds were excluded from this analysis due to their small vertical extent and limited literature on multi-phase reactions of DMS and DMS-products with ice clouds. We find that only a minor fraction of trajectories had a second cloud encounter (less than 30% at each step and mostly <10% for those with convective uplift in the last 24 h, see Fig. 2b). Even if the fraction of cloud encounters was high (e.g. MSA 6 and 9 hours after marine convection, Fig. 2a) the impact on the mean SA and MSA concentrations is minimal. A reason for this could be that a large fraction of these clouds will be ice or at least mixed-phase clouds, and aqueous-phase processes are expected to occur at a much lower rate there, if at all.

Overall, we expect the influence of aqueous-phase processes in the free troposphere to be much weaker than under boundary layer conditions.

The original sentence was changed to:

The model output corresponds to the total production of SA and MSA by gas-phase oxidation of DMS. It can be directly compared to the measured data if aqueous-phase processes are disregarded.

Additionally, we added a paragraph discussing aqueous-phase reactions in more detail:

A shortcoming of the box model setup is the lack of aqueous-phase reactions, which have been shown to play a major role in DMS processes [20, 9]. In the BL, cloud processing is a crucial process promoting MSA formation and reducing SO_2 concentrations through the uptake of HPMTF [20, 9, 24, 38]. It can occur in the convective cloud during the uplift and the resulting MSA and SA may be injected in the particle phase into the upper troposphere. This could be the reason for the enhanced values shortly after contact to a convective cloud. However, the case study of RF14 and previous aircraft campaigns indicate low particle concentrations in cloud outflows [1, 10]. More detailed modelling would be required to quantify this pathway. Cloud processing can additionally happen after the injection of gas-phase DMS into the upper troposphere if another cloud is encountered. Using backtrajectories and Himawari satellite data we estimate that less than 30% and in many cases less than 10% of data points had a second cloud encounter after convective uplift (Fig. A23). Excluding these data from the analysis changes the results only marginally. Furthermore, the majority of the encountered clouds are likely ice clouds, where aqueous-

phase reactions are expected to be considerably less important than in liquid-phase cloud droplets. We have hence no evidence that aqueous-phase reactions strongly impact our results.

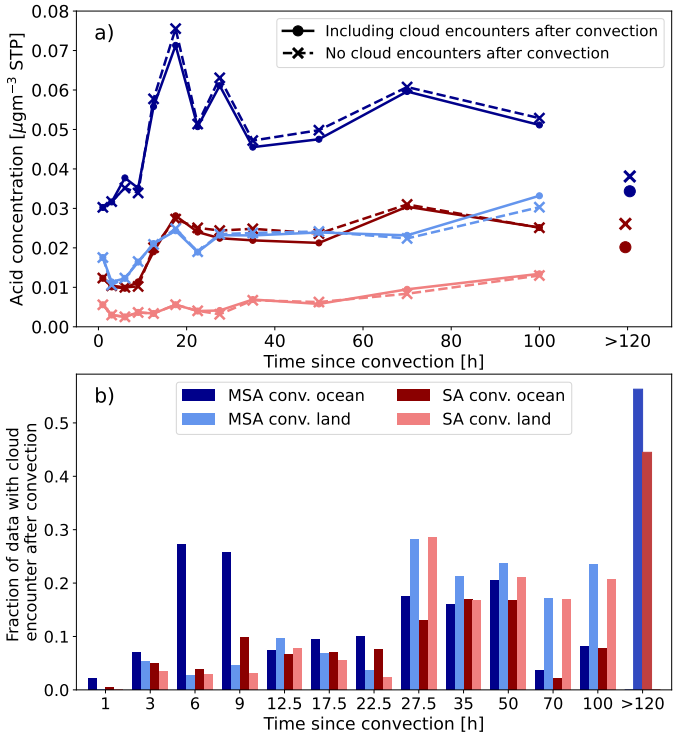


Figure 2: Analysis of cloud encounters after the initial convective uplift. (a) shows the data in Fig. 9 and (solid lines, round markers) and additionally the data excluding second cloud encounters after the convection (dashed lines, cross markers). (b) shows the fraction of data points with a second cloud encounter at each time bin. Note that the x-axes are not aligned.

Minor comments:

1. Since DMS concentrations were obtained from CAMS reanalysis, the plotting of NO_x and SO_2 would also be useful to investigate other sources of MSA and SA. In line 444 the influence of NO_x (and O_3) has been mentioned, however how that affects the results is lacking. This would be particularly helpful in the boundary layer runs with trajectories coming from land (and the comparison of Shen et al. 2022 simulations in lines 232-240).

The plot of NO_x and SO_2 mixing ratios (Fig. 3) was added to the appendix.

The NO_x mixing ratios in the marine BL of the Indo-Pacific are rather low away from main shipping routes (below 100 pptv), and in the warm pool region often below 10 pptv. In the upper troposphere significant NO_x concentrations are only expected in vicinity of convection over northern Australia and Papua New Guinea caused by lightning, whereas values above the ocean stay mostly around 50–100 pptv. Direct measurements during CAFE-Pacific report NO values between 17–28 pptv and often below the limit of detection above the warm pool [39], indicating CAMS might be slightly overestimating the values.

NO_x can impact the DMS oxidation at different points. Through the formation of NO_3 predominantly at night, the initial oxidation can be shifted towards the abstraction pathway, which favours SO_2 formation [28, 8]. On the other hand, NO_x can also impact the CH_3SO_x radical

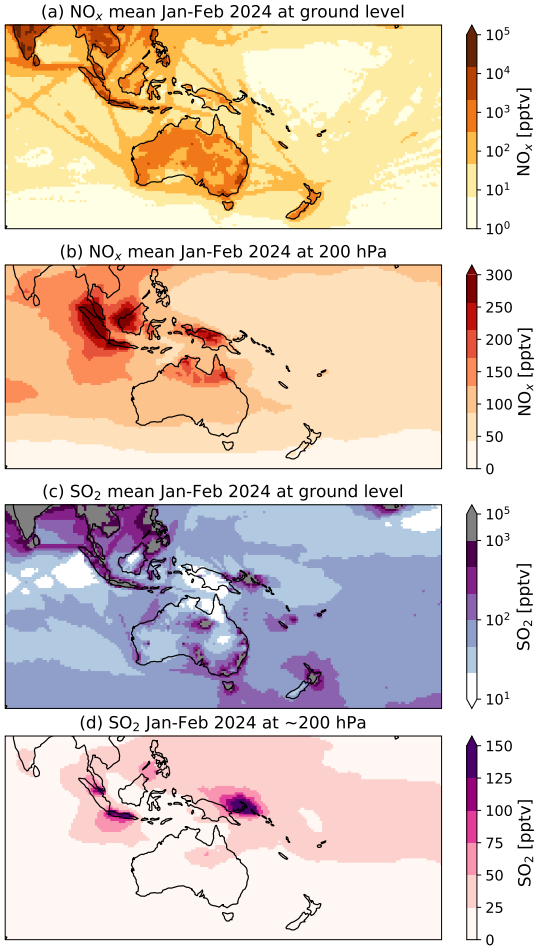


Figure 3: CAMS data for January and February 2024. The panels show the NO_x mixing ratios at ground level (a) and 200 hPa (b) (sum of NO and NO_2), the SO_2 mixing ratios at ground level (c) and approximately 200 hPa (d). The figure was generated using data by the Copernicus Atmosphere Monitoring Service (2020): CAMS global reanalysis (EAC4). Copernicus Atmosphere Monitoring Service (CAMS) Atmosphere Data Store, DOI: 10.24381/d58bbf47 (Accessed on 26-05-2026) [21].

chemistry and its products SO_2, MSA and SA. The effect of NO_x on the MSA/SA ratio is not consistent between publications. Koga and Tanaka [32] and Ye et al. [50] report higher MSA yields under high NO_x conditions, whereas Librando et al. [33] observed the opposite (at ppb to ppm levels of NO_x). Shen et al. [43] report a small increase in SA/MSA ratio at higher NO_x but the effect is much weaker than the impact of temperature. Additionally, Jacob et al. [22], Ye et al. [50] state that under low-NO_x conditions the HPMTF pathway is more important which could lead to a higher but delayed SO_2 formation. Hence it is difficult to estimate the effect of NO_x on the DMS oxidation in the Indo-Pacific. The NO_x values outside of direct sources are likely too low for significant DMS oxidation by NO_3 [7, 44]. The low-NO_x and low-O_3 conditions in the Warm Pool could reduce the formation of CH_3SO_3 from CH_3SO_2 and instead favour the formation of CH_3SO_2OO which continues to react to MSA. On the other hand, strong production of HPMTF and its cloud processing in the BL could reduce the concentration of gas-phase SO_2 and SA [38]. However, determining the exact mechanism under the conditions would require further studies. Especially the conditions of the free troposphere are not well covered by current literature and typically include the extrapolation of reaction coefficients to low temperatures.

The following sentences were added to the manuscript:

The effect of NO_x on the chemistry should be negligible due to the low mixing ratios above the ocean (mostly below 100 pptv, see Fig. A10). Even higher mixing ratios closer to the coast or shipping routes would only slightly enhance the SA/MSA ratio according to experimental results in Shen et al. [43], although the literature on the effect of NO_x on DMS oxidation is not conclusive [32, 33, 50].

and in the box model section:

However, NO_x is expected to have a minor influence (Fig. A10, [43, 39]) and the approximate agreement in MSA concentrations rules this out as an explanation for the discrepancy unless the initial DMS concentration is significantly higher.

SO_2 values in the region show hotspots in densely populated areas, but especially around volcanoes, where mixing ratios of several hundred ppbv can be reached. However, in the marine BL, the values stay mostly below 100 pptv. In the upper troposphere, hotspots above PNG and Indonesia are caused by volcanic emissions. Above the ocean the SO_2 mixing ratios remain below 50 pptv, likely mostly originating from DMS oxidation.

We have added to the manuscript:

CAMS shows hotspot regions for volcanic sites in Papua New Guinea and in the denser populated south east of Australia. However, none of the trajectories in Fig. 2 overlap with these regions indicating minor influence of non-DMS-derived SO_2.

Technical comments:

Abstract: Space needed in dimethyl sulfide

Line 29: Space needed in dimethyl sulfide

The spelling was corrected in both cases.

Line 32: Slightly misleading, DMS can be oxidised to many other products (including SO_2). Should have more depth in the atmospheric chemistry of DMS here, and cite more papers

The sentence was changed to:

Main oxidation products of DMS are SO_2, SA, MSA and hydroperoxymethyl thioformate (HPMTF).

Additionally, a more detailed discussion of DMS oxidation was added as described above.

Line 31: Missing a reference for 'The MSA formation from DMS is strongly temperature-dependent, with higher formation rates at cold temperatures'. If it is the Shen 2022 reference, this should be made clearer

The paragraph was rewritten and more references were added:

Temperature plays and important role in the MSA/SA ratio from DMS oxidation. At cold temperatures the addition pathway is favoured over the abstraction pathway [7, 44]. Additionally, decreased thermal decomposition reduces fragmentation towards SO_3 and SO_2. Overall, this leads to an enhanced MSA production at cold temperatures resulting in higher yields at high latitudes [43, 7, 44].

Line 34: Again, slightly misleading, as from these papers it is the dominant source of MSA in

cloud droplets/aerosol (which will be in a different form, MS^-)

The sentence was changed to:

This pathway contributes substantially to total atmospheric MSA (predominantly present as methanesulfonate, MS^-, in cloud and aerosol water) and DMS-derived sulfate, especially in cloud-processed marine air [9, 20, 27].

Figure 3, line 3: I don't think 'Marine' should be capitalised here

The spelling was changed.

Line 209: Instead of saying 'these rather high values', which is quite subjective, it would be better to compare where they lie within the literature/other observations.

The sentence was replaced by:

These values are comparable but slightly higher than measurements in the Arctic [5], Antarctica and the Southern Ocean [26, 3, 40], while similar values have been reported in the tropical Indian Ocean [42].

Line 224: This is a confusing sentence, you should expand on it to make it more understandable

We changed the sentence to:

Air of purely marine origin shows slightly higher concentrations up to 2 km. Above this altitude, the data points are too scarce to determine a reliable trend.

Line 231: I am not sure what a 'good' agreement between the model and observations are, can you quantify this? Additionally, although there is a similar trend with temperature, the modelled values mostly lie outside the 75th percentile, which I would not consider 'good'

The sentence was replaced by:

Both in the model and the measurements, a clear shift towards MSA at colder temperatures can be confirmed. However, the measured SA/MSA ratios consistently fall below the model prediction, with the largest discrepancy in the BL.

It is followed by a more detailed discussion of the potential reasons for the deviation, which was already shown in response to the second comment.

Line 375: This is misleading, as the oxidation of DMS is a chemical source of SO_2. If you are specifically referring to volcanic/anthropogenic SO_2 sources, this should be clearer

The statement was limited to DMS, since the direct transport of SO_2 is not expected to be a major pathway due to the high losses within clouds [34].

There are no chemical sources for DMS in the upper troposphere.

Line 450: Again, not sure that you can say that the observations have 'good' agreement with the model, especially considering the limitations in comparing gas-phase chemistry to observed gas-phase and particle phase concentrations

We adjusted the statement and refrain from calling the overall agreement 'good'.

The simple box modelling here is limited in its representation of atmospheric processes, specifically due to the lack of aqueous-phase reactions and dynamic processes. Nevertheless, we achieve good agreement regarding the time scale of DMS oxidation and produced MSA concentrations. Discrepancies in the SA concentrations should be addressed in further studies using more advanced modelling and potentially chemical reaction coefficients optimised for upper tropospheric conditions.

Reviewer 2

This paper presents MSA and H_2SO_4 concentrations measured from the HALO aircraft over the Indo-Pacific. The dataset is highly valuable, the figures are beautifully presented, and the discussion is for the most part thorough and well judged. My comments below are offered in the spirit of strengthening an already strong manuscript, and once addressed I highly recommend this for publication in ACP.

Major comments

Title. My main comment is largely stylistic. The title makes a firm assertion: "Methanesulfonic and sulfuric acids are major contributors to tropical Indo-Pacific aerosol", yet throughout the manuscript the authors appropriately hedge their conclusions with "potentially", "suggesting", and similar, and discuss the relevant uncertainties carefully. The "are" in the title therefore reads as a slight overstatement, particularly when set against the abstract's own phrasing: "...suggesting that free-tropospheric particles particularly over the Indo-Pacific Warm Pool may be dominated by MSA." This does not undermine the paper in any way, but the title conveys a degree of certainty that the text itself is careful not to claim. I would encourage softening it to match the measured tone of the body.

We agree with the reviewer and have changed the title of the manuscript to:

Aircraft observations suggest an important contribution of methanesulfonic and sulfuric acids to tropical Indo-Pacific aerosol

This concurs with the statements made in the manuscript and emphasises the method of aircraft measurements.

Evaporation. My second comment concerns the substantial discussion given to evaporation of the particle phase into the gas phase, which for SA and MSA can cause gas-phase concentrations to increase by three orders of magnitude (and approaches four in Fig. 1/A2). This is framed largely as a positive in the manuscript, and to a degree it is, as it is what allows the authors to comment on aerosol composition below the lower cut of the C-AMS at all. However, the group's recent isoprene-nitrate work using the same instrument feels like something of an elephant in the room. I appreciate that this paper is not about isoprene nitrates, and that Curtius et al. 2024 (https://doi.org/10.1038/s41586-024-08192-4) concerns a different campaign, but it would be valuable to discuss whether comparable evaporative behaviour would be expected for isoprene nitrates. Curtius et al. in fact present data at the same altitudes at which the authors here expect their MSA and SA particles to evaporate entirely, so the question of cross-instrument artefact seems difficult to avoid. I recognise the volatilities of acids and organic nitrates may well differ, and that the evaporation here is tied to the particles' acidic state (Fig. A3) so the answer may be that nitrates behave differently; but that is for the authors to demonstrate rather than leave unaddressed.

The reviewer states correctly that the same instrument and conditions are present in the measurements of IP-OOMs reported in Curtius et al. [10]. This paper states in the methods section clearly that:

'Although the inlet residence time at these conditions is rather short (0.38 s), this temperature increase could lead to the evaporation of molecules from the aerosol to the gas phase, which could enhance our measured gas-phase signals. At -58 °C, the main IP0,1,2N products are ELVOCs, whereas they shift to the low-volatility organic compounds range at -13 °C (Fig. 3). Especially for

freshly nucleated particles with diameters in the size range in which the Kelvin effect has a role, this could cause evaporation. Therefore, the measurements are considered to be a combination of pure gas-phase concentration and potentially re-evaporated aerosol-phase molecules. However, even if a large fraction of our signal would originate from evaporation of freshly nucleated particles, this still highlights the crucial role of isoprene-derived oxidation products for the NPF process in the upper troposphere over the Amazon.' Curtius et al. [10]

The evaporation was already addressed in the paper and shown not to affect the conclusions drawn regarding the importance of IP-OOMs, specifically isoprene nitrates. After the analysis conducted in the present study, we can say that evaporation is definitely occurring and the elevated MSA concentrations measured outside the convective outflow in Curtius et al. [10] originate most likely from the particle phase. While we see indications for the evaporation of organics in the CLOUD chamber comparison, quantifying the effect for these remains difficult due to the limited data, numerous species and their individual volatilities.

We have added a short statement in the conclusion:

This effect was already implied in the publication about isoprene oxidation products in the Amazon's upper troposphere [10]. While we cannot quantify the impact on all the organic species detected, the enhanced MSA concentrations reported in that publication can be attributed to particle evaporation. However, this does not alter the conclusions drawn in the study concerning the role of isoprene nitrates in particle formation.

Minor comments

Particle neutrality. The assumption that the sampled particles are acidic is fundamental to the paper, and is probably correct, but a few sentences setting out explicitly why the authors believe this would strengthen the argument. A demonstrated absence of NH4+ in the C-TOF-AMS data, for instance, would help support it directly.

NH_3 is expected to be low in the upper troposphere with the exception of the Asian Monsoon region [19] and a previous study has found a high fraction of acidic particles in the tropical Pacific [14]. In fact, recent global modelling indicates that the Pacific Warm Pool is the region on Earth with the lowest influence of anthropogenic ammonia on upper tropospheric CCN concentrations [48].

The AMS measurements during the campaign indicate that ammonium was below the limit of detection in more than 90% of observations. Among the subset of data with reliable sulfate measurements, acidic particles were more prevalent than neutralised particles. However, the generally low aerosol mass concentrations in the free troposphere prevent a robust determination of particle acidity.

We have added a brief overview of the AMS measurements in the appendix and extended the text:

The neutralisation state of particles below 40 nm could not be measured during CAFE-Pacific. For particles above this diameter the composition measurements remain highly uncertain due to the very low mass concentrations, especially of ammonium (Fig. A5 and Fig. A6). Therefore, the values shown in the subsequent analysis represent lower limits for the total MSA and SA concentrations. However, outside the Asian monsoon region, very low ammonia concentrations are expected in the upper troposphere [14, 19, 37, 25] and large parts of the measurement region

show the lowest impact of anthropogenic NH_3 on CCN concentrations globally [48].

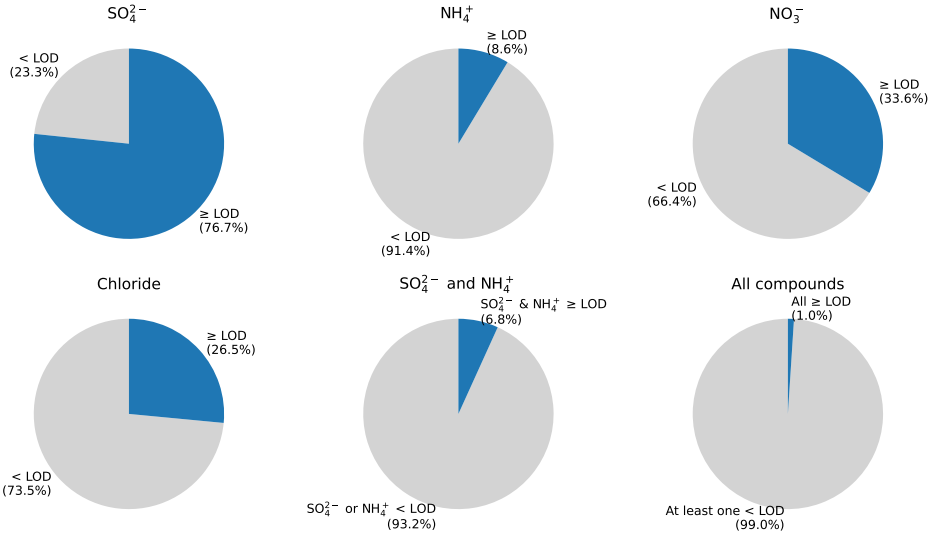


Figure 4: Fraction of C-TOF-AMS measurements during CAFE-Pacific above and below the limit of detection. Only data points in the middle and upper troposphere are considered, where SCORPION is expected to measure particle phase as well.

Gas-phase model. Two points here. Firstly, I broadly agree with Reviewer 1, though I would regard an exhaustive revision of the underlying mechanism as somewhat out of scope here. Reviewer 1 is nonetheless right that the gas-phase-only model is not strictly comparable to measurements that capture both gas and particle phases, especially given the strong linear fit in Fig. 4, which itself demonstrates how much of the signal is evaporated particle.

The box modelling and chemical mechanism here is limited to the gas-phase oxidation of DMS, which is transported to the upper troposphere by deep convection. It was set up under the assumption that the particle concentration in convective outflows is low (as shown in RF14). The MSA and SA produced in aqueous-phase reactions in the convective cloud are hence assumed to be a minor contribution.

The model does not include nucleation or condensation processes of the produced MSA and SA. However, the SA concentrations estimated by the model are sufficiently high to initiate nucleation at the cold temperatures of the upper troposphere [12]. MSA readily condenses on the newly formed particles [2]. Therefore, in the real atmosphere, MSA and SA will likely be in the particle phase. Since no transformation of MSA and SA is expected in the particle phase (unless cloud processing occurs), the acids produced in the model are still representative of the total acids measured in SCORPION, if direct injection of SA and MSA formed in the convective cloud and subsequent cloud processing is disregarded. We confirm that the latter is of minor importance by tracing back trajectories to cloud encounters after convective uplift (Fig. 2).

The initial sentence was adjusted to make this point clearer. Additionally, the discussion on the shortcomings of the box model was extended (already in response to comments by Reviewer 1).

The model output corresponds to the total production of SA and MSA by gas-phase oxidation of DMS. It can be directly compared to the measured data if aqueous-phase processes are disregarded.

A shortcoming of the box model setup is the lack of aqueous-phase reactions, which have been shown to play a major role in DMS processes [20, 9]. In the BL, cloud

processing is a crucial process promoting MSA formation and reducing SO_2 concentrations through the uptake of HPMTF [20, 9, 24, 38]. It can occur in the convective cloud during the uplift and the resulting MSA and SA may be injected in the particle phase into the upper troposphere. This could be the reason for the enhanced values shortly after contact to a convective cloud. However, the case study of RF14 and previous aircraft campaigns indicate low particle concentrations in cloud outflows [1, 10]. More detailed modelling would be required to quantify this pathway. Cloud processing can additionally happen after the injection of gas-phase DMS into the upper troposphere if another cloud is encountered. Using backtrajectories and Himawari satellite data we estimate that less than 30% and in many cases less than 10% of data points had a second cloud encounter after convective uplift (Fig. A23). Excluding these data from the analysis changes the results only marginally. Furthermore, the majority of the encountered clouds are likely ice clouds, where aqueous-phase reactions are expected to be considerably less important than in liquid-phase cloud droplets. We have hence no evidence that aqueous-phase reactions strongly impact our results.

Secondly, one point I would like the authors to clarify: they note that "incorporating dilution losses into the model would substantially reduce the predicted acid concentrations [and] would imply a missing source." Does this not suggest that the model only reproduces the observations because losses were omitted, i.e. that making the model more physically realistic would degrade, rather than improve, the agreement? If so, the agreement cannot also serve as validation of the measurements, and I would ask the authors to reconcile these two uses of the comparison.

Dilution is an important process when observing several-day-old convective outflows. We have added a simple calculation to estimate the decrease in acid concentrations by dilution. The dilution time scale in convective outflows was estimated at <2 days by Wang et al. [47], while Hernández Pardo et al. [16] estimates 3.7 days for combined dilution and wet scavenging of aerosols. Correspondingly, we use dilution time scales of 2 and 3 days. These are only applied to the MSA and SA concentrations produced by the model, not to DMS or intermediate compounds and only the results of the Jacob scheme were used. The results can be seen in Fig. 5. Instead of increasing continuously, the MSA concentrations decrease after 1–2 days. The SA concentrations still show an increase during the first two days, then the values stay flat, counteracting the delayed production of SA from SO_2.

The incorporation of dilution does not lead to a larger deviation from our measurements. The decrease in MSA is, in fact, similar to what is observed during CAFE-Pacific and also the absence of the continuous increase in SA in our measurements compared to the box model could be a result of dilution. However, the modelled SA concentrations remain well below the measured concentrations. For longer time scales (>3 days), the measured concentrations of both acids are significantly above the modelled, one reason could be the mixing in of air masses which contain MSA and SA instead of clean air as assumed by the dilution calculation. Especially in the marine FT, this is not unlikely due to the high frequency of convective transport and ubiquitous presence of MSA and SA.

We added the figure to the appendix and the following discussion in the main text:

Another important drawback is the lack of dilution processes. After release into the upper troposphere, the air parcel undergoes dilution through mixing with the surrounding air. The decrease in MSA and SA by dilution can be calculated assuming an e-folding timescale of 2 or 3 days [47, 16], as shown in Fig. A22 in the appendix. The resulting decrease does not negatively affect the agreement with

MSA measurements. For SA, the dilution counteracts the slow increase, improving the comparison in the temporal trend with our measurements, while the absolute concentrations remain significantly lower in the model.

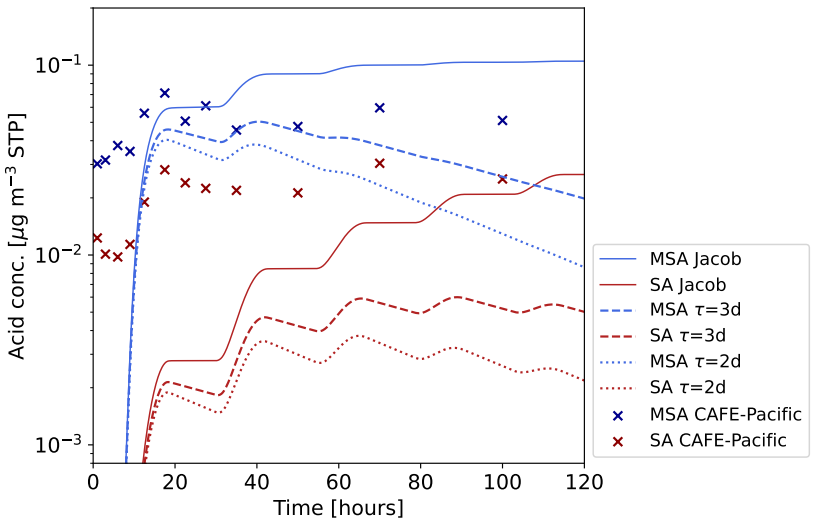


Figure 5: Box modelling results of upper tropospheric DMS oxidation including dilution of MSA and SA with two different time scales of 2 and 3 days (dotted and dashed lines). Only the Jacob et al. chemical scheme results from Fig. 10 were used and the dilution was only applied to MSA and SA concentrations not to intermediate products or DMS.

K-means clustering (Fig. 8). It would be helpful to describe how the K-means clustering was applied. The two standard approaches are to cluster on the Euclidean distance between trajectories or on differences in trajectory angle (as offered, for example, by the two options in trajCluster in R), and the choice materially affects cluster membership. A note on the distance metric used, the trajectory features clustered on, and the basis for selecting nine clusters would aid reproducibility.

To prepare the clustering, a subset of trajectories was selected that satisfied the conditions for MSA and SA evaporation and for which measurements were conducted above 8 km altitude. The longitudes are unwrapped to avoid a jump in sign when crossing 180° longitude. Subsequently, each trajectory is represented as one long vector containing latitude and longitude. Trajectories are not normalised to a common origin point to preserve different air mass origins in different flight regions.

Standard Euclidean mean clustering is used, which compares each trajectory to the mean trajectory of a cluster. It is sensitive to spatial, temporal and transport-path similarity because every hourly coordinate contributes to the Euclidean distance. The Euclidean distance clustering was selected instead of the trajectory angle because the focus was on the identification of different source regions rather than the similarity of motion directions.

The number of clusters was determined empirically to achieve the best representation of distinct but representative airmass origins.

The following sentences were added to the manuscript:

Euclidean distance clustering is applied to account for spatial, temporal and transport-path similarity. The number of clusters was determined empirically to achieve an optimal representation of distinct but representative airmass origins.

Convection. On p.20 the authors use five-day back-trajectories combined with satellite cloud observations to identify convective influence. This could usefully be fleshed out for us chemists with limited meteorological background. We would benefit from a clearer account of how convective contact is actually established from these two data sources.

The last contact to convection is established using a combination of HYSPLIT trajectories and Himawari satellite data. For each hourly position of the air parcels along the 5-day trajectories, the location was compared with the corresponding satellite-derived cloud-type classification. 15 different cloud/ground types can be identified, with type 9 (very high opaque) corresponding to the core of a deep convective cloud. If the mode cloud type in a 15 km radius around the location of the trajectory is 9, an encounter with a convective cloud is assumed. Finally, only if the altitude of the trajectory during the encounter lies between 8 and 15 km, convective outflow is inferred. Lastly, the location of the convection is classified as above land or above ocean.

We added a short explanation to the main text:

This is done by computing the mode cloud type in a 15 km radius around the location of the air parcel for every hour. The first encounter of a very high opaque cloud (type 9) is considered convective outflow if the air parcel is in an altitude between 8 and 15 km. The convection is subsequently characterised as over land or over ocean.

Reviewer 3

This manuscript presents airborne measurements of methanesulfonic acid (MSA) and sulfuric acid (SA) over the tropical Indo-Pacific during the CAFE-Pacific aircraft campaign. The dataset is valuable because direct observations of MSA and SA in the tropical upper troposphere remain limited. To interpret the observed MSA and SA, the manuscript combines aircraft observations with inlet-characterization experiments, HYSPLIT trajectories, satellite cloud fields, and box modelling. The study indicates a potentially important role of deep convection and DMS oxidation products in upper-tropospheric aerosols. The paper is well written and scientifically important. However, several aspects require further discussion, e.g., the total MSA/SA measurements at high altitude, and the interpretation of DMS oxidation chemistry using observations and box modelling. I recommend publication after the comments below are addressed.

General comments

1. A central idea of this study is that adiabatic heating in the SCORPION inlet can cause acidic particles to evaporate at high altitude, allowing the instrument to detect total MSA and SA (combined gas- and particle-phase). However, uncertainties associated with total MSA and SA should be further discussed.

1) Figure 1 defines the "total" regime using thresholds in ΔT (Tinlet - Tambient) and inlet RH. Does this classification assume that the evaporation efficiency of acidic particles is effectively complete and stable within the selected "total" regime? Please discuss whether evaporation efficiency could influence the observed profiles of MSA and SA at high altitude.

Based on the comparison with the AMS in Fig. 4 (original manuscript) and the experience in the laboratory, we assume that the evaporation of small acidic particles is complete. However, we do not have a way to prove this for all flight conditions since there are no particle mass measurements available below the cutoff diameter of the AMS. Since this assumption is critical for the interpretation of the results presented in the paper, we added a discussion and another comparison to strengthen the argument.

The additional CLOUD experiment (Fig. 6) shows an injection of pure SA particles with a mean diameter of approximately 30 nm produced in a flow tube. The CLOUD chamber is at a temperature of 2.5 °C and RH of less than 1%. The RH in the inlet of SCORPION is even lower (<0.3%) due to the temperature increase, which lies around 11 °C. At these low RH values, SA evaporates strongly in SCORPION. The mass detected by SCORPION is almost identical to the mass calculated from the particle size distribution (measured by two SMPS), assuming a density of 1.8 $\mathrm{g\,cm^{-3}}$ for SA particles. The gas-phase SA values recorded by the LTOF are two orders of magnitude lower than the SCORPION measurements showing that SCORPION measures almost exclusively particle phase. This experiment indicates that at low RH in the inlet, sulfuric acid particles can be measured quantitatively by SCORPION.

It is not possible for us to map out the entire phase space of RH, ΔT and particle mass to determine exactly under which conditions complete evaporation occurs. The classification used is our best estimate based on the lab experiments and aircraft observations. However, we cannot fully exclude incomplete evaporation. In this case, the strength of evaporation would depend on the temperature difference and RH in the inlet. Both would lead to weaker evaporation at lower altitudes and hence an underestimation of total acid concentrations there. Additionally, if MSA evaporates more easily than SA, as suggested by its higher volatility [18], our measurements could yield too high MSA/SA ratios. We tried to account for this effect by choosing different RH and ΔT thresholds for MSA and SA. Wherever MSA/SA ratios are stated, only data points

are considered which fulfil the conditions of evaporation for both acids.

We refined the text in the manuscript:

For low enough RH and high enough values of ΔT we expect the evaporation to be complete. An experiment with injection of pure SA particles at <0.5% RH in the inlet is shown in Fig. A3 in the appendix. The comparison between the SA mass concentration measured by SCORPION and the mass calculated from the particle size distribution shows almost identical values, hence confirming the total evaporation of the particles. [..]

Since it was not possible to reproduce the full phase space of ΔT, RH and particle mass in the chamber experiments and the impact of bases remains largely unquantified in the aircraft measurements, we cannot fully rule out incomplete evaporation. This would primarily impact measurements in the middle troposphere where ΔT is lower and the RH in the inlet higher, which could lead to an underestimation of acid concentrations there. The values reported here therefore represent a lower limit of total MSA and SA.

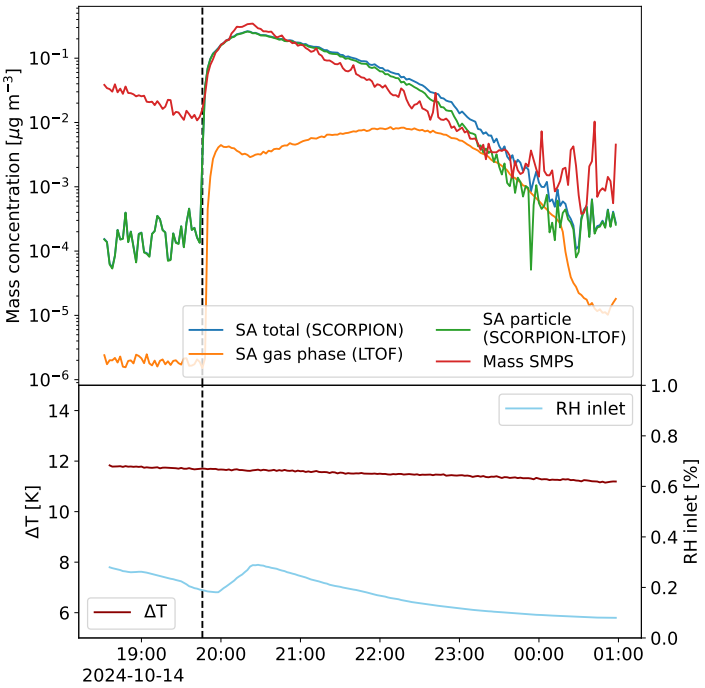


Figure 6: Time series of an experiment at the CLOUD chamber showing the injection of approximately 30 nm pure SA particles from a flow tube into the CLOUD chamber. (a) shows the mass of SA detected by SCORPION, the gas-phase SA detected by the LTOF, the difference between both and the mass calculated from the particle size distribution recorded by the SMPS assuming a density of 1.8 g cm^{-3}.

2) The authors state that acidic particles evaporate efficiently, whereas neutralized or partially neutralized particles do not. However, the neutralization state of small particles during CAFE-Pacific could not be directly measured. What fraction of the measurements was associated with particles too small for reliable AMS composition measurements? For particles within the AMS size range, can the authors provide information on the degree of neutralization?

Aerosol composition measurement with the AMS is strongly limited in the free troposphere.

Figure 7 shows the fraction of data points where measurements above the LOD of the AMS are available for the corresponding high-altitude measurements in the manuscript. The highest coverage is achieved for sulfate with more than 76% followed by nitrate with 34%. Chloride data is available for approximately a quarter of the measurements however these values have a high uncertainty due to the fragmentation of other compounds on the peaks associated with chloride. Ammonium could only be measured in less than 9% of the data points. This is likely due to the lower presence as well as the higher limit of detection compared to sulfate. For reliable acidity estimation at least sulfate and ammonium data need to be above the LOD, which is the case for only 7% of the measurements. A full analysis with all compounds required is only possible for 1% of the data.

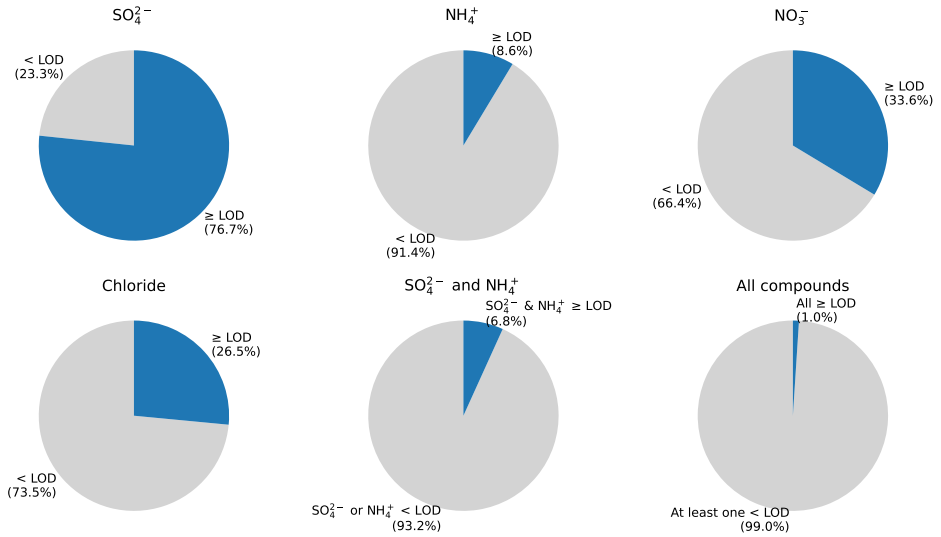


Figure 7: Fraction of C-TOF-AMS measurements during CAFE-Pacific above and below the limit of detection. Only data points in the middle and upper troposphere are considered, where SCORPION is expected to measure particle phase as well.

The neutralisation ratio of particles can be estimated using all these compounds to calculate the ratio between measured NH_4^+ and the NH_4^+ required for neutralisation according to eq. 1 from Zhang et al. [52].

$$R_{neutralisation} = \frac{NH_4^+}{18 \times \left(\frac{2 \times SO_4^{2-}}{96} + \frac{NO_3^-}{62} + \frac{Cl^-}{35.5}\right)} \tag{1}$$

Figure 8 shows the fraction of data points with acidic and neutral particles. We assume that particles with a neutralisation ratio below 0.75 are acidic and between 0.75 and 1.25 neutral. Frequently, we calculate values strongly above 1.25, these are likely a result of the high uncertainty of the measured values or potentially an overestimation of the ammonium concentration. Dividing two small concentrations barely above the LOD, can lead to high ratios which are not realistic. While these particles could be identified as neutral, we consider the uncertainty too high for a reliable identification. If all data with measurable sulfate are considered (Fig. 8a), we see a strong dominance of acidic over neutralised particles, however the unidentified fraction is high. For the data points with sulfate and ammonia above the LOD, neutral particles are slightly more frequent with more acidic values above the ocean and more neutralised particles above land. However, the largest fraction remains unidentified. Even if the concentrations are above the LOD, almost 60% of the calculated neutralisation ratios in Fig. 8b have uncertainties of more than 50% due to the low mass loading.

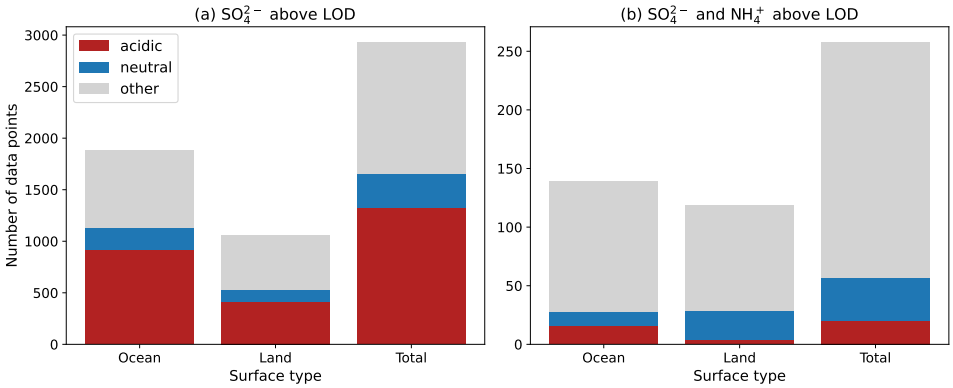


Figure 8: Acidity measurements by the C-TOF-AMS during CAFE-Pacific. Shown are the amount of 1-minute-measurements with acidic and neutral particles. Acidic particles are identified by a neutralisation ratio below 0.75 calculated according to Zhang et al. [52]. Neutral particles have a neutralisation ratio between 0.75 and 1.25. Ratios above 1.25 are considered artefacts of low overall concentrations and high uncertainties and cannot be classified with certainty. Data was separated by surface type based on the location of the measurement, not the airmass origin. (a) shows the data set used for the high altitude measurements in the main manuscript with sulfate above the LOD. (b) shows the subset with sulfate and ammonium measurements above the LOD. Note that only considering these measurements reduces the available data strongly and does not provide a representative picture of the conditions during the campaign.

Overall, the identification of particle acidity with AMS data is rarely possible and highly uncertain in the free troposphere.

We added both figures to the appendix of the manuscript and adjusted the text:

The neutralisation state of particles below 40 nm could not be measured during CAFE-Pacific. For particles above this diameter the composition measurements remain highly uncertain due to the very low mass concentrations, especially of ammonium (Fig. A5 and Fig. A6). Therefore, the values shown in the subsequent analysis represent lower limits for the total MSA and SA concentrations. However, outside the Asian monsoon region, very low ammonia concentrations are expected in the upper troposphere [14, 19, 37, 25] and large parts of the measurement region show the lowest impact of anthropogenic NH_3 on CCN concentrations globally [48].

2. In the RF18 case, the observations convincingly show high total MSA concentrations together with enhanced 10 60 nm particles in aged convective outflow after OH exposure. The authors then made mass estimates using assumed particle diameter, concentration and density, to suggest that MSA dominated the observed enhanced particle amounts. In contrast, the RF14 case, where the convective outflow had limited OH exposure, showed much lower acids and particle concentrations. This supports an important role of DMS oxidation products. However, the conclusion that DMS oxidation products are the dominant component of these particles seems stronger than what is directly demonstrated.

1) The total MSA and SA from SCORPION represent combined gas- and evaporated particle-phase signals. The relative contributions of gas- and particle-phase could not be separated. Unless the authors can demonstrate that the gas-phase contribution is negligible in the RF18 case, comparing total MSA with aerosol mass estimates may not directly indicate MSA contribution

to aerosols.

Recent measurements at the CLOUD chamber have proven that MSA condenses kinetically on particles at temperatures of 9°C and below at high relative humidities (50%) [2, 51]. This is the case even at minute levels of ammonia. These conditions apply in the upper troposphere during RF18. Therefore, we expect MSA to rapidly condense on the particles detected during the flight. Assuming a particle size of 20 nm and a concentration of 5,000 cm^{-3} (STP), calculations yield a condensation sink of 4×10^{-3} s^{-1} if all variables are converted to ambient conditions (215 K, 175 hPa). This condensation sink would lead to a decrease in MSA concertation of 90% within 9 min or 99% within 17 min. Consequently, concentrations observed during RF18 could not be sustained in the gas phase. While we cannot fully exclude a small contribution of gas-phase MSA, it can only make up a minor fraction of the detected signal.

We added the following reasoning to the text:

Due to the kinetic condensation of MSA onto particles [2, 51], the measured concentrations could not be sustained in the gas phase and represent almost exclusively particle phase measurements.

2) Is RF18 the only case with enhanced 10–60 nm particles? If similar events were observed during other flights, were they consistently associated with high total MSA concentrations and marine convective outflow after OH exposure? Or were the cases with high total MSA always associated with particle enhancement?

Answering this is complex, since most of the flights were not focused on measuring the outflow of marine convection. There are several occasions where high MSA concentrations were detected, especially in air masses from the Warm Pool as shown in Figure 8 of the manuscript. However these do not necessarily correlate with particle number concentrations. Instead, they will depend on the particle mass. A detailed analysis of this would require full particle size distributions throughout the whole campaign, which are not available at the current stage. Small concentrations of large particles can quickly overwhelm the mass of several thousand newly formed particles. Additionally, tracing back measurements to convection is significantly more uncertain if the aircraft passed through an air mass in a straight line instead of measuring it repeatedly as in RF14 and RF18.

However, we do not consider RF18 a special or singular occasion. High particle mass concentrations in combination with high MSA concentrations were also measured during RF08, which is used for the comparison between C-TOF-AMS and SCORPION in the paper (Figure 4). Unfortunately, no size resolved particle measurements are available during this flight but the good agreement with the AMS sets a lower limit of 40 nm. The trajectories are clearly of marine origin and the Himawari satellite indicates high convective activity around the flight area during the past 24 hours (see fig. 9). These convective systems represent a possible origin for the particles. However, the comparably high ozone mixing ratios of 60 ppb and low RH (20%) in the air mass indicate that the convection could have occurred further in the past. Simply based on the trajectories and the satellite data, the exact airmass origin cannot be determined conclusively.

Another example is RF21, where we detect high MSA and SA concentrations in the Pacific Warm Pool (fig. 10 and fig. 11). In the same region, we measured enhanced particle number concentrations for nucleation mode (2–12 nm) and Aitken mode (12–60 nm) particles. The 22-hour-backward trajectories indicate a possible connection to convection east of the flight track. The low O_3 mixing ratios around 15 ppbv agree with convective origin. The RH is highly variable between 20 and 80% indicating that potentially different air masses were measured along the flight path. Overall the correlation between the acids and the particle number concentration

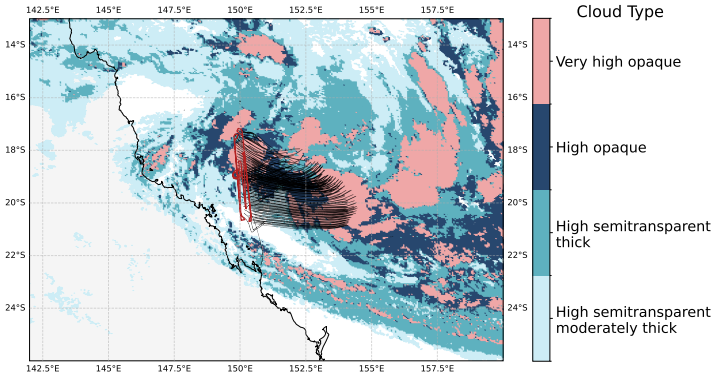


Figure 9: Trajectories during a section of RF08 over the ocean close the the Australian coast with a Himawari satellite picture indicating the cloud types. The flight path is shown in red, the 10-hour backward trajectories in black. The satellite picture shows the situation at 23.01.2026 15:00 UTC which corresponds to 8–12 hours before the measurement. This convection is a possible origin of the particles, however low RH and comparably high O_3 mixing ratios could hint towards a more aged convective outflow.

is not perfect, likely caused by the encounter of different air masses with different aerosol size distributions and origins. The enhanced MSA concentrations in the beginning of the flight could for example be caused by low concentrations of larger particles or a larger contribution of gas-phase MSA.

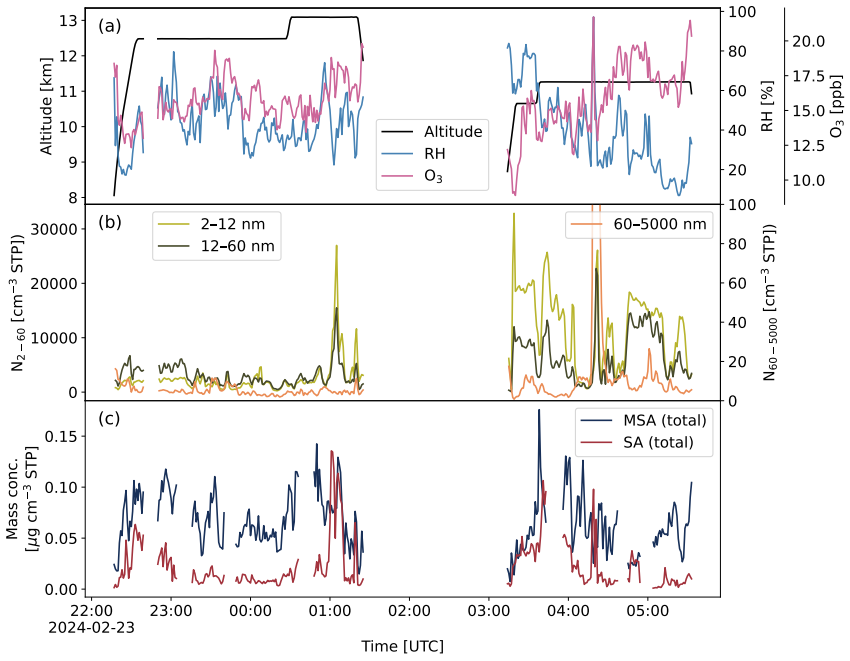


Figure 10: Time series of RF21 with (a) showing the altitude, RH and O_3 mixing ratios, (b) the particle number concentrations in different size ranges and (c) the mass concentration of total MSA and SA.

We would like to refrain from adding a deeper analysis on multiple other flights, as we consider this outside the scope of this study. In particular, since another publication is planned, focusing on the particle number concentration measurements in different size ranges. This will also include

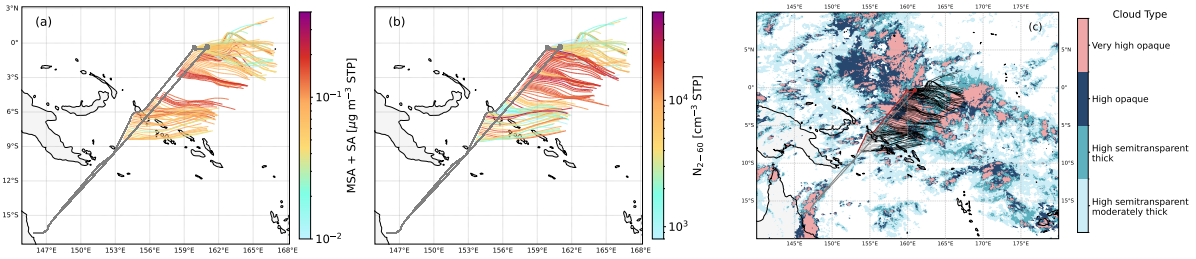


Figure 11: Flight track (grey) and 24-hour backward trajectories for a section of RF21. The trajectories are coloured by the sum of MSA and SA mass concentrations (a) and the concentration of particles with diameters between 2 and 60 nm (b). In (c) a Himawari satellite picture is shown from 23.01.2024 03:00 UTC which corresponds to 22–26 hours before the measurements in (a) and (b).

the potential different origins and compositions of these particles. In the present publication, we focus on the direct MSA and SA measurements conducted by the nitrate CI-APi-TOF.

We added these figures to the appendix of the manuscript and the following statement in the conclusion:

Enhanced MSA concentrations were also observed during several other research flights. RF08 provides another example of elevated particle-phase MSA associated with a marine air mass (Fig. 4, Fig. A24). During RF21 in the Warm Pool region up to 20,000 cm^{-3} nucleation mode and 10,000 cm^{-3} Aitken mode particles were detected coinciding with enhanced SA and MSA concentrations of around 0.05 $\mu\mathrm{g\,m}^{-3}$ and 0.1 $\mu\mathrm{g\,m}^{-3}$ respectively (Fig. A25). These could originate from a convective system observed approximately 24 hours before the flight (Fig. A26). A more comprehensive analysis of the particle size distributions throughout the entire campaign and their sources will be presented in a future publication.

3) It would be useful to show the time series of other AMS species for RF18 and RF14. Although the AMS does not cover particles below 40 nm, these data would provide complementary constraints on larger particle composition, acidity, and the possible contribution of other aerosol components.

We have added the time series for the AMS species for RF14 and RF18 to the appendix (Fig. 12 and Fig. 13). Most compounds are below or very close to the detection limit of the instrument in agreement with the low particle number concentration of particles above 60 nm. We added the two following sentences to the discussions of the respective case studies:

All other compounds measured by the AMS stay below or close to the limit of detection (Fig. A14).

All other compounds measured by the C-TOF-AMS also barely exceed the limit of detection (Fig. A15).

3. The chemical box model provides useful support for the timescale of DMS oxidation after convective uplift. However, additional details and discussion are needed.

1) Please provide more details on the initial model setup. How were the initial DMS concentra-

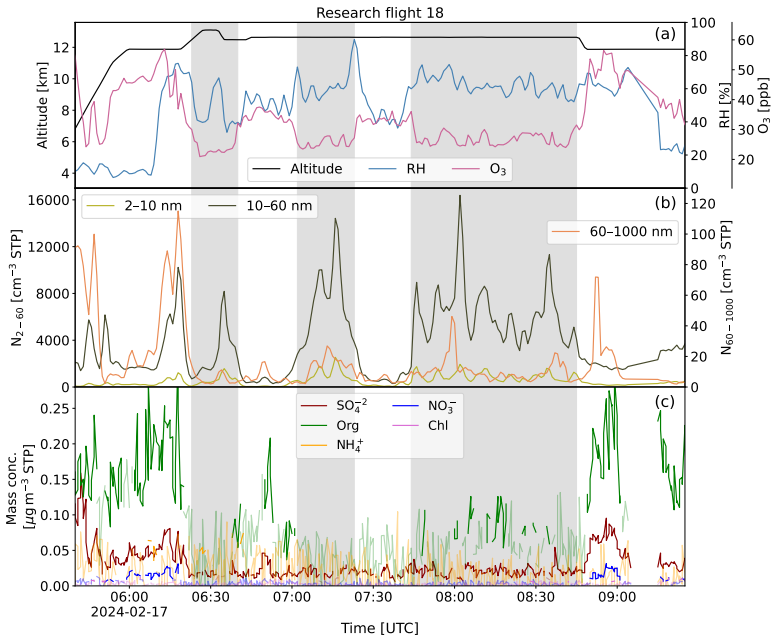


Figure 12: Similar to Fig. 5 in the main text but with all species detected by the C-TOF-AMS plotted in (c). The faded out colour indicates values below the limit of detection of the instrument. In the air mass of interest ammonium, nitrate and chloride are not detected at all, whereas sulfate and occasionally organics reach values slightly above the LOD.

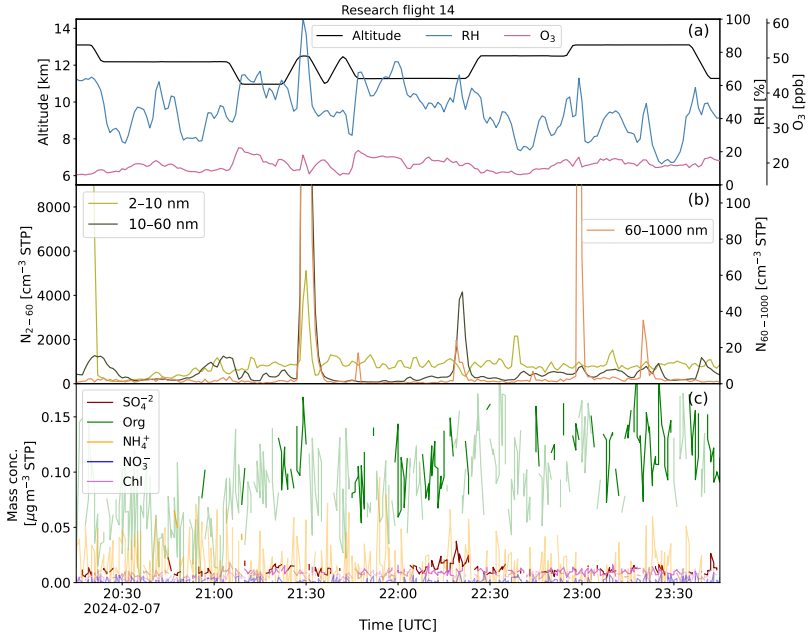


Figure 13: Similar to Fig. 6 in the main text but with all species detected by the C-TOF-AMS plotted in (c). The faded out colour indicates values below the limit of detection of the instrument. Ammonium and nitrate are not detected at all, whereas sulfate, chloride and organics occasionally reach values slightly above the LOD.

tion, temperature, pressure, relative humidity, and background gases selected?

The model was set up to represent condition of upper tropospheric convective outflow, specifically those encountered during RF14 and RF18. 200 hPa and 223 K are representative for an altitude

of 12 km, which is the approximate flight altitude of case studies for RF14 and RF18, and generally within the typical outflow region of 10-17 km [13]. An RH of 60% is reached frequently in RF18, and less consistently also in RF14. The O_3 mixing ratio is fixed to 30 ppbv comparable to RF18 values, a bit higher than RF14 and representative for O_3 values encountered above the ocean during the campaign [39]. Similarly low NO values of 40 pptv were chosen to represent the low mixing ratios in the Pacific region and the low production by lightning in marine convection [39]. The DMS mixing ratio was selected based on the CAMS EAC4, specifically on the 99th percentile values in Fig. A11 since we expect these to show the maximum concentrations available directly in convective outflows.

We added details of the set up to the box model section:

The simulations were performed under fixed meteorological upper-tropospheric conditions. The model temperature was held constant at 223 K, the pressure at 200 hPa, and the relative humidity at 60%. These values are representative for an outflow altitude of 12 km similar to the case studies of RF14 and RF18 and within literature values of tropical outflow altitudes of 10–17 km [13]. The mixing ratios of major background constituents O_2, N_2, CO, O_3 and H_2O were kept constant, to isolate the chemical evolution of the sulfur and radical species of interest. O_3 was set to 30 ppbv comparable to the values in RF18. NO was initialised with 40 pptv corresponding to the low values encountered in the marine upper troposphere [39]. NO_x and all other species in the mechanism were allowed to evolve freely according to the coupled gas-phase chemistry and photolysis.

The DMS mixing ratio is initialised with 50 pptv. After initial equilibration, it is around 45 pptv, a value that can be reached in convective outflows in the region and during the time of the campaign according to the CAMS global reanalysis (EAC4), see Fig. A11 in the appendix.

2) The DMS mechanism from Shen et al. (2022) is used in the modelling work. Please explain why this mechanism was selected, and how it differs from other available DMS oxidation mechanisms?

The mechanism by Shen et al. [43] was initially selected because it is based on chamber experiments, focuses on low-NO_x conditions and explicitly contains the temperature dependence of MSA/SA formation. Since many other mechanisms have been published in the past years, we decided to include a second, more recent scheme. Jacob et al. [22] is based on the MCM and multiple other mechanisms. It performed best in a reanalysis of several other chamber studies. In particular it likely presents an improvement to the Shen mechanism in terms of HPMTF and SO_2 concentrations. In our box model it enhances the formation of MSA and SA while DMS is oxidised slightly slower compared to Shen et al. Overall, both models predict concentrations within a similar range, indicating that no large discrepancies are expected when using different current DMS oxidation schemes.

The discussion of the box model was extended to both mechanisms, as was already described in the response to Reviewer 1.

The DMS mixing ratio decreases more slowly in the Jacob mechanism than in the Shen mechanism. However, more MSA and SA are produced using the Jacob scheme, which indicates a more efficient conversion of DMS to acids. The DMS in the Shen mechanism is largely converted to HPMTF, which has mixing ratios 3–4 orders of magnitude higher than in the Jacob scheme (Fig. A20). This could be caused by the low OH reaction rate in the Shen scheme or the additional HPMTF photolysis included by Jacob et al. The higher SA formation in the Jacob scheme

likely results from the enhanced production of SO_2 (Fig. A20), while the Shen scheme has been shown to underpredict SO_2 formation [22]. Overall, acid production is comparable between the two schemes, with the Jacob mechanism yielding 10–60% more SA than the Shen mechanism. MSA concentrations are initially up to 20% higher using the Jacob scheme but later remain up to 20% lower than using the Shen scheme.

3) The model considers gas-phase OH chemistry only, and excludes dilution, nucleation, condensation, multiphase processing, and removal. Other DMS oxidation pathways should be further discussed to avoid over-interpreting the role of gas-phase OH chemistry. For example, the relative importance of different DMS oxidation mechanisms (OH vs. halogen vs. NO_3) have been discussed in some studies (e.g. Fung et al., 2022; Tashmim et al., 2024). In addition, aqueous-phase reactions can be an important or even dominant source of MSA (e.g. Chen et al., 2018; Hoffmann et al, 2016). Although cloud chemistry is not resolved in the box model, the authors could evaluate whether the back trajectories overlap with cloud fields after convective, and discuss how unresolved cloud chemistry may affect the inferred MSA/SA production.

We agree that a more detailed discussion on the limitations of the box modelling is required, especially regarding the complex DMS chemistry pathways. Apart from OH, DMS can be oxidised by NO_3, O_3 and halogen compounds, mostly BrO [9, 44]. Oxidation by O_3 is expected to be a minor pathway and O_3 mixing ratios in the Warm Pool region are very low [44, 39]. NO_3 chemistry is included in the model. However, due to the low NO_x values in the marine UT the values stay low (<0.05 pptv) and it is expected to play only a weak role in DMS oxidation. Otherwise, we would also expect higher values in the early morning flight, since NO_3 concentrations are highest at night. This is also in agreement with global models, which predict NO_3 only as a major oxidant for DMS in polluted regions [7, 44].

Halogen species are not included in the original chemical schemes by Shen et al. [43] and Jacob et al. [22]. To estimate the impact of halogen species, we have added reactions from Burkholder et al. [6] and conducted two more model runs initialised with 5 pptv of Br_2 and 5 pptv of Cl_2. Both compounds are rapidly converted to ClO and BrO. Inorganic Br_y in the target region has been measured during a previous aircraft campaign [31] in the range of 0-10pptv. The value of 5 pptv was chosen to represent this range and is at the upper limit of the BrO observations. ClO observations in the tropical UT are sparse. Hobe et al. [17] report mostly mixing in the low pptv range (at or below LOD) in tropospheric air. Higher values were observed only for stratospheric influence or heterogeneous reactions in connection with cirrus clouds.

The results in Fig. 14 show slight changes in the DMS oxidation and product concentrations. DMS mixing ratios decline faster in both chemistry schemes due to the higher oxidant concentrations. MSA concentrations are enhanced compared to the model run without halogens with almost twice as high values. This is likely due to the enhanced formation of MSA intermediate compounds retaining the methyl group rather than fragmentation to SO_2/SA. In the Shen mechanism, less SA is produced, whereas SA concentrations in the Jacob mechanism are only weakly affected, mostly at longer time scales. Overall, the inclusion of halogen compounds at realistic values does not significantly impact the comparison of model and observations. While MSA concentrations remain within the uncertainty range of the measurement, observed SA concentrations still exceed modelled values strongly.

We extended the discussion on the box modelling by the following paragraph:

Oxidation of DMS by halogens is not considered in the box model. To estimate the potential impact, a sensitivity run was initialised with 5 pptv of Br_2 and 5 pptv Cl_2 which are rapidly converted to BrO and ClO. The results in Fig. A21 show a more

rapid oxidation of DMS an enhanced production of MSA (50% in Shen and 90% in Jacob) while SA concentrations are affected less. As these halogen mixing ratios are within the upper range of ambient observations [17, 31], their omission in Fig. 10 is unlikely to have a major impact. NO_3 and O_3 oxidation are included in the chemical scheme but play negligible roles due to the low concentrations.

Aqueous-phase processes were already discussed in a response to reviewer 1. They play an important role in atmospheric DMS chemistry. DMS, DMSO or MSIA can be efficiently oxidised in the aqueous phase. Overall, this leads to a higher MSA yield in global simulations compared to the omission of these reactions [9, 20]. Similarly, van Rooy et al. [46] also observed higher MSA/SA ratios under more humid conditions. The loss of MSA by reactions with OH in the aqueous phase, on the other hand, is low (12% globally [9]) compared to dry and wet deposition. It is expected to occur on a timescale of days at room temperature and likely much slower in the free troposphere [36]. Another important pathway is the uptake of HPMTF, which is converted with almost unit efficiency to sulfate [24]. However, it should be considered that most clouds encountered in the upper troposphere would be ice clouds. To our knowledge, there are no systematic studies on the interaction of DMS or its oxidation products with ice clouds, but it can be assumed that any reactions would be much weaker than in the liquid phase. While cloud processing is highly important and well studied in low liquid clouds, it is likely much less impactful in the mid and upper troposphere.

Nevertheless, to estimate the impact of cloud processing, the trajectories and cloud type field by the Himawari satellite were used to investigate cloud encounters of different cloud types. For this, it was assumed that the trajectory encountered different clouds if it was located in the respective altitude: very low cloud: 0–2 km, low cloud: 2–3.5 km, medium cloud: 3.5–6.5 km, high cloud: 6.5–10 km. Due to the small vertical extent and limited literature on interactions of DMS with ice clouds, cirrus clouds were excluded from this analysis.

The data in figure 9 was then plotted again, omitting the trajectories which encountered another cloud after the convective uplift (dashed lines in Fig. 2). For convection over the ocean, slightly higher acid values are detected if no cloud was encountered. However, only minimal differences can be seen in the mean MSA and SA values. Figure 2b shows the fraction of data points in each bin where cloud encounters were identified, split up by the different acids and convection locations. The fraction of increases with increasing time since convection as expected due to the longer time spend in the atmosphere. Not more than 30% of the data points with convection in the past 5 days were impacted by a second cloud encounter. Even those with high fractions (e.g. MSA 6-9 hours after convection over the ocean) show only minimally different mean concentrations. While there are certainly limitations due to the accuracy of the trajectories and vertical extent of the clouds, we still conclude that aqueous-phase reactions after convective transport do likely not majorly impact our results.

The figure was added to the appendix and the following paragraph to the discussion:

A shortcoming of the box model setup is the lack of aqueous-phase reactions, which have been shown to play a major role in DMS processes [20, 9]. In the BL, cloud processing is a crucial process promoting MSA formation and reducing SO_2 concentrations through the uptake of HPMTF [20, 9, 24, 38]. It can occur in the convective cloud during the uplift and the resulting MSA and SA may be injected in the particle phase into the upper troposphere. This could be the reason for the enhanced values shortly after contact to a convective cloud. However, the case study of RF14 and previous aircraft campaigns indicate low particle concentrations in cloud outflows [1, 10]. More detailed modelling would be required to quantify this pathway.

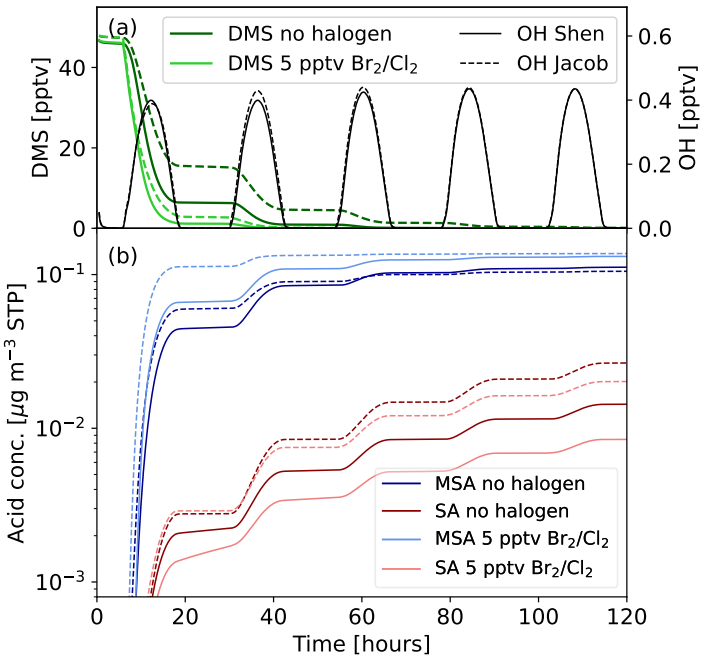


Figure 14: Box model simulations of upper tropospheric DMS oxidation including halogen reactions. The model set up is identical to Fig. 10 for the darker colours and of 5 pptv Br_2 and 5 pptv Cl_2 were added for the lighter lines. The chemistry schemes by Shen et al. [43] (solid lines) and Jacob et al. [22] (dashed lines) were used with additional halogen reactions from Burkholder et al. [6]. For better readability the OH mixing ratios are only plotted without halogens.

Cloud processing can additionally happen after the injection of gas-phase DMS into the upper troposphere if another cloud is encountered. Using backtrajectories and Himawari satellite data we estimate that less than 30% and in many cases less than 10% of data points had a second cloud encounter after convective uplift (Fig. A23). Excluding these data from the analysis changes the results only marginally. Furthermore, the majority of the encountered clouds are likely ice clouds, where aqueous-phase reactions are expected to be considerably less important than in liquid-phase cloud droplets. We have hence no evidence that aqueous-phase reactions strongly impact our results.

Minor comments:

Please define SA at the first use and ensure consistent use of the abbreviation throughout.

We corrected the manuscript and use the abbreviation consistently.

Line 132. Please explain how the authors define that "the values are dominated by evaporated particles even at low particle concentrations."?

A simple calculation assuming pure MSA particles shows that the evaporation of only 10 particles per cm^3 with a diameter of 60 nm, or a single particle per cm^3 with a diameter of 150 nm, produces MSA concentrations exceeding typical gas-phase values. Consequently, even a small number of large particles can dominate the measured signal. This relationship is illustrated in Fig. 15. An analogous calculation for SA yields nearly identical results because of the similar molecular masses and densities of MSA and SA.

The text was changed to:

However, the concentrations are dominated by evaporated particles even at relatively low particle number concentrations. For example, as few as 10–20 pure MSA particles cm^{-3} with diameters of 60 nm or larger are sufficient to produce MSA concentrations that exceed typical gas-phase MSA levels (see Fig. A7).

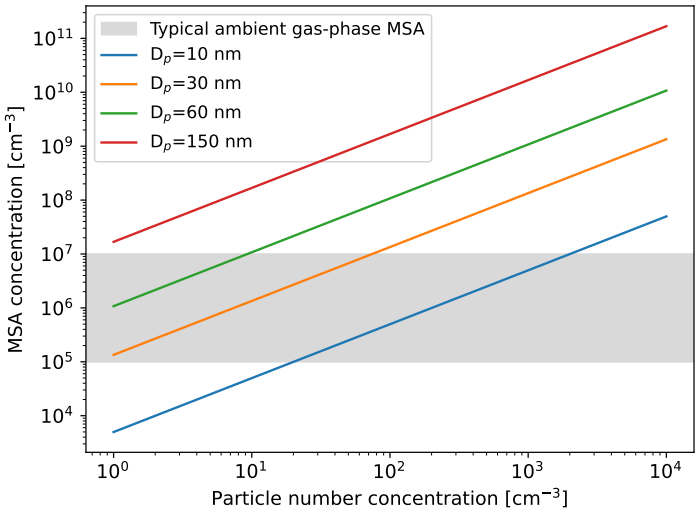


Figure 15: Calculation of gas-phase MSA produced by the evaporation of pure MSA particles of different diameters and concentrations. The grey area shows typical gas-phase concentrations of MSA.

Line 157. "The FASD instrument consists of 10 ultrafine Condensation Particle Counters (CPC) with different fixed cut-off diameters and measures particles in the range of 2–20 nm. The larger particles between 60 and 1000 nm are measured by an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)." How were the "10–60 nm" particles shown in Figs. 5 and 6 defined, without the range between 20 to 60 nm?

The FASD instrument consist of 10 CPCs which can measure the particle concentration above their set cut off diameters. These can in principle lie in the range of 2–20 nm. However, the exact cut off diameters depend on the instrumental settings and vary slightly between flights. The UHSAS measures particle number concentrations in size bins spanning diameters from 60 to 1000 nm. These measurements can be converted into cumulative number concentrations by summing all size bins above a given diameter. The particle number concentration in the 10–60 nm size range is then obtained by subtracting the cumulative concentration above 60 nm from the cumulative concentration above 10 nm. Deriving concentrations from the difference between two instruments results in greater uncertainty due to potential systematic uncertainties. We tried to minimize this effect by cross-checking accumulation mode particles of the UHSAS with a CCN counter.

The text in the methods section was adjusted:

The larger particles between 60 and 1000 nm are measured by an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) and can be integrated over the respective diameter range to obtain a cumulative size distribution similar to the FASD data. Particle number concentrations in different size intervals are subsequently obtained by taking the difference between cumulative concentrations at the corresponding diameter thresholds.

Line 204. Low-altitude measurements of "gas-phase" SA and MSA are shown...

This was corrected in the manuscript.

Line 209. The authors state that "These rather high values can be explained by the strong marine influence...". Please clarify what values are being referred to, and what they are higher relative to.

We have changed the sentence to make the statement more objective:

The concentrations of MSA and SA in the BL vary from below our limit of detection ($<$3–4$\times10^6$ cm^{-3}) to several 10^7 cm^{-3}. These values are comparable but slightly higher than measurements in the Arctic [5], Antarctica and the Southern Ocean [26, 3, 40], while similar values have been reported in the tropical Indian Ocean [42].

Line 224. Recommend adding the expected lifetime of sulfuric acid and references.

The lifetime of SA depends mainly on the condensation sink which is determined by the particle sizes and concentrations. It yields SA lifetimes from seconds in the polluted BL to several hours in the free troposphere [41, 45]. A typical value in coastal conditions is around 8 min (condensation sink of 2×10^{-3}s^{-1} [11]).

For SA, the highest concentrations are measured closest to the ground in all air masses indicating the diverse sources and its short lifetime (seconds to few hours depending on the condensation sink [11, 41, 45]).

Line 236. Recommend adding the expected lifetime of MSA and references.

Reports of MSA lifetime in the gas phase are sparse. Chemical reactions of MSA are slow [4], hence the lifetime is typically determined by condensation. However, this depends on the volatility of MSA which is still an ongoing area of research and likely RH dependent [18, 51]. The sentence was removed from the manuscript and the discussion focused on limitations of the chemical scheme and evaporation instead.

Line 238. The authors state that "Another important source could be evaporation of MSA from particles at low ambient RH values." It would be useful to show the profiles of observed ambient RH.

We have added an altitude profile of the relative humidity in the appendix (Fig. 16). It shows high values in the boundary layer, mostly exceeding 60% and a larger spread in the middle and upper troposphere.

It is referenced in the main text:

While the RH in the BL is mostly high (∼80%, Fig. A9 in the appendix), the range of values in the free troposphere is broad.

Line 239. The authors state that "This could also explain the high MSA values measured for low marine fractions in Fig. 3a." Should this be Fig. 3b?

Yes, it was corrected in manuscript

Line 264. The authors state that "This indicates that under these conditions...". Please specify what "these conditions" refer to. Recommend adding the approximate range of ΔT and inlet RH for this RF08 example.

The conditions were added:

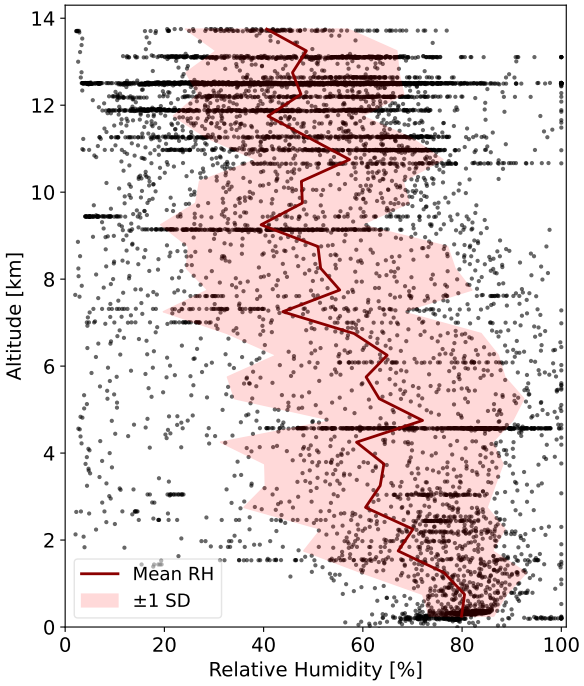


Figure 16: Altitude profile of the measured relative humidity (above water) during all 17 research flights of CAFE-Pacific. The red line shows the mean values with the standard deviation as shaded area. Data points with values above 100 % were recorded during cloud encounters and for simplicity set to 100 % in this plot.

This indicates that under these conditions (3–7% RH in the inlet and $\Delta T \approx 30$ K), SCORPION efficiently evaporates particles, which are simultaneously measured by the C-TOF-AMS.

Line 267. The authors should clarify how they infer "mostly acidic particles" from the AMS measurements.

The acidity is typically calculated based on the measured NH_4^+ and what would be required to neutralise the particles based on SO_4^{2-}, NO_3^- and Cl^- measurements according to [52] and eq. 1. However, during the measurements shown in Figure 4, the ammonium concentration remained below the limit of detection. We have adjusted the sentence to:

The C-TOF-AMS confirms this by showing no detectable ammonia even during the periods with higher mass loadings, which is consistent with the low ammonia levels expected in the upper troposphere [19, 25].

Line 275. Recommend adding values or a supporting profile figure, to show the "much higher particle concentrations at low altitudes compared to high altitudes."

This statement is based on a comparison between the measurements during flight RF08 (Fig. 4) and RF17 (Fig. A7). The sentence was changed to:

This is the case despite the substantially higher particle mass concentrations at lower altitudes, as indicated by the AMS sulfate measurements and OPC concentrations, both of which are approximately one order of magnitude greater than those shown in Fig. 4.

Fig. 5c. Total MSA and SA are both high around 06:30 UTC, while only MSA was largely enhanced during other shaded periods. What may cause this difference?

The measurements with higher SA concentrations were obtained during the aircraft's ascent and at a slightly higher altitude than the following encounters. While the low O_3 values indicate that it is a convective outflow, the lower aerosol concentrations and lower MSA/SA ratio could hint at a different origin than the air mass that was measured later in the flight.

Line 314. The authors state that "hence concentrations which are one order of magnitude higher than during other flights." Please clarify what type of flights or conditions these "other flights" refer to.

The sentence was changed to:

This leads to efficient oxidation of the transported DMS to MSA and SA and hence concentrations which are one order of magnitude higher than during other flights such as the fresh outflow encountered during RF14, which is discussed in the following section.

Line 356. The authors should provide more details on the method of "K-means clustering". Please check whether the cluster numbering is consistent between Figs. 8, A9, and A10 (now A17 and A18).

We have added a brief explanation on the K-means clustering method used (see below) and corrected the cluster numbers in A9 and A10.

Five-day HYSPLIT backward trajectories and K-means clustering [35] were used to determine the main source regions. Euclidean distance clustering is applied to account for spatial, temporal and transport-path similarity. The number of clusters was determined empirically to achieve an optimal representation of distinct but representative airmass origins.

Additional changes to the manuscript

We noticed a mistake in the value for the residence time. Instead of a constant value, it varies with altitude between 0.45 and 1.7 s. The sentence was corrected:

Due to the long inlet line with a residence time between 0.45 and 1.7 s, this can influence sampled particles and, in some cases, lead to their evaporation.

Additionally, the CAMS data in the appendix of the previous submission was given in mass mixing ratios instead of volume mixing ratio. We corrected this in the revised manuscript.

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