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 (T_{inlet} - T_{ambient}) 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.

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?

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.

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?

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.

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 concentration, temperature, pressure, relative humidity, and background gases selected?
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?
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. NO3) 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.

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

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

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?

Line 204. Low-altitude measurements of “gas-phase” SA and MSA are shown…

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.

Line 224. Recommend adding the expected lifetime of sulfuric acid and references.
Line 236. Recommend adding the expected lifetime of MSA and references.

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.

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?

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.

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

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

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?

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.

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.

References

Chen, Q., Sherwen, T., Evans, M., and Alexander, B.: DMS oxidation and sulfur aerosol formation in the marine troposphere: a focus on reactive halogen and multiphase chemistry, Atmos. Chem. Phys., 18, 13617–13637, https://doi.org/10.5194/acp-18-13617-2018, 2018.

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