General.

We would like to appreciate the referee for providing the valuable comments to improve the manuscript. We have revised our manuscript by fully taking the editor's comments into account. Responses to specific comments are described below. All the changes made and appeared in the revised text are shown in red. All detailed answers to comments are displayed in blue.

Comments of Referee #4 and our responses to them

This manuscript presents an observational study on organosulfates (OSs) and nitrogen-containing organic compounds (NOCs) in fine particulate matter in an island in the South China Sea. The source of marine emissions has been emphasized though other source contribution of long-range transport could still be found. This study highlights the contribution of marine biogenic emissions for OSs and NOCs in the oceanic area. I would recommend it to be published in ACP after moderate revision. My comments/suggestions are listed below.

Response: We deeply appreciate your valuable suggestions and time spent reviewing our manuscript.

General comments:

1. Line 254-255, Page 12: As the proportions of OSa in the total OSs are higher at both inland and ocean sites, how can we differentiate between the anthropogenic and oceanic sources of OSa based on its percentage in aerosol samples?

Response: The mass fractions of OS_a (aromatic and aliphatic SOA) in the total OSs in inland China were smaller than that in Sansha Island (a region with minimal anthropogenic pollution), although the OS_a concentration was lower in Sansha Island (Figure 1a and Table S4). The attribution of this phenomenon to the long-range transport of pollutants from coastal cities in China may not be a reasonable assumption. This is due to the fact that the transmission effect was incapable of inducing an increase in the proportion of OS_a from coastal cities with developed industries (e.g., Shanghai and Guangzhou) to Sansha Island (see section 3.3 for further evidence). In addition, the SOA markers we detected showed a significant positive correlation with ozone ($P<0.01$, Figure 5), indicating that these SOA markers were formed locally (Lines 245–264).

2. Line 295-297, Page 14: The assertion that atmospheric oxidation capacity does not constrain OS formation is contradicted by the significant correlation between OSs and O3 mentioned in 3.2.

Response: We are very sorry for the confusion caused by our description. Here, what we want to express is that atmospheric oxidation capacity is not a limiting factor for OS formation at the site of this study. We have rephrased the sentence in the revised draft.

Lines 306–307: …sulfate availability and atmospheric oxidation capacity would not be limiting factors for OS formation…

3. Ling 330, Page 16: Given that both marine biological activities and anthropogenic sources contribute to atmospheric sulfate, what are the non-sea salt and sea salt proportions of the total sulfate in aerosol samples, and are there any differences in *correlations of OSs versus these sulfate fractions?*

Response: If sea salt sulfates are defined as 0.060 times the sodium ion concentration based on previous report (Chen et al., 2010), the calculated average proportion of non-sea-salt sulfates is approximately 93% of the total sulfates. However, this method does not seem to be suitable for calculating the actual concentration of sea-salt sulfates. This is because the sulfates in the $PM_{2.5}$ sample are mainly formed through secondary processes associated with precursors emitted from the ocean (e.g., DMS transformation), and sea-salt sulfates and sodium should be mainly distributed in coarse particles. Therefore, we cannot accurately distinguish the ratio of sea-salt and non-sea-salt sulfates here. However, this study can at least demonstrate that abundant marine aerosol sulfates are not a limiting factor for OS formation by comparing observation cases of coastal cities and Sansha.

The following content is what we want to emphasize

Lines 345–349: OSs showed significant correlations ($r > 0.6$, $P < 0.01$) with local O_3 levels. The above results suggest that aerosol OSs in the Sansha area may be mainly formed locally and are tightly associated with precursor emission levels (e.g., abovementioned phytoplankton emissions) rather than sulfates.

4. Line 388-389, Page 19: How to explain the coincidence between the maximum of biomass burning intensity and elevated levels of OSs and aromatic NOCs during winter and spring months, assuming the SCS atmosphere is unaffected by air mass long-range transport?

Response: The analysis of air mass backward trajectories shows that although biomass combustion activity is more intense in inland areas during winter and spring, the main air masses arriving at study stie have not passed through the combustion intensive areas. Furthermore, if we assume that biomass burning in inland or coastal areas transferred OSs to Sansha, this will only lead to an increase in OS_a abundances, rather than a highly consistent increase in OS_i, OS_m (biological origin), and OS_a abundances.

We have added more descriptions in the revised manuscript.

Lines 410–413: Even if we assume that biomass burning in inland or coastal areas transported OSs to the Sansha region, it would only lead to a significant increase in OS_a abundances but not induce a synchronous increase in OS_i, OS_m, and OS_a abundances…

5. Line 392-394, Page 19: The inconsistent variation trends between nss-K+ and the surrounding continental fire point density alone cannot conclusively demonstrate that the SCS atmosphere is not impacted by air mass long-range transport. Are there additional anthropogenic or terrestrial tracer concentration patterns strengthening this assumption?

Response: If we assume that biomass burning in inland or coastal areas transferred OSs to Sansha, this will only lead to an increase in OS_a abundances, rather than a highly consistent increase in OS_i, OS_m (biological origin), and OS_a abundances (Lines 410–413).

6. Line 415-417, Page 20: Does the inverse relationship between isoprene/monoterpenes and OSs/aromatic NOCs contradict the previous allusion that the predominance of isoprene and monoterpenes-derived NOCs over aromatic NOCs results from marine *emissions of isoprene and monoterpenes?*

Response: Here we refer to the emissions of isoprene and monoterpenes from ships. Their lack of significant positive correlation with OSs and aromatic NOCs indicates that ship emissions are not the main factor driving the temporal changes in OSs and NOCs in the Sansha area.

7. Line 439-441, Page 22: Could the author provide the exact Mantel's r and p value from the Mantel test, since the threshold of >4 is too broad for meaningful interpretation?

Response: The code we used cannot directly input all specific Mantel's r and p values. Thus, we performed PCA analysis and correlation analysis between specific markers ($C_6H_5O_4S^-$ and $C_7H_7O_4S^-$) and key parameters (**Figure 6**) to further support our inference.

8. Line 487, Page 25: How can Ca2+ indicate marine sources given its negative correlation with Na+ and Cl- in Figure 5?

Response: We have removed the inappropriate statements. Indeed, calcium ions, as mineral ions that can be transported over long distances and are mostly present in coarse particles, should not be hastily determined for their oceanic origin.

9. Line 502, Page 25: Did these two aromatic OSs dominate in the total aromatic OSs? How do other aromatic species correlate with these marine emission indicators?

Response: On average, $C_6H_5O_4S^-$ and $C_7H_7O_4S^-$ accounted for only about 6% of the total aromatic OS_s (**Table S3**). The total aromatic OSs were significantly correlated with the marine emission indicators. We chose these two SOA markers because their structures have been resolved and we have a clear understanding of their precursors.

The detailed discussion is as follows:

Lines 508–518 : The release of benzene and toluene from marine organisms has been suggested to be important precursors for marine SOA (Wohl et al., 2023). It has been established that the reaction of benzene and toluene with sulfate radicals in the aqueous phase can lead to the formation of two aromatic OSs, namely $C_6H_5O_4S^-$ and $C_7H_7O_4S^-$ (Huang et al., 2020). We detected both aromatic OSs in aerosols collected in the Sansha area, the structures of which were shown in **Figure 6b**. Moreover, we found that these two benzene- and toluene-derived OSs showed significant positive correlations with multiple indicators of marine emissions (e.g., Chl a, surface seawater isoprene, Na^+, OS_i, and OS_m) (**Figure 6b**). This further corroborates the important effect of marine emissions on the formation of aromatic-derived SOA in the Sansha area…

Once again, we deeply appreciate the time and effort you've spent in reviewing our manuscript.

References

Chen, H. Y., Chen, L. D., Chiang, Z. Y., Hung, C. C., Lin, F. J., Chou, W. C., Gong, G. C., and Wen, L. S.: Size fractionation and molecular composition of water - soluble inorganic and organic nitrogen in aerosols of a coastal environment, J. Geophys. Res.: Atmos., 115, doi: 10.1029/2010JD014157., 2010.

Huang, L., Liu, T., and Grassian, V. H.: Radical-Initiated Formation of Aromatic Organosulfates and Sulfonates in the Aqueous Phase, Environmental Science & Technology, 54, 11857-11864, 10.1021/acs.est.0c05644, 2020.

Wohl, C., Li, Q., Cuevas, C. A., Fernandez, R. P., Yang, M., Saiz-Lopez, A., and Simó, R.: Marine biogenic emissions of benzene and toluene and their contribution to secondary organic aerosols over the polar oceans, Science Advances, 9, eadd9031, doi:10.1126/sciadv.add9031, 2023.