Responses to the comments

We appreciate the editor's additional comment on the manuscript. Our response to the comment and the change made to the manuscript (Supplement) are given below.

The editor's comment and our response

From the pre-print manuscript and answers to the reviewers, it is clear that 3-MBTCA and 2-methyltetrols were not the only organic compounds being determined. Glucose, pinic acid and pinonic acid were also mentioned. Please add those to Table S2 for completeness. Would any discussion point or conclusion need to be changed as a result of this requested addition?

Reply: According to the comment, the data of the three organic compounds (glucose, pinic acid, and pinonic acid) have been added to Table S2 in the Supplement. This revision will not affect any points or conclusions in the manuscript.

Comments by the referee and our responses

The authors have addressed almost all of my comments and I am content for the manuscript to be published. I would suggest a couple of changes that might be made.

Line 80 explain briefly the "blank procedure", since there are several ways to do this.

Reply 1: Four field blanks were collected with quartz-fiber filters mounted on the impactor without running the HVAS, which were obtained on the ship during the expedition. This has been additionally described in the revised manuscript (L. 80).

Line 223 I would prefer molar units, but if you use mass units please specify them more ng/m3 SO4 for example should specify if the weight is as S or SO4.

Reply 2: The mass unit is used for sulfate, because the mass concentrations are compared with those of the other chemical component (i.e., OM, etc.) in this context. Also, as we clearly mention "Sulfate (SO_4^{2-})" at the beginning of a sentence, we believe it is apparent that the concentration is for the sulfate mass rather than S. Therefore, we decided to keep them as they are.

The paper usefully demonstrates that fine mode organic aerosol composition in this area is dominated by marine probably gaseous sources. I would however, suggest that the argument about the mechanism can be clarified a bit.

Firstly in line 220 the data seems to suggest that aerosol WSOC is similar before the bloom, so are the results really bloom period specific?

Reply 3: First, the WSOC concentrations shown in Section 3.1 are those for all the data obtained during each period. Meanwhile, the difference in the WSOC concentrations of "marine origin" between the bloom (803±555 ngC m^{-3}; Table 1) and the pre-bloom (545±332 ngC m^{-3}) periods is larger than that for all the data. In addition, the contribution of sea spray aerosols to WSOC was more significant during the pre-bloom period (Miyazaki et al., 2018) compared to the bloom period in this study. Therefore, these results support our conclusion that aerosol WSOC and WSON of marine origin were likely affected by secondary formation from precursors of marine origin rather than primary emissions of sea spray aerosols.

Secondly there seems some contradiction in the suggested sources of some of the WSOC and WSON in aerosols.

In lines 325 and 370 there is discussion of proteins in seawater, but I assume the authors are not suggesting that these are volatile.

The discussion around line 335 links the seawater DOC and DON to the aerosol WSOC and WSON, but most of the DOC and DON in seawater is not volatile – indeed we know it is high molecular weight and recalcitrant- and the authors acknowledged in the response to reviewers that amines while volatile form seawater are at very low concentrations.

I would suggest the authors clarify (or remove) the mechanisms they are proposing

Reply 4: We do not intend to mention that the seawater DOC and DON compositions were preserved during their sea-to-air emissions, but some parts of them may have been affected by photodegradation and/or biodegradation in the air-sea interface to produce more volatile compounds. Although it is difficult to provide a clear explanation of the exact mechanism for the aerosol WSON formation including the processes in the air-sea interface, we have added the following statement in the revised manuscript, taking account of the referee's comment:

L. 407: ***"It is noted that the majority of DOC and DON discussed in this study are generally high molecular weight compounds and have low volatility. Therefore, photodegradation and/or biodegradation of DOC and DON in the air-sea interface are likely important to produce more volatile compounds for the atmospheric emissions, which needs further investigation in future studies."***

Reference

Miyazaki, Y., Yamashita, Y., Kawana, K., Tachibana, E., Kagami, S., Mochida, M., Suzuki, K., and Nishioka, J.: Chemical transfer of dissolved organic matter from surface seawater to sea spray water-soluble organic aerosol in the marine atmosphere, Scientific Reports, 8(1), 14861, https://doi.org/10.1038/s41598-018-32864-7, 2018.