Responses to the comments of Referee#2

Comments: This is a review of the manuscript: *"Formation of marine atmospheric aerosols associated with the spring phytoplankton bloom after sea ice retreat in the Sea of Okhotsk"* by Miyazaki et al. The manuscript refers to samples collected during a field campaign in April 2021 during the bloom and post-bloom periods. The authors want to investigate the effects of marine phytoplankton emissions on the atmospheric organic aerosol composition. The results show an increase in WSON (and an associated decrease in the C:N ratio) and changes in WSOC, although bloom values are very similar to pre-bloom values from a previous study.

While the field of research is extremely interesting, I am afraid that the experimental set-up is not ideal, specifically because a pre-bloom period should have been included to better understand the effects of phytoplankton on altering the molecular composition of OA. Considering the limited number of days investigated (six) during the bloom-decay period, who can tell if the observed changes in WSON and WSOC are related to phytoplankton activity or simply internal variability (e.g., changes in air-mass sources)?

On top of that, I think that the manuscript suffers from key methodological issues: the authors used sonication to extract their organics. However, it is known that organics can degrade during sonication. Did the authors can prove that degradation of the targeted organic molecules is absent? Also, where the filter punches were located before ultrasonication? In plastic vials? How were they cleaned? This is critical as ultrasonication can release impurities from the vial walls if not properly cleaned. How many blanks were collected? How they looked like (this holds for both inorganic and organic species)? Other missing methodological details include: a) at which temperature and vacuum the rotary evaporator was operating? b) How the method for organics was validated (i.e., what is the recovery and reproducibility of the extractions)?

More general and specific comments are reported below together with some suggestions that the authors can use to improve their manuscript.

Overall, my recommendation is **major revisions** and reconsideration for publication when the concerns are properly addressed.

Reply 1: We appreciate the referee's valuable comments on our work. We provide a point-by-point response to each comment/question raised above as follows.

-While the field of research is extremely interesting, I am afraid that the experimental set-up is not ideal, specifically because a pre-bloom period should have been included to better understand the effects of phytoplankton on altering the molecular composition of OA. Considering the limited number of days investigated (six) during the bloom-decay period, who can tell if the observed changes in WSON and WSOC are related to phytoplankton activity or simply internal variability (e.g., changes in air-mass sources)?

Reply 2: We have the aerosol data obtained during the pre-bloom period in the Oyashio region (Miyazaki et al., 2018; 2020), a nearby oceanic region of the southern Sea of Okhotsk. Taking account of the comments, we have provided the major aerosol chemical parameters of "marine origin" observed during the pre-bloom period to compare them with those during the bloom and bloom-decay periods of this study in the revised manuscript (as Table S1). At least, the differences in the WSON concentration, WSOC:WSON ratio, and Chl *a* concentrations in surface seawaters between the pre-bloom and bloom periods were significant ($p < 0.05$), supporting our conclusion in this study. Also, we made an additional statement on this in the revised manuscript.

L.320: "*..Furthermore, the WSON concentration (WSOC:WSON ratio) was higher (lower) ($p < 0.05$) than those during the pre-bloom period in the Oyashio region (Miyazaki et al., 2018; 2020), a nearby oceanic region of the southern Sea of Okhotsk (Table S1)*."

- On top of that, I think that the manuscript suffers from key methodological issues: the authors used sonication to extract their organics. However, it is known that organics can degrade during sonication. Did the authors can prove that degradation of the targeted organic molecules is absent?

Reply 3: Please see our Reply 22 to the duplicated comment below.

-Also, where the filter punches were located before ultrasonication? In plastic vials? How were they cleaned?

Reply 4: The filter punch was located in glass and plastic vials for the analysis of organic and inorganic species, respectively. All the vials were cleaned with detergent, followed by being washed with distilled water. The glass vials were then baked at 410℃ for two hours to remove VOC contaminants prior to the extraction process. The following statement has been added to the text in the revised manuscript:

L.80: ***"The filter punch was located in glass and plastic vials for the analysis of organic and inorganic species, respectively. All the vials were cleaned with detergent and then washed with distilled water. The glass vials were then baked at 410 °C for two hours to remove contaminants prior to the extraction process."***

-How many blanks were collected? How they looked like (this holds for both inorganic and organic species)?

Reply 5: Blank filters were collected four times on the ship during the expedition. The blank value was calculated from their average mass, which accounted for the mass of <30% of WSOC and WSTN, <27% of inorganic ions, and <3% of the organic compounds measured in this study. All the mass concentrations were calculated by subtracting the original values from these blanks. The statement on the blanks is now additionally made in the revised manuscript (L.80, 120, 127, 168).

-a) at which temperature and vacuum the rotary evaporator was operating?

Reply 6: The rotary evaporator was run at 25°C, and the gauge on the vacuum pump read 80 mmHg. This sentence is added to the text in the revised manuscript (L.158).

- b) How the method for organics was validated (i.e., what is the recovery and reproducibility of the extractions)?

Reply 7: The recovery of each organic compound is higher than 81% for all the compounds measured, and the reproducibility of the extraction is <13%. This information is added to the text in the revised manuscript (L.167).

Further responses to the specific comments and details of the changes made to the manuscript are given below.

General comments:

- The authors used an HVAS and an Andersen to collect atmospheric aerosol samples. However, it is not clear in the text which filters were analyzed for what.

Reply 8: Basically, the analytical results of the bottom filter samples (submicrometer aerosol samples) obtained by HVAS are shown throughout the manuscript, whereas the results obtained by the Andersen impactor are only shown in Fig.7 (the 1st half of section 3.3) to emphasize the importance of the submicrometer size range in terms of secondary formation of WSOC in this study. This statement has been now added to the text of the revised manuscript as follows:

L.92: "*In this study, the results of submicrometer particles collected by the HVAS are mainly shown, whereas the results of aerosol particles collected by the cascade impactor are presented only to show the size distributions of water-soluble fractions of organic matter (section 3.3)*."

- Comparisons between averages were done mainly qualitatively. However, differences must be either significant or not significant with associated p-values. I suggest the authors implement the appropriate statistical tests to discuss differences between averages throughout the entire manuscript. Also, p-values must be provided for all the correlation values discussed in the text.

Reply 9: According to the comment, we have just provided p-values for all the relevant values to discuss each difference in the text. Further details are given in each reply below.

- From my understanding at L138-140, the authors say that they analyze oxidation products of a-pinene (3-MBTCA, pinic acid and pinonic acid) and of isoprene (2-methyltetrols). Also glucose was mentioned, but not analyzed. Why do they only discuss 3-MBTCA and 2-methyltetrols?

Reply 10: In this manuscript, a representative compound of each tracer is shown. 3-MBTCA and 2-methyltetrols are shown as representative compounds of oxidation products of α-pinene and isoprene, respectively. Glucose was not shown here as its characteristics were similar to those of sodium, which serves as a tracer of primary emission from the sea surface. Taking into account the comments, the compounds for which the results are not shown in the manuscript (i.e., pinic acid, pinonic acid, and glucose) have been deleted from the text.

- There is a lot of discussion regarding WSOM, but not much regarding the WIOM. Whether I can understand the reason why WSOM increases during the bloom period (more oxidation of volatile precursors), why WIOM increases during the bloom-decay period (almost doubling)?

Reply 11: As the referee pointed out, the increase in the WIOM concentration was observed during the bloom-decay period, particularly on April 23–25 (Fig. 3a). Considering the large contribution of marine source based on the stable carbon isotope ratios, one possible explanation for the increase is that the contribution of sea spray aerosol to WIOM was likely significant compared to that during the bloom period. This is partly supported by the increase in the Na^+ concentration, which ranged from ~60 to 300 ng m^{-3} during the corresponding period (Fig.S2a), although a positive relationship between WIOM and Na^+ concentrations is not clear due to the limited number of samples.

- I don't fully agree with the conclusions: "the current study highlights the importance of this atmospheric formation process of OM originating from the sea surface after ice melting in the subarctic region". How, in the absence of data regarding the pre-bloom period? I see that isotope analyses and correlations with MSA can be supportive of this conclusion, but at the same time we do not have a clear chemical characterization of the conditions before the bloom.

Reply 12: Please see our Reply 2 to the duplicated comment above.

Specific comments:

L17-18: I would downsize the emphasis of this sentence: "Relations between WSOC and molecular tracers suggested that the majority of WSOC of marine origin was affected […] instead of primary emissions of sea spray aerosol". Here the authors based their interpretation on a linear correlation between WSOC and two molecules (3-MBTCA and MSA). Considering that WSOC is composed of several hundreds of different compounds, I think that this is a bit over interpretation.

Reply 13: According to the comment, we have weakened the statement as follows:

L.17: "*Relations between water-soluble OC (WSOC) and molecular tracers suggested that aerosol WSOC of marine origin was likely affected by secondary formation from precursors such as α-pinene and DMS-relevant compounds rather than primary emissions of sea spray aerosols.*"

L27–28: I would reformulate the sentence. Oceans are sources of volatile organic compounds, not of "secondary organic aerosols," which indeed form in the atmosphere through reaction with atmospheric oxidants.

Reply 14: Taking into account the comment, the sentence has been revised as follows:

L.27: *"Ocean surfaces are a significant source of atmospheric organic aerosols (OAs), which are formed by direct emissions of particles (i.e., sea-spray aerosols) or by volatile organic compounds (VOCs) followed by particle formation (i.e., secondary OAs)."*

L65: could you also add a sea-ice map for the corresponding year, showing where the sea-ice was and at which concentration (March-April would be enough).

Reply 15: According to the comment, maps showing the temporal change in the sea-ice extent in the Sea of Okhotsk from March to April 2021 (every 10 days) have been added as Fig. S1 in the Supplement.

L68–69: I would put letters a, b, c in the panels of Figure 1 and refer to Fig. 1a etc. in the main text.

Reply 16: The letters a, b, and c have been inserted in Figure 1, and they are referred to as Fig. 1a, 1b, and 1c in the text as suggested.

L80: Can the authors better clarify which sizes the nine-stage size-segregated aerosol samples refer to?

Reply 17: The size ranges of aerosol particles collected by the Andersen impactor are now shown in section 2.2 of the revised manuscript (L.86).

L70–87: It is not clear to me which aerosol fractions were analyzed for what, as in the results the authors do not refer to any specific aerosol fraction. Also, from which fractions were the organic tracers MSA, 3-MBTCA, 2-MTL, and Na+ analyzed? Based on what the authors write at L101 ("submicrometer sample"), I guess these analyses were done from the HVAS filters, but I would appreciate better clarification.

Reply 18: Throughout the manuscript, we focused on the analytical results obtained from the bottom stage of the impactor, which collected submicrometer particles. The only exception is Figure 7, which shows the mass size distributions of WSOC and WSON across all size ranges to support the discussion on the secondary formation of those parameters. In the revised manuscript, we have clearly mentioned this point as follows:

L.76: "…*we focused on the analytical results obtained from the bottom stage of the impactor, which collected particles with an aerodynamic diameter (Dp) of < 0.95 μm and are referred to as submicrometer particles*." …

L.92: *"In this study, the results of submicrometer particles collected by the HVAS are mainly shown, whereas the results of aerosol particles collected by the cascade impactor*

are presented only to show the size distributions of water-soluble fractions of organic matter (section 3.3).”

Also in the captions of Figures 5, 6, and 9, the words “submicrometer aerosols” have been added.

L88–L95: The authors claim that they did sampling at the surface and at 5-m depth and it seems that they report surface values for DOC and DOM and 5-m depht values for phytoplankton taxa. Why this choice?

Reply 19: All the DOC, DON, and Chl *a* concentrations were measured using the same seawater samples obtained either from the sea surface or at the 5-m depth, which are defined as “surface seawater”. This is to secure a certain amount of data shown in this study.

L91: with which acid? Which concentration? For how long?

Reply 20: The bucket was precleaned with 10% (v/v) hydrochloric acid and then washed with Milli-Q water. This procedure was done for a while, not all night. This information is now additionally described in the revised manuscript (L.99).

L113: “another cut of filters”, not clear which particle size is taken into account.

Reply 21: The sentence has been revised as “*Another cut of filters for each submicrometer aerosol sample (28.3 cm^2) as well as samples obtained by the Andersen impactor (3.14 cm^2) was extracted...*” (L.123)

L115 and L140–143: Ultrasonication is usually not good for the analysis of organics as they can decompose. Can you please provide some sort of evidence that the targeted organic species are not influenced by sonication? For example, you could test pure water spiked with a known amount of targeted compounds and test it over different sonication times (5–10–20 min). It would be even better if you still have a filter and you can compare extraction just with shaking or with sonication at different times.

Reply 22: We conducted an extraction experiment of the target organic compounds from the sample filter under different conditions: a) extraction just with shaking for a few seconds, or ultrasonication for b) 5 mins, c) 10 mins, and d) 20 mins. The results for two representative compounds, namely 2-methyltetrols and 3-MBTCA, are shown in the figure below.

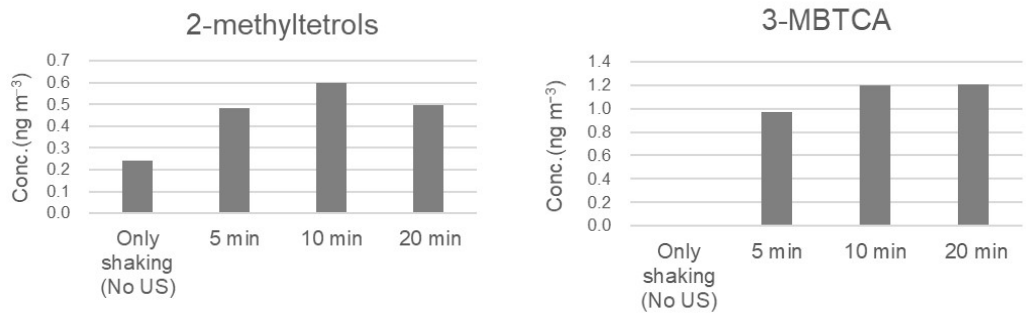

Fig. Concentrations of 2-methyltetrols and 3-MBTCA derived from the sample filters under different conditions of extraction: only shaking with no ultrasonication (US), 5 mins, 10 mins, and 20 mins of US.

Ultrasonication is an important procedure for the complete extraction of target compounds from a sample filter. As evidence, extraction with just shaking and ultrasonication for a short time (5 mins in this case) resulted in the lower concentration measured, which suggests incomplete extraction rather than decomposition of compounds. At least the results did not show the effect of decomposition of the target compounds. Based on this experiment, we believe that ultrasonication for 10 mins (as our method) is an appropriate condition to extract the compounds in our study.

L129: Filter cut from HVAS? Andersen? Please specify here and elsewhere.

Reply 23: It is a filter cut from the HVAS. In the revised manuscript, it is now clearly mentioned in that part (L.142) as well as in all the other parts (L.109, 122, 132, 154, 172).

L139–141: As far as I can understand, the authors report that tracer compounds such as 3-MBTCA, pinic acid, and pinonic acid were analyzed. I was wondering why only 3-MBTCA is reported and not pinic acid and pinonic acid. Ratios between pinic acid and 3-MBTCA could also be particularly interesting from an atmospheric chemistry point of view to investigate how atmospheric aging proceeds.

Reply 24: As we explained in Reply 10, a representative compound of each tracer is shown in this manuscript. 3-MBTCA and 2-methyltetrols are shown as representative compounds of oxidation products of α-pinene and isoprene, respectively. In fact, the temporal variations in the concentrations of 3-MBTCA, pinic acid, and pinonic acid were similar, and 3-MBTCA is shown as a representative compound of oxidation products of α-pinene. Also, because ratios of pinonic acid (or pinic acid) to 3-MBTCA did not show any statistical difference between the bloom and bloom-decay periods, we did not include pinic acid or pinonic acid in our manuscript. As we mentioned in Reply 10, the compounds for which the results are not shown in the manuscript (i.e., pinic acid, pinonic acid, and glucose) have been deleted from the text.

L151: Which internal standard? At which concentration? Was an internal standard also added before rotary evaporation to assess any potential loss of the analytes or was it only used to monitor instrument performances?

Reply 25: As the internal standard, *n*-Tridecane (C_{13}) with a concentration of 1.43 ng μl^{-1} in hexane was added just before the injection into GC-MS to determine the mass of target compounds, considering the status of the instrument. We have just added this information to the text as follows:

L.163: ***"After the derivatization, the derivatives were diluted with hexane containing the internal standard (n-Tridecane (C_{13}) with concentration of 1.43 ng μl^{-1} in hexane) and then injected into a capillary gas chromatograph (GC8890, Agilent) coupled to a mass spectrometer (MSD5977B, Agilent)…"***

L203-204: What can be the interpretation of these sulfate values? Can be a contribution from fossil sources a reason?

Reply 26: Based on the stable carbon isotope ratios together with the positive correlation between WSOC and sulfate, the increase in sulfate concentration was

attributable to marine sources (i.e., DMS) rather than anthropogenic sources such as fossil fuel combustion. Indeed, the concentration of sulfate showed a positive correlation with that of MSA (R^2 =0.61; $p < 0.05$) during the entire period, which supports this explanation.

L206: what is the correlation coefficient between sulfate, and OC and WSOC? This information should be added to add consistency to the discussion.

Reply 27: As the referee pointed out, the coefficients of determination for sulfate vs. OC (R^2 = 0.60; $p < 0.05$), and sulfate vs. WSOC (R^2 = 0.45; $p < 0.05$) have been added to the text (L.228).

L224–225: While the isotope analyses are undoubtedly interesting, I would integrate also some back-trajectory studies in order to corroborate these findings for both investigated periods (e.g., 72 hrs). This would provide additional evidence to the isotope results in clearly showing the air-mass provenance, and proving that changes in WSON are associated to changes in the marine environment and not to changes in air mass provenance.

Reply 28: As the referee suggested, we added representative backward trajectories for the bloom and bloom-decay periods (as shown in Figure S2) to illustrate the origins of the typical air masses transported in this study. The backward trajectory frequencies showed that air masses with frequencies >40% were indeed transported or originated over the southern Sea of Okhotsk, with minor contributions (e.g., <20%) from land surfaces, such as Hokkaido and eastern Eurasian continent. This supports the results of stable carbon isotope ratios in this study.

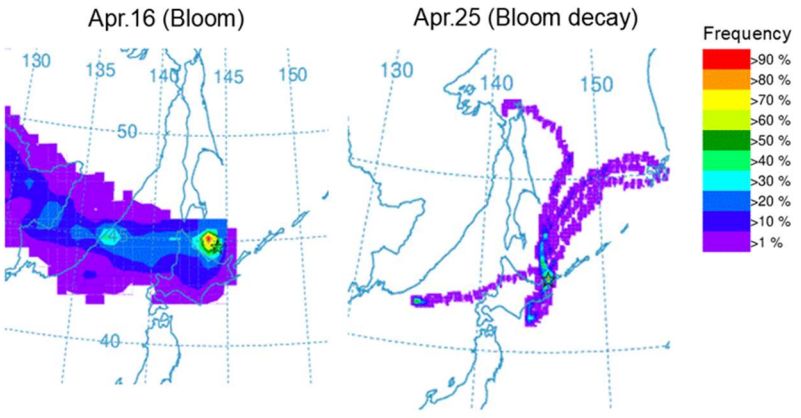

Figure S2: Typical 48-hour backward trajectory frequencies calculated from the sampling points for the bloom and bloom-decay periods.

The corresponding statement on the trajectory has been added to the text.

L.257: "*To support the source apportionment, Fig. S2 shows typical 48-hour backward trajectory frequencies calculated from the sampling points for the bloom and bloom-decay periods. The backward trajectory frequencies showed that air masses with frequencies >40% were indeed transported or originated over the southern Sea of Okhotsk, with minor contributions (e.g., <20%) from land surfaces, such as Hokkaido and eastern Eurasian continent. The trajectory supports the results of $\delta^{13}C$ values in this study.*"

L227–232: I would probably avoid a pie chart considering the great variability of the dataset. Discussing average values without their standard deviation doesn't say much about whether the observed differences are actually significant.

Reply 29: We believe that the pir charts shown in Fig. 4 are still useful for identifying the average fraction of chemical components shown in Fig. 3a, so we have decided to keep them.

For Fig.4, what we intend to mention is:

1) **WSOM was dominant OM during the bloom period (75.8±15.0%; $p < 0.05$).**
2) **The difference in the OM/submicrometer mass ratio during the bloom (48.5±13.2%) and bloom-decay (44.0±11.5%) periods was insignificant ($p = 0.23$).**

As the referee pointed out, we have added the standard deviations to indicate statistical significance in the corresponding statement as follows:

L.252: *"For aerosols of marine origin, OM accounted for 48.5±13.2% and 44.0±11.5% of the submicrometer aerosol mass during the bloom and bloom-decay periods, respectively, where the difference is insignificant ($p = 0.23$). In particular, WSOM was dominant OM (75.8±15.0%; $p < 0.05$) in submicrometer aerosol during the bloom period;.."*

L243 and L250 and L257: when the authors mention the word “significant” or “insignificant” they should also provide a p-value.

Reply 30: Among the lines, the word “insignificant” was used only in L.243, but not in L.250 and 257 in the original manuscript. Because p-values were 0.54 and 0.12, we modified the sentence as follows:

L. 271: *“The WSOC concentrations did not show positive correlations with Na^+ concentrations (R^2 = 0.05 with p = 0.54 during the bloom period and R^2 = 0.30 (negative correlation) with p =0.12 during the bloom-decay period).”*

L249–252: It would be nice to see scatter plots of the correlations between WSOC and MSA and 3-MBTCA as well as the time series. Also, this sentence should be downsized: a simple correlation with WSOC and MSA, while suggesting a process, cannot really be generalized in the way the authors claimed (“greatly affected”). As I wrote elsewhere, organic aerosol is constituted up to several thousands of compounds.

Reply 31: According to the comment, scatterplots of WSOC vs. MSA and WSOC vs. 3-MBTCA have been added to the supplement as Fig. S4. In addition, the sentence has been revised (downsized) as follows:

L. 279: “*The WSOC concentration showed positive correlations with those of MSA (Fig. S4a; R2 = 0.62 and 0.73 (p < 0.05) during the bloom and bloom-decay periods, respectively), suggesting that WSOC, which dominated the OC mass, was affected by the secondary production through the oxidation of DMS or DMS-relevant precursors.*”

L259–L261: I believe that this sentence should be downsized, as it is very hard to say if the lack of correlation with methyltetrols can really be associated with a lack of isoprene emissions from the ocean water. While being a tracer for isoprene emissions (I agree), it should be acknowledged that organic aerosols can consist of up to thousands of different compounds.

Reply 32: At least the results indicate that the observed WSOC was more influenced by the emissions of α-pinene or DMS rather than primary sea spray emissions based on the differences in each coefficient of determination. Taking account of the comment, we have modified the sentence as follows (L.289): “*The overall results*

***suggest that the observed ocean-derived WSOC was affected by secondary formation from DMS-relevant compounds and α-pinene rather than primary sea spray emissions*.”**

L262–263: “higher” and “lower” should be avoided. Were the differences in MSA and 3-MBTCA significant or not?

Reply 33: The differences in the MSA and 3-MBTCA concentrations were insignificant between the bloom and bloom-decay periods (p = 0.16 and 0.24, respectively). We modified the statement as follows:

L. 292: *“The average MSA and 3-MBTCA concentrations during the bloom-decay period were similar to those during the bloom period (Table 3), where the differences are insignificant (p = 0.16 and 0.24 for MSA and 3-MBTCA, respectively).”*

L264: The authors mention “increased sunlight intensity.” Can the authors provide information about radiation in the area during the investigated period (e.g., from reanalysis products) to prove this claim?

Reply 34: The average direct solar radiation measured at Abashiri, a coastal city in the southern edge of the Sea of Okhotsk, showed that the intensity during the bloom-decay period (24.7±14.4 MJ m^{-2}) was indeed higher ($p < 0.05$) than that during the bloom period (11.1±7.84 MJ m^{-2}) (Japan Meteorological Agency). We believe that the difference in the solar radiation intensity between the two periods supports our measurement result. In the revised manuscript, this statement has been added to the text:

L.296: *”Indeed, the average direct solar radiation measured at Abashiri, a coastal city in the southern edge of the Sea of Okhotsk, showed that the intensity during the bloom-decay period (24.7±14.4 MJ m^{-2}) was indeed higher ($p < 0.05$) than that during the bloom period (11.1±7.84 MJ m^{-2}) (Japan Meteorological Agency).”*

L351–352: Could a correlation between WSON and Na help in discerning between direct emissions of N-containing compounds or secondary organic aerosol formation?

Reply 35: Yes, the R^2 value between WSON and Na^+ concentrations was below 0.01 (p = 0.05) during the bloom, as expected from the relationship between WSOC and Na^+ concentrations. This suggests that the primary emission of sea spray made a minor contribution to WSON, supporting the large contribution of SOA to WSON in this study. We have made an additional description of this in the revised manuscript.

L.334: "*In this study, the R^2 value between WSON and Na^+ concentrations in submicrometer aerosols was below 0.01 (p = 0.05) during the bloom, as expected from the relationship between WSOC and Na^+ concentrations. This also suggests the minor contribution of primary emission of sea spray to WSON, and the major contribution of SOA to WSON in the current study.*"

L365–366: Please provide statistics in support of this claim. To me, it appears that WSOC and WIOC are influenced by great internal variability, meaning that simply comparing averages can be misleading (unless a statistical test supports the claim).

Reply 36: In the revised manuscript, the p-value ($p < 0.05$) for the difference in the WSOM:OM and WIOM:OM has been provided to support the statement (L.254).

L368–370: Could you please provide the uncertainties associated with OM? Are these values significantly different?

Reply 37: The measurement uncertainty of WSOC was below 8% and the calculated uncertainty of WIOC was below 13%. Because we used empirical factors to derive WSOM and WIOM, the uncertainty of OM is larger than each uncertainty. As we wrote in the original text as well as in Reply 29, the difference in the OM:submicrometer mass ratio between the bloom and bloom-decay periods was insignificant (p = 0.23).

L370–372: I would water down this sentence. Considering that OA is composed of thousands of different compounds, I think it is too generalizing to attribute sources of WSOA only based on correlations with three chemical species.

Reply 38: At least a lack of correlation between WSOC and Na^+ concentrations, together with the correlation with MSA and 3-MBTCA, suggests the larger

contributions of secondary formation to WSOC rather than primary emissions of sea spray aerosols. Taking into account the comment, we have modified the sentence as follows:

L.410: ***"Correlations between concentrations of WSOC and those of molecular tracers suggested that the ocean-derived WSOC observed in this study was likely affected by secondary formation rather than primary emissions of sea spray aerosols during the study period."***

L373: "significantly lower": how did you justify this "significantly"?

Reply 39: In the revised manuscript, we added $p < 0.05$ for the difference in the WSOC:WSON ratios between the bloom and bloom-decay periods, together with their average values (L.320).

Table 1: I would also add values referring to WSON and C:N ratios.

Reply 40: The average values of WSON concentrations, WSOC:WSON, and DOC:DON ratios are added to Table 1 as suggested.

Table 2: "terretrial" is a typo.

Reply 41: It has been corrected.

Figure 3d: could you consider the possibility to add above -22 something like: "marine sources", and below -22 "terrestrial sources".

Reply 42: According to the suggestion, the terms "marine sources" and "terrestrial sources" have been added to Figure 2 with arrows to indicate their ranges.

References

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.

Miyazaki, Y., Suzuki, K., Tachibana, E., Yamashita, Y., Müller, A., Kawana, K., and Nishioka, J.: New index of organic mass enrichment in sea spray aerosols linked with senescent status in marine phytoplankton, Scientific Reports, 10(1), 17042, https://doi.org/10.1038/s41598-020-73718-5, 2020.