

**We appreciate the referee's valuable comments on our work. Our responses to the specific comments and details of the changes made to the manuscript are given below.**

**Responses to the comments of Referee #1:**

*Comment 1:* The authors convincingly show that regions R1 and R2 are not affected by terrestrial air masses; however, Figure 1 (right) shows that the 5-day back trajectories for R3 all recently had crossed the continent. How can it be convincingly stated that the increase in WSOC/Na<sup>+</sup> and ozone can be attributed to aging and secondary production of aerosol from local marine organics rather than transport from other regions? Adding a vertical cross-section of the back trajectories in Figure 1 (right) might help to make this case.

***Reply 1:*** We have now added a vertical cross-section of the back trajectories in Figure 1 as suggested. The air masses observed in R3 had been transported by low-level air flow from the Atlantic. In fact, they had passed over the Isthmus of Panama at higher altitudes as evident from the new figure, followed by descent to the sampling point in R3. This is consistent with the results from the isotopic analysis of WSOC, which suggest that the influence of land surface on the observed WSOC was insignificant.

**In the revised manuscript, the following sentences have been added to the text and the caption of Figure 1:**

***(P.7, L.30–34) “It is noted that the observed aerosols in R3 had been transported by low-level air flow from the Atlantic, as indicated by the back trajectories in Figure 1. In fact, the trajectories had passed over the Isthmus of Panama at higher altitudes, followed by descent to the sampling point in R3 as seen in Figure 1, indicating less influence from the land surface. This is consistent with the results from the isotopic analysis of WSOC, which suggest that the influence of the land surface on the observed WSOC was insignificant.”***

***(Figure 1 caption) “A vertical cross-section of the back trajectories is shown in the right bottom panel.”***

*Comment 2:* Similar to Point #1, the percentage exposures given in Figure 7 and discussed on Pg. 7, Lines 8-13, seem of little value to me as even a short exposure of a given air mass to the strong aerosol emissions sources in the terrestrial mixed layer would be enough to likely overcome marine influences over subsequent days. A better treatment of the air mass back trajectories including the past horizontal and vertical transport would be more

informative here. In addition, does the model provide any information about cloud processing or rainout over the transport period?

**Reply 2:** As shown in the vertical cross-section of the back trajectories added to Figure 1, the air masses had passed over the Isthmus of Panama at higher altitudes, suggesting that the influence of the land surface on the observed aerosols was likely insignificant. This is also clearly presented in Figure 7, which shows little exposure of the observed aerosols on the land surface and is consistent with the results from the isotopic analysis. The trajectory calculation shows that relative humidity (RH) in the air mass along the back trajectories was typically within the range of 60–80%, with a maximum of 95%. This indicates that cloud processing or rainout of aerosols was likely insignificant over the period of transport in our study.

*Comment 3:* Please add error bars to compositional traces in Figures 2, 4, 6, and 8 that reflect the uncertainty associated with each measurement.

**Reply 3:** Because adding error bars to the panels, particularly in Figures 4 and 8, reduces the clarity of the figures, we decided not to add them. Instead, we have added the uncertainty of each measurement including the blank subtraction in Table 1.

*Comment 4:* Non-normal observational distributions need to be treated more carefully than just a simple arithmetic mean and standard deviation. This is apparent from the large standard deviations reported for some species in Table 1 and the non-physical result of  $90 \pm 25\%$  reported in the abstract and conclusions – the latter of which is particularly glaring. The authors should reassess the distribution of the data that go into the summary statistics and evaluate the appropriateness of geometric means and geometric standard deviations (if logarithmically distributed) or another functional form for reporting the data or, if there is not a good functional form, then median and percentile values should be reported.

**Reply 4:** Besides the mean values, we have added median and percentile values to Table 1.

*Comment 5:* The WSOC field blank concentrations are discussed on Page 3, Lines 30-31, but similar values for the speciated organic species and inorganic ion concentrations are not included. Please add these values to this paragraph. Is there any contamination associated with storing these samples in glass containers, which can leach inorganic cations? Also, please report the uncertainty associated with the  $^{13}\text{C}$  and  $^{15}\text{N}$  values in the subsequent paragraph on Page 4.

**Reply 5:** We have added the field blank levels for the inorganic ions ( $\text{Na}^+$ ,  $\text{Cl}^-$ ,  $\text{Mg}^{2+}$ , and MSA) and organic molecular tracers measured in this study, as follows:

(P.4, L.11–13) *“The MSA value of field blanks corresponded to less than ~12% of the concentrations of the ambient samples, whereas the blank values of  $\text{Na}^+$ ,  $\text{Cl}^-$ , and  $\text{Mg}^{2+}$  were less than 1% of the ambient concentrations.”*

(P.4, L.20–21) *“The values of a field blank were less than ~24% of the concentration of these molecular compounds in the ambient samples.”*

As the reviewer has pointed out, there was some contamination from the glass vials, particularly in the  $\text{Na}^+$  concentration, as evident from the blank levels. However, the ambient concentrations of  $\text{Na}^+$  in our samples were generally sufficiently high to neglect this contamination (<1%). The uncertainties associated with the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values have been added to Table 1.

*Comment 6:* I don't understand the value of Figure 6 and associated discussion on Pg. 6, Line 36 – Pg. 7, Line 7. Are the authors concluding that there is some sort of relationship between  $^{13}\text{C}$  and  $^{15}\text{N}$ ? The data do not seem to support this.

**Reply 6:** It was not our intention to discuss the linear relationship between  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ , but rather to show the ranges of the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values compared with those in seawater. In the revised manuscript, we have modified the sentences relevant to this point, both in the text and in the caption of Figure 6, as follows (P. 7, L.16–17):  
*“Figure 6 shows the ranges of the nitrogen isotope ratio of the water-soluble total nitrogen ( $\delta^{15}\text{N}_{\text{WSTN}}$ ) and  $\delta^{13}\text{C}_{\text{WSOC}}$  in the submicron aerosols for each oceanic region.”*

*Comment 7:* On Page 4, Line 34, and throughout the text, a “correlation coefficient ( $r^2$ )” is reported, which is confusing and needs to be fixed. Typically, a correlation coefficient is denoted as “ $r$ ” and a coefficient of determination is reported as “ $R^2$ ”. Which type of coefficient is being calculated and reported here?

**Reply 7:** Yes, a correlation coefficient is defined in our manuscript as “ $r$ ” and a coefficient of determination as “ $r^2$ ”. This point has been clarified in the revised manuscript.

*Comment 8:* The final line of the manuscript states that “This study provided direct evidence that the contribution of DOC was the dominant control on the submicron WSOC mass regardless of the oceanic areas over the study region.” Similar statements are elsewhere in the manuscript (e.g., Pg. 1, Ln. 30; Pg. 10, Ln. 5). While I agree that there is

indeed a compelling correlation between the concentrations of water-soluble sugars and the overall aerosol WSOC concentration and less compelling correlations with MSA and fatty acids, I do not think that this supports the strong assertion that DOC is the dominant control on submicron WSOC. This conclusion should be reworded to be more consistent with what is actually being demonstrated by this study – a “strong correlation”, not a “dominant control”.

**Reply 8:** As suggested, the final sentence in the conclusion section has been reworded as (P.11, L.12–14), *“This study provided direct evidence that the contribution of DOC was significantly correlated with the submicron WSOC mass across the study region regardless of the oceanic area.”* The other two sentences have been also modified as follows:

(P.1, L.31–34) *“The combined analysis of the  $\delta^{13}\text{C}$  and monosaccharides, such as glucose and fructose, demonstrated that DOC concentration was closely correlated with the concentration levels of submicron WSOC across the study region regardless of the oceanic area. The result implies that DOC may characterize background organic aerosols in the MBL over the study region”*

(P.10, L.29–31) *“Nevertheless, the present study demonstrates that DOC is closely correlated with the submicron WSOC aerosol concentration and implies that it may characterize background OA in the MBL over the study region.”*