

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: Page (P) 1, Line (L) 17. I do not think it is correct to associate marine aerosol WSOC to Ice Nucleating Particles in such a straightforward way. Usually, water soluble aerosol components are not considered good ice nucleators (Kanji et al., 2017) and there is no proof that WSOC contributes to the ice nucleating properties of sea-spray aerosol. On the other hand, it is likely that the ice nucleating properties of sea-spray is related to cell fragments or algal exudates (McCluskey et al., 2017; 2018; Wilson et al., 2015), which are typically insoluble (Orellana et al., 2011). I do not have the same problem with other parts of the text, like P2L3, where ice nuclei are associated to marine aerosol organics more in general (i.e., without a direct reference to the water soluble fraction).

Kanji, Z. A. et al., Measurement and Modeling Challenges, 58, 10.1175/amsmonographs-d-16-0006.1, 2017.

McCluskey, C. S. et al., Journal of the Atmospheric Sciences, 74, 151-166, 10.1175/jas-d-16-0087.1, 2017.

McCluskey et al., Journal of the Atmospheric Sciences, 75, 2405-2423, 10.1175/jas-d-17-0155.1, 2018a.

Orellana, M. et al., PNAS, 108, 3313612–13617, 2011.

***Reply 1:* We agree to the referee's comment. We did not intend to directly connect WSOC with IN, but just intended to associate WSOC to cloud formation. According to the comment, the sentence has been revised as follows (P.1, L.17):**

“...which are important factors relevant to cloud formation of aerosol particles.”

Comment 2: Sect. 2. The manuscript would benefit of some extra information. For instance, I noticed that the sampling times are not the same for day and night samples. This affects the detection limits (LOD) in terms of concentration per cubic meter. LODs will correspond to lower atmospheric concentrations in day samples than in night ones. To evaluate the impact of this difference on the reported concentrations, the authors should provide more information:

how many samples have been analyzed in total? How many are MBL, FT, wet and dry? How many samples have concentrations below LOD per each analysed species? What is the detection limit (in atmospheric concentration units) per species, in day and night samples? How the samples below detection limits have been treated to calculate the averages and st. deviations reported in Table 1?

For instance, Br⁻ represents an extreme case. Br⁻ presents always concentrations below LOD in night samples. Without more information, it is impossible to understand which of the following situation is represented by the data:

- there is a huge concentration gap between day and night samples for Br⁻
- Br⁻ concentrations are pretty similar but in night samples they fall below LOD as an effect of the lower sampling times.

Reply 2: We agree that more information about the measurements should be added, including LOD, the numbers of samples analyzed in each category, etc. The numbers of samples and LOD in each category have been provided in Table 1 of the revised manuscript. Moreover, the number of samples that showed the values below LOD are also given in each category in Table 1. If the measured concentrations were below LOD, the concentrations were treated as zero when the averages and standard deviations were calculated. The description of this has been added to the caption of Table 1. With regard to bromide, the mass concentrations in all of the FT data were below the detection limit, whereas there was indeed the difference in the concentrations between MBL and FT conditions. The additional descriptions in the caption of Table 1 (P.13) are as follows:

“...The numbers in the parenthesis show the lower detection limit of mass concentration and the number of samples that showed values below the lower detection limit (LOD) in each category. If the measured concentrations were below LOD, the concentrations were treated as zero when the averages and standard deviations were calculated.”

Comment 3: P3L10. This sentence needs revision: subject and verb are not consistent. Removing “The term” at the beginning of the sentence may solve it.

Reply 3: The word has been deleted as suggested (P.3, L.16).

Comment 4: P4L8. What does “significant” mean? This paper is based on comparing different conditions: MBL vs FT, wet vs dry season; therefore, the statistical significance of the described differences should be addressed in a quantitative way. I invite the authors to report the result of the appropriate statistic tests, when presenting and discussing these differences (number of samples in the compared subsets, confidence interval, ...).

Reply 4: According to the comment, we have now mentioned the information about the statistical tests. Regarding the difference in the water vapor mixing ratios between wet and dry seasons, “significant” means that the confidence interval of 95% for the difference is higher than 0. As stated in Reply 2, the numbers of samples in each category have been provided in Table 1. At the end of the corresponding sentence, the following statement has been added (P.4, L.25):

“..., where the confidence interval of 95% for the difference is higher than zero.”

Comment 5: P4L23-24. “The variation of the mass fractions was similar in MBL and FT in both seasons”: the meaning of this sentence is not clear.

Reply 5: The sentence has been revised as follows (P.5, L.5–6):

“The pattern of the temporal variation of the mass fractions was similar in MBL and FT in both seasons.”

Comment 6: P4L30. Maybe the difference in WSOC concentration is indicative of some level of local contamination. This is an important issue and I invite the authors to discuss it. If possible, the authors should demonstrate that their measurements are representative of uncontaminated marine conditions. For instance, Fig. 3 shows a couple of samples with important NO₃ contribution. According to my experience, NO₃ indicates potential anthropogenic influence in marine aerosol.

Reply 6: Regarding larger concentrations of WSOC in this study compared to those reported by Sciare et al. (2009), Sciare et al. (2009) observed air masses that had experienced much longer-range transport compared to the aerosols observed in our data. As is also stated in Sciare et al. (2009), atmospheric dilution induced by the long-range

transport of marine aerosols in their study resulted in the lower concentrations of WSOC and may partly explain the difference in the concentrations of WSOC between the two studies.

Indeed, two tropical cyclone events (March 13-21 and April 18-26) can explain the increased concentrations of nitrate in two samples, which was attributable to the effect of local anthropogenic sources. However, our stable carbon isotope analysis suggests that the majority of the observed WSOC originated from marine sources during this period. Therefore, possible contributions of local contamination to the WSOC concentrations are likely minor during the entire period of our study. The following statement has been added to the revised manuscript:

(P.5, L.13–15) *“Sciare et al. (2009) suggested that atmospheric dilution induced by the long-range transport of marine aerosols resulted in the lower concentrations of WSOC in their study, which may partly explain the difference in the concentrations of WSOC between the current study and their study.”*

(P.7, L.28–34) *“It is noted that a few samples showed some contributions of terrestrial source mainly explained by nitrate (Figure 7), which was attributable to the effect of local anthropogenic sources. In fact, two tropical cyclone events (March 13–21 and April 18–26) can explain the increased concentrations of nitrate in those limited samples (Figure 3), when air masses on a local scale were vertically transported rapidly to the sampling site. However, our measurement of $\delta^{13}C_{WSOC}$ suggests that the majority of the observed WSOC originated from marine sources particularly during wet seasons. Therefore, possible contributions of local contamination to the WSOC concentrations are likely minor in our study.*

Comment 7: P5L1-5. Provide a reference that links bromide emissions to sea-spray aerosol and biological productivity.

Reply 7: The following reference has been added (P.5, L.27) according to the comment.

Zhu, L., Jacob, D., Eastham, S., Sulprizio, M., Wang, X., Sherwen, T., Evans, M., Chen, Q., Alexander, B., Koenig, T., Volkamer, R., Huey, L. G., Le Breton, M., Bannan, T. and Percival, C.: Effect of sea-salt aerosol on tropospheric bromine chemistry, *Atmos. Chem. Phys.*, 19(9), 6497–6507, doi:10.5194/acp-19-6497, 2019.

Comment 8: P5L5-8. A correlation analysis between the different aerosol species would make this part more quantitative.

Reply 8: We agree to the comment. We have now added relevant number outputs of the correlation analysis as follows (P.5, L.27–30):

“The temporal variation of the concentration of MSA is similar to that of WSOC ($R^2 = 0.71$, $p < 0.01$), suggesting that the dominant source of WSOC is similar to MSA. The concentrations of 2-methyltetrol, ..., although their temporal variations are generally different from those of WSOC ($R^2 < 0.01$) with exceptions of a few samples.”

Comment 9: Sect. 3.4. From the plots in Figure 9, I noticed a weird finding, which is never discussed in the manuscript. The contribution of “Marine PA” to WSOC is higher in FT samples than in MBL ones, both during the wet and dry seasons. This result should be addressed and discussed.

First of all, this seems in disagreement with the Br^- results. Br^- was presented by the authors as a tracer for biogenic organic matter from primary sea-spray and, accordingly, it presents higher concentration in MBL samples than in FT ones. How can this result be reconciled with the PMF output?

Absolute concentrations data for the WSOC contributing sources are not presented, so it is difficult to judge properly, but I have the impression that this high PA contribution to WSOC is driven by a few samples, while on the rest of the time series the signal is almost negligible. I invite the authors to analyse this aspect more in detail. I think this is an indication of the limits of the WSOC source apportionment performed by PMF, with such a limited number of samples and analysed species. I am not discussing the general PMF results, but maybe the authors are overconfident on the WSOC source attribution potential of this approach. For instance, I notice that the most consolidated organic aerosol source apportionment technique by PMF, the one based on Aerosol Mass Spectrometry data, makes use of much larger input matrixes, both in terms of number of samples and analytic information (mass spectra peaks) to resolve the organic aerosol sources.

Considerations on these aspects should be added in the text. I do not think this spoils the work, as the WSOC source attribution is supported soundly by the carbon isotope results, but still, I would like to see the limits of the PMF clearly stated in the text.

Reply 9: The higher “average” contribution of “Marine PA” to the WSOC mass in FT than in MBL is just due to two samples which showed dominant or higher contributions of Marine PA to WSOC in FT at the beginning of both wet and dry seasons. These can be explained by two events of tropical cyclones occurred during March 13-21 and April 18-26, when marine PA was rapidly transported to FT. This Marine PA factor is mainly reflected by substantially large concentrations of sea salt (i.e., sodium and magnesium). It is difficult to interpret the lower concentrations of bromide in the corresponding samples in FT, and the source apportionment of WSOC only by PMF has a limitation with a limited number of samples as the referee pointed out. However, our measurements of stable carbon isotope ratios and FLEXPART in this study were consistent with the output of PMF. Therefore, we believe that the result of PMF is supported by the stable carbon isotope ratios and outputs of FLEXPART and is convincing. Taking account of the referee’s comment, we have made additional statements on these points in the revised manuscript as follows (P.7, L.35-40):

“It is also noted that the average contribution of marine primary aerosol to the WSOC mass was higher in FT than in MBL (Figure 9). This was attributable to the two samples which showed dominant or higher contributions of marine primary aerosol to the WSOC mass in FT at the beginning of both wet and dry seasons. These can be explained by the two cyclone events as mentioned above, when marine primary aerosols were rapidly transported to FT. Although the source apportionment of WSOC only by PMF has a limitation with a limited number of samples in the current study, the overall result is supported by the measurements of $\delta^{13}C_{wsoc}$ as well as the result of FLEXPART, which are consistent with the output of PMF.”

Furthermore, regarding bromide as a tracer in this study, the following statement has been added to the text (P.6, L.36-39):

“It is interesting to note that bromide had also large contribution to F1 (marine SOA). Bromide is expected to be recycled rather quickly via the gas-phase and redistributed among all types of aerosols on time scales of minutes to few hours after emission (e.g., Zhu et al., 2019). Therefore, primary bromide, which is emitted with sodium as a part of sea spray aerosols, is expected to appear in F1 (marine SOA).”

Comment 10: P6L33-36. This would be clearer by showing a regression analysis.

Reply 10: According to the comment, we have added some numbers by the regression analysis, namely, correlations of WSOC with pinic acid ($R^2 = 0.53$, $p < 0.01$), pinonic acid ($R^2 = 0.27$, $p < 0.01$), and 3-MBTCA ($R^2 = 0.26$, $p < 0.01$). The corresponding sentences haven't been modified as follows (P.7, L.19–21):

“It is noted that F1 had also large contributions of oxidation products of α -pinene (i.e., pinic acid, pinonic acid, and 3-MBTCA; Figure 7). The regression analysis showed that R^2 of WSOC with pinic acid, pinonic acid, and 3-MBTCA were 0.53 ($p < 0.01$), 0.27 ($p < 0.01$), and 0.26 ($p < 0.01$), respectively. This result is also attributable to local terrestrial biogenic emissions of VOCs during the transport from the ocean to the observatory.”

Comment 11: P713-14. Please provide a reference. According to my experience, “models” do not consider either marine POA.

Reply 11: We added the following reference to the revised manuscript (P.8, L.14), according to the comment. Indeed, some global models consider marine POA, but those models rarely take account of marine SOA.

Gantt, B., Glotfelty, T., Meskhidze, N. and Zhang, Y.: Simulating the impacts of marine organic emissions on global atmospheric chemistry and aerosols using an online-coupled meteorology and chemistry model, *Atmospheric and Climate Sciences*, 5, 266-274, doi: 10.4236/acs.2015.53020, 2015.

Comment 12: Figure 8. Change the colour palette. “Marine SOA” and “Terrestrial source” are too similar to each other.

Reply 12: The color of Factor 5 (Terrestrial source) has been changed (now shown in gray) to be clearly distinguished from Marine SOA. This change has been also made in Figures 7 and 9.

Comment 13: Data availability statement. To me, this seems not in line with the journal policy: “The output of research is not only journal articles but also data sets, model code, samples, etc. Only the entire network of interconnected information can guarantee integrity, transparency,

reuse, and reproducibility of scientific findings. Moreover, all of these resources provide great additional value in their own right. Hence, it is particularly important that data and other information underpinning the research findings are "findable, accessible, interoperable, and reusable" (FAIR) not only for humans but also for machines.

Therefore, Copernicus Publications requests depositing data that correspond to journal articles in reliable (public) data repositories, assigning digital object identifiers, and properly citing data sets as individual contributions. Please find your appropriate data repository in the registry for research data repositories: re3data.org. A data citation in a publication resembles a bibliographic citation and needs to be included in the publication's reference list. To foster the accessibility as well as the proper citation of data, Copernicus Publications requires all authors to provide a statement on the availability of underlying data as the last paragraph of each article (see section data availability)".

Reply 13: As long as the data will be provided according to the journal policy, we do not think that our statement is not in line with the policy. Moreover, a number of the ACP paper published even in the last one month made the same or similar statements in "data availability" (e.g., <https://doi.org/10.5194/acp-21-12443-2021>, <https://doi.org/10.5194/acp-21-12479-2021>, <https://doi.org/10.5194/acp-21-12173-2021>, <https://doi.org/10.5194/acp-21-12155-2021>, <https://doi.org/10.5194/acp-21-11815-2021>, etc.). Therefore, we believe that our statement is acceptable.

Responses to the comments of Referee #2:

General comments:

This study presents chemical analyses of submicron aerosols sampled at Maïdo observatory on La Réunion Island in the Indian Ocean during March – May 2018. This period covers part of the wet and dry season. There is a clear need for improved knowledge on the physicochemical properties of aerosols in remote locations, especially the undersampled Southern Hemisphere. I therefore support eventual publication of this manuscript in ACP, however comments detailed below should be addressed first. Overall, the manuscript is well written, and the analytical procedures appear sound, with the exception of PMF, where more details on the choice of factors should be given.

***Reply:* 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.**

Specific comments:

Comment 1: P. 1, L. 23 – 24; P. 2., L. 33 - 34: The separation into FT and MBL is usually not that simple (see e.g. Collaud Coen et al., 2018), and also the references given (Guilpart et al., 2017, seems to be missing in the reference list) state that no observational evidence is available to confirm the assumption of FT conditions at night. A short discussion and more information about the uncertainties related to the potential influence of MBL or residual layer on the nighttime samples need to be given.

***Reply 1:* To avoid the influence of residual layer on the separation between MBL and FT conditions, the daytime was defined as 0700–1800 LT and nighttime as 2200–0500 LT, where 1800-2200 LT and 0500-0700 LT were set to be transition time. Moreover, we have shown the water vapor mixing ratios in day and nighttime to support the validity of the definition. Indeed, Guilpart et al. (2017) showed a one-year long record of the isotopic composition of water vapor at the same observatory to demonstrate the validity of the assumption of FT conditions at night. Taking account of the comment, we have made additional statement on these points with the addition of Collaud Coen et al. (2018), as pointed out by the referee. Moreover, Guilpart et al. (2017) is now listed in the reference, which was missing in the original manuscript. We made additional statement in**

subsection 2.1 as follows (P.2, L.34–36):

“Although Collaud Coen et al. (2018) pointed to influence of residual aerosols in daytime on nighttime conditions at high-altitude stations, Guilpart et al. (2017) showed a one-year long record of the isotopic composition of water vapor at the same observatory to demonstrate the validity of the assumption of FT conditions at night.”

Additionally, the following statement has been added (P.3, L.8–9):

“To avoid the influence of residual layer on the separation between MBL and FT conditions, 1800-2200 LT and 0500-0700 LT were set to be transition time.”

Reference:

Collaud Coen, M., Andrews, E., Aliaga, D., Andrade, M., Angelov, H., Bukowiecki, N., Ealo, M., Fialho, P., Flentje, H., Hallar, A. G., Hooda, R., Kalapov, I., Krejci, R., Lin, N.-H., Marinoni, A., Ming, J., Nguyen, N. A., Pandolfi, M., Pont, V., Ries, L., Rodríguez, S., Schauer, G., Sellegri, K., Sharma, S., Sun, J., Tunved, P., Velasquez, P., and Ruffieux, D.: Identification of topographic features influencing aerosol observations at high altitude stations, *Atmos. Chem. Phys.*, 18, 12289–12313, <https://doi.org/10.5194/acp-18-12289-2018>, 2018.

Comment 2: P. 1, L. 26; P. 2, L. 31 – 32; P. 4, L. 13 - 26: The separation between wet and dry season is described in “methods” as “[...] wet (typically from November to April) and dry seasons (from May to October).” How do you justify the separation of your data into “dry” and “wet” with the change taking place within one day? More information needs to be given, e.g. a time series of the water vapor mixing ratios should be shown. Also, in section 3.1 you mention “significant” differences in water vapor mixing ratios at several instances, however, are the differences really statistically significant? If not please use another word than “significant”. Also, without more context and analyses the values do not prove or confirm the separation into FT and MBL. Please revise.

Reply 2: With regard to the separation between the wet and dry seasons, time series of the water vapor mixing ratio is shown below. It is clearly seen that the levels of water mixing ratio substantially decreased just after April 24. In the revised manuscript, this figure has been added as a supplement figure (Figure S1), and we believe that this justifies the definition of the two seasons. As the differences in the water vapor mixing ratio shown in the text are statistically significant where the confidence interval of 95% for the difference is higher than zero, we use the word “significantly.” This point is now

mentioned in the revised manuscript (P.4, L.24–26):

“... reflecting the significant difference in the water vapor mixing ratios ..., where the confidence interval of 95% for the difference is higher than zero. Indeed, it is apparent that the level of the water vapor mixing ratios substantially decreased just after April 24 (Figure S1).”

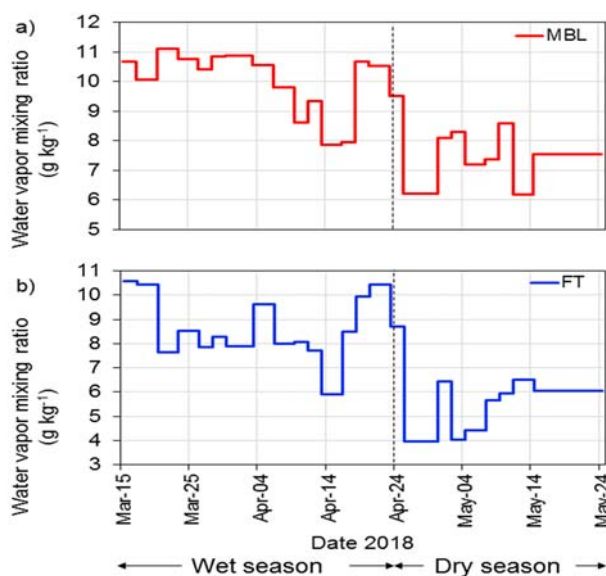


Figure S1: Time series of the water vapor mixing ratios under the (a) MBL and (b) FT conditions.

Comment 3: P. 1, L. 26, P. 5, L. 5: The outbreak of the volcano coincides with the start of the dry season. The sulfate is therefore not due to a change in season, but due to the volcano. This needs to be stated very clearly, otherwise the interpretation is very misleading.

Reply 3: According to the comment, we have now clearly described about the volcanic eruption as follows (P.1, L.25–27):

“.. during the wet season (March 15–April 23). On the other hand, sulfate dominated (~77±17%) during the dry season (April 24–May 24), most of which was attributable to the effect of volcanic eruption.”

Comment 4: P. 1, L. 33: “[...] which may be responsible for cloud formation [...]” - This is too strong of a statement, I suggest toning it down, similar for P. 7, L. 38 – 39

Reply 4: According to the comment, we have modified the statement as “*may affect cloud formation*” both in P.1 (L.34) and P.8 (L.34).

Comment 5: P. 2, L. 8: The bubble bursting process also produces particles larger than 1 micrometer

Reply 5: We do not intend to say that the bubble bursting process does not produce supermicrometer particles. As we focus on OM in SSA in this context, we intend to mention that OM is substantially included in the submicrometer particles produced by the bubble bursting process. We believe that readers can understand the meaning and the context as it is.

Comment 6: P. 3, L. 5 – 8: How were the filters stored after sampling and before analysis? Please add this information.

Reply 6: After the collection, the filter samples were stored in the glass vial with a Teflon-lined screw cap at -20°C in a freezer prior to the analysis. This information has been added to the revised manuscript (P.3, L.9–10).

Comment 7: P. 4, L. 3 – 10, section 3.4: These sections need more information for the reader to be able to understand the PMF results; e.g. how were the analytical measurement uncertainties calculated? Based on what criteria did you choose a six-factor-solution, especially given the uncertainties related to F5 and F6? You state that PMF reproduced 86% of the measured WSOC mass – what about the other aerosol components that you used as input in PMF? You state in L. 4 - 5 that you investigate the sources of WSOC using PMF, however your input also contains inorganic ions. Please clarify.

Reply 7: Analytical uncertainties were derived in each individual analysis (by including accuracy and lower detection limits) and were used as uncertainties in the PMF model. We explored PMF outputs by varying number of factors systematically, to check Q values

(i.e., values reached by the objective function which is minimized by the PMF model), distribution of residuals, physical sense of source profiles and contributions. Then we found out the most reasonable solution of 6 factors as done in many other previous studies. As the purpose of this PMF calculation was to derive source contributions to the WSOC mass, reproducibility was calculated only for WSOC. Inorganic ions were used as “tracers” of possible sources of WSOC as well as the other compounds in this study, which is a general method used in many other previous studies. We have now mentioned these points in the revised manuscript. The statement of PMF in subsection 2.6 has been revised as follows (P.4, L.13–17):

“...The analytical measurement uncertainties of each component were derived in each individual analysis and were used as uncertainties in the PMF model. The calculation was performed with 20 runs. We explored PMF outputs by varying number of factors systematically, to check Q values (i.e., values reached by the objective function which is minimized by the PMF model), distribution of residuals, physical sense of source profiles and contributions. As a result, 6 factors were the most appropriate number of factors corresponding to meaningful sources. ...”

Comment 8: P. 5, L. 1 -2: Why? Can you give some reasoning here?

Reply 8: While both our data and Sciare et al. (2009) showed the dominant contributions of marine SOA to the WSOC, Sciare et al. (2009) observed air masses that had experienced much longer-range transport compared to the aerosols observed in our data. As is also stated in Sciare et al. (2009), atmospheric dilution induced by the long-range transport of marine aerosols in their study may partly explain the difference in the concentrations of WSOC between the current study and Sciare et al. (2009). The following sentence has been added to the revised manuscript (P.5, L.13–15):

“Sciare et al. (2009) suggested that atmospheric dilution induced by the long-range transport of marine aerosols resulted in the lower concentrations of WSOC in their study, which may partly explain the difference in the concentrations of WSOC between the current study and their study.”

Comment 9: P. 5, L. 26, 29: Given the stated uncertainty range, these values are very close to

the limit value. Please comment.

Reply 9: In the wet season, while the average values of $\delta^{13}\text{C}_{\text{WSOC}}$ are close to the lower limit, the individual data with the large concentrations of WSOC ($>300 \text{ ngC m}^{-3}$) showed much larger values than the limit value. In the dry season, the average values of $\delta^{13}\text{C}_{\text{WSOC}}$ are almost similar to the limit value, which is consistent with the FLEXPART and PMF results. We have not mentioned that the average values of $\delta^{13}\text{C}_{\text{WSOC}}$ in the dry season are different from the limit value. Taking account of the comment, we have modified the sentence as follows (P.5, L.40–P.6, L.1):

“...showed the $\delta^{13}\text{C}_{\text{WSOC}}$ larger than -24% , with averages of $-23.2 \pm 1.0\%$ (MBL) and $-23.5 \pm 2.5\%$ (FT). While these average values with the uncertainty were close to -24% , the larger concentrations of WSOC (e.g., $> 300 \text{ ngC m}^{-3}$ in MBL; Figure 4a) corresponded to a higher $\delta^{13}\text{C}_{\text{WSOC}}$ ($> -24\%$).”

Comment 10: P. 5, L. 31 – 34: Give details here on the mass balance equation. Also, the $\delta^{13}\text{C}$ values you used in the equation – how well do they represent your dataset? Please discuss.

Reply 10: We added the following statement on the details about the mass balance equation in the revised manuscript (P.6, L.6–11):

“To estimate the relative contributions of marine and terrestrial OC sources to the observed WSOC, a mass balance equation (e.g., Turekian et al., 2003) was applied using the following equations:

$$\delta^{13}\text{C}_{\text{WSOC}} = F_{\text{marine}} \times \delta^{13}\text{C}_{\text{marine}} + F_{\text{terrestrial}} \times \delta^{13}\text{C}_{\text{terrestrial}}$$

where F_{marine} and $F_{\text{terrestrial}}$ are the fractions of marine and terrestrial carbon respectively, and $\delta^{13}\text{C}_{\text{marine}}$ and $\delta^{13}\text{C}_{\text{terrestrial}}$ are the reported isotopic values for marine and terrestrial carbon, respectively. A $\delta^{13}\text{C}$ value is assumed to be -21.5% for marine OC (Turekian et al., 2003; Miyazaki et al., 2010), and -28% for terrestrial OC (e.g., Cachier et al., 1986).”

Regarding the assumed $\delta^{13}\text{C}$ values, the result obtained by the mass balance equation is quantitatively consistent with the PMF results as we stated in the manuscript. Therefore, we believe that this supports the validity of the assumed $\delta^{13}\text{C}$ values.

Comment 11: P. 6, L. 38 - 39: “[...] attributable to local terrestrial biogenic emissions of VOCs

during the transport from the ocean to the observatory [...]” – unclear, how can terrestrial emissions be transported from the ocean?

Reply 11: Local terrestrial biogenic emissions of VOCs occurred in the surface slope of the island, and the aerosols of F1 were likely transported along the island slope. This is clearly mentioned in the previous sentence in the original manuscript (now in P.7, L.16–18).

Comment 12: P. 7, L. 11 – 12: I don’t understand how you come to this conclusion.

Reply 12: As we stated in the text, insignificant contribution of aged POA to the SOA mass can be explained by the low contribution of sea salt (as a tracer of marine primary emission) in the submicrometer particles. Therefore, we came to this conclusion which is supported by the PMF result.

Technical comments:

Comment 13: P. 1, L. 16 – 17: Revise sentence; CCN and IN are aerosol particles, and “important factors relevant to” therefore is unclear

Reply 13: According to the comment, the sentence has been revised as follows (P.1, L.17):
“...which are important factors relevant to the cloud formation of aerosol particles.”

Comment 14: P. 2, L. 2: The ocean is a source of aerosols, the ocean surface is simply the interface to the atmosphere. Suggest deleting.

Reply 14: According to the suggestion, the word “surface” has been deleted (P.2, L.2).

Comment 15: P. 3, L. 19: Should read “were transferred”

Reply 15: Because the subject of this statement is “40 μ L of each sample,” “was

transferred” is correct as originally stated which was confirmed by a native language check.

Comment 16: P. 4, L. 1: ECMWF: Give full name

***Reply 16:* ECMWF is an abbreviation for the European Centre for Medium-Range Weather Forecasts, which is now given in the revised manuscript (P.4, L.6) .**

Comment 17: P. 5, L. 11 – 12: “Substantially larger” – compared to what?

***Reply 17:* We intend to say that the concentrations were substantially larger in the MBL than in the FT. We have added “*than in the FT*” in the sentence of the revised manuscript (P.5, L.25).**

Comment 18: P. 5, L. 19 – 22: Give a short explanation here how this is done, and the accuracy and uncertainty range of the limit values in L. 24

***Reply 18:* The way of source apportionment using isotope ratios is now given with detailed explanation of a mass balance equation (please see Reply 10). Uncertainty of the limit value ($\pm 1.0\%$) is now given in the revised manuscript (P.5, L.39).**

Comment 19: P. 4, L. 38: “typical” for what?

***Reply 19:* We suppose that the referee mentioned P.5 (not P.4), L.38, which includes the word “typical.” In order to clarify it, the word “typical” has been replaced by “representative.” (P.6, L.16)**

Comment 20: P. 6, L. 10: “Enrichment” compared to what?

Reply 20: Enrichment means that the relative contribution to certain factor is larger compared to the other factors. We have added the following explanation (P.6, L.29–30):
“...by the enrichment of each tracer compound in a factor compared to the other factors.”

Comment 21: P. 7, L. 15: “additional” to what?

Reply 21: We intended to say additional contribution to the OA mass. As we have already described that SOA “contributes up to 60% of additional OA mass” in the same sentence, the word “additional” here has been removed (P.8, L.10).

Comment 22: Caption Figure 2: “Extreme data”?

Reply 22: The word “extreme” was wrong and the description is now corrected to
“medians and the upper/lower 25 percentile of data are shown.”

Comment 23: Figure 5: Dotted lines are not visible

Reply 23: In the original manuscript, it was mistakenly shown by dashed line. Now we have shown it by bold dotted line.

Comment 24: Figure 6: Specify 10-day period. Color scale is missing.

Reply 24: We have added the color scale to the figure according to the comment.