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 night-time 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:

"*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 in subsection 2.2:

"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."

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 rations 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. The following statements have been added in the subsection 3.1.:**

"… 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 mixing ratios substantially decreased just after April 24 (Figure S1)."

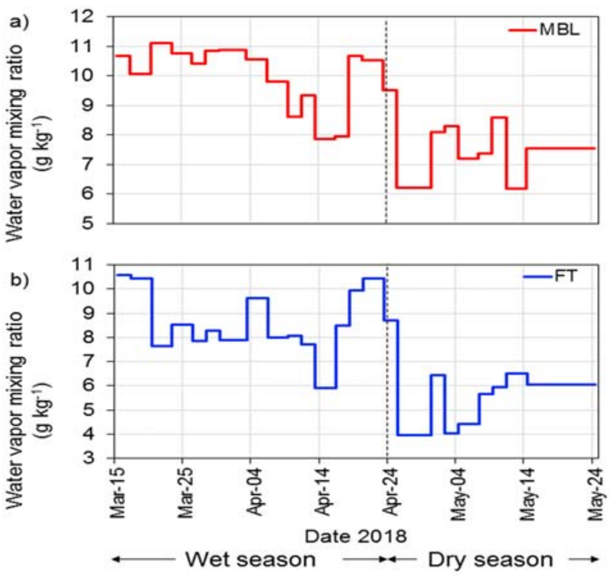

Figure S1: Time series of the water vapor mixing ratios under the (a) MBL and (b) FT conditions.

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.26): *"... 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 and P.7.**

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.**

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 2.6 has been revised as follows:**

"…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:**
"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 $\delta^{13}C_{WSOC}$ are close to the lower limit, the individual data with the large concentrations of WSOC (>300 ngC m^{-3}) showed much larger values than the limit value. In the dry season, the average values of $\delta^{13}C_{WSOC}$ 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 $\delta^{13}C_{WSOC}$ in the dry season are different from the limit value. Taking account of the comment, we have modified the sentence as follows: "*…showed the $\delta^{13}C_{WSOC}$ larger than −24‰, with averages of −23.2 ± 1.0‰ (MBL) and −23.5 ± 2.5‰ (FT). While these average values with the unceratinty were*"**

***close to −24‰, the larger concentrations of WSOC (e.g., > 300 ngC m^{-3} in MBL; Figure 4a) corresponded to a higher $\delta^{13}C_{WSOC}$ (> −24‰).*"**

Comment 10: P. 5, L. 31 – 34: Give details here on the mass balance equation. Also, the d13C values you used in the equation – how well do they represent your dataset? Please discuss.

Reply 10: **Taking account of the comment, we have added the following details about the mass balance equation in the revised manuscript:**

"*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:*

$$\delta^{13}C_{WSOC} = F_{marine} \times \delta^{13}C_{marine} + F_{terrestrial} \times \delta^{13}C_{terrestrial}$$

where F_{marine} and $F_{terrestrial}$ are the fractions of marine and terrestrial carbon respectively, and $\delta^{13}C_{marine}$ and $\delta^{13}C_{terrestrial}$ are the reported isotopic values for marine and terrestrial carbon, respectively.

***A $\delta^{13}C$ 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 $\delta^{13}C$ 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 $\delta^{13}C$ 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.**

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: "…*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.**

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

Reply 15: **Because the subject of this statement is "40 µL 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.**

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.**

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 (±1.0‰) is now given in the revised manuscript.**

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."**

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: "*…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.**

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.**

References:

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.