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, 201.

***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: "…*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.**

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.**

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: "..,** ***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: "*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 (in the subsection 3.4): "*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 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: "*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:

"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."

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 modified as follows: "*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, 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.