

We thank the Reviewer #2 for the helpful and constructive comments, which have led to significant improvements in the manuscript. We have carefully revised the text. Our point-by-point replies are given below (blue), following the referees' comments (black). Changes to the manuscript are marked with green.

Reviewer 2:

General comments

This paper presents a one-year data set of PM₁ samples obtained in Ny-Ålesund with the bulk chemical analytical results of H-NMR spectroscopy and off-line HR-ToF-AMS. The authors show that the analytical results of the two measurements are consistent in terms of source contributions to OAs. Their data suggested that the observed OAs in winter and spring are dominated by long-range transport of anthropogenic pollution in Eurasia, while the aerosol in summer is characterized by biogenic aerosols from marine sources. Overall, the paper provides new insights into our understanding of the seasonality in the source contributions to OAs in the Arctic region and confirms some of the findings already reported in previous studies. While the data presented are valuable and interesting, there are some issues that need to be clarified before its publication in ACP.

Response: We appreciate the general Reviewer's positive feedback on the manuscript relevance.

Specific comments

(1) From the text, the contributions of terrestrial BSOA traced by oxidation products of terpenes are not clear. The authors attributed these BSOAs to emissions from wildfires but this is not always the case. Indeed, Moschos et al (2022) reported that significant or non-negligible amounts of BSOA (not necessarily related to biomass burning) from forests for the observed OA in pan-Arctic in summer. I think that the authors should add more discussions on this point (e.g., if the author's result is different from Moschos et al., why?).

Response: we thank the Referee for the careful examination and suggestion. We decided to attribute wildfires as main source for F2 because of a combination of its spectral features (HULIS features for NMR, many oxygenated fragments at high m/z for AMS), resembling very aged continental biomass burning emissions, and its temporal/geographical pattern (sporadic very high concentrations corresponding to fire events). But we wanted also to spot out that in NMR spectra of specific samples associated to F2, we see signals related to BSOA for which in any case the PMF was not able to separate a specific factor in NMR nor in AMS. So, we cannot separate and quantify a specific BSOA contribution within our dataset. We interpreted it as the result of a strong co-variation between the BSOA signals and the others signals of F2 more characteristics of aged BB. We made the hypothesis that what we sporadically traced at the receptor site of GVB were the BSOA co-emitted during fire events (characterized by higher temperature/convection facilitating the subsequent transport to long distances). The alternative hypothesis (also considered plausible and reported in the main text) is that BSOA signals identified in our dataset represent oxidation products of forest emissions (terpene and isoprene) moving to Ny-Alesund from the same area (boreal forests in Eurasia), contributing to a variable fraction of our F2 but also to F4 (Background mix) that shows some biogenic terrestrial signals as well and has a slight increase during polar-day months (Fig. 4).

Given these considerations, to answer the Referee, we add that:

1) the overall picture that emerges from our hypotheses and descriptions is not actually inconsistent with that of Moschos et al. (2022): in Moschos during the polar day for both Gruebadet (9%) and Zeppelin (19%), bioSOA is a minor contributor to OC in PM10. We believe that in Moschos' study the Authors managed to separate such a specific BSOA factor because there were included stations from the continents (e.g. Pallas), where bioSOA is a major contributor. Whether the attributed concentrations (9-19%) are that reliable in Svalbard or not would probably need more future confirmations. In any case the picture emerging in our study is consistent with a portion of 9-19% of BSOA included partially in F2 and/or in F4 (representing up to 28% and up to 34% of organic PM1, for F2 and F4 respectively, as summertime averages).

2) the time series, air mass origin and spectral features of F2 show pretty clearly that aged fires are the main source attributable to this factor, supporting our interpretation. Compounds identical/similar to bioSOA can also be emitted during fires and we very likely traced them in F2.

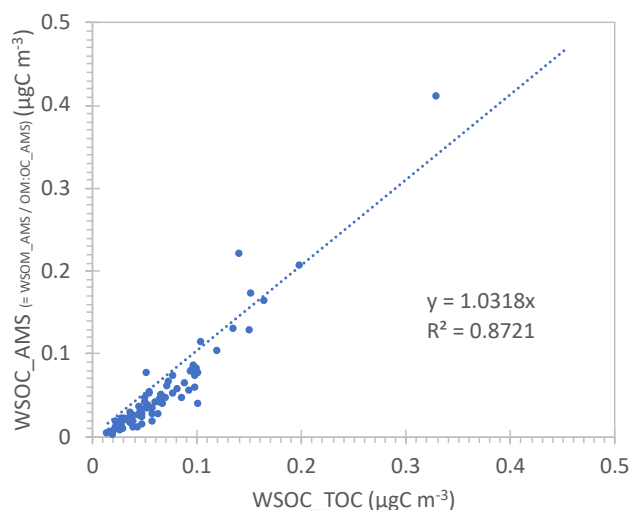
3) we agree with the Referee that we should explicitly refer to Moschos et al. findings in our discussion and we should clarify better the possibility that BSOA can contribute partially also to F4. Aiming these objectives, we added few sentences in Section 3.3 and Section 4.

(2) P.7, L.215: $WSOM = WSOC \times (OM:OC)_{AMS}$

I understand that the advantage of the use of OM:OC at the time of each sampling is to be able to expect more realistic abundance of WSOM rather than by use of a constant value of the factor. Meanwhile, the composition of OM minus OC can also include water-insoluble compound mass. How can this use of OM:OC ratio be verified (or is there any evidence) to represent water-soluble mass? The authors should add some more description including uncertainty in this calculation.

Response: given that AMS analyses were done off-line on water-extracts of the filters (as clearly explained in Section 2.2.2), the OM:OC ratios refer to the water-soluble fraction of OA by definition. We do not believe it is necessary to add more explanations/evidences of that in the text.

Nonetheless, an evidence of the consistency of OM:OC ratios is the fact that if we apply OM:OC ratios to the WSOM mass measured by AMS (i.e., $WSOC_{AMS} = WSOM_{AMS} / OM:OC_{AMS}$), the resulting $WSOC_{AMS}$ corresponds quite well with the WSOC measured by TOC-analyzer (as shown in the following plot for the Referee)



(3) P.10, L.295: “The major chemical mass ..., followed by..”

I think that this statement may cause misunderstandings of readers: some may think that sulfate was the most abundant and the second most is seasalt, followed by OM. However, there seems to be no statistical difference in the fraction among sulfate, seasalt, and OM. As the authors described in the conclusion section, these three components had similar contributions to the PM1 mass. Please modify the sentence.

Response: we thank the Referee for the suggestion. We rephrased the sentence accordingly, as follows:

“On yearly average the PM1 was mainly constituted in similar proportions of nss-sulfate (representing $33 \pm 13\%$ of the total), seasalt ($29 \pm 13\%$) and OM ($28 \pm 16\%$, of which $22 \pm 14\%$ represented by WSOM), the rest being accounted for by much smaller contributions of ammonium ($4 \pm 2\%$), nitrate ($1 \pm 1\%$), eBC ($2 \pm 2\%$) and other non-sea salt ions (i.e., nss-K, nss-Mg and nss-Ca, amounting to $3 \pm 4\%$ in total).”

(4) P.11, after L. 329: Regarding the statement starting with “MSA and ...,” which figure is referred to? Maybe Fig. 4? Please clarify it.

Response: no, it is referred to Figures 3 and S4, as mentioned on the line above. We added in the revised version more references to the proper Figures along the description.

(5) Figure 5: This figure is very hard to see. For example, the color code is not clear what it represents. The authors explain it in the caption with quantitative information, but they should show the color code in the figure panel in addition to describing it in the caption. Moreover, geographical lines (map) in the figure are not clear at all.

Response: we thank the Referee for the suggestion. We modified the Figure accordingly, adding the color scale to each map and changing colors of the continental edges lines in order to improve readability.

(6) Figure 6 and P.16: As the author described, the scaled contributions of the NMR and AMS factors to total OC showed generally good agreement. However, the relative contributions of background OA, Arctic haze OA, and Aged wildfires OA between NMR and AMS particularly in spring are significantly different. The authors should add more discussion on the possible reason for this difference.

Response: we already openly acknowledged in the text the discrepancies and attributed them mainly to the different sensitivity of the two techniques to specific classes of organics present in the complex mixtures of compounds that constitute OA; for instance, NMR seems to be more sensitive to fatty acid chains attributing more mass to marine POA (see Fig.4), while it "misses" something of the aged continental BBOA and the Arctic haze compared to AMS, which are enriched of branched polysubstituted oxygenated species. This depends on the fact that actually H-NMR is measuring directly the resonances of hydrogen atoms (-H) of functional groups (meaning specific kind of bonds and molecular structures) bearing not-exchangeable protons, like aliphatic -CH₂ chains of fatty acids, but cannot quantify directly carbon atoms not protonated (or bearing exchangeable protons), like aliphatic carbonyl/carboxyl groups. AMS on the other hand is really sensitive to the latter (and less to the former). However, this level of detailed discussion is extremely technical and would remain basically speculative without extensive laboratory tests which, honestly, go beyond the scope of the present manuscript. For this reason, we prefer not to address this with an extensive additional discussion; instead, we modified the text to highlight that evidencing such discrepancies in the OA source apportionment is a further important finding emerging from the present study, which provides a measure of the level of bias one may encounter when relying on a single technique to characterize OA.

We changed the last part of Section 4 as follows:

“Although the AMS and NMR showed an overall good agreement in OC source apportionment, some discrepancies could be noticed in the relative contributions of specific components to the aerosol OC (Table 1). While the total marine and the continental aged BB & BSOA fractions agreed quite well, a greater contribution for Factor 4 (background OA) respect to Factor 3 (Arctic haze OA) was derived by AMS when comparing to NMR (for background OA 40 versus 28% and for Arctic haze OA 28 versus 33% on yearly average, for AMS and NMR respectively). We believe that such discrepancies, likely related to the different sensitivity of the two instruments to specific organic mixtures (see Sect. 2.2.1 and 2.2.2), provide a measure of the level of bias one may encounter when relying on a single technique to characterize OA, representing a further relevant output of this study. And despite these discrepancies, the overall agreement between NMR and AMS characterizations highlights the robustness of the study's findings and reveals a consistent picture of the main organic submicron aerosol sources in Ny-Ålesund and their seasonality.”