In the following, the referee comments are in black, whereas the author's replies are in red.

Reply to referee 1:

General comments

This is an interesting study which addresses important topics within the scope of ACP. I believe it should be published in the journal after revision. The work is of high scientific quality, but the presentation could be improved, and further discussion is needed regarding the very large differences in results between the different source apportionment methods used in the study.

Reply by authors: We would like to thank the referee for his/her very thorough and constructive review of our work and for the suggested corrections and improvements to the submitted manuscript; it is highly appreciated!

We have addressed issues raised regarding improved presentation. The wish for an extended discussion on the differences between the source apportionment methods has been met. Text on this topic has been added to the abstract, section 2.8, section 3.2.3, and section 3.5 (See further down for authors replies and added text).

Specific comments

• It would be good to add the years covered in the study (2017–2020) in the title.

Reply by authors: We have added the years covered by the study in the title.

Changed from: *Composition and sources of carbonaceous aerosol in the European Arctic at Zeppelin Observatory, Svalbard*

Changed to: [2] *Composition and sources of carbonaceous aerosol in the European Arctic at Zeppelin Observatory, Svalbard (2017 – 2020).*

• The abstract is a bit too long. The first part is unnecessary; I suggest starting with "We analyzed long-term measurements..." (line 46) and skipping the part before that. Also, the last sentence (lines 79-80) "FF combustion was the prevailing..." can be removed since the same information is already given on lines 70-72.

Reply by authors: We have removed the sentences suggested by the referee.

• Lines 60–62, Unclear sentence: "Primary biological aerosol particles (PBAP) tracers (various sugars and sugar-alcohols) were elevated in the NH-season but evolved differently, whereas cellulose was completely decoupled from the other PBAP tracers." What did the PBAP evolve differently from?

Changed from: Primary biological aerosol particles (PBAP) tracers (various sugars and sugar-alcohols) were elevated in the NH-season but evolved differently, whereas cellulose was completely decoupled from the other PBAP tracers.

Changed to: [57-59] "Tracers of primary biological aerosol particles (PBAP), including various sugars and sugar-alcohols, showed elevated levels in the non-heating season, albeit with different seasonal trends, whereas cellulose had no apparent seasonality."

• Lines 91–92: You mention increasing sea salt aerosol in the Arctic; is this a general increase of SSA in the Arctic or just at the Zeppelin site?

Reply by authors: The reference on increased SSA levels is that of Heslin-Rees et al. (2020), which showed that the increased presence of SSA at Zeppelin resulted from changes in air circulation patterns, i.e., increased influence from marine western air masses. Although the change was documented at Zeppelin where the measurements took place, such a change in transport pattern will have a regional character affecting a larger region, although not stated explicitly.

• Line101–102: The statement (and references) about the knowledge about Arctic INP and CCN being rudimentary seems a bit outdated. This is a very active research field and newer references may be more appropriate, such as Creamean et al., 2022 (Nat Commun 13, 3537, 2022, https://doi.org/10.1038/s41467-022-31182-x)

Reply by authors: We have changed the sentence and included the reference suggested.

Changed from: Knowledge regarding concentration, activation temperature, composition, sources, origin, and seasonality of Arctic INP and CCN is rudimentary (Creamean et al., 2018; 2019; 2020; Hartmann et al., 2019; 2020).

Changed to: [102-105] Knowledge regarding concentration, activation temperature, composition, sources, origin, and seasonality of Arctic INP and CCN has a noticeable focus given its relevance to the Arctic climate (Creamean et al., 2018; 2019; 2020; 2022; Hartmann et al., 2019; 2020; Freitas et al., 2023).

• Regarding Sec 2.2 Sampling, handling, and storage of ambient aerosol filter samples: Lines 187–188: you state that you collected aerosol particles "for one week". Do you mean that you took weekly samples (i.e., collected the aerosol particles for one week for each sample)? The LHS-analysis presented in Table S3 seems to have varying sample length from 7, 10, 26, 20, 8, 18, 8, 8, 10, 18, 16, 10 days. I assume that the longer time periods are pooled but according to lines 251–252 you "pooled two consecutive samples" for six of the months (including December) but this is not clear enough (e.g. for Dec. it seems like only three days were included for 2018 and N/A for eight days in 2017). You have estimated the positive sampling artifacts for OC (by the QBQ method). Is there any risk of non-negligible negative sampling artifacts in your measurements?

Reply by authors: We have rewritten the sentence starting on line 182 (original version) to clarify its content.

Changed from: We collected aerosol particles on pre-fired (850 °C; 3 h) quartz fiber filters (PALLFLEX Tissuequartz 2500QAT-UP; 150 mm in diameter) for one week.

Changed to: [200-202] We collected aerosol particles on pre-fired (850 °C; 3 h) quartz fiber filters (PALLFLEX Tissuequartz 2500QAT-UP; 150 mm in diameter) at a weekly time resolution. There was some variability in sampling time, typically due to harsh weather conditions."

Reply by authors: We have rewritten the sentence starting on line 249 (original version), which combined with the change made to the sentence starting on line 200, should clarify the sampling time for the samples listed in Table S3.

Changed from: We picked a filter sample for each month of the year from samples collected from 2017 to 2018 to capture the seasonal variability in source composition, resulting in a data capture range of 20% in February to 67% in September.

Changed to: [263-266] We picked 1 – 2 filter samples for each month of the year from samples collected from 2017 to 2018 (Table S3) to capture the seasonal variability in source composition. No valid solution was found for the sample 05 – 21.12.2017, using the Latin Hypercube Sampling (LHS) approach (Sect. 2.8), hence the low coverage for December (6%) compared to September (77%.)

Reply by authors: Without a rigorous sampling set up one cannot estimate the negative sampling artifact of OC, thus any guess on the magnitude of the negative sampling artifact in this or other studies becomes speculative. A sampling line including a denuder can estimate the negative sampling artifact of OC from a back filter, but again, this would just be an estimate and would not be representative for the negative sampling artifact for a single filter set up. As stated in the text, the QBQ approach provides a conservative estimate of particulate OC (OC_P).

• Regarding the "Selection criteria for samples subject to radiocarbon analysis" (lines 249–251): The first sentence of Sec2.5.1 is unclear. Are the samples "chosen" those from the time periods given in Tables S3–S4? If so, it would be good to refer to one of these tables to specify which samples were used. If the time periods are different please provide a table in the Supplement specifying which samples were used for radiocarbon analysis. How were the specific weeks selected? What was the actual selection criteria used to decide which samples were used for radiocarbon analysis for each month? The statement about the "data capture range of 20% in February to 67% in September" is also unclear; what does it mean? In Table S3, June 2017 seems to be covered to a larger extent than September (25/30 days compared to 20/30 for Sep)? What about the "data capture" for December? From Table S3, it seems like no values are given for the December week in 2017 and the LHS only includes 3 days of December 2018.

Reply by authors: We have clarified which samples were analyzed with respect to ^{14}C and thus used as input to the LHS approach in our reply to your previous question. This included rewriting sentences and referring to Table S3, as suggested. We have changed the word "capture" to "coverage" in the revised sentences referring to the percentage of a month covered by the sample(s).

The coverage for the various months is Jan (26%), Feb (21%), Mar (29%), Apr (33%), May (35%), June (60%), July (65%), Aug (45%), Sep (77%), October (74%), Nov (27%) and Dec (6.5%).

The higher percentage for June vs. September that you point to, is due to a typo in Table S3 and S4. For all months where two samples were pooled these were consecutive samples, except for June. We also found a typo for the end date of the sample 05.12.2017 – 13.12.2017.

Changed from: 31.05.2017 – 26.06.2017 (In Table S3 and Table S4)

Changed to: 31.05.2017 – 09.06.2017 (In Table S3 and Table S4)
16.06.2017 – 26.06.2017

Changed from: 05.12.2017 – 13.12.2017 (In Table S3 and Table S4)

Changed to: 05.12.2017 – 21.12.2017 (In Table S3 and Table S4)

Reply by authors: Concerning "How were the specific weeks selected?". In the text we state that the selection criteria were set to "meet the LOD (3 µg C) for EC" and that "the front/back filter carbon content ratio >3". These criteria are hard to meet for the low loading Arctic, and as stated in the text the front/back filter carbon content >3 criterion was not met for all samples selected; thus, we chose those that came closest to the criteria set. Further, the pool of samples to choose from was small, e.g., only two for February for the actual period.

• Sec 2.6: Please specify the MAC values used to convert the absorption data to eBC concentrations in this study. Were there significant differences in MAC values between different seasons or years? How well correlated were the absorption data with the EC measurements? This information can be provided in the Supplement.

Reply by authors: For this study we did not directly calculate any MAC values, due to the complexity of correcting the multiple scattering parameter c as a function of tape type and cross sensitivity to scattering, itself a function of the aerosol single scattering albedo (Yus-Díez et al., 2021; 2022). Rather, we adopted the pragmatic approach of converting the absorption coefficient directly to eBC using the EC. This also accounts for any bias in the lateral flow losses above the tape and spot size. We have clarified this in section 2.6.

Changed from: (…) yielding equivalent black carbon (eBC) by normalization with the mass absorption cross section (MAC) via co-located EC measurements.

Changed to: [278-279] (…) yielding equivalent black carbon (eBC) by normalization with co-located EC measurements.

• Sec 2.8: It is interesting that you changed the LHS S-A compared to Yttri et al., 2011a. Instead of using levoglucosan to estimate TCbb, OCbb and ECbb, you now use 14C-EC. Considering the uncertain atmospheric lifetime of LG, this seems like a good idea but please explain your reasoning in the manuscript. A comparison of the bb-contributions according to your revised LHS S-A and a similar analysis based on the LG-based methodology (from Yttri et al., 2011a) should be included in this article. It would be interesting to see how different the source apportionment becomes when using 14C data instead of levoglucosan data. A comparison of the two approaches is valuable when using S-A data from earlier studies.

Reply by authors: Zeppelin Observatory is a remotely situated Arctic Observatory, experiencing biomass burning (BB) emissions originating from source regions far away, typically in Eurasia. Without going into details about how levoglucosan might be deleted in the atmosphere, it is likely that the level of depletion will increase with increasing atmospheric residence time, thus causing an underestimation of the BB aerosol when used at a site like Zeppelin. Consequently, we have used ^{14}C-EC to apportion the BB aerosol instead of levoglucosan. We have added a sentence in section 2.8 about why we are using ^{14}C-EC instead of levoglucosan for Zeppelin in the revised manuscript.

As stated in your comment, we previously have published results based on a the LHS S-A approach, using levoglucosan to quantify the BB aerosol. Notably, the paper that you refer to (Yttri et al., 2011a) is not about Zeppelin. Note also that LHS S-A is a versatile approach where different variables and macro tracers can be used as input to a varying number of equations, and there is no default solution using levoglucosan, hence the deliberate use of ^{14}C-EC and not levoglucosan in the present study.

As requested, we have run the LHS S-A using levoglucosan for comparison with the LHS S-A using ^{14}C-EC to compare how different the BB contributions would be. These results are discussed in section 3.5. The referee states that "*A comparison of the two approaches is valuable when using S-A data from earlier studies.*" It is not clear to us which earlier studies you refer to and we would like to emphasize

that the comparison between levoglucosan and ^{14}C-EC asked for is only valid for the Zeppelin Observatory and not urban or rural background sites addressed in previous studies.

Added text (Section 2.8; [318-320]): At a remote site like Zeppelin where BB emissions originate from distant source regions, ^{14}C-EC seems a better option for apportioning BB emission than levoglucosan, assuming significant depletion of levoglucosan under such conditions.

Added text (Section 3.5; [779-784]): Rerunning the analysis using levoglucosan instead of ^{14}C-EC for apportionment of BB emissions, lowered the contribution from WF in the NH-season from 26% to 4.4%. However, the contribution from natural sources remained consistent, as BSOA accounted for the modern carbon redistributed from WF. In the H-season, the contribution from RWC decreased from 46% to 8.9%, making BSOA (62%) and natural sources (64%) the major fractions even in the H-season. Hence, the choice of tracer (levoglucosan vs. ^{14}C-EC) would lead to different results and conclusions for Zeppelin.

• Results and discussion, Sec 3.1; lines 367–369: The very high CA levels in 2020 compared to the other years need to be discussed, not just stated. What is the reason for the large differences?

Reply by authors: The reason for the very high levels of CA in 2020 is discussed in Section 3.6.1. We have added the following sentence in section 3.1 to clarify this and included a reference to section 3.6.1.

Changed from: (…) 2020, with EC and OC (…)

Changed to: [395-396] (…) 2020 due to a major LRT episode in July (Sect. 3.6.1), with EC and OC (…)

• Sec 3.2.2, lines 433–438: It is interesting that your results show that non-fossil EC dominates in the H-season and that the contribution is much higher than the one found for 2012–2013 by Winiger et al. Do you have any indications that the relative emissions of fossil and non-fossil EC may have changed substantially during the time period 2012–2020? You mention “differences in sample preparation and in 14C analytical protocol” between the studies but do not provide details about that. Are these differences substantial? Please provide information if you think they are large enough to possibly explain the differences in the results.

Reply by authors: The EC fraction attributed to non-fossil EC in 2017 – 2018 (57 ± 18%) in the present study is comparable to that of the H-season in 2009 (57 ± 21%) (Winiger et al., 2015), and both studies showed higher non-fossil fractions than for the in-between H-season in 2012 – 2013 (26 – 39%) (Winiger et al., 2019). This could indicate pronounced annual variability, but more and long-term measurements are needed to conclude upon this.

The approach used in the present study (Rauber et al., 2023) and that of Winiger et al. (2015 and 2019) are somewhat different from each other, e.g., Winiger et al. used acid fumigation to treat the samples before separation with the NIOSH protocol. Even a minor difference can be the origin of a substantial change. The most recent developments of the method used by Rauber et al. (2023) has not yet been subject to ILC with other methods, moreover, to find out which step(s) in a method are responsible for the observed difference compared to another method is a major work and we will not speculate about that.

• Sec 3.2.3. It is interesting that your PMF-results for the fossil – non-fossil contributions to eBC differ so much from previous studies, with a very dominant contribution from FF in your study. This is also in contrast to your LHS-results based on 14C-measurements, which indicate that non-fossil EC

dominates. The comparison presented in Table 4 is especially important since it shows results from the same time-periods using the two different S-A methods (+ the FLEXPART-model based S-A). The documentation of the PMF-method Platt et al. (2023) is still "in preparation", which it was already in 2021. Since the PMF-based source apportionment in your study gives such different results, you should provide a more detailed description of the methodology (possibly in the supplementary material).

Reply by authors: A detailed description of the PMF is provided in Yttri et al., 2021, and it is a reference to that one in section 2.6 **[280].**

• One of the most interesting and important results of this study is the completely different results for the BB contribution to EC/eBC from the LHS and PMF S-A. This indicates serious problems with either one or both of the S-A approaches. This should be highlighted more clearly in the abstract and a more thorough discussion of these differences and possible explanations is needed before this manuscript can be published in ACP. Could there be problems with the 14C-measurements and/or the EC-OC split in the TOA, leading to some non-fossil OC being detected as EC? Are there any potential problems with the eBC-PMF that could misrepresent eBCBB as eBCFF? If no explanations can be found, it is also an important result – that it is currently not possible to determine the relative contributions of FF and BB to BC/EC with any meaningful accuracy at the Zeppelin site (and presumably also not at other sites where long-range transport is the main source of BC).

Reply by authors: Firstly, why is there a difference between ^{14}C-EC and eBC by PMF? Some comments regarding the ^{14}C-measurements have already been made in our reply to your previous question and we will not repeat them here.

Given the very low concentrations in the Arctic environment differences between co-located or duplicate measurements would be expected due to the relatively high uncertainties. However, eBC- and ^{14}C-measurements are not even duplicates. The physical set up (inlets, flow rates, sample storage location) and the measurement principle itself are different: The eBC derived from Aethalometer measurements does not contain information on the age of the carbon undergoing combustion, rather the wavelength dependence of the absorption, which is linked to conditions under which combustion occurred. Consequently Factor 1 obtained by the PMF, labelled as fossil fuel, could include biofuels, which would be observed as modern fraction (^{14}C). Conversely, coal combustion might appear in Factor 2 labelled biomass burning. Ideally, one might reflect this when naming the factors, e.g., liquid fuel instead of fossil fuel and solid fuel instead of biomass burning, however we leave it as it is for comparability with the ^{14}C-based apportionment and with other studies.

Moreover, applying the Aethalometer-model approach with and Ångstrøm exponent pair of {1,2} (Sandradewi et al. 2008) and {0.9,1.68} (Zotter et al., 2017) we find annual biomass eBC_{BB} contributions of 32% and 29%, respectively. These numbers support our more novel PMF approach and have been included in the main text.

Finally, while there is indeed a difference in the size of the estimated contribution from biomass burning vs. fossil fuel sources between ^{14}C-EC measurements and eBC by PMF, it is important to note that important conclusions drawn are unaffected by this. For example, both ^{14}C measurements and PMF indicate the presence of residential wood combustion and wildfire sourced black carbon at Zeppelin, and with a higher relative BB contribution during the non-heating season.

Secondly: Are there any potential problems with the eBC-PMF that could misrepresent eBC_{BB} as eBC_{FF}?

Problems leading to misattribution in the PMF can arise for two main reasons: 1) The Aethalometer does not measure "biomass burning" or "fossil fuel", and 2) The PMF algorithm does not directly apportion biomass burning and fossil fuel.

On point 1), the wavelength dependence of absorption is mainly influenced by the combustion conditions (Garg et al., 2016). Combustion is considered poorer in a residential heating setting or during a wildfire such that high wavelength absorption is relatively higher at low wavelengths and hence we attribute the high Ångstrøm source from PMF as biomass burning. Other sources with low combustion efficiencies (e.g., residential combustion of coal, waste, peat) may contribute. We have added this point in the extended discussion requested for section 3.2.3. Another example would include the use of biofuels in vehicles, which should appear in the low Ångstrøm factor.

On point 2) it is important to note that the PMF algorithm identifies factors based on variability. It is hypothetically possible that variability is driven, or partly driven, by two well mixed sources with similar composition but varying transport patterns. However, we note that the Ångstrøm exponents obtained from the PMF {1,1.6} are consistent with Ångstrøm exponents observed at numerous sites globally (e.g., Tobler et al., 2018).

As requested, we have extended section 3.2.3 with a discussion on the potential differences between the BB contribution to EC/eBC from the LHS and PMF source apportionment. We have also added a section to the abstract.

Added text (Abstract; [74-78]): Notably, the BB fraction of EC was twice as high as that of eBC, reflecting methodological differences between source apportionment by LHS and PMF. However, important conclusions drawn are unaffected, as both methods indicate the presence of RWC- and WF-sourced BC at Zeppelin, with a higher relative BB contribution during the non-heating season.

Added text (section 3.2.3; [523-548]): Comparing PMF results to the few samples subjected to ^{14}C measurements and source apportionment by tracer based LHS showed that these two approaches were on opposite ends of the scale, with FLEXPART in between (Table 4). Radiocarbon measurements and LHS estimated a BB fraction twice as high as the PMF approach, but all three methods agreed on a higher BB fraction in the NH-season than in the H-season. Notably, BB and FF fractions of eBC derived from PMF were more aligned with those from radiocarbon measurements at Zeppelin in 2012 to 2013 (Winiger et al., 2019) and with fractions derived from levoglucosan measurements at Zeppelin in winter 2008 to 2009 (Yttri et al., 2014). However, inter-annual variability makes such a comparison indicative only. Consideration of methodological differences is essential. Crucial steps of ^{14}C-EC measurements include preventing EC loss during OC removal and avoiding OC mixing with the minor EC fraction, impacting its modern vs. fossil fuel signature. The advancements in the analytical approach used in this study (Rauber et al., 2023) specifically aimed to improve these critical steps. Additionally, eBC derived from Aethalometer measurements provides no information on the age of carbon undergoing combustion but reflects the wavelength dependence of the absorption linked to the combustion condition (Garg et al., 2016.). Consequently, the eBC_{FF} factor obtained by PMF could also contain emissions from combustion of biofuels, observed as modern carbon by ^{14}C-measurements. Conversely, emissions from coal combustion might contribute to the eBC_{BB} factor. Terms like liquid fuel instead of fossil fuel and solid fuel instead of biomass burning could be more appropriate, but we maintain the notation for comparability with ^{14}C-based apportionment and Aethalometer-model studies (Sandradewi et al., 2008). Applying the Aethalometer-model approach with an Ångstrøm exponent pair of (1,2) (Sandradewi et al. 2008) and (0.9,1.68) (Zotter et al., 2017), yields annual eBC_{BB} contributions of 32% and 29%, supporting our more novel PMF approach (27±14%) (Table 4). Further, we note that the Ångstrøm exponents obtained from the PMF (1,1.6) are consistent with the distribution of Ångstrøm exponents observed at various sites across Europe (e.g., Tobler et al., 2021). Despite the differences in the estimated contribution size from biomass vs. fossil fuel sources between

^{14}C-EC measurements and eBC by PMF, important conclusions drawn are unaffected. For instance, both methods indicate the presence of residential wood combustion and wildfire-sourced black carbon at Zeppelin, with a higher relative BB contribution during the non-heating season.

• Sec 3.2.4 (Lines 506–516). This whole (short) section about Seasonal footprints for eBCBB and eBCFF is confusing and does not contain substantial information. It should either be completely rewritten or removed from the manuscript. Removal may be the best choice. Additionally, Fig. 6 is difficult to understand. What are these plots really showing? What is meant by "and all footprints for this season"? Why do you use "relative" plots here instead of showing the footprints directly?

Reply by Authors: We have removed section 3.2.4 and Figure 6

• Sec 3.3 (BSOA tracers) and 3.4 up to 3.4.2 (PBAP) are interesting and well-written. The only objection (apart from a few minor typos) is that Sec 3.4.3 (Source apportionment by LHS) should be put in another Section (probably a new Sec 3.5) since it is not only about PBAP.

Reply by authors: **[756]** Section 3.4.3 has been changed to Section 3.5.

• Sec 3.4.3 (to be changed to Sec 3.5) Lines 728–731: The sentence about the Zwaaftink (2022)-based tests is unclear. "We increased the PBAP fraction to 11%..." sounds odd; do you mean that the LHS-based PBAP fraction increased to 11% when you used the alternative OC-to-PBAP ER from Zwaaftink et al.? Please specify clearly that the Zwaaftink-based PBAP-estimate is based on another set of tracers than the ones used in the LHS here. As far as I understand, Zwaaftink et al. used the sum of mannitol, arabitol, glucose and trehalose (and no cellulose at all!) while you use mannitol and arabitol (for fungal spores) and cellulose (for debris) in the LHS. Does the 11% that you "increased the PBAP fraction to" exclude the cellulose tracer – or did you include it together with the mannitol+arabitol+glucose+trehalose?

Changed from: We increased the PBAP fraction to 11% using an OC-to-$PBAP_{Tracers}$ emission ratio (ER) of 14.6 ± 2.1 (Zwaaftink et al., 2022), noting that the ER was obtained from measurements in the boreo-nemoral zone, and thus more representative of LRT than local PBAP sources.

Changed to [765-768]: The PBAP fraction increased to 11% when using an OC-to-$PBAP_{Tracers}$ emission ratio (ER) of 14.6 ± 2.1 (Zwaaftink et al., 2022) including the sum of arabitol, mannitol, glucose, and trehalose. Note that this ER was obtained from measurements in the boreo-nemoral zone, and thus more representative of LRT than local PBAP sources.

• Carbonate carbon. In Sec 3.5.3 (Episode 3) you mention the presence of carbonate carbon (CC) during the episode. How frequently do you observe CC in the TOA at Zeppelin? It would be interesting to see the amount of CC in Table 1.

Reply by authors: CC is not regularly measured at Zeppelin, indeed only for the Zwaaftink et al. (2022) paper, so there is no data available to be presented. Such data would certainly be interesting.

• Sec. 4 Implications: You suggest/encourage trend studies and a pan-Arctic investigation of the spatial variability of eBCBB and eBCFF based on the aethalometer-based PMF approach used in your study. This could be interesting but considering the widely different results from the PMF S-A and the tracer-based LHS S-A, do you really trust that the eBC PMF gives correct (reliable/useful) results for the source apportionment? If so, does it mean that you think that the tracer-based S-A is incorrect? To me, your results indicate that we should be careful with using aethalometer-based

source apportionment data and not trust that they give an accurate split between BB and FF. I would really like it if you could prove me wrong on this point!

Reply by authors While acknowledging a noticeable difference in the estimated fractions of biomass burning vs. fossil fuel sources between PMF and ^{14}C-measurements, we emphasize that crucial conclusions remain unaffected. Despite lower eBC_{BB} values and higher eBC_{FF} values compared to ^{14}C-measurements, they remain valuable for trend studies assessing relative changes. Consistent measurement and apportionment methods ensure comparability across Arctic sites. We concur on the need for further investigation to understand the causes behind the observed deviations in source-apportioned eBC and EC.

Technical corrections

The manuscript could benefit from thorough language editing. Some parts of the text are well-written and most of it is understandable, but it would be easier and more pleasant to read if the text was revised from a language perspective. According to the Author contribution statement, "All co-authors contributed to writing, reviewing, and editing the final article". Since there are almost 20 authors, some of whom seem to have English as their primary language, I am surprised about the language and the editing of some parts of the manuscript. Did all co-authors really check the entire manuscript before submission? Although ACP may include language editing of accepted manuscripts, it would be helpful if all co-authors reviewed the language and structure of the text before resubmission. I will not go through every part that I found awkwardly formulated or correct all minor language mistakes (I think that is the job of the author-team) but will only mention some examples and some of the unclear parts.

• The authors use the word "prevail" (and prevailing) frequently and in a strange way. It is used more than ten times in the manuscript and I think incorrectly in most (or all) cases. One example (lines 653–655): "Mannitol and arabitol were highly correlated in the NH-season (r2 = 0.983) when levels were elevated, and mannitol to arabitol ratio variability minor, suggesting one common source prevailing." – I think you mean that there was one common dominant source?
Similar cases are found on lines 70, 79, 142, 442, 461, 465, 514, 699, 760, 820 – in most or perhaps all of these cases I think you mean dominant or primary rather than prevailing.

Reply by authors: The sentences mentioned have been rewritten:

Line 67: changed to: (…) and BSOA (25%) in the H-season, whereas BSOA (56%) dominated over WF (26%) and FF (15%) in the NH-season.

Line 142-147 changed to: Their study identified three factors dominated by anthropogenic sources (Oxygenated Organic Aerosol, Arctic Haze, and Primary Organic Aerosol) and three factors associated with natural emissions (Methane Sulfonic Acid-Related Organic Aerosol, Primary Biological Organic Aerosol, and Biogenic Secondary Organic Aerosol). These factors exhibited distinct seasonal patterns, with the first three dominating in winter and the latter three in summer.

Line 471 changed to: (…) it cannot be excluded that that BB emissions can dominate for an entire week.

Line 493 changed to: Neither eBC_{WF}, nor eBC_{RWC} (eBC_{BB} in H-season), dominated on a monthly basis (Fig. S3), although (…)

Line 498 changed to: FLEXPART predicted an almost equal share of BC from BB and FF annually, whereas BC_{FF} (56 ± 3.6%) dominated in the H-season and BC_{BB} (61 ± 3.3% (…)

Line 692 changed to: , suggesting one common source dominating.

Line 735-736 changed to: Lack of comparable seasonality between nearby sites indicates that local sources dominate (Brighty et al., 2022), (…)

Line 802-803 changed to: Flaring was the dominating fossil fuel source category according to FLEXPART, (…)

Line 865 changed to: (…), i.e., natural sources, particularly BSOA, dominating in the non-heating season (…)

• Abstract, line56–60: The sentence "Intrusions of warm air masses from Siberia..." is too long and needs to be restructured.

Changed from: Intrusions of warm air masses from Siberia in summer caused three- and ninefold increases in 2-methyltetrols compared to 2017 to 2018, in 2019 and 2020, respectively, warranting investigation of the local vs. the long-range atmospheric transport (LRT) contribution, as certain Arctic vegetation has highly temperature sensitive biogenic volatile organic compounds (BVOC) emission rates.

Changed to [54-57]: Warm air masses from Siberia led to a substantial increase in 2-methyltetrols in 2019 and 2020 compared to 2017 to 2018. This highlights the need to investigate the contribution of local sources vs. long-range atmospheric transport (LRT), considering the temperature sensitivity of biogenic volatile organic compounds emissions from Arctic vegetation.

Introduction

• Line 97: It might be better to write "Some PBAP are efficient ice nucleating particles" – the reference given (Tobo et al., 2019) is perhaps not the best regarding general INP properties for PBAP since the article focus on dust as a source of INP?

Reply by authors: The reference to Tobo et al. (2019) is correct.

From Tobo et al. (2019): *"Nevertheless, as relatively abundant microbial communities adapted to severe temperature and nutrient regimes are known to exist in supra- and proglacial ecosystems in Svalbard[44,45,46], it is assumed that highly ice nucleation active organic matter contained in the glacial outwash sediments may be attributable to certain microbial sources."*

Changed from: PBAP are efficient ice nucleating particles (INP) at high temperatures (Tobo et al., 2019), (…)

Changed to [99]: Some PBAP are efficient ice nucleating particles (INP) at high temperatures (Tobo et al., 2019), (…)

• Line 106–108: The sentence starting with "Overall, elucidating developments in local aerosol emissions and or formation," is a bit awkward and (at least) the "or" should be removed.

Changed from: Overall, elucidating developments in local aerosol emissions and or formation, changes in LRT of aerosols, and, in turn the aerosol chemical profile, is crucial to understanding a changing Arctic and its regional and global climate impact.

Changed to [109-111]: Understanding changes in local aerosol emissions and formation, shifts in LRT of aerosols, and consequently, alterations in the aerosol chemical composition are essential for understanding the evolving Arctic environment and its regional and global climate impacts.

• Line 108–112: The sentence “Meanwhile, understanding and validating these changes...” needs revision – perhaps it is best to split into at least two sentences. Replace “excepting” with “with the exception of” and add “impact on” before “climate and albedo”.

Changed from: Meanwhile, understanding and validating these changes in atmospheric composition requires high quality, long-term observations, which are particularly lacking for carbonaceous aerosol (CA), excepting black carbon (BC), a focus of attention due to its direct climate and albedo (Clarke and Noone, 1985; Pueschel and Kinne, 1995; Hansen and Nazarenko, 2004; Elefteriadis et al. 2009; Hirdmann et al., 2010).

Changed to [111-115]: Confirming and understanding these changes in atmospheric composition requires high-quality, long-term observations. This is particularly true for carbonaceous aerosol (CA), except for black carbon (BC), a focus of attention due to its impact on climate and albedo (Clarke and Noone, 1985; Pueschel and Kinne, 1995; Hansen and Nazarenko, 2004; Elefteriadis et al., 2009; Hirdmann et al., 2010).

• Line 117: “mostly only” – either “mostly” or “only”

Reply by authors [120]: Removed “only”.

• Line 122: AH abbreviation not explained the first time it is used

Reply by authors: AH is explained in line 119.

• Lines 137–143; The long sentence “Moschos et al. (2022) applied...” ends in a confusing way – it is unclear which of the six factors are “prevailing in winter and summer, respectively” and also which of them have “equally large contributions”. This sentence needs to be reformulated in a clearer way.

Changed from: Moschos et al. (2022) applied positive matrix factorization (PMF) to spectral data of water-soluble organic carbon extracts (offline analysis using an aerosol mass spectrometer, AMS), finding three anthropogenic dominated factors: oxygenated organic Aerosol (OOA), Arctic Haze (AH); and primary organic aerosol (POA)), and three from natural-dominated emissions: methane sulfonic acid-related organic aerosol (MSA-OA), primary biological organic aerosol (PBOA), biogenic secondary organic aerosol (BSOA)), dominating in winter and in summer, respectively, with equally large contributions.

Changed to [141-147]: Moschos et al. (2022) used positive matrix factorization (PMF) on spectral data derived from water-soluble organic carbon extracts, analyzed off-line using an aerosol mass spectrometer. Their study identified three factors dominated by anthropogenic sources (Oxygenated Organic Aerosol, Arctic Haze, and Primary Organic Aerosol) and three factors associated with natural emissions (Methane Sulfonic Acid-Related Organic Aerosol, Primary Biological Organic Aerosol, and Biogenic Secondary Organic Aerosol). These factors exhibited distinct seasonal patterns, with the first three dominating in winter and the latter three in summer.

• The map shown in the right panel of Figure 1 is not of high enough resolution. E.g. the Arctic circle line is hardly visible.

Reply by authors: A new Figure 1 with improved resolution has been added.

Experimental

• Line 290–292: Consider referring to Table S3 here to provide information about which 13(?) samples were used. Also, clarify what happened to the 05.12.2017–13.12.2017 sample.

Reply by authors [312]: We added a reference to Table S3. Concerning sample 05.12.2017 – 13.12.2017, see authors reply to the referee's question regarding "Selection criteria for samples subject to radiocarbon analysis" (lines 249–251, original manuscript):

• Table S1: Typo in the notes a "0" instead of a "+": TC × F14C-TC = OC × F14C-OC 0 EC × F14C-EC – change 0 to +

Reply by authors [Table S1]: "0" has been replied by "+"

• Table S3 and S4: Extend the table headers to explain what N/A means. Also, consider adding information about the start and stop hours of the sampling.

Reply by authors: The table header of S3 and S4 has been extended to include the explanation of N/A: (N/A = Not Available)

• Lines 319–341: Please add a reference or URL for the ECLIPSEv6b emission data. Also, clarify whether the ECLIPSE emissions were estimated for the actual modelled years (2017–2020) or if they were the same for all years? How were the temporal variations of the anthropogenic emissions handled? Did you include some temperature dependent emissions from the residential sector? What about seasonal variations for other sectors?

Reply by authors: We use the following reference for the ECLIPSEv6b, as there is no peer revieved paper on the 6b version yet published:

[344] Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J. and Sch.pp, W.: Global anthropogenic emissions of particulate matter including black carbon, Atmos. Chem. Phys., 17, 8681–8723, doi:10.5194/acp-17-508681-2017, 2017.

Added text [357-360]: The ECLIPSEv6b dataset provides emission data at 5-years intervals. These emissions are then interpolated to annual emissions according to the trend in geographical areas considered in ECLIPSE (Klimont et al., 2017). The temporal variation in the emissions of all sectors was provided by IIASA (Klimont et al., 2017).

Reply by authors: Concerning the temporal variation of the anthropogenic emissions, see figure below:

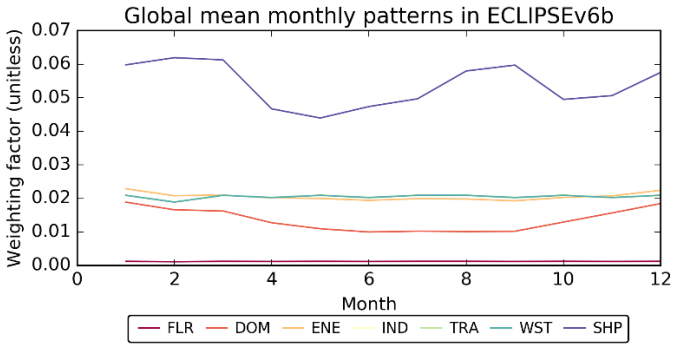

Fig. Monthly weights used in each emissions sector in ECLIPSEv6b averaged globally.

Line 331: Clarify what is meant by “in-land waters”. Does this refer to lakes and rivers? Did you not include ocean-going shipping?

Reply by authors: We made the two following changes:

Changed from: Emissions from shipping activities in in-land waters (SHP)

Changed to [354]: Emissions from international shipping activities (SHP).

Changed from: Transport sector (TRA) – emissions from all land-based transport of goods, animals, and persons on road networks and off-road activities.

Changed to [352-353]: Transport sector (TRA) – emissions from all land-based transport of goods, animals, and persons on road and off-road networks, including domestic shipping and aviation.

Results and discussion

• Table 1: Add an explanation of OCB in the Table header and specify which months are included for the H-S and NH-S in the table notes

Changed from: Table 1: Annual and seasonal mean concentrations of OC, OC_B, EC, TC, and organic tracers at Zeppelin Observatory, 2017 to 2020.
Changed to [Table 1]: Table 1: Annual and seasonal mean concentrations of OC, OC_B (OC on backup filters) EC, TC, and organic tracers at Zeppelin Observatory, 2017 to 2020.

Changed from: Notations: H-S = Heating season NH-S = Non-heating season

Changed to [Table 1]: Notations: H-S = Heating season (November to May); NH-S = Non-heating season (June to October)

• Figure 3. The figure caption is not completely correct: b) 2-MT is not shown only 2-MET and 2-MT/2-MET; c) the ratio mannitol/arabitol is not shown

Reply by authors: We have corrected the Figure 3 caption accordingly.

Changed from: Figure 3. Panels show monthly mean concentrations for 2017 to 2020 and box plots (mean, 25, 50, 75 percentiles and IQR) for 2017 to 2020 at Zeppelin Observatory for a) Levoglucosan and levoglucosan/mannosan; b) 2-methylerythritol (2-MET), 2-methylthreitol (2-MT), and 2-MT/2-MET; c) Mannitol, arabitol, and mannitol/arabitol; d) Fructose and glucose; e) Trehalose; f) Cellulose. All variables are measured in the PM_{10} size fraction.

Changed to [Figure 3]: Panels show monthly mean concentrations for 2017 to 2020 and box plots (mean, 25, 50, 75 percentiles and IQR) for 2017 to 2020 at Zeppelin Observatory for a) Levoglucosan and levoglucosan/mannosan; b) 2-methylerythritol (2-MET) and 2-MT/2-MET; c) Mannitol and arabitol and mannitol/arabitol; d) Fructose and glucose; e) Trehalose; f) Cellulose. All variables are measured in the PM_{10} size fraction.

• Line 348: Table 2 incorrect, it should be Table 4

Reply by authors [375]: We have changed from Table 2 to Table 4.

• Figure 5. I suggest that you start the Figure caption with "Source apportionment using the LHS approach (Sect. 2.7)" instead of having this at the end. Also specify that the relative fractions are given in percent.

Reply by authors: Change made as suggested. "%" included on secondary y-axes in revised figure.

Changed from: Figure 5. Panels show monthly mean concentrations and relative contributions for samples collected in 2017 to 2018 at Zeppelin Observatory for A) Biogenic Secondary Organic Aerosol (OC_{BSOA}) and OC_{BSOA}/TC; B) Primary Biological Aerosol Particles (OC_{PBAP}), being the sum of fungal spores (OC_{PBS}) and plant debris (OC_{PBC}), and OC_{PBAP}/TC; C) Biomass burning (OC_{BB}, OC_{BB}/TC) and fossil fuel sources (OC_{FF}, OC_{FF}/TC; D) Fossil fuel (EC_{FF}, EC_{FF}/TC) and biomass burning (EC_{BB}, EC_{BB}/TC), source apportioned using the LHS approach (Sect. 2.7).

Changed to [Figure 5]: Figure 5. Source apportionment using the LHS approach (Sect. 2.7). Panels show monthly mean concentrations and relative contributions for samples collected in 2017 to 2018 at Zeppelin Observatory for A) Biogenic Secondary Organic Aerosol (OC_{BSOA}) and OC_{BSOA}/TC; B) Primary Biological Aerosol Particles (OC_{PBAP}), being the sum of fungal spores (OC_{PBS}) and plant debris (OC_{PBC}), and OC_{PBAP}/TC; C) Biomass burning (OC_{BB}, OC_{BB}/TC) and fossil fuel sources (OC_{FF}, OC_{FF}/TC; D) Fossil fuel (EC_{FF}, EC_{FF}/TC) and biomass burning (EC_{BB}, EC_{BB}/TC).

- Line 356: Table S6 should be Tables S5 and S6
- Line 357: Table 6S should be Table S5

Changed from: Comparison is made with Birkenes Observatory (Southern Norway), representative of the lowest CA levels in regional background Europe (Yttri et al., 2021) (Table S5), with Ispra, a regional background site in the Po Valley (Northern Italy), one of Europe's most polluted regions (Table 6S), (…)

Changed to [383-384]: Comparison is made with Birkenes Observatory (Southern Norway), representative of the lowest CA levels in regional background Europe (Yttri et al., 2021) (Table S5 and S6), with Ispra, a regional background site in the Po Valley (Northern Italy), one of Europe's most polluted regions (Table 5S), (…)

• The first sentence of Sec. 3.1 (lines 361–363) needs to be rephrased for clarity. and
• Line 365: "second lowest only to levels observed in Antarctica" – rephrase this sentence to improve readability

Changed from: The interannual variabilities of EC (34%) and OC (38%) were comparable to SO_4^{2-} (40%), which like OC can be either primary or secondary, or of LRT or local origin, and originate from natural as well as anthropogenic sources, and having a time series dating back to 1991 (Platt et al., 2022). The annual mean concentrations ranged from 6.5 to 16.3 ng Carbon (C) m^{-3} (EC) and from 90.3 to 197 ng C m^{-3} (OC), second lowest only to levels observed in Antarctica (1.9 ng EC m^{-3}; 12.2 ng OC m^{-3}) (Table S7) (Rauber et al., in prep.)

Changed to [388-393]: The interannual variabilities of EC (34%) and OC (38%) were comparable to SO_4^{2-} (40%). Like OC, SO_4^{2-} can have both primary or secondary sources, originate from LRT or local emissions, and stem from natural as well as anthropogenic sources. Notably, its time series spans back to 1991 (Platt et al., 2022). The annual mean concentrations ranged from 6.5 to 16.3 ng Carbon (C) m^{-}

^{3} for EC and from 90.3 to 197 ng C m^{-3} for OC. These levels are amongst the lowest globally, still notably lower compared to Antarctica (1.9 ng EC m^{-3}; 12.2 ng OC m^{-3}) (Table S7) (Rauber et al., in prep.).

• Table 2. Specify in the header that all components refer to concentrations in PM10. I also think it would be good to add a column with total PM10 concentrations to the Table.

Reply by authors: We have included the following text in section 2.7 for documentation of the sampling approach for SO_4^{2-}, Cl^-, Na^+, K^+, Mg^{2+}, Ca^{2+}, Al, Fe, Mn, and Ti.

Added text [**287-292**]: "Inorganic anions and cations were obtained using a NILU stacked filter unit (SFU) collecting aerosol particles on Teflon filters (2 µm pore, 47 mm Zefluor Teflon, Gelman Sciences). The SFU has a downward-facing inlet that effectively reduces the sampling efficiency for aerosol particles with an equivalent aerodynamic diameter (EAD) larger than 10 µm (Zwaaftink et al., 2022). Elements were obtained from paper filters (Whatman 41) using a high-volume air sampler with an inlet discriminating against aerosol particles with an EAD larger than 3 µm."

We have included the following notation to Table 2:

[Table 2]: "1) 3 µm EAD size fraction (10 µm EAD for other variables)."

Reply by authors: PM_{10} mass concentration is not measured at Zeppelin Observatory and thus cannot be added to Table 2.

• Line 376: "EC dropped by a factor of two" – should be "EC increased by a factor of two"

Changed from: EC dropped by a factor of two during the H-season compared to the NH-season, due to more efficient transport of air masses to the Arctic in winter (Ottar et al., 1989) and by AH accumulation in winter and spring (Shaw, 1995).

Changed to [405-407]: EC increased by a factor of two during the H-season compared to the NH-season, due to more efficient transport of polluted air masses to the Arctic in winter (Ottar et al., 1989) and by AH accumulation in winter and spring (Shaw, 1995).

• Lines 393–406: This last part of Sec. 3.1 is a bit disorganised and would benefit from a restructuring or rewriting
• Lines 396–397: ", as seen for eBCFF." what do you mean by that?
• Lines 397–399: the sentence about Marine MSA is confusing. What do you mean by "for calculation"? What calculation did you do for MSA?
• Lines 399–400: The sentence about marine heterogenic polymer-gels seems misplaced here. It is really not part of your results and here it just makes the text more disorganised and confusing.

Changed from: The BB tracer levoglucosan decreased greatly from February to March, indicating that OC from fossil sources became more important as the AH period progressed. However, we speculate that there was a substantial degradation of levoglucosan from the end of the polar night (15 February) or as day length became significant, as eBC_{BB} persisted until the end of the AH period in April, as seen for eBC_{FF}. Marine MSA starts increasing in April but contributed <6% to OC for April to August 2017 to 2020, using monthly mean MSA data from Zeppelin for 1998 to 2004 for calculation (Sharma et al., 2012). Marine heterogenic polymer-gels are likely to contribute to Arctic CA, but levels are low (Karl et al., 2013), and are not addressed in this study. BB, BSOA and PBAP tracers typically peaked in July and August, but whereas BSOA tracers decreased abruptly in early fall, PBAP tracers persisted to late fall, whereas BB tracers, EC, eBC_{BB}, and eBC_{FF} started increasing again towards the end of the year.

Changed to [422-430]: The BB tracer levoglucosan experienced a significant decrease from February to March, suggesting that OC from fossil sources became more prominent as the AH period progressed. However, we speculate that there was a substantial degradation of levoglucosan starting from the end of the polar night (15 February) or as daylight hours increased, as no similar decrease was observed for eBC_{BB}. This degradation might be due to factors such as aerosol particle scavenging by low-level Arctic clouds, which is known to peak in early spring for BC (Zieger et al., 2023), and subsequent depletion by water phase reactions. BB, BSOA and PBAP tracers typically peaked in July and August, but whereas BSOA tracers decreased abruptly in early fall, PBAP tracers persisted to late fall, whereas BB tracers, EC, eBC_{BB}, and eBC_{FF} started increasing again towards the end of the year.

• Lines 426–427: "but the very high L/M ratios (occasionally > 40) in spring argued to be emissions from crops residue burning in Asia were not observed." – please rephrase this sentence

Changed from: Our findings correspond with L/M ratios <5 in summer at Gruvebadet (Ny-Ålesund) (Feltracco et al., 2020), but the very high L/M ratios (occasionally > 40) in spring argued to be emissions from crops residue burning in Asia were not observed.

Changed to [453-456]: Our findings correspond with L/M ratios below 5 in summer at Gruvebadet (Ny-Ålesund) (Feltracco et al., 2020). However, we did not observe the very high L/M ratios, occasionally exceeding 40, that have been attributed to emissions from crops residue burning in Asia during the spring.

• Lines 431–433: the sentence starting with "61 ± 15% of EC, " is incomplete.

Changed from: Tracer based LHS source apportionment found that BB was the primary source of EC in all but one of the 13 samples analyzed (Table S3 to S4); i.e., 61 ± 15% of EC, with the percentage varying by season: 67 ± 5% in the NH-season when EC levels were low and influenced by WF, and 57 ± 18% in the H-season when RWC dominates.

Changed to [459-462]: Tracer based LHS source apportionment found that BB was the primary source of EC in all but one sample (Table S3 and S4). On average, 61 ± 15% of EC was attributed to BB, with this percentage varying by season: 67 ± 5% in the NH-season when EC levels were low and influenced by WF, and 57 ± 18% in the H-season when RWC dominated.

• Lines 444–445: "was unprecedented compared to Winiger et al. (2015)" – what do you mean by this?

Changed from: The weekly minimum BB fraction for Jan. 2018 (21%) unprecedented compared to Winiger et al. (2015) (39%).

Changed to [475-476]: The weekly minimum BB fraction for Jan. 2018 (21%) was much lower compared to the lowest percentage reported by Winiger et al. (2015) (39%).

• Sec 3.2.3 (Line 451): Add "and FLEXPART modelling" to the Section title

Changed from: eBC from biomass burning and fossil fuel combustion obtained from PMF.

Changed to [482-483]: eBC from biomass burning and fossil fuel combustion obtained from PMF and FLEXPART modeling.

• Lines 458–463: The three sentences starting with “By the crude, but still realistic, assumption” and ending by “emphasizing RWC as a larger source of eBC than WF” need to be reformulated – they are unclear and the language needs improvements.

Changed from: By the crude, but still realistic, assumption that all eBC_{BB} in the NH-season comes from WF (here: eBC_{WF}), then the 27 to 42% contribution of eBC_{WF} calculated for the 2017 to 2020 NH-seasons is lower than previous results. Neither eBC_{WF}, nor eBC_{RWC} (eBC_{BB} in H-season), dominated monthly (Fig. S3), although by a short margin for October 2017, July 2020, and October 2020 (46 to 48%). WF was estimated to contribute 5.4 to 12% to eBC annually, and RWC 20 to 26%, emphasizing RWC as a larger source of eBC than WF.

Changed to [490-496]: In our study, 27% – 42% of eBC was attributed to WF emissions in the NH-season, which is lower than previous findings. Our estimate assumes that all eBC_{BB} in the NH-season comes from WF emissions (eBC_{WF}) and from residential wood combustion emissions (eBC_{RWC}) in the H-season. Neither eBC_{WF}, nor eBC_{RWC} dominated on a monthly basis (Fig. S3), although they came close in October 2017, July 2020, and October 2020, accounting for 46% to 48%. The annual contribution of eBC_{WF} to eBC was estimated to be 5.4% to 12%, while eBC_{RWC} contributed 20 to 26%, highlighting that RWC is a larger source of eBC compared to WF.

• Lines 482–483: Confusing sentence – what do you mean by “contributing 64% in 2020”?

Reply by authors: We removed “contributing 64% in 2020”.

• Lines 484–486: There is a discussion of the degradation of levoglucosan during LRT as a possible explanation of the differences in results but would a degradation not rather decrease the estimated BB-fraction (and thus you would have expected it to be lower rather than higher than found by the PMF and FLEXPART)?

Reply by authors: Note that we are referring to the fraction of levoglucosan observed in the NH-season vs. the H-season, not EC_{WF} and EC_{RWC} derived from levoglucosan multiplied by some emission ratio. We have removed the three last lines from Table 3 and chosen to refer to these values in the text only.

Changed from: Using the levoglucosan BB tracer, the WF fraction (36 to 64%; Table 3) was higher than seen for both eBC_{WF} (17 to 35%) (PMF) and BC_{WF} (32 to 45%) (FLEXPART).

Changed to [515-517]: For the BB tracer levoglucosan, the fraction observed in the NH-season (36 to 64%), corresponding to the WF fraction, was higher than seen for both eBC_{WF} (17 to 35%) (PMF) and BC_{WF} (32 to 45%) (FLEXPART).

• Lines 651–653: the sentence “The annual mean mannitol to arabitol ratio…” is a bit confusing. Either remove the numerical value for Zeppelin (and according to Table S6 it is 1.1±0.5 rather than 1.1±0.2) or add the corresponding value for Birkenes. The end of the sentence “and fungal spores (Bauer et al., 2008)” is unclear.

Changed from: The annual mean mannitol to arabitol ratio was comparable between Zeppelin (1.1 ± 0.5) and Birkenes (1.0 ± 0.0) (Table S6), to values reported for the Nordic countries (Yttri et al., 2011b) and to fungal spores (Bauer et al., 2008).

Changed to [689-691]: The annual mean mannitol to arabitol ratio was comparable between Zeppelin (1.1 ± 0.5) and Birkenes (1.0 ± 0.0) (Table S6), and to values reported for the Nordic countries.

• Lines 675–676: “no correlations...were obtained” but you give a r2-value of 0.423 for the NH-season; this is not a very high correlation but I would not consider it as no correlation?

Changed from: Indeed, no correlations between levoglucosan and sugar-alcohols ($r^2_{NH\text{-}season} < 0.423$; $r^2_{H\text{-}season} < 0.056$) were (…)

Changed to [712-174]: Changed from: Indeed, low correlations between levoglucosan and sugar-alcohols ($r^2_{NH\text{-}season} < 0.423$; $r^2_{H\text{-}season} < 0.056$) were (…)

• Lines 707 and 711: the “(here ng C m-3)” seem unnecessary and just confusing?

Reply by authors: We agree that omitting the unit (ng C m^{-3}) would make the sentence clearer. However, we are referring to concentrations of cellulose with the unit ng m^{-3} in the beginning of the section and we suspect that the reader might be in doubt if we do not make this clarification.

• Lines 714–717: The sentence about the plant debris contribution at low OC levels should be rephrased for clarity. The “and were correlated” part of the sentence is not totally clear about what was correlated but I assume it must be the plant debris contribution and total OC? I think the part “and elevated cellulose levels” is redundant since cellulose is the tracer used to determine the debris contribution. Is it really correct to say that the “plant debris drives observed OC levels” when the contributions are only 5 to 12%?

Changed from: Weekly samples (n = 23) with a high (5 to 12%) plant debris contribution were associated with low OC levels (mean: 53 ng C m^{-3}; 22 percentile) and elevated cellulose levels (mean: 3.1 ng m^{-3}; 80 percentile) and were correlated ($r^2 = 0.707$), suggesting that plant debris drives observed OC levels at low concentrations.

Changed to [751-753]: Weekly samples (n = 23) with a high (5 to 12%) plant debris contribution were associated with low OC levels (mean: 53 ng C m^{-3}; 22 percentile). Plant debris and OC were correlated ($r^2 = 0.707$), suggesting that plant debris is a driver of observed OC levels at low concentrations.

• Lines750–751 and 795–797: Clarify how the Episode 1 OC-level (2172 ngC/m3 for 5 days) can “explain” 17% of the “annual OC loading” (for 2020) while the Episode 3 OC-level (818 ngC/m3 for 8 days) “explain” 16% of the annual OC loading (also 2020)? If I understand this correctly the integrated OC level for the Episode 1 is ca 66% larger than that for Episode 3, that is a much larger difference than 17 to 16%?

Reply by authors: There has been a mistake in the calculations made. We have sorted this out and made the following correction:

Episode 1:

Changed from: These levels were the highest in four years of observations, explaining 17% of the annual OC loading, but only 6% of EC.

Changed to [793-794]: These levels were the highest in four years of observations, explaining 21% of the annual OC loading, but only 7% of EC.

Episode 2:

Changed from: Air masses with a history over south-western Russia, eastern, central, and northern Europe (including Scandinavia) (Fig. 8) increased the OC (549 ng C m^{-3}) and EC (52 ng C m^{-3}) concentrations at Zeppelin to levels corresponding to 13% of their annual loading.

Changed to [823-825]: Air masses with a history over south-western Russia, eastern, central, and northern Europe (including Scandinavia) (Fig. 8) increased the OC (549 ng C m^{-3}) and EC (52 ng C m^{-3}) concentrations at Zeppelin to levels corresponding to 14% of their annual loading.

Episode 3:

Changed from: Briefly, the EC level (78 ng C m^{-3}) was the highest in four years of observations, whereas the OC level (818 ng C m^{-3}) was much lower than observations made during the July 2020 (Sect 3.5.1) episode, explaining 19% and 16% of the annual EC and OC loading, respectively (Fig. 9).

Changed to [837-840]: Briefly, the EC level (78 ng C m^{-3}) was the highest in four years of observations, whereas the OC level (818 ng C m^{-3}) was much lower than observations made during the July 2020 (Sect 3.5.1) episode, explaining 15% and 13% of the annual EC and OC loading, respectively (Fig. 9).

Section 3.4.1: Correction of miscalculation as seen for episode 1 to 3:

Changed from: However, maximum concentrations of sugars and sugar-alcohols were observed for the LRT episode 22 – 27 July 2020 (Sect. 3.6.1), explaining 24% of the annual sugars and sugar-alcohols loading.

Changed to [666-668]: However, maximum concentrations of sugars and sugar-alcohols were observed for the LRT episode 22 – 27 July 2020 (Sect. 3.6.1), explaining 29% of the annual sugars and sugar-alcohols loading.

• Figure 7. No data are given for T<0 so this can be removed from panel B. What does T average represent in panel B? Specify this in the Figure caption.

Reply from the authors [Fig. 6A, Fig. 6B, Fig. 6C]: We have updated Figure 6A (Originally Figure 7 A) accordingly. “T average” is the mean temperature. “T average” has been changed to “mean T” for a specified period in the legends of Figures 6A to 6C (e.g., “mean T sept 2017-2020 in Figure 6A).

• Lines 770–772: The sentence about “nutrient-bearing aerosol from Boreal WF” is out of place here! If it is relevant to include in the article it should probably be moved to the introduction.

Reply from the authors: We have removed the sentence.

• Line 784: “corresponding to 13% of their annual loading” – is this for both OC and EC?

Reply by authors: Yes, it is. Note that 13% has been changed to 14%.

• Table 4: Rephrase “Means are based on identical time stamps” – I guess you mean that the BB and FF fractions in the table are for the same time periods for all three methods.

Changed from: Means are based on identical time stamps (See Table S3).

Changed to [table 4]: Means are based on identical time periods (See Table S3).

• Table S8: The information presented in this table is unclear, in regards to the data provided. The header should be rephrased to better convey what is shown in the table. The last column “Mean conc. and percentile levoglucosan (ng m-3) (percentile)” is particularly confusing. It would be helpful

to specify what is being shown and why it is relevant, especially given the unusual selection of samples. If necessary, add a short text section could be added before the table to explain why this information is being investigated.

Reply by Authors: We appreciate the suggestion of a short text to explain the importance of Table S8 and have included such text before Table S8. We have also attempted to clarify the content of Table S8 with a new table caption and new headers in the columns of Table S8.

Background information to Table S8:

Added text [Table S8]: The samples with the highest PBAP and BSOA tracer concentrations were collected in the non-heating Season (June to October). A major fraction of these samples experienced elevated concentrations of the biomass burning tracer levoglucosan, as shown in Table S8. This points to the importance of wildfires for observed concentrations of PBAP and BSOA in the Arctic.

Changed from: Table S8: Mean PBAP and BSOA tracer concentrations for those of the top ten concentration samples collected in the non-heating season and with a levoglucosan concentration higher than the long-term annual mean at Zeppelin Observatory (2017 to 2020).

Changed to [Table S8]: Table S8: Mean concentration of PBAP and BSOA tracers for the top ten highest concentration samples (second column). Mean concentration of PBAP and BSOA tracers for those of the top ten samples collected in the non-heating (NH) season and with a levoglucosan concentration exceeding the long-term mean (third column). Mean concentration of levoglucosan for the samples listed in the third column and their percentiles compared to the long-term mean (Fifth column). Zeppelin Observatory, 2017 – 2020.

References:

Garg, S., Chandra, B. P., Sinha, V., Sarda-Esteve, R., Gros, V., and Sinha, B.: Limitation of the Use of the Absorption Angstrom Exponent for Source Apportionment of Equivalent Black Carbon: a Case Study from the North West Indo-Gangetic Plain, Environmental Science & Technology, 50, 814-824, 10.1021/acs.est.5b03868, 2016

Tobler, A. K., Skiba, A., Canonaco, F., Mocnik, G., Rai, P., Chen, G., Bartyzel, J., Zimnoch, M., Styszko, K., Necki, J., Furger, M., Rózanski, K., Baltensperger, U., Slowik, J. G., and Prevot, A. S. H.: Characterization of non-refractory (NR) PM_1 and source apportionment of organic aerosol in Krakow, Poland, Atmospheric Chemistry and Physics, 21, 14893-14906, 10.5194/acp-21-14893-2021, 2021.

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Yus-Díez, J., Bernardoni, V., Močnik, G., Alastuey, A., Ciniglia, D., Ivančič, M., Querol, X., Perez, N., Reche, C., Rigler, M., Vecchi, R., Valentini, S., and Pandolfi, M.: Determination of the multiple-scattering correction factor and its cross-sensitivity to scattering and wavelength dependence for different AE33 Aethalometer filter tapes: a multi-instrumental approach, Atmos. Meas. Tech., 14, 6335–6355, https://doi.org/10.5194/amt-14-6335-2021, 2021.

Zieger, P., Heslin-Rees, D., Karlsson, L., Koike, M., Modini, R., Krejci, R. Black carbon scavenging by low-level Arctic clouds. *Nat Commun* **14**, 5488 (2023). https://doi.org/10.1038/s41467-023-41221-w

Reply to referee 2:

We would like to thank the referee (R2) for his/her thorough and constructive review of our work and for the suggested corrections and improvements to the submitted manuscript.

A very large number of abbreviations are used in the manuscript of which many are introduced already in the introduction. I suggest to reduce the number of abbreviations as it makes the manuscript difficult to read. Please also check that:

Reply by authors: Quite often abbreviations are more commonly known and used than spelling the word out. Also, these abbreviations are used as index and in equations, such as OC_{PBAP}, which does not favor full spelling of the word. PBAP (= Primary Biological Aerosol Particles) is mentioned 30 in the text, BSOA (= Biogenic Secondary Organic Aerosol) 50 times, WF (Wildfires) 74 times, RWC (Residential wood combustion) 33 times. These abbreviations account for 187 words, whereas spelling them out amounts to 567.

CCN (= Cloud condensation nuclei) and INP (Ice nucleating particles) together are mentioned 20 times and we have chosen to drop the abbreviations for these to meet the request from the referee, as for BVOC (Biogenic Volatile Organic Aerosol), mentioned 6 times. We have also spelled out sea-salt aerosol (SSA) and mineral dust (MD) where possible, but kept the abbreviations where appropriate, such as for equations and in relation to equations. We have also dropped the abbreviation for AMS (Aerosol mass spectrometer).

We also changed one sentence:

Changed from: Their study identified three factors dominated by anthropogenic sources [Oxygenated Organic Aerosol (OOA), Arctic Haze (AH), and Primary Organic Aerosol (POA)] and three factors associated with natural emissions [Methane Sulfonic Acid-Related Organic Aerosol (MSA-OA), Primary Biological Organic Aerosol (PBOA), and Biogenic Secondary Organic Aerosol (BSOA)]. These factors exhibited distinct seasonal patterns, with MSA-OA, PBOA, and BSOA dominating in summer, and OOA, AH, and POA in winter.

Changed to [142-147]: Their study identified three factors dominated by anthropogenic sources (Oxygenated Organic Aerosol, Arctic Haze, and Primary Organic Aerosol) and three factors associated with natural emissions (Methane Sulfonic Acid-Related Organic Aerosol, Primary Biological Organic Aerosol, and Biogenic Secondary Organic Aerosol). These factors exhibited distinct seasonal patterns, with the first three dominating in winter and the latter three in summer.

1. all abbreviations are only introduced once (e.g. LRT is explained twice in the abstract).

Reply by authors: We have removed the excess LRT explanation.

2. all abbreviations are actually used later in the text. If not, they are not necessary and just add to difficulty of reading the manuscript.

Reply by authors: We have gone through the manuscript and checked this.

Line 116-152: These paragraphs argue about the importance of the present work, but the authors have chosen a somewhat critical tone towards previous studies to highlight the importance of the

present work. The readers will appreciate this work without the negative tone towards previous studies. I suggest to revise accordingly.

Reply by authors: It is by no means our intension to be critical towards other studies and highlight our own. To document why we wrote the actual paper, we need to demonstrate scientific knowledge gaps, and when possible, refer to other published papers. We have made modifications to the actual paragraphs, as requested.

Changed from: Even short-term, direct measurements of organic carbon (OC) or OM are scarce (e.g., Hansen et al., 2014; Barrett et al., 2015;119 Ferrero et al., 2019) and not suited to establish seasonality, annual mean, or inter annual variability.

Changed to [121-124]: Even short-term, direct, measurements of organic carbon (OC) or OM are scarce (e.g., Hansen et al., 2014; Barrett et al., 2015; Ferrero et al., 2019), limiting our understanding of even basic parameters such as seasonality, annual mean, or inter annual variability.

I suggest to add more information to introduce the source specific tracers in the introduction. This will be helpful for the readers to understand the results and discussion about e.g. ratios of levoglucosan and mannosan.

Reply by authors: We extended the section referred to by the referee (116-152 in original manuscript) to include introductions to the tracers used in the present study. This has also caused rewriting part of the text highlighted by the referee. Concerning the levoglucosan to mannosan ratio, we refer to your next point.

Changed from: Source specific organic tracers measured in the Arctic, include levoglucosan for BB (e.g., Schneidemesser et al., 2009; Fu et al., 2013; Zangrando et al., 2013; Hu et al., 2013a; Yttri et al., 2014; Feltracco et al., 2020), sugars and sugar-alcohols for PBAP (e.g., Fu et al., 2009b; Fu et al., 2013; Feltracco et al., 2020), and different oxidation products of isoprene (e.g., 2-methyltetrols), monoterpenes (e.g., 3-Methyl-1,2,3-butane-tricarboxylicacid) and sesquiterpenes (e.g., β-caryophyllinic acid), for BSOA (Fu et al., 2009a; Fu et al., 2013; Hu et al., 2013). Most of these studies were for short time periods, or a part of the year, largely failing to address seasonal, annual and interannual variability of sources and their impact on the Arctic CA, excepting the one-year study of Yttri et al. (2014), and the multi-seasonal study of Feltracco et al. (2020).

Changed to [148-164]: Source specific organic tracers identified in the Arctic include levoglucosan, mannosan and galactosan (e.g., Schneidemesser et al., 2009; Fu et al., 2013; Zangrando et al., 2013; Hu et al., 2013a; Yttri et al., 2014; Feltracco et al., 2020), which are combustion products of cellulose and hemi-cellulose serving to trace biomass burning emissions (Simoneit et al., 1999). Sugars, sugar-alcohols (here: glucose, fructose, trehalose, arabitol, and mannitol) and cellulose are used for tracing PBAP (Graham et al., 2003; Elbert et al., 2006; Sanchez-Ochoa et al., 2007), with sugar-alcohols typically associated with yeast and fungal spores, and sugars linked to pollen, fern spores and other giant bioaerosol (Graham et al., 2003). Cellulose, a primary component of plant cell walls, is used to trace plant debris (Sanchez-Ochoa et al., 2007). Sugars and sugar-alcohols have previously been detected in Arctic aerosol (e.g., Fu et al., 2009b; Fu et al., 2013; Feltracco et al., 2020), but cellulose has not been reported in these studies. Oxidation products of isoprene (e.g., 2-methyltetrols), monoterpenes (e.g., 3-Methyl-1,2,3-butane-tricarboxylic-acid), and sesquiterpenes (e.g., β-caryophyllinic acid) are all BSOA species previously detected in the Arctic aerosol (Fu et al., 2009a; Fu et al., 2013; Hu et al., 2013). Most studies measuring organic tracers in the Arctic have been limited to short time periods or specific seasons, lacking a comprehensive understanding of the seasonal, annual and interannual variability of sources and their impact on Arctic CA. Notable exceptions are the one-

year study of Hansen et al. (2014) and Yttri et al. (2014), along with the multi-seasonal investigation of Feltracco et al. (2020).

L415-427: Please also explain the significance of the levoglucosan to mannosan ratio.

Reply by authors: Varying L/M ratios have been reported across the world. When sources of L/M are known, such as for prescribed fires, certain studies have used these results to make interpretations about the origin of BB emission at other places around the world measuring these two species. One example is the attempted use to separate softwood and hardwood burnt. With mixing of air masses, possible differences in lifetimes of levoglucosan and mannosan, and differences in various analytical methods applied, one should be careful what to interpret from this ratio. Here we simply demonstrate that the L/M ratio has a seasonal variability with increased levels in winter when RWC dominate compared to summer when wildfires are more frequent. At Zeppelin, the L/M ratio thus has a potential role as a qualifier for these two (RWC vs. WF) different BB emission sources. As the time series at Zeppelin Observatory continues, the L/M ratio is one piece of metadata that can assist in our interpretation of the BB source influencing the site.

We have already expressed this view in the following sentence included in the paper **[451-453]:**

"The levoglucosan to mannosan ratio (L/M) was lower for the NH-season (4.8 ±1.2) compared to the H-season (7.5 ± 1.9) (Fig. 3; Table S6) and might reflect a shift from WF and AWB in the NH-season to RWC in the H-season."

L440-449: Can local influence be completely excluded, in particular for RWC?

Reply by authors: Some local influence of RWC emissions cannot be excluded. RWC at Svalbard is largely limited to cabins at Nordenskiölds land, primarily south of Longyearbyen, and for a few older buildings in the circumference of Longyearbyen. Around 20 cabins are located around Kongsfjorden where Ny-Ålesund is located. There are also a few research stations in the Kongsfjorden basin using wood for heating. It is allowed to burn driftwood, although not recommended due to its sea salt content, having a negative effect on the combustion unit, hence log wood is imported to Svalbard. An overview of the amount of wood burnt at Svalbard is not available (Personal communication: Governor of Svalbard). Although RWC emissions at Svalbard are deemed low, future effort should be made to allocate its contribution.

Added text [473-475]: Notably, Kongsfjorden has around twenty cabins and a few research stations, and wood is used for heating in these facilities when in use. Hence, emissions from these sources cannot be excluded.

In general, it is useful to include standard deviations with average concentrations and contributions e.g. line 568-569.

Reply by authors: We agree that when presenting mean values the standard deviation would be nice to include. The results given in the two actual lines are "up to" and thus not mean values.

Minor comments:

Line 43-45: This introduction is confusing – "lack of long-term observations for many components such as OA" but then it is stated that the exception is eBC and MSA. I suggest to change "many" to "important" or similar.

Reply by authors: This part of the abstract has been removed.

L57-58 "compared to 2017 to 2018, in 2019 and 2020" – please clarify.

Changed from: Intrusions of warm air masses from Siberia in summer caused three- and ninefold increases in 2-methyltetrols compared to 2017 to 2018, in 2019 and 2020, respectively, warranting investigation of the local vs. the long-range atmospheric transport (LRT) contribution, as certain Arctic vegetation has highly temperature sensitive biogenic volatile organic compounds (BVOC) emission rates.

Changed to [53-57]: Warm air masses from Siberia led to a substantial increase in 2-methyltetrols in 2019 and 2020 compared to 2017 to 2018. This highlights the need to investigate the contribution of local sources vs. long-range atmospheric transport (LRT), considering the temperature sensitivity of biogenic volatile organic compounds emissions from Arctic vegetation.

L73-74 more important source to eBC?

Changed from: Both FLEXPART and the PMF analysis concluded that RWC is a more important source than WF.

Changed to [72-73]: Both FLEXPART and the PMF analysis concluded that RWC is a more important source of (e)BC than WF.

L86: Please add references to all statements.

Changes from: These rapid changes affect atmospheric transport and removal of Arctic aerosols (Jiao and Flanner, 2016), aerosol relative source contributions (Heslin-Rees et al., 2020), vegetation, and the carbon cycle.

Changed to [86-88]: These rapid changes affect atmospheric transport and removal of Arctic aerosols (Jiao and Flanner, 2016), aerosol relative source contributions (Heslin-Rees et al., 2020), vegetation and the carbon cycle (Kramshoj et al., 2016).

L95-96: It is not clear to me how the Ng and McFiggans papers support this statement. Please clarify in your response.

Reply by authors: The Hallquist et al. (2009) paper ought to be sufficeint, thus we have removed the Ng et al. (2017) and the Mc Figgans et al. (2019).

L96: These changes may? Otherwise a reference is needed.

Reply by authors: We are not entirely sure what the referee is asking. In line 91 – 96 we list high-latitude sources that have had a documented increase in recent years, or which are predicted to experience a change. The sentence starting on line 96 refers to this, hence: *"These changes in sources are also changing Arctic aerosol physical/chemical properties and hence their climate impact.* Are you requesting a reference for different climate impact of aerosol particles as a function of their physical and chemical properties?

L148: tricarboxylic acid (please add a space before acid)

L149: Hansen et al. ACP 2014 (already a reference) reported monoterpene oxidation products in aerosols collected at two Arctic sites during a full year.

Reply from authors: Yes, we are aware of this and have added Hansen et al. (2014) as one of the references.

Changed from: (…) (e.g., 3-Methyl-1,2,3-butane-tricarboxylicacid) and sesquiterpenes (e.g., β-caryophyllinic acid), for BSOA (Fu et al., 2009a; Fu et al., 2013; Hu et al., 2013).

Changed to [158-160]: (…) (e.g., 3-Methyl-1,2,3-butane-tricarboxylic acid) and sesquiterpenes (e.g., β-caryophyllinic acid), for BSOA (Fu et al., 2009a; Fu et al., 2013; Hansen et al., 2014; Hu et al., 2013).

Changed from: Most of these studies were for short time periods, or a part of the year, largely failing to address seasonal, annual and interannual variability of sources and their impact on the Arctic CA, excepting the one-year study of Yttri et al. (2014), and the multi-seasonal study of Feltracco et al. (2020).

Changed to [163-164]: Notable exceptions are the one-year study of Hansen et al. (2014) and Yttri et al. (2014), along with the multi-seasonal investigation of Feltracco et al. (2020).

L224: Please add information about where standards were obtained.

Reply by authors: This information is already provided in the manuscript in relation to the line that you are referring to, i.e., (Table S1 in Yttri et al., 2021).

[237-239]: "We identified all species based on retention time and mass spectra of authentic standards, using isotope-labelled standards of levoglucosan, galactosan, mannitol, arabitol, trehalose, and glucose as recovery standards (Table S1 in Yttri et al., 2021)."

L355-358: Please include the numbers here to make the comparison more clear. Were the Antarctica measurements part of the present study?

Reply by authors: Lines 344 – 358 (in the original manuscript) is an introduction and an overview of where the various results can be found and is not to contain any results. The results for Zeppelin are compared to those of Birkenes (Norway), Ispra (Italy) and Trollhaugen (Antarctica) in the following section (3.1), e.g.,:

[391-393]:*"The annual mean concentrations ranged from 6.5 to 16.3 ng Carbon (C) m^{-3} (EC) and from 90.3 to 197 ng C m^{-3} (OC), second lowest only to levels observed in Antarctica (1.9 ng EC m^{-3}; 12.2 ng OC m^{-3}) (Table S7) (Rauber et al., in prep.)."*

The measurements from Antarctica were included in the following study to put the levels observed at Zeppelin into context, similarly as we included levels observed at Birkenes and Ispra for the same purpose. The data for Antarctica has not previously been published, unlike those of Birkenes and Ispra. Forthcoming papers, of which the paper by Rauber et al. (in prep.) is the first in line, will describe the actual study in more detail, while the sampling approach is described in association with Table S7.

L363: Time series of EC, OC and sulfate?

Changed from: The interannual variabilities of EC (34%) and OC (38%) were comparable to SO_4^{2-} (40%), which like OC can be either primary or secondary, or of LRT or local origin, and originate from natural as well as anthropogenic sources, and having a time series dating back to 1991 (Platt et al., 2022).

Changed to [388-390]: The interannual variabilities of EC (34%) and OC (38%) were comparable to SO_4^{2-} (40%). Like OC, SO_4^{2-} can have both primary and secondary sources, originate from LRT or local emissions, and stem from natural as well as anthropogenic sources.

L377: more efficient transport of polluted air masses?

Changed from: EC dropped by a factor of two during the H-season compared to the NH-season, due to more efficient transport of air masses to the Arctic in winter (Ottar et al., 1989) and by AH accumulation in winter and spring (Shaw, 1995).

Changed to [405-407]: EC increased by a factor of two during the H-season compared to the NH-season, due to more efficient transport of polluted air masses to the Arctic in winter (Ottar et al., 1989) and by AH accumulation in winter and spring (Shaw, 1995).

L393: greatly. I suggest to change to considerably or significantly

Changed from: The BB tracer levoglucosan decreased greatly from February to March, indicating that (…)

Changed to [422-423]: The BB tracer levoglucosan experienced a significant decrease from February to March, suggesting that OC from fossil sources became more prominent as the AH period progressed.

L415-427: Please also explain the significance of the levoglucosan to mannosan ratio.

Reply by authors: We have already replied to this question (See further up).

L439-440: Of EC?

Reply by authors: The entire chapter is about EC, hence the title *"EC_{BB} and EC_{FF} obtained from radiocarbon measurements and LHS"*. We have added "of EC" to the actual sentence to emphasize this.

Changed from: The weekly maximum BB fraction in Feb. 2017 (81%) was somewhat lower than the extremely high (95 to 98%) daily BB fractions during AH at Zeppelin in 2009 (Winiger et al., 2015).

Changed to [468-469]: The weekly maximum BB fraction of EC in Feb. 2017 (81%) was somewhat lower than the extremely high (95 to 98%) daily BB fractions during AH at Zeppelin in 2009 (Winiger et al., 2015).

L522: isoprene is the BVOC emitted in highest amount, not the most abundant.

Changed from: (…), which are important low-NOx oxidation products of isoprene (Paulot et al., 2009), the most abundant BVOC (500 Tg C yr-1) globally (Williams and Koppmann, 2007), and an important source of BSOA (Hallqquist et al., 2009; Noziere et al., 2015).

Changed to [559-562]: (…) which are important low-NO_x oxidation products of isoprene (Paulot et al., 2009), the biogenic volatile organic compound (500 Tg C yr^{-1}) emitted in highest amount globally (Williams and Koppmann, 2007), and an important source of BSOA (Hallqquist et al., 2009; Noziere et al., 2015).

L533: Please direct the reader to these data.

Changed from: (…) the time series at Zeppelin is delayed by half a month compared to Birkenes, 2021), although concentrations drop by mid-October at both sites.

Changed to [571-572]: (…) the time series at Zeppelin is delayed by half a month compared to Birkenes (Fig. 6 Yttri et al., 2021), although concentrations drop by mid-October at both sites.

L580-584: I suggest to divide into two sentences.

Changed from: With the exceptions mentioned, the mean ratio for the NH-season at Zeppelin agrees with the upper range (0.25 to 0.58) reported by others (Claeys et al., 2010), and thus, relate to the formation mechanism of 2-methylterols outlined by Bates et al. (2014), which shows a 1:2 relationship between cis-ß-IEPOX and trans-ß-IEPOX, accounting for >97% of observed IEPOX, and which are the precursors of 2-methylthreitol and 2-methylerythritol, respectively.

Changed to [619-623]: With the exceptions mentioned, the mean ratio for the NH-season at Zeppelin agrees with the upper range (0.25 to 0.58) reported by others (Claeys et al., 2010). This relates to the formation mechanism of 2-methylterols outlined by Bates et al. (2014), which shows a 1:2 relationship between cis-ß-IEPOX and trans-ß-IEPOX, accounting for >97% of observed IEPOX, and which are the precursors of 2-methylthreitol and 2-methylerythritol, respectively.

L588-592: This part seems to need additional editing. The dashes seem to indicate that this is from a draft version.

Changed from: - If the 2-methyltetrols formation was exclusively abiotic, resulting from atmospheric oxidation of isoprene (Claeys et al., 2004), there would be a racemic mixture of the 2-methyltetrols - This is consistent with the known production of the 2-methylerythritol D-form by plants, algae, and microorganisms (Anthonsen et al., 1976, 1980; Dittrich and Angyal, 1988; Ahmed et al., 1996; Duvold et al., 1997; Sagner et al., 1998; Enomoto et al., 2004).

Changed to [628-632]: If the 2-methyltetrols formation was exclusively abiotic, resulting from atmospheric oxidation of isoprene (Claeys et al., 2004), there would be a racemic mixture of the 2-methyltetrols. This is consistent with the known production of the 2-methylerythritol D-form by plants, algae, and microorganisms (Anthonsen et al., 1976, 1980; Dittrich and Angyal, 1988; Ahmed et al., 1996; Duvold et al., 1997; Sagner et al., 1998; Enomoto et al., 2004).

L631: Larger contribution to PBAP in the Arctic?

Changed from: but LRT likely makes a larger contribution to the Arctic than for more vegetated southerly biomes.

Changed to [669-670]: but LRT likely makes a larger contribution to PBAP in the Arctic than for more vegetated southerly biomes.

L641: Where can the reader see this data?

Changed from: The composition of sugars and sugar-alcohols at Zeppelin and Birkenes varied, reflecting different biomes.

Changed to [679-680]: The composition of sugars and sugar-alcohols at Zeppelin (Table 1) and Birkenes (Table S5) varied, reflecting different biomes.

L740: I suggest to include a more recent reference such as Glasius et al., Atmospheric Environment, 173, 127-141 (2018)

Reply by authors: The suggested reference has been included:

Reference added [777]: Glasius, M., Hansen, A.M.K., Claeys, M., Henzing, J.S., Jedynska, A.D., Kasper-Giebl, A., Kistler, M., Kristensen, K., Martinsson, J., Maenhaut, W., Nøjgaard, J.K., Spindler, G., Stenström, K.E., Swietlicki, E., Szidat, S., Simpson, D., Yttri, K.E. (2017) Composition and sources of carbonaceous aerosols in Northern Europe during winter. Atmospheric Environment, 10.1016/j.atmosenv.2017.11.005.

L793- : Please provide brief information about what was discussed in Zwaaftink et al. compared to the current study.

Changed from: This episode is discussed in detail by Zwaaftink et al. (2022).

Changed to [835 - 837]: This episode was studied by Zwaaftink et al. (2022), combining surface and remote sensing observations and transport model simulations to understand its origin and development, whereas we in the present study focused on its carbonaceous aerosol content.