

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

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 intention 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; 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-tricarboxylic acid) 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, arabinol, 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 Nordenskiöld's 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 sufficient, 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-tricarboxylic acid) 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-NO_x 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 (Hallquist 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 (Hallquist 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-β-IEPOX and trans-β-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-β-IEPOX and trans-β-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.