We thank Referee #2 for her/his comments and suggestions, which helped to improve the manuscript.

Our response is formatted as follows:

Reviewer's comments
Author's reply
Changes to the manuscript.

All page, line, section and figure numbers in bold refer to the original manuscript, all others to the revised version.

Kollner et al present summertime aircraft-based online aerosol chemical composition data from two mass spectrometers, providing both single-particle and bulk composition information, which is quite unique. These novel data were collected as part of the Canadian NETCARE campaign and significantly contribute to understanding of Arctic summertime aerosol composition, knowledge of which is observationally limited, especially when considering non-ground-based measurements. This is an important dataset to publish. Overall, the paper is well-done and contributes significantly to the field. The vertical profiles of aerosol composition and source region analyses using backward air mass trajectories are a good analysis approach to correlate composition with air mass source regions. It is excellent that the authors scaled the ALABAMA data to number concentrations and conducted uncertainty analysis. The supplementary material is comprehensive.

However, I have one major data analysis concern, as described below, about the significant fraction of unexamined single-particle data, the omission of which potentially impacts the presentation and interpretation of the results.
My major data analysis concern regards the approach in the classification of the ALABAMA single-particle mass spectra. As stated on lines 140-141, "the ion marker method classified particles based on the presence of pre-selected species that are of interest." This means that the authors have chosen what chemical species to focus on, which is not necessarily problematic; however, only 54% of the single-particle mass spectra were classified. This means, as shown in Figure 6 and stated in the Figure S9 caption and on line 320, that 46% of the single-particle mass spectra are not included in the discussed data/results. Being nearly half of the ALABAMA data and the fact that this paper is meant to be an overview/summary of the results during NETCARE, I am concerned that the authors may inadvertently be biasing their results by not classifying the remaining data, and I do not consider 46% to be an acceptable fraction of unclassified/ real spectra. Figure S9 shows that at least the majority of these mass spectra are real, and therefore, they need to be classified and discussed. Figure 6 shows that this requires further ion marker searches to classify the particles, and a good starting point for this is the examination of the mass spectra shown in Figure S9. In their previous paper (Kollner et al 2017, ACP), which focused on trimethylamine-containing particles, they included an additional particle type called "K-containing" (potassium-containing). Figure S9 shows the mean mass spectrum of the "Others" (non-classified) particles, and this shows a dominant K+ peak, carbonaceous ions, and sulfate. Figure S9 is also nearly identical to the mean K-containing mass spectra in Figure 4e of Kollner et al (2017, ACP). Since this particle type is consistent with biomass burning (the mass spectrum of which is discussed on lines 441-444), it seems even more important to include a K-

containing, or similar, particle type to classify more of the remaining data. Given the similarity of Figure S9 to biomass burning, it is necessary for these data to be included in the evaluation of the vegetation fire influence portion of the manuscript in particular. How does the PF.N>320 vs vegetation fire PES fraction compare for when all K-containing particles are included, or how does it compare currently for this "Others" category as a starting point? Conducting a K+ search on the remaining mass spectra will also be informative to then examine the mean mass spectra of the still unclassified particles, which can be further classified to potentially unearth another particle type that may provide greater insights. It is possible that by including more of the available data that some of the presented trends may change, or new trends may emerge, and without including more of the available data, I am concerned about the results being biased by the chosen ion markers.

> Potassium was included in more than 60 % of all particles analyzed during the NETCARE 2014 measurements. Moreover, potassium is thus largely internally mixed with other substances, such as TMA, nitrate, and DCA (see modified Fig. 5), making it difficult to draw conclusions on the source regions/sectors of this substance. However, with 22 %, potassium-containing particles make up a large fraction from the group of "Others". We therefore decided to work with this K-containing sub-type (22 %). The analysis of this particle type is presented in Figs. 5, 6f, 10, 12, 13, S11, and S14. The new results are discussed in the respective sections.
>
> Further, we classified the rest (24% of all particles) based on marker species ammonium, methanesulfonic acid, and sulfate and show this classification in the Supplement Fig. S8. By this analysis, the reader can comprehend the composition of the remaining particles, even though these particle sub-groups are not further analyzed. Having done this, 16 % of all particles still remain unclassified. The mean spectrum of "Others" thus changed and is shown in the Supplement Fig. S9. We added a sentence in Sect. 3.1, Lines 315-317:
>
> "24% of all mass spectra are not considered for the further analysis (gray filling in Fig. 5), however, those remaining mass spectra are further sub-classified with marker species sulfate, ammonium, MSA etc. (see Figs. S8 and S9)."

Section 2.5.2 & Figure 3: Why does the category of "Arctic open water" not include the full Arctic Ocean, including the Beaufort and Chukchi Seas? Figure S36 shows some surface influence from this region, so inclusion of only open water north of 73.5 deg means that additional Arctic open water is instead classified as "Alaska" and missed in the "Arctic open water" source sector analysis. The authors could sub-categorize the High Arctic, but the full Arctic Ocean should be considered as "Arctic" for consistency with its geographic definition and for comparison to other work.

In addition, as described below, the authors' definition of "Arctic" as north of 73.5 deg is inconsistent with its geographic definition and norms of other studies, and impacts the source region attribution and discussion. Otherwise, the majority of my other comments are minor.

This is also true with respect to the definition of 73.5 deg north as "Arctic", when the Arctic Circle is at 66 deg 34' N. Given little population north of the Arctic Circle, it would seem more appropriate to either sub-divide the Arctic into two categories, or include 66-73 N in the Arctic category. The categories of Alaska, northern Canada, Europe, Greenland & the

Atlantic, and Siberia extend much, much further north, and therefore, air mass influence still in the "Arctic" (i.e. >66 deg N) would be discussed as being from a further south influence with the categories shown. The full geographic domain of the Arctic should be considered in the source region categorization. This is important since Lines 297-298 state that "The cumulative contribution of all regions outside the Arctic dominated air mass history within the lower troposphere, making up to 97% of the PES", but this is likely partially due to the definition of "Arctic" in this work. For example, if the Beaufort and Chukchi Seas were included in the "Arctic open water" in Figure 8, would the trend be even stronger?

We do not agree that the usage of the word *Arctic* when discussing transport related atmospheric and meteorological processes is being fixed or defined by the definition of the Arctic circle (> 66° N). Earlier studies demonstrated that the Arctic lower troposphere (LT) is characterized by upward-sloping isentropes that form a dome-like structure (Carlson, 1981; Iverson, 1984; Barrie, 1986). This structure later became known as the polar dome (Klonecki et al., 2003; Law and Stohl, 2007). The location of the polar dome boundary, and thereby the transport barrier that isolates the Arctic LT from lower latitudes, is often characterized by the location of the Arctic front (Klonecki et al., 2003; Law and Stohl, 2007).

In our recent study (Bozem et al., 2019, ACP), we applied measurements of trace gas gradients for the identification of the polar dome boundaries. As a result, the polar dome boundary during the NETCARE 2014 campaign was located at around 73.5° N, which is in agreement with a recent study by Crawford and Serreze (2015), demonstrating the more northern location of the Arctic front in summer. However, the location is variable in space and time and thus needs individual analysis for different measurement campaigns. We argue that the definition of the polar dome latitudinal boundary is more appropriate to use when studying atmospheric processes in high latitude regions. Along with this, there is no consensus in literature about the definition of Arctic regions or a convention to define Arctic regions in atmospheric studies. As examples: Liu et al. (2015) and Stohl (2006) defined the Arctic as being north of 70° N; Shindell et al. (2008) used a definition of 68° N; Yang et al. (2018) and Xu et al. (2017) described the Arctic as being north of 66.5° N, whereas Zhu et al. (2020) and Stohl et al. (2013) used > 66° N.

We added Table 2, specifying the latitudinal and longitudinal boundary of the source regions. This table should provide a better overview for the reader and show clearly show that the method used here to define Arctic region is specifically applied for the July 2014 measurements.

Figure 4b: What do the areas "not categorized in the selected geographic regions" correspond to? At >3 km, this corresponds to >20% of the data, and yet Figure 2 shows that all of the Northern Hemisphere north of 25 deg, except northern Africa, should be included in the source sector categorization.

"Not categorized" corresponds to regions in Mexico and northern Africa, as given in Fig. 3. For the second period, data on aerosol composition above 3 km are sparse. Thus, the further data analysis will not consider altitudes above 3 km.

We added the following sentences in Sect. 3.3.1:

> "Regions that were not categorized (Mexico and northern Africa both north of 25° N, see Fig. 3) contribute significantly to air mass history at altitudes above 3 km (Fig. 14). However, our data on aerosol composition at altitudes above 3 km are sparse, with the result that our further data analysis is limited to altitudes below 3 km."

I'm also concerned about the use of 15 days of backward trajectories in terms of the associated spatial uncertainties in these trajectories, which I couldn't find to be addressed. Have the authors done analysis to validate or assess the uncertainty in the trajectories?

> We did not perform a dedicated uncertainty analysis, in terms of analysing the variability given by the meteorology, e.g., through using meteorological ensemble data. However, our approach provides a certain uncertainty analysis with respect to the release of the individual tracer particles. In total, we released an ensemble of 20,000 individual air particles (i.e., trajectories) in each 10-min flight interval within a 3-dimensional air volume with maximum and minimum boundaries in space given by the flight track. Thus, we do not analyse individual trajectories, but the averaged (correct: statistically anticipated) value of 20,000 individual calculations, which together provide a certain probability distribution.
>
> Of course, 15 days is a long time period where many bifurcation points can alter the path of the trajectories. However, this is partially considered in the analysis by the vast ensemble of trajectories. More so, prior Arctic studies give us confidence that our analysis, using 15-days backward trajectories, is well suited to study the Arctic summertime composition with respect to transport processes (e.g., Hirdmann et al., 2010; Stohl, 2006; Stohl et al., 2007)

Additional comments: Intro, Lines 58-61: This sentence states that the references provided are "recent studies demonstrat[ing] gas flaring and shipping", but several of these studies examined oil and gas extraction activities and not specifically flaring.

> We updated the list of references and separated references including results on oil/gas extraction and flaring:
>
> "Regarding high-latitude anthropogenic sources, recent studies demonstrated oil/gas extraction activity and shipping to significantly impact the lower tropospheric BC, organic, and sulfate aerosol burdens (e.g., AMAP, 2010; Eckhardt et al., 2013; Breider et al., 2014; Ferrero et al., 2016; Gunsch et al., 2017; Creamean et al., 2018; Kirpes et al., 2018). The contribution of high-latitude flaring emissions to Arctic BC concentration is controversially discussed. While some studies suggest gas flaring to be an important source of BC, particularly in winter and spring (Stohl et al., 2013; Xu et al., 2017; Leaitch et al., 2018; Zhu et al., 2020), others provide evidence that flaring plays a minor role (Winiger et al., 2017, 2019)."

Further, the cited Winiger et al 2017 (PNAS) study shows flaring to be a minor BC source, and the more recent Winiger et al 2019 (Sci Adv, not currently cited in this work) also showed that the BC isotopes were not consistent with flaring. Therefore, this sentence needs to be revised to more accurately reflect the results in the manuscripts cited.

As mentioned above, we modified the sentences according to your suggestion as follows:

"The contribution of high-latitude flaring emissions to Arctic BC concentration is controversially discussed. While some studies suggest gas flaring to be an important source of BC, particularly in winter and spring (Stohl et al., 2013; Xu et al., 2017; Leaitch et al., 2018, Zhu et al., 2020), others provide evidence that flaring plays a minor role (**Winiger et al., 2017, 2019**)."

Intro, Lines 64-66: A missing and important summertime Arctic airborne study is ARCTAS-B campaign, which included flights over the Canadian Arctic during July 2008. See Matsui et al (2011a&b, JGR), which also included SP2 BC data and air mass source analysis, making it well-suited to also be considered by the authors for comparisons to the results herein.

We added the references Matsui et al. (2011a,b) in the Introduction and the Discussion as follows:

Introduction:

"Boreal fires and to a lesser extent anthropogenic activities in North America and northern Eurasia can strongly influence the organic aerosol burden in the summer Arctic free troposphere (FT) (Hirdman et al., 2010; Schmale et al., 2011; **Matsui et al., 2011a;** Lathem et al., 2013; Breider et al., 2014)."

"Sulfate concentrations in the summertime Arctic FT are largely influenced by anthropogenic sources in northern Eurasia, North America, and East Asia (Shindell et al., 2008; Hirdman et al., 2010; Kuhn et al., 2010; Bourgeois and Bey, 2011; Schmale et al., 2011; **Matsui et al., 2011a;** Breider et al., 2014; Yang et al., 2018; Sobhani et al., 2018)."

"In particular, airborne studies that attribute aerosol physical and chemical properties to sources are sparse, especially in summer (Radke and Hobbs, 1989; Brock et al., 1989; Paris et al., 2009; Schmale et al., 2011; Quennehen et al., 2011; **Matsui et al., 2011a, b;** Kupiszewski et al., 2013; Creamean et al., 2018)."

Discussion Sect. 3.2.3:

"This is in agreement with previous Arctic measurements of BC (or EC) at different seasons (e.g**., Matsui et al., 2011b**; Winiger et al., 2019), demonstrating particularly low concentrations during summer compared to the rest of the year. **Matsui et al. (2011b)** could show the influence of Asian anthropogenic emissions on the BC concentration in Arctic summer, in line with our results during the Arctic air mass period."

"It has previously been demonstrated that anthropogenic emissions in northern Eurasia and East Asia contribute to enhanced sulfate concentrations in the summertime Arctic FT (e.g., **Matsui et al., 2011a;** Breider et al., 2014; Yang et al., 2018). "

Discussion Sect. 3.3.2:

"It is thus conceivable that this transport pathway was linked to intensive particle wash-out events (Garrett et al., 2010, 2011; **Matsui et al., 2011a**; Browse et al., 2012; Sato et al., 2016)."

"This finding is in line with modeling and observational studies (Hecobian et al., 2011; Brock et al., 2011; Lathem et al., 2013; Kondo et al., 2011; **Matsui et al., 2011a**; Breider et al., 2014), reporting on the response of nitrate, ammonium, and organic concentrations in the Arctic on the transport of large fire emissions from sub-Arctic regions."

"Consistently, it has previously been shown that the sulfate burden in the summertime Arctic FT is dominated by transport of a wide variety of anthropogenic emissions in northern Eurasia, North America, and East Asia (e.g., Shindell et al., 2008; Hirdman et al., 2010; Kuhn et al., 2010; Bourgeois and Bey, 2011; Schmale et al., 2011; **Matsui et al., 2011a**; Breider et al., 2014; Yang et al., 2018; Sobhani et al., 2018)."

Intro, Lines 66-68: Please consider revising this sentence. The ground-based studies can report the influence of long-range transport on the BL composition, but the strength of this work is the examination of the varying influence across the vertical column.

This sentence had been removed.

Methods, Lines 156-157: This needs to be rephrased because scaling to number concentrations allows quantitation but it does not "allow for assigning particle types to different sources" as stated here, as chemical composition and air mass trajectory analysis, not the concentrations themselves, are used to assign sources.

We modified this sentence as follows:

"In the following, we present the conversion of unscaled ALABAMA measurements into quantitative particle number concentrations."

Figure 4 caption: Please add the dates of the Arctic and southern air mass periods in the caption for clarify and also refer the reader in the caption to Figure S1 for flight maps for context.

Done.

Figure 5: Please clarify whether an air mass can correspond to more than one source sector, as this seems quite possible.

Yes, air mass history is typically influenced by different sources; in particular, if source sectors are in close proximity. We added Section 9 in the Supplement to show comparisons of PES fractions between different source sectors for the first period (Figs. S37a-c) and for the second period (Figs. S37d-f). It is obvious that air masses with high Arctic open water PES fraction are largely isolated from other sources (Fig. S37a); whereas anthropogenic sources (industrial and populated areas) often contribute on either side to air mass history (Figs. S37 c and f). However, a pre-processing of the PES fractions was applied if possible, in order to differentiate different source sector contributions from each other.

We added the following sentences in Sect. 2.5.2:

"Different source sectors can contribute to air mass history within the 15 days prior to sampling. The Supplement Sect. 9 shows comparisons of PES fractions between different source sectors. It was found that air masses with high Arctic open water PES fraction are largely isolated from other sources; whereas anthropogenic sources (industrial and populated areas) can contribute on either side to air mass history by their close proximity. However, a pre-processing of the PES fractions was applied, if possible, in order to differentiate different source sector contributions from each other. For example, we could separate contributions of vegetation fires to air mass history from anthropogenic sources. Further details can be found in the Supplement Sect. 9."

Please also clarify what is meant by: "The PES above vegetation fires refers to the 1-5 km vertical range." Are air masses than pass within the BL above a fire included? This is confusing as worded.

Air masses that were injected between 1 and 5 km (so-called footprint layer) above vegetation fires are included in the analysis. Injection heights of vegetation fires typically vary with fuel type, temperature etc. Several studies show injection heights for boreal fires typically between 1 and 5 km. We discussed this topic briefly in Sect. 2.5.1 and in more detail in the supplementary material (Sect. 2.1).

The caption of Figs. 7 and 14 had been changed as follows:

"The FLEXPART derived contribution is expressed as a fraction of the potential emission sensitivity (PES) in the model domain lowest vertical level (0 – 400 m, except for vegetation fires) over a 15-days backward simulation. A vertical injection layer between 1 and 5 km is applied for vegetation fires (see details in Sect. 2.5.1)."

Lines 278-279: Based on Figure 4a, it appears that Siberia should be the largest contributor at >3 km, but that isn't mentioned here.

We added the following sentence:

"The contribution of Siberian regions to air mass history is highest in altitudes above 3 km."

Lines 297-298: Figure 4b shows 30% Arctic in the BL, which seems to contradict the 97% outside of the Arctic quoted in this sentence.

We agree. This is confusing as worded. We removed the part "making up to 97% of the PES".

Lines 303-304: It is stated that "the contribution of air masses that had resided above Arctic open water was significantly lower during the southern air mass period (Fig. 5b), compared to the Arctic air mass period." However, it looks pretty similar at <200 m in Fig 5b.

We changed the sentence as follows:

"Also, the contribution of air masses in the lower FT that had resided above Arctic open water was significantly lower during the southern air mass period above (compare Figs. 7b and 14b), compared to the Arctic air mass period."

Line 324: Note that the mass spectra show oxalate, malonate, and succinate, rather than oxalic acid, malonic acid, and succinic acid (i.e. paired cation is not definitively known through the mass spectra – or at least it isn't discussed). Please use the appropriate wording throughout.

The ALABAMA mass spectra typically show ion fragments of the analyzed particles, owing to the high energy laser-ablation process. We thus use laboratory measurements to analyze the fragmentation pattern of atmospheric compounds. The laboratory study by Silva and Prather (2000) demonstrated that mass spectra of carboxylic acids show deprotonated negative parent ions, such as oxalate for measured oxalic acid. We added the following sentence for clarification:

"It should be noted that the ALABAMA detects oxalate-, malonate-, and succinate ions that most likely originate from oxalic, malonic, and succinic acid, respectively (Silva and Prather, 2000)."

Line 325: Clarify here and elsewhere that you are referring to number percentages, since mass fractions are more commonly reported in the literature.

We added the following two sentences:

Lines 206-207: "To note, the following use of the word *fraction* always refers to the number fraction measured by the ALABAMA."

Lines 314-315: "To note, percentages given in this study always refer to number percentages measured by the ALABAMA."

Also, clarify whether 86% here refers to oxalate being present in 86% of the DCA-containing particles, as this is not clear as stated here, and please clarify similar wording throughout.

This was changed as follows:

"The term particulate DCA implies the presence of oxalic, malonic, and/or succinic acid (see Table 1) with oxalic acid as most abundant (in 86 % of DCA particles), followed by succinic acid with 41 % of DCA particles and malonic acid with 38 % of all DCA particles (not shown)."

Line 328 & Fig S8: To aid in the interpretation of this sentence, please label chloride and nitrate in this figure.

Done.

Figure 8: This figure and other similar figures show really nice analysis!

Thanks.

Lines 350-351: This sentence refers to air residence time but I don't see time in Figure 8, so I can't see how to evaluate this statement.

Thanks for making us aware of this mistake. All respective figures show the PES fraction on the x-axis. We modified this sentence as follows:

"Particulate TMA was predominantly abundant when the air resided for more than 50% of the 15 days (PES fraction) prior to sampling over Arctic open water areas (Fig. 8)."

All respective figure captions in the main document and supplementary part had been changed accordingly.

Lines 377: This statement is strange because the previous ground-based studies weren't sampling the FT, so this seems to be an unnecessary statement. It can either be removed, or the authors can simply refer to the ground-based data as being less influenced by long-range transport, whereas this work sampled both the BL and FT, which is a strength.

The sentence had been deleted.

Line 427: This reference seems to be missing from the reference list.

The reference is listed in line 1164.

SI Section 1. Definite hit rate for the non-single-particle mass spec reader.

We added the following sentences in the SI:

"The instrument hit rate is defined as the number of particles that are successfully ionized by the ablation laser and that create a mass spectrum relative to the number of laser shots (Clemen et al., 2020). The triggered shot requires that the particle velocity was prior successfully determined and that the laser was ready to shoot (Brands et al., 2011). The maximum shot repetition rate of the Nd:YAG laser was set to 12 Hz."

Fig S8: Please clarify whether this statement means that 60%, by number of the sea spray aerosol particles containing at least one DCA, as the statement is confusing as written.

The caption had been modified as suggested:

"Expanded mean anion spectrum of 60 % (by number) of the sea spray particles containing at least one DCA."

Figures S11 and S14: Add missing a and b labels to figure to agree with caption.

Done.

Figure S18: This seems to show that DCA-containing particles were not observed for the southern air masses. Is that correct?

We see a clear vertical trend for DCA fraction during the first period. However, this is not the case for the second period. A reason might be that meteorological conditions during the second period led to more mixing between the BL and FT. We thus decided to discuss the DCA abundance in relation with the first period, where the separation between BL and FT air is clearer.

SI Equation 5 (page 24): I believe there is a typo here, as the definition of uncertainty for binomial statistics should be: sigma = sqrt((PF(1-PF))/N). If this typo impacts the data uncertainties shown, then please fix throughout.

The equation you noted above is the same as Eq. 5 in the SI, because 1/sqrt(N)= sqrt(N)/N.

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