We thank Referee #1 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.

Köllner et al. provide a detailed summary on Arctic boundary layer and lower free tropospheric aerosol composition during the summer using multiple analytical methods, satellite data, and air mass trajectory analyses. Two main air sectors were defined during their observations including Arctic air from the north and southerly transport from lower latitudes. Sources reported include wildfire, industrial, and other combustion sources in addition to marine sources such as sea spray. While this detailed information fills a key gap in aerosol observations during the Arctic summertime, there are a few issues that should be addressed prior to publication as delineated below.

The abstract is essentially a list of findings and contains no information on how the current work ties into broader implications for Arctic climate or why these results are novel. The authors should consider adding a couple sentences to demonstrate the importance of their work.

We added the following sentences in the abstract:

"Aerosol particles impact the Arctic climate system, both directly and indirectly by modifying cloud properties, yet our understanding of their vertical distribution, chemical composition, mixing state, and sources in the summertime Arctic is incomplete. In-situ vertical observations of particle properties in the high Arctic, combined with modeling analysis on source attribution are in short supply, particularly during summer."
[…]
"Our findings improve our knowledge of mid-latitude and Arctic regional sources that influence the vertical distribution of particle chemical composition and mixing state in the Arctic summer."

The authors provide a very comprehensive, detailed account of their results, but how their findings fit into the bigger picture is not apparent. A synopsis of why assessing the aerosol composition to this level of detail is needed to provide a clear picture on the importance of such measurements. For example, why do we care about knowing the abundance of DCA, sulfate, BC, and organic aerosols, specifically? What has previous work elucidated in terms of these specific aerosols and their impacts on radiation and cloud formation? What observations exist and why are the limited? What are models lacking that require data such as those from the current work? These are key questions for Arctic aerosol-cloud-radiation

studies that should be elaborated upon to motivate the purpose of this work. Surely, any Arctic aerosol scientist would know why this work is important; however, to emphasize the importance to a larger crowd (and, say, a cloud physicist looking for information on why aerosols are important for clouds), a broader context and implications should be included in the introduction and revisited in the conclusions.

We re-formulated and re-structured the introduction, such that the impact of particle composition and mixing state on aerosol-radiation (ARI) and aerosol-cloud interactions (ACI) is discussed:

[…]
The coupling between aerosol particles, clouds, and radiation strongly depends on particle chemical composition and mixing state (e.g., Haywood and Boucher, 2000; Boucher et al., 2013). Light-absorbing aerosol, such as black carbon (BC) and mineral dust, can impact the regional Arctic climate by a combination of different aerosol-radiation processes. First, light-absorbing particles when deposited on snow and ice can lead to increased sea ice melt by reducing the snow/ice albedo (e.g., Hansen and Nazarenko, 2004; Flanner, 2013; Jiao et al., 2014; Schacht et al., 2019). Second, the presence of aerosol in Arctic tropospheric layers leads to cooling of the surface beneath by absorbing incoming solar radiation and by reflecting radiation back to space. The result is an increase in tropospheric stability (e.g., Flanner, 2013; Schacht et al., 2019). Third, increased radiative forcing by black carbon in mid-latitudes leads to increasing meridional temperature gradient and subsequent increased northward heat transport (e.g., Sand et al., 2013a, b). However, recent studies show that the magnitude of aerosol-radiation interactions of BC is largely dependent on the particle mixing state (Bond et al., 2006; Kodros et al., 2018). In contrast to light-absorbing aerosol, scattering aerosol species, such as sulfate, of both anthropogenic and biogenic origin, exert a net negative shortwave radiative forcing on the Arctic surface (Quinn et al., 2008; Yang et al., 2018). Overall, modeling studies focusing on aerosol radiation interactions demonstrate that reductions in Arctic anthropogenic aerosol (mainly BC and sulfate) likely contributed to the observed Arctic surface warming in recent decades (e.g., Shindell and Faluvegi, 2009; Najafi et al., 2015; Acosta Navarro et al., 2016; Breider et al., 2017).
Aerosol particles, serving as nuclei for water condensation or nucleation of the ice phase, are fundamental to cloud formation. The effects of these particles on clouds are important drivers of the Arctic surface energy budget. It is known that the net radiative effect of Arctic low-level clouds varies significantly with season. Arctic low-level clouds warm the Arctic surface through most of the year. However, for a short period in summer when the incoming solar radiation is maximum over regions with a low albedo, clouds exert a negative radiative forcing on the Arctic surface (Intrieri et al., 2002; Shupe and Intrieri, 2004). Particularly, in the summertime pristine Arctic environment with low aerosol concentrations, cloud formation and properties are sensitive to the available cloud nuclei concentration (Mauritsen et al., 2011; Moore et al., 2013; Leaitch et al., 2016). Particle sources, formation, and atmospheric processing control particle composition, mixing state, and size distribution, which in turn significantly impact the ability of particles to act as cloud nuclei (e.g., Junge and McLaren, 1971; Haywood and Boucher, 2000; Dusek et al., 2006; McFiggans et al., 2006; Moore et al., 2011; Martin et al., 2011; Boucher et al., 2013). In the summertime pristine Arctic boundary layer (BL), marine emissions can contribute significantly to low-level cloud nuclei concentrations (Orellana et al., 2011; Leaitch et al., 2013; Willis et al., 2016; Burkart et al., 2017; Dall'Osto et al., 2017, Baccarini et al., 2020), whereas the episodic transport of anthropogenic and biomass burning aerosol from southern latitudes can have a crucial

impact on cloud formation and cloud properties (e.g., Moore et al., 2011; Zamora et al., 2016; Coopman et al., 2018; Norgren et al., 2018). Along with long range transport and aerosol aging processes, oxygenation of the organic aerosol enhances aerosol's hygroscopicity and thus the ability of the particles to form cloud droplets (e.g., Furutani et al., 2008; Jimenez et al., 2009; Chang et al., 2010; Moore et al., 2011). Together, it is important to know the particle composition, mixing state, and size distribution as well as related sources and formation processes to accurately predict impact of aerosol on the Arctic climate system."

[…]

"DCA are highly water-soluble, thus, the presence of particulate DCA can result in a more hygroscopic aerosol population, which can affect cloud formation and cloud properties (e.g., Giebl et al., 2002; Ervens et al., 2004; Abbatt et al., 2005; Chang et al., 2007). Besides the importance for aerosol-cloud interactions, little is known on the vertical distribution of DCA in Arctic aerosol and on its related sources."

[…]

"Although considerable advances have been achieved in recent years, the majority of our current understanding of aerosol properties in the summertime Arctic is obtained from ground-based and shipborne measurements. 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). However, detailed knowledge on the vertical structure of aerosol composition and mixing state as well as on related particle sources, formation, and aging processes are necessary to have a predictive understanding of our Arctic climate system; yet, the vertical distribution of summertime Arctic aerosol is not well represented in Arctic models (e.g., Quinn et al., 2008; Moore et al., 2011; Eckhardt et al., 2015; Sato et al., 2016; Sand et al., 2017; Willis et al., 2018, Abbatt et al., 2019). Our study thus focuses on processes and sources controlling summer Arctic aerosol, using vertically resolved measurements of aerosol properties and trace gases, together with Lagrangian air mass history analysis. To our knowledge, this is the first comprehensive source attribution study of summertime Arctic aerosol composition, combining airborne single particle and bulk chemical composition methods, with focus on the vertical structure."

While the authors do compare their findings a couple of times to Schmale et al. (2011) and reference others like Shaw et al. (2010), Kawamura et al. (2012), and Leaitch et al. (2018), the comparisons are quite limited, even to the extent where they state "This finding is consistent with previous ground-based and shipborne studies…" and only provide references but not what the findings from those studies. In order to put their results into the context of others, and demonstrate why their results fill key observational gaps, a mentioning of how their results compare to other analogous studies by briefly describing what those studies concluded is needed. Also, other key studies such as those by Quinn et al. (2002, 2009) and Winiger et al. (2019), to name a few, that describe aerosol composition at various sites throughout the Arctic should be included in the discussion and comparison of results.

Studies by Quinn et al. (2002) and Winiger et al. (2019) were added in the Introduction and Discussion as follows:

Introduction:

"The summertime Arctic BC burden is mainly controlled by vegetation fires, whereas anthropogenic sources contribute less to the overall transport of BC to Arctic regions in summer (Bourgeois and Bey, 2011; Stohl et al., 2013; Breider et al., 2014; Xu et al., 2017; **Winiger et al., 2019**; Zhu et al., 2020)."

"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**)."

Discussion Sect. 3.2.3:

"This is in agreement with previous Arctic measurements of BC (or EC) across seasons (e.g., Matsui et al., 2011b; **Winiger et al., 2019**), demonstrating particularly low concentrations during summer compared to the rest of the year."

Discussion Sect. 3.3.2:

"This result is in line with earlier studies, analyzing the source attribution of BC (or EC) in the summertime Arctic and suggesting a dominant influence of biomass burning on the BC (or EC) burden in summer (e.g., Hirdman et al., 2010; Bourgeois and Bey, 2011; Stohl et al., 2013; Breider et al., 2014; Xu et al., 2017; Sobhani et al., 2018; **Winiger et al., 2019**; Zhu et al., 2020)."

"Given that the majority of nss-nitrate-containing particles were internally mixed with potassium (see Sect. 3.1), we have additional indications for their biomass burning origin (Silva et al., 1999; Hudson et al., 2004; Pratt and Prather, 2009; Pratt et al., 2011; **Quinn et al., 2002**)."

"Earlier measurements show that background concentrations of ammonium and nitrate are generally low in Arctic summer (**Quinn et al., 2002**; Kuhn et al., 2010; Chang et al., 2011; Schmale et al., 2011; Quennehen et al., 2011; Hamacher-Barth et al., 2016; Lange et al., 2018), which we can confirm under the pristine conditions during the Arctic air mass period (Fig. 10)."

We added more comparisons and more detailed comparisons as follows:

Discussion Sect. 3.2.3:

"These results are in line with findings of an Arctic airborne study by Schmale et al. (2011), who demonstrated the presence of low-volatility highly oxygenated organic aerosol in the summertime Arctic FT that was transported over long distances from lower latitudes, irrespective of the source sector and regions. The molecular identity of the observed organic aerosol in Schmale et al. (2011) is not known. Direct observations of DCA in Arctic regions exist, however, up to now confined to measurements in the BL (e.g., Shaw et al., 2010; Kawamura et al., 2012; Leaitch et al., 2018). These earlier studies suggest summer minimum concentrations of carboxylic acids, due to a combination of diminished transport of precursors and more efficient aerosol wet

removal compared to other seasons. We provide new data on the vertical profile of DCA. It is demonstrated that a significant fraction of DCA was present in the summertime Arctic FT, influenced by long range transport from sources outside Arctic regions. Besides the presence of particulate DCA in the FT, DCA-containing particles internally mixed with TMA and MSA (see Sect. 3.1) were largely present in the stable stratified BL (Fig. S12). Further, these particles were detected when the residence time within the Arctic was high (Fig. S13), indicating sources of particulate DCA in the Arctic. This finding is consistent with previous ground-based and shipborne studies (e.g., Kawamura et al., 1996; Kerminen et al., 1999; Kawamura et al., 2010, 2012), partly linking the abundance of DCA in the summertime Arctic to regional sources."

"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). In line with this, Schmale et al. (2011) observed an increasing sulfate fraction and decreasing organic fraction along with increasing anthropogenic influence from southern latitudes."

Discussion Sect. 3.3.2:

"Earlier measurements show that background concentrations of ammonium and nitrate are generally low in Arctic summer (Quinn et al., 2002; Kuhn et al., 2010; Chang et al., 2011; Schmale et al., 2011; Quennehen et al., 2011; Hamacher-Barth et al., 2016; Lange et al., 2018), which we can confirm under the pristine conditions during the Arctic air mass period (Fig. 10). However, our vertical profile measurements during the southern air mass period (Fig. 15) show that episodic and localized transport of vegetation fire emissions can perturb background concentrations of nitrate, ammonium, and organic matter. 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. Also gas-phase measurements demonstrate the large input of ammonia (precursor gas of particulate ammonium) by vegetation fire emissions to the summertime Arctic lower troposphere (Lutsch et al., 2016)."

"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)."

Specifically, for the air mass source assessment, a more direct link to Stohl (2006) is warranted, given that study evaluated air mass sources of the Arctic using FLEXPART in detail. Are the current results consistent or contradictory to previous work?

We added more detailed comparisons to Stohl (2006) and Klonecki (2003) in Sections 3.2.1 and 3.3.1:

[…]
"Air mass history during the first period reflects the concept of isentropic transport to Arctic regions during summer. **Klonecki et al. (2003) and Stohl (2006)** provide comprehensive analyses on the role of transport pathways into the Arctic troposphere across seasons. It was found that the Arctic summertime lower troposphere is quite isolated from southern latitude sources as diabatic low-level transport into the polar dome is largely absent. This is in line with our results. We found that the near-surface regions are largely isolated from the rest of the atmosphere, whereas regions aloft are episodically influenced by air originating from southern latitudes (Fig. 7a)."
[…]
"However, contributions from southern latitude regions, i.e. Europe, Siberia, northern Canada, Greenland, and the Atlantic Ocean, increased with altitude due to enhanced quasi-isentropic transport from these regions into the high Arctic (Fig. 7a). The abovementioned studies by **Stohl (2006) and Klonecki et al. (2003)** demonstrate that emissions in Siberia, Europa, and Asia can influence Arctic summertime composition in altitudes above the BL, which is consistent with our results."
[…]
"This period is referred to as southern air mass period, since air mass history shows the prevalence of air masses originating from southern latitudes. Lagrangian air mass history analysis suggests a pronounced impact of southern latitude sources on Arctic composition (Fig. 14). The cumulative contribution of all regions outside the Arctic dominated air mass history within the lower troposphere (Fig. 14a). By comparing the synoptic situation during the southern air mass period with climatological mean (see Supplement Sect. 3), we found that the presence of the low-pressure system led to a significantly anomalous synoptic situation. This finding explains the discrepancy between our results on air mass history during the second period and results on Arctic transport climatology by **Stohl (2006)**, revealing the largely unperturbed Arctic lower troposphere during summer."

The results and discussion section seems a bit fragmented as the discussion goes back and forth between the north and south influences, and describing single particle composition for both is combined. Perhaps the entire section would be easier to follow if all data (composition, FLEXPART, and satellite data) were discussed in tandem but organized by source region (north versus south).

Thanks for the helpful suggestion. We re-structured the "Results and discussions" section as follows:

3.1 Single particle chemical composition
3.2 Arctic air mass period
    3.2.1 Meteorological overview and air mass history
    3.2.2 Arctic marine influences on particle composition
    3.2.3 Long range transport influences on particle composition
3.3 Southern air mass period
    3.3.1 Meteorological overview and air mass history
    3.3.2 Vegetation fire and anthropogenic influences on particle composition

Also, why are vegetation fires and anthropogenic sources a separate section? Aren't they technically a south air mass influence sources and aren't they technically long-range transported? Not clear why these are segregated from the section on the southerly sources.

The section 3.3.2 now includes both the discussion on source regions and sectors during the southern air mass period.

Seems like Figure S1 should be in the paper. There is a description of the site locations in text, but it is not clear to those unfamiliar with the area. I would think this is important to show for a clear link between sources and transport pathways, and the type of land they cover.

The figure has been added to the main manuscript.

It was not clear to me until later in the methods why the UHSAS size range was restricted. Please provide the size range of the ALABAMA in section 2.2.1. Also, what is the size range of the SP2 data used?

The ALABAMA size range is given in Sect. 2.2.1. To make it more clear, we added the following sentence in Sect. 2.3:

"For the conversion of the ALABAMA fraction into number concentration (see Sect. 2.4), UHSAS number concentrations in a size range between 320 nm and 870 nm ($N_{>320}$) are used (see Sect. 2.2.1). "

The size range of the SP2 had been added in Sect. 2.2.3.

The statement on the top of page 16 indicates the aerosol data presented are from "sampling outside of clouds". But does that mean the measurements were conducted during cloud free periods or were the aerosols sampled below/above clouds during cloudy periods, or some combination of each? I assume based on the generalized north and south air mass descriptions that clouds were mostly present during the latter half of the study, but it is not clear when exactly the aerosols were sampled during cloudy versus clear conditions. This would be important to discuss, at least briefly, because aerosol composition can be quite different in the Arctic boundary layer during clear versus cloudy conditions.

We analyzed measurements that were conducted outside clouds, irrespective if clouds were present or absent. The selection "sampled outside clouds" was applied because the aerosol inlet was not suitable for in-cloud sampling. Thus, measurements inside clouds were discarded by using data from an under-wing FSSP (Forward Scattering Spectrometer) probe (Leaitch et al. (2016)).

It is correct. The Arctic air mass period was characterized by generally clear skies with occasionally broken stratocumulus clouds, whereas the southern air mass period was characterized by overcast sky (see Sections 3.2.1 and 3.3.1). Further details on the presence and properties of clouds during the NETCARE 2014 airborne study can be found in Leaitch et al. (2016).

However, given that air mass history changed during the course of the study and at the same time we observed different cloud characteristics, it is difficult to study solely the interaction between aerosol composition and clouds. Further, the ALABMA particle counting statistics during the NETCARE 2014 study is not sufficient to conduct a cloud-by-cloud analysis.

During the Arctic airborne campaign ACLOUD in summer 2017, the ALABAMA was operated with an improved setup providing a better counting statistic (Clemen et al., 2020, AMT). In addition, a counterflow virtual impactor was used to sample and analyze cloud residuals. We will thus announce an upcoming publication (Eppers et al, in prep.) that will focus on the coupling between aerosol and clouds during this campaign.

What is the relative contribution of the Arctic versus southern air mass periods? It is obvious for the beginning and end, but what about the 5-day transitional period? The authors could provide the % of each air mass contribution for the entire study at the bottom of page 11.

We added the following information in Sect. 3.2:

"The synoptic conditions changed over the course of NETCARE 2014 from an initial Arctic air mass period (July 4-12, **~26 measurement hours**) to a southern air mass period (July 17-21, **~19 measurement hours**) with a transition in between (Burkart et al., 2017; Bozem et al., 2019)."

We did not consider to study the air mass history of the transitional period, because flying was impeded during the transitional period. Thus, the focus of this paper is on the Arctic and southern air mass periods, which provide airborne measurements on particle chemical composition.

Why is organic:sulfate only shown in select panels/figures (i.e. Figures 10, 11, 14, and 15)? The ratio is also shown as a different color in Figure 11a but should be consistent with the rest of the figures.

We had chosen different colors for different parameters. Dark green refers to the organic-to-sulfate ratio. Pink refers to the sulfate-to-organic ratio.

The organic-to-sulfate ratio as a function of altitude and as a function of residence time over Arctic open water was presented in Willis et al. (2017). To differentiate from our previous work, we will not show these results again.

For consistency, we added the sulfate-to-organic ratio in Fig. 10, Fig. 13b, and Figs. 17b and c to analyze the anthropogenic influence on particle composition, according to Schmale et al. (2011). In Fig. 18b, we added the organic-to-sulfate ratio to analyze the influence of biomass burning on particle composition. The new results and trends are discussed in the respective sections.

For Figure 15, there does not seem to be major differences between the PES sources for fires, population, and industry. The authors describe each separately, but some discussion on why they look so similar is needed.

We re-scaled the y-axes to better view the differences. However, we agree, trends look similar, but differentiation in the absolute values can be seen. For example, highest sulfate concentrations were observed in the Arctic at maximum PES fraction over anthropogenic sources. Further, the organic-to-sulfate ratio was increasing with vegetation fires PES fraction, which is not the case for anthropogenic sources (compare with sulfate-to-organic ratio). We added these points in Sect. 3.3.2.

Similar trends for nitrate, sulfate, organics, and ammonium might be caused by different reasons. First, precursor gases (such as NO_x, SO_4, NH_3, VOCs) are emitted by anthropogenic sources and vegetation fires on either side. Second, during long-rang transport (irrespective of the source region/sector), we would expect that particles are exposed to intensive atmospheric aging processes, which predominantly affect particle composition. Third, different source sectors can contribute to air mass history 15 days prior to sampling (see comment and reply Reviewer#2 and Supplement Sect. 9). As a result, industrial sources cannot be differentiated from populated areas, partly due to their close proximity.

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