High–Arctic aircraft measurements characterising black carbon vertical variability in spring and summer

We would like to thank the referees for their detailed and constructive comments, which helped us to improve our manuscript. While the referee comments are given in **black bold,** our answers are given below in blue letters. Additionally, we added the changes we made in the revised manuscript in **blue bold** letters.

Answers of the authors to anonymous Reviewer#1

Anonymous Review of Manuscript acp-2018-587 GENERAL REMARKS

This paper presents vertical distributions of black carbon aerosol from two aircraft campaign in the high arctic during the spring and summer seasons. They look at BC loading, BC fraction of total aerosol, BC mass median diameter and BC/CO and they run back trajectories. The instrumental methods and the writing are fine though the analysis is a rather qualitative and the conclusions basic. The main finding of the paper, as I read it, was that there are seasonal differences in BC sources within and transported to the arctic in the spring and summer that drive marked differences in BC loadings between the two seasons. I don't think that's a particularly surprising finding and the larger motivations, outlined in the introduction, of connecting these observations to deposition rate to the surface and atmospheric heating are not fully realized. Nor is it a paper that can serve to constrain sources of BC to the arctic; as such I am struggling a bit to define what exactly this paper is about or how it might be used by the community.

The authors would like to point out that the referees raised questions concerning the interpretation of the BC/CO ratio as indicator for wet scavenging and encouraged us to verify the subsequent hypothesis and conclusions. Due to the high number of comments on this specific topic, we prefer to provide here a general and common answer to all reviewers. As a consequence of the above-mentioned reasons, Section 3.4 was substantially modified. The discussion now focusses on the importance of transport patterns on the observed BC concentration. Thus, Figure 7 and Figure 8 were modified. The discussion on potential impact of wet scavenging on BC and BC/CO ratio is now substantially reduced. However, additional analysis of back trajectories, including encounter with clouds, is now presented in the supplementary material.

Specific comments of Reviewer#1

Below are a few specific instances of where the analysis fell short of conclusive for me.

Due to its complexity, some comments were split into two or more parts. This allowed responding to the specific issues and making our answers clearer.

1)The authors posit (in section 3.2) that the data imply that there is increased wet removal of BC in the summer relative to the spring driving the lower concentrations and smaller size distributions. While this may be true I don't follow the logic of how they have isolated wet deposition from dry deposition within the arctic and different convective processes driving transported airmasses in different seasons. It seems to me that a wide variety of combinations of transport pathways and in-arctic processing could lead to the observed trends.

This issue was indicated by all reviewers. We are now aware of the limits of the interpretation based on our data. The discussion on potential wet removal is now supported by an enlarged set of references, while additional back trajectory data were analysed in order to better understand any influence of wet removal. However, separating wet from dry removal or identifying the main removal mechanism is beyond the scope of the present work. Hence, the discussion on wet removal is now substantially reduced due to the absence of strong evidence supporting the hypothesis.

2.1) In Section 3.3.1 the authors say "this suggests that the rBC mass is contained in fewer, larger particles." I don't think that this is actually true and I don't know what conclusion we can draw from this statement without any point of comparison.

We agree with the referee that the statement might be confusing and, in its current form, it appears to indicate that most of the rBC mass was found in larger particles. Although this might be potentially true if there was a pronounced mode of large particles in the size distribution. But, such a situation was not indicated by the analysis of mass—size distributions in Sec. 3.5. The statement was intended to emphasize the smaller number of rBC particles relative to the number of total aerosol compared to upper levels, while the mass-mean diameter was largest in the lowest atmospheric level. As this statement confused the main message, it was removed and the consistent part of the text (P13, L9-16) was reformulated as:

[…] The profiles in Fig. 5 show a homogeneous distribution of rBC with a mean M_{rBC} of 32 ng m^{-3} (IQR: 13–48 ng m^{-3}) that was present in a temperature gradient capped surface layer (level I). This layer held the coldest air encountered with temperatures of 255 K down to 245 K. The observed M_{rBC} across the lowest flight sections matches well with the mean ground–based rBC observations performed in Alert for spring seasons of the years 2011 to 2013 with 30±26 ng m^{-3} (Sharma et al., 2017). Moreover, level I showed the highest average MMD of 204 nm (IQR: 153–250 nm). Such large mean rBC core diameters were already observed at the surface in the European Arctic in spring (Raatikainen et al., 2015; Zanatta et al., 2018) and are distinctly different from freshly emitted rBC (MMD ≈ 100 nm) in urban areas (Laborde et al., 2013). […]

2.2) rBC is stated to be a small fraction of total aerosol (3.8%) but relative to what? I believe that anything over 1-2% is actually a large fraction by number when considering most continental locations and many remote ones as well.

The number fraction of rBC was calculated over the number concentration of total aerosol measured by the Ultra High Sensitivity Aerosol Spectrometer (UHSAS) in the optical diameter range of 85-1000 nm (see P7, L1-10). The referee is actually right, the particle number concentration provided by the UHSAS is a relatively small fraction of the total particles, especially considering that particles below 100 nm constitutes the majority of the aerosol number concentration. As a consequence, the here presented R_{numTA} represent a higher estimation of rBC number fraction. However, our ratios are lower compared to those published in (Kodros et al., 2018; Raatikainen et al., 2015; Sharma et al., 2017), due to their different restriction of the overlapping size range. Nevertheless, R_{numTA} is a useful proxy for assessing the BC relative presence and for isolating any eventual smoke events. The statement now reads:

[…] Although rBC represented a minor component of the total aerosol in the respective size range by number, with an averaged R_{numTA} of 3.8% that was low with respect to higher levels (II-V), rBC mass was comparably high relative to co–emitted CO with a mean R_{CO} of 5.7 ng m^{-3} $ppbv^{-1}$ (IQR: 2.7–10.5 ng m^{-3} $ppbv^{-1}$). […]

The description of BC number fraction in Section 2.2.1 was improved:

[…] In the here presented work, the number ratio of rBC over TA particles, R_{numTA}, was used to identify atmospheric layers influenced by combustion generated aerosol. It must be noted that, due to the restricted detection range of the UHSAS, the TA number is biased low and R_{numTA} must be considered as an upper estimate of the number fraction of rBC particles. […]

2.3) The MMD observed is rather large but not dramatically larger than observed in other studies for some biomass burning emissions, including residential burning or for inefficient fossil fuel combustion. I don't think there are a great number of published size distributions for BC aerosol produced from shipping sources, especially in open waters so they could be from that, for example.

Although the diameter range reported in the present study might not be extreme, it is at the upper end of BC diameters observed under different conditions by (Laborde et al., 2013): traffic BC=100 nm; biomass burning BC = 130 nm; aged BC = 160 nm; continental BC = 200 nm. The comparison with other observations is now included in the text, see answer to comment 2.1. Considering the ship emissions, we cannot exclude a priori the influence of large BC emitted by ships. Nevertheless, the frozen sea precludes a consistent presence of vessels in the Canadian Arctic during spring. As a consequence, the local influence of ship emission may be considered negligible. Nevertheless, part of the text in Section 3.4.1 was modified in order to better treat this topic:

[…] In level I, the highest MMD and R_{CO} might suggest entrainment of pollution from the marginal Arctic which underwent no or inefficient wet scavenging. In fact, high R_{CO} values were already associated in the past with low precipitation during transport to the Arctic (Matsui et al., 2011). On the other hand, longer atmospheric processing undergone by rBC sampled in the highest atmospheric levels might favour wet removal of larger rBC particles due to increased hygroscopicity (Moteki et al., 2012), and thus potentially explains the decrease of MMD from the surface to level V. […]

Section 3.5 was also modified with the same goal, see answer to comment 7.

3. I'm not sure I see the utility of the labored analysis of the BC/CO relationship. As noted by the authors, sources themselves are known to have high variability in this ratio due to different combustion efficiency and the behavior of this ratio, even for a single source, as a function of processing time, is not well characterized. I think it's probably true that if BC and CO are uncorrelated, one can assume that BC has undergone a long period of transport but trying to make sense of a non-zero number to tell you things about unknown sources and unknown processing is hard unless you know something about either the source or the processing. In this case the authors seem to be trying to make statements about both at the same time and I'm not sure the data can really tell us much.

The potential role of BC/CO ratio as indicator of wet removal was largely modified in the current version of the manuscript. Several parts of the text were modified accordingly, especially in Section 3.4. Finally, we might argue that the BC/CO ratio alone is not an appropriate tool to investigate wet removal in remote locations, due to competing factors affecting the relative presence of BC and CO such as dry deposition and source type. A part of section 3.4.1 now reads:

[…] The vertical profiles plotted in Fig. 5 showed a gradual decrease of R_{CO} with altitude, excluding a sharp enhancement in level II. Assuming R_{CO} as a useful indicator of wet removal, we could argue that transport patterns involving the lifting of air might have caused preferential removal of aerosol via wet scavenging. Such an approach was already used in the past, combined with accumulated precipitation along trajectories to investigate the impact of wet scavenging on BC concentration in the Arctic (Matsui et al., 2011). Nevertheless, R_{CO} might be also affected by emission type. The enhancement of R_{CO} in level II might be predominantly caused by entrainment of pollution emitted by different sources. In fact, the low-level air transport from Eastern Russia struck areas influenced by both, biomass burning and gas flaring. Similarly, the second RCO peak observed in level IV might be caused by a change in source types linked to the extended influence of airmasses coming from Northeast Asia. However, R_{CO} did not show any clear correlation with the transport patterns and with the occurrence of liquid and ice clouds along the trajectories (Fig. S2). Due to the complexity of the transport pathways, potentially entraining pollution from different source types, R_{CO} alone proved itself to be insufficient in order to assess the impact of atmospheric processing on BC variability in the Canadian Arctic in spring. The parallel interpretation of R_{CO} and accumulated precipitation along the trajectories might be a better tool to investigate the impact of wet removal on BC presence in the Arctic, as already proposed by (Matsui et al., 2011). However, a complete investigation on the efficiency of BC removal mechanism is beyond the scope of the present work. […]

4. In all of the discussion of the vertical distributions of BC properties in the springtime polar dome I can't summarize what is learned. Looking at Figure 5, I would guess that the springtime arctic in levels I and II is most impacted by local sources because the MMD is large and the BC/CO ratio is relatively high. Local biomass burning sources and possibly ship emissions could have those characteristics but I don't know if the expected sources are large enough to explain the observed loadings. At the end of this section I guess the conclusion is that different sources are contributing to BC loadings at different atmospheric levels but I don't know what the community can do with that information as presented.

Conclusions based solely on the vertical profiles may be premature, we can, nevertheless, learn some interesting things out from Figure 5. Sources at the margins of the Arctic Ocean, such as those related to resource extraction, contribute to BC concentrations in the lower dome – especially in level II. Subsidence and long-term accumulation in the dome may also contribute to the background concentrations. High-Arctic local sources might not represent a strong contributor to BC atmospheric load. This is particularly true when we consider that the highest BC mass concentration and number fraction was found way above the surface due to influence from long-range transport. Moreover, the strong decrease of MMD with altitude implies that atmospheric processing or different source types play an important role on BC microphysics. This is of high interest for radiative forcing

estimations, since the mass absorption cross-section of BC is also a function of particle diameter (Kodros et al., 2018). BC at low levels can especially cause strong surface radiative forcing. Finally, in Section 3.3.1 we aimed to describe all the features observed during the flights and to intrude all the properties, such as MMD and R_{CO}, which will be used for further discussion in Section 3.3.1, 3.4 and 3.5. However, we recognize that the section needs some adjustments in order to improve the readability and understanding of our findings. In order to avoid redundancy and merely bibliographical writing, some parts of the text were shortened (R_{CO} introduction: P13L25-32) or removed (BC-CCN behaviour: P13L10-P14L7).

5. In section 3.4, I don't understand why the authors choose to only run back trajectories for times associated with higher concentration observations. It seems that it would be more conclusive if you were to compare back trajectories across a range of observed BC loadings and see which patterns emerge that are common to the high and low concentrations observations respectively.

Following the comments of anonymous reviwer#1 and other referees, the analysis of back trajectories is now extended to all the considered measured points. The modified approach allowed a better interpretation of the results presented in Section 3.4.

6. In section 3.4.1 on page 19, there is a mention of the data implying "more efficient wet removal in the upper polar dome". But, again, I think it could also be that the convective processes that lifted the air parcel near the sources were different. What does this really tell us about the polar dome that is useful? Similarly, in section 3.5 the authors try to tie differences between layers to differences in local removal within the arctic but I don't know how local effects can be resolved distinct from source differences and transport effects.

All the referees shared similar doubts on the interpretation of the BC/CO ratio results provided by the authors. As a consequence, a substantial number of sections were modified, this applies to Section 3.4.1 and to Section 3.5 as well.

7. On page 23 there is a brief discussion of how MMD might affect the radiative properties of BC in the arctic. Yet the present study doesn't actually report BC coatings or hydrophilicity so this paragraph serves mostly as a literature review which is an odd note to end the results section. Also, just because BC contributes a small amount of aerosol by number doesn't mean it can't be an important absorber. I agree with the authors that it is probably a negligible contribution to extinction but absorption is what you're really interested in and you have very little information about possible absorption enhancements for these particles.

The authors understand the points risen by the reviewer, which are partly shared by the other reviewers. The above-mentioned section was heavily reworked, and now focus mainly on the observed differences in the BC size distribution. A reduced discussion on the potential causes and impact are finally presented, mostly as an outlook. Section 3.4 now reads:

[…] Especially in the Arctic region, where import of BC with an airmass and cloud formation driven removal were found to be a synergistic process (Liu, Fan, Horowitz, & Levy, 2011), it became clear that the BC core size distribution alone is not sufficient to determine the dominant removal process or source type. Even though the present dataset does not allow a complete decoupling of factors controlling the seasonal and altitudinal change of BC diameter, the latter might influence the BC optical properties and subsequent radiative forcing. The mass absorption cross-section of pure BC varies as function of BC diameter (Bond & Bergstrom, 2006), and a shift from ~200 nm to ~250 nm of D_{rBC} causes a decrease of the mass absorption cross section from 6.7 m2 g-1 to 4.9 m2 g-1 (Zanatta et al., 2018). Though the direct BC forcing in the Arctic is dominated by the absolute BC concentration and mainly affected by BC mixing state (Kodros et al., 2018), the change of the BC core diameter is rarely considered in radiative forcing estimations. […]

Minor comments of Reviewer#1

P9, line 7: "southern boarder" should be "southern border".

Changed.

P17, line 9: "Highest values" should be "The highest values"

Changed.

P18, line 31: "The minimal latitude" should be "The minimum latitude"

Changed.

P18, line 34: "Which is reaching up to" should be "Which reaches"

Changed.

Towards the end of the paper the authors have several instances of the acronym MSD instead of MMD. Please make the terminology consistent.

In most part of the manuscript the discussion on the vertical variability of the BC size is based on the vertical profiles of the mean mass diameter (MMD) presented in Figures 4-5-6. In Section 3.5 the discussion is mainly based on the mass size distribution (MSD; Figure 9) averaged within the atmospheric layer of main interest. The use of "MSD" as acronym is, therefore, justified.

P22, line 32: "Likeliness" should be "likelihood"

Changed.

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