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#2

Anonymous Review of Manuscript acp-2018-587 GENERAL REMARKS

This paper presents novel data of black carbon (BC) aerosols obtained by aircraft observations over the Canadian Arctic in summer 2014 and spring 2015. The authors report clear seasonal variations of BC mass concentrations, mass-mean diameter, BC/total aerosols number ratio, and BC/CO ratio, and then try to understand the mechanisms that controlled vertical profiles of BC properties based on the analysis of potential temperature and back-trajectory. The instrumentation is clearly written in the Method section. Because the data of the vertical profiles of BC in the Arctic is very important to assess their radiative impacts and validate numerical models, the novel data presented in this paper is valuable to the community. One aspect of the paper that I thought could be improved is the discussion on the mechanisms controlling vertical profiles. In many parts of the paper, the authors try to interpret the profile variations by "wet removal" or "nucleation scavenging". However, their conclusion is rather speculative and not well supported by data. I understand that the wet removal process is complex (especially ice-cloud scavenging) and difficult to examine, but if the authors can use any supporting data such as precipitation amount or humidity that air masses had experienced along the back-trajectory paths, more quantitative discussion on the observed variability (and also related new insights) may be achieved in the paper.

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#2

P6, Line 5: "avalanche photo-diode" should be "photomultiplier tube"?

The referee is right, the statement was consequently changed:

[…] The incandescence light detector, a photomultiplier tube […].

P7, Line17: "STP" is used here, but defined later in Line 18.

Lines 17 and following now read:

[…] The SP2 and UHSAS shared one bypass line off the main aerosol inlet and sampled with constant 120 ccm (volumetric) and 50 ccm (at standard temperature and pressure, STP), respectively. […] The rBC mass and

number concentrations presented in this study refer to standard temperature and pressure of 273.15 K and 1013.25 hPa, respectively, […]

P8, Line 31–33: I could not fully understand what the authors meant here (i.e., the meaning of "threshold filtering"). Please clarify this sentence.

In the present version of the manuscript no BC or BC/CO threshold was applied to the back trajectories. The text included in P8L31-33 and P9L1-3 now reads:

[…] The time series of trajectories along the track of the aircraft were correlated with in–situ measurement values, in order to relate individual features in the vertical profiles to an ensemble of trajectories (see Sec. 3.4). Due to the potential influence of wild fires and gas flaring on BC presence in the Arctic region, the spatial distribution of gas flaring sites from the ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short–Lived Pollutants) emission inventory (Klimont et al., 2017; Stohl et al., 2015) and active fires from the MODIS level 2 satellite product (Giglio et al., 2003) were also considered for the interpretation of trajectory pathways. […]

P11, Figure 4: I suggest adding BC/CO data to Figure 4 so that it can correspond to Figure 5.

The BC/CO data were mainly used to understand the potential impact of wet removal on BC load and properties. The mutual dependency of the BC/CO ratio on source type and on wet removal, and thus the transport history of BC laden air to different height levels of the polar dome, makes a clear discussion and univocal interpretation of its seasonal variability quite delicate. We preferred to reduce the potential to confuse the reader with the superimposed variability of the BC/CO ratio when shown as function of pressure—altitude in order to rather discuss it along with potential temperature as vertical coordinate in Sec. 3.3. As a consequence, we decided to maintain Figure 4 as it is.

P12, Line 15: I could not understand what the "partitioning of rBC particle size within polluted layers" meant. Please clarify it.

With this sentence we meant to underline that the peaks of rBC mass concentration did not directly imply an enhancement of the rBC number fraction. This might involve different removal mechanisms for different aerosol types during transport. On the other side, other processes as different emission sources might play a role. Due to its speculative character the present statement was removed.

P14, Figure 5 and 6: In Figure 5, are the colored profiles examples chosen from all the spring fights? (because they are indicated as like "YYYYMMDD_F1" in the legend). On the other hand, in Figure 6, the colored profiles are simply indicated as "YYYYMMDD". Is there any difference?

During the spring campaign 2015, but not for the summer campaign 2014, two different flights took place on the same day (8 April). The notations "F1" and "F2" refer to the first and second flight of the day respectively. In order to avoid confusion, the notation "Fx" was removed in the legend of Figure 5 for all flights excluding the two occurred on the 8 April. The caption of Table 1 now describes explicitly the meaning of notations "F1" and "F2".

P15, Line12–13, The low R_numTA does not necessarily suggest that rBC has been depleted by nucleation scavenging, because total aerosols can be also depleted by nucleation scavenging and thus modify the R_numTA value.

The authors agree with the referee's comment. The statement now reads:

[…] Highest variability in rBC abundance and its properties was present between 265 to 277K (level III). At the beginning of the observation period (7 April), low mean M_rBC of 17 ngm^{-3} (IQR: 4–22 ngm^{-3}) was measured, while the two flights on 8 April encountered significantly higher concentrations up to 111 ngm^{-3} (IQR: 65–151 ngm^{-3}). The overall average concentration of BC in level III was 49 ngm^{-3}. The enhancement of black carbon mass concentration, together with a decrease in the mean R_CO and MMD potentially suggests different transport or removal regimes compared to the lower atmospheric levels. The ratios R_numTA and R_CO as well as MMD were significantly below average on the 7 April, while supply of polluted air set in on 8 April and lasted over the course of the observation period with variable intensity. […]

P17, Line 18–19: In Figure 6, the level (III) starts at 294 K. Is it correct?

The statement directly refers to the work of Bozem et al. (2018), who defined the upper limit of the polar dome during the summer NETCARE campaign within the potential temperature range of 299-303.5 K. In order to improve its clarity, the statement was modified as:

[…] The highest investigated level (III) of the atmosphere was characterized by potential temperature above 294 K, and most probably represented a strong temperature gradient separating the polar dome from free tropospheric conditions. In fact, Bozem et al. (2018) identified the upper boundary of the summer polar dome in the potential temperature range of 299-303.5 K. […]

P21, Line 2: "Sec. 6" should be "Sec. 3.3.2"?

Changed accordingly.

P22, Line 19: Sahu et al. (2012) reported mass "median" diameter by applying lognormal fit to the observed size distributions. On the other hand, the present paper reports mass "mean" diameter. For comparison of the data, it should be noted that the definition of "MMD" is different between these studies.

The authors agree with the comment of the referee. After major changes implemented to Section 3.5, the "Sahu et al. (2012)" reference is now only used to underline the sensitivity of BC diameter to different sources. The specific part of the updated Section 3.5 now reads:

[…] BC size distribution was found to be extremely sensitive to both emission type and atmospheric processing. For example, while rBC particle diameters increase when switching from fresh urban emissions to biomass burning emissions (Sahu et al., 2012; Laborde et al., 2013), ship emitted rBC can show a bimodal size distribution characterised by a second mode above 600 nm of D_{rBC} (Corbin et al., 2018). […]

P23, Figure 9: X-axis should be log-scale?

Changed.

P23, Line 4: "asses" should be "assess".

Changed.

P23, Line 9–15: Moteki et al. (2012) discussed coating volume (mass), not shell/core diameter ratio. Because a BC particle with core of 220 nm and shell/core diameter ratio of 1.4 actually has larger coating volume than a BC particle with core of 140 nm and shell/core diameter ratio of 1.6, the results explained here are not contrasting but rather consistent with Moteki et al. (2012).

The referee is right. The coating mass of the considered BC-containing particles was calculated assuming a fixed density of 1 g cm^{-3} according to Moteki et al. (2012). Small BC cores (140 nm of diameter) having a core/shell ratio of 1.6 showed a coating mass of 4.4 fg. On the contrary, the coating mass of larger BC cores (220 nm of diameter) showing a core/shell ratio of 1.4 was quantified to be 9.7 fg. Thus, the coating-core mass dependency presented by Moteki et al. (2012) is here confirmed. However, after major changes implemented to Section 3.5, the discussion on mixing state of BC was largely reduced due to its mere literature review character. Now the mixing state is briefly introduced as a factor influencing the optical properties and radiative forcing of BC as:

[…] 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 et al., 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 and 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 m^2 g^{-1} to 4.9 m^2 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. […]

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