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#3

Anonymous Review of Manuscript acp-2018-587 GENERAL REMARKS

This paper discusses and analyzes the measurements of the vertical distribution of refractive black carbon (rBC) in the high Arctic during the spring and summer measured with a single particle soot photometer (SP2) during the NETCARE project. The mean and variance of the vertical profiles of total rBC mass, mass-mean diameter, and the ratio of rBC to CO and total aerosol number are discussed, along with the changes in transport patters and sources that lead to the distinct vertical layers observed in the profiles. The data gathered in this campaign helps to fill an important gap in previous observations of the Arctic. The work presented in the paper is an excellent, detailed analysis of the sources and mechanisms (e.g., wet deposition) leading to the observed vertical profiles of rBC, helping to provide a conceptual model that explains key features displayed in the observations. Generally, the conclusions of the study are well-justified by the results shown and the potential for alternate explanations is appropriately discussed. I don't have any major concerns with the methodology, results, or conclusions of the study. Most of my comments below focus on either unclear wording in the manuscript text or issues with the figures that made it difficult to identify some of the features discussed in the text. I've only focused on cases where it was unclear to me what the authors intended to say. There are numerous other language choices that struck me as odd, but rather than list them here I would suggest that these be addressed by an English language copy-editor before publication.

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#3

P1, L3: You might discuss what you mean by "high Canadian Arctic" here to clarify the region your results apply to.

The "Canadian High Arctic" definition was already adopted in most of the publications resulted from the NETCARE projects (Abbatt et al., 2018). Generally, High Arctic can be defined as the area ensemble located at latitudes higher than 70°N (AMAP, 2015). In the present study the Canadian High Arctic includes the northernmost Canadian research stations which mainly experience Arctic conditions all year long, being included in the Polar Dome both during the cold and warm season. P1L3 was modified as:

[…] high Canadian Arctic (>70°N). […]

P1, L6: "caused and changed" is an odd phrase. I think you mean the cyclonic disturbances caused additional transport of pollution, correct?

The statement was modified, now it reads:

[…] The observation periods covered evolutions of cyclonic disturbances which favored the transport of air pollution into the High–Arctic, as otherwise the air mass boundary largely impedes entrainment of pollution from lower latitudes. […]

P3, L5 "spread of more than one order of magnitude" in what? rBC concentrations, deposition, or emissions? Please clarify what you are referring to here.

The statement was clarified, now it reads:

[…] the balance of these effects in the Arctic can only be estimated in models as accurately as vertical distributions of BC is known. However, profiles of BC concentration show a spread of more than one order of magnitude amongst different state-of-the-art models as well as between models and observations (AMAP, 2015). […]

P5, Table 1: Please add a note to the table explaining that the "station" is the location the plane left from.

Added.

P10, Figures 2 and 3: The black triangles are very hard to see against the blue and green colors. Consider making the triangles white instead?

Changed

P11, Figure 4: The two blue shades for Alert and Eureka are very hard to tell apart. Can you make them easier to distinguish?

The colors were chosen to clearly distinguish the spring and summer measurements. In order to improve the readability of the lines, the color of the Alert profiles was changed to green.

P12, L1-2: I'm not sure the ARCTAS and NETCARE observations discussed in this paragraph are enough to conclude that "wet removal becomes more efficient during summer within the polar dome, but as well already during northward transport outside the dome." Do you have other evidence supporting that the changes seen in both campaigns are due to more efficient wet removal?

Our interpretation of wet removal was found weak or inconsistent by other reviewers. In order to improve the interpretation of our observations, the frequency of liquid and ice cloud encounter by the air parcels during transport was calculated for each flight and discussed in Section 3.4. Even with this additional tool, it was difficult to properly estimate the effective impact of wet removal on BC concentration and its properties. Thus, the interpretation of wet scavenging based on the BC/CO ratio was substantially reduced. The mentioned statement was removed. The previous statement about the balance of rBC supply and removal is stressed by:

[…] This might be due to the fact that their observations were from a Sub-Arctic region (northern Alaska), where pollution supply and removal are not necessarily in the same balance as within the polar dome. The balance between supply and removal of rBC appears to have a pronounced seasonality, based on the ARCTAS and NETCARE observations. […]

P12, L7-9: Are you saying that mixed Asian outflow is a source of the Arctic haze you observed in this campaign, or is it just an example to show that the haze concentration is usually lower than near the source?

The statement was meant to compare the mass concentration of BC found in the Arctic with other locations, inside and outside the Arctic. However, the comparison with Asian outflow conditions was removed because of its low relevance in the present context. The sentence was modified as following:

[…] The overall range of BC concentrations is similar to previous spring observations reported for the European Arctic (Liu et al., 2015) and comparable to measurements from the mixed boundary layer over Europe (McMeeking et al., 2010). […]

P12, L15-16: I'm not clear what you mean by "could indicate a partitioning of rBC particle size within polluted layers." Can you please clarify what you mean by this sentence?

Anonymous referee #2 already reported this issue. 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 hand, other processes, as different emission sources, might play a role. Due to its speculative character, the present statement was removed.

P12, L21-22: I think you need to be careful with the writing here. rBC could have a significant impact on solar light extinction (measured in W/m^2) even if the number fractions of rBC particles relative to total aerosol was low. However, the low ratio, combined with the low rBC mass concentration, means the impact in this case is negligible. Thus, I think you have to mention the low mass concentration of rBC here before concluding rBC has a negligible impact.

We agree with the referee's comment. The text was changed in accordance to other modifications made in Section 3.5.

[…] In contrast to the spring, the summer MMD showed a slight increase from the surface (129 nm) to about 600 hPa (140 nm; Fig. 4 b). As pointed out in Bond and Bergstrom (2006), the mass absorption cross-section of BC particles depends, also, on the particle's diameter. As a consequence, the concentration of rBC mass in small particles could potentially contribute to the enhancement of the absorption coefficient of the total aerosol. Nevertheless, the low values of (average of 2 ng m^{-3} with IQR 0—12 ng m^{-3} throughout the column) and R_{numTA} (average of 0.75%) makes BC a minor contributor to the total aerosol light extinction. A more detailed description of seasonal and vertical variability of the BC core diameter will be provided in Section 3.5[…]

P17, L31-32: How does this choice of only using trajectories that encountered above average M_rBC potentially impact your analysis and results? Is there a potential for bias from this choice?

Our choice of using a M_{rBC} threshold was also questioned by other referees. Initially we wanted to focus on the most intense plumes, which most likely cause high but local forcing. We now understand the limits of our choice, which might systematically remove air parcels that originated in pristine regions or that experienced precipitation. For this reason, the trajectories presented and discussed in Section 3.4 include all points at which measurements were made.

P18, L1-2: Why did you not use the ECMWF boundary layer heights to determine the hatching instead?

The ECMWF data were used to run the LAGRANTO back-trajectories. A flag indicating whether the trajectory was traveling within the boundary layer was however not part of the available model output. Linking the trajectory position at each time step again with the boundary layer height information from datasets (e.g. ERA-interim) would be possible in post—processing, however only as a complicated approach relaying on several assumptions. A detailed evaluation of the trajectories' interactions with the boundary layer was beyond the scope of the maps in Fig. 7 and 8 and we believe that detailed information would have been blurred in these multiple day average maps. The text was changed and now stresses that the hatching highlights the presence of trajectories moving at atmospheric pressures >920 hPa in a grid cell in contrast to trajectories already lifted up from the lower atmosphere.

[…] A hatching highlights grids where trajectories travelled at atmospheric pressures >920 hPa, which is equal to less than about 0.5 km. Climatological boundary layer heights over Europe are typically <1 km during daytime [Seidel et al., 2012], thus pollution uptake from surface sources may be possible in a well-mixed atmosphere in the hatched areas in contrast to trajectories moving in the upper atmosphere or being lifted already due to vertical motion in synoptic scale systems (Sec. 3.1). […]

P19, L1-2: I don't see the difference between Levels II and III discussed in this sentence in Figure 7. What should I be looking for in the figure?

The authors agree with the referee's comment. The interpretation of the Figure 7b and Figure 7c was not accurate. In fact, back trajectories suggested that the motion of airmasses from the Eurasian sector to Level II and Level III was quite similar. Insights on transport and origin of air parcels higher in M_{rBC} are now more evident after extending the trajectory study to the entire dataset and focusing the discussion on M_{rBC} instead of R_{CO}. The entire Section 3.4 was substantially modified, we thus encourage the reviewer to consider the changes implemented in the entire section.

P20, Figure 7 and P21, Figure 8: These figures are very difficult to see, maybe due to their small size in the combined figure. The color bar for the overpass frequencies should also be included in both figures. I'm also wondering if the color for high rBC/CO values is too similar to the red colors used for high overpass frequencies, so in Figure 7 a, I'm not sure if the red near the surface sites is just high overpass frequencies or also high rBC/CO ratios at the endpoints.

The authors are aware of the interpretational limits of both Figure 7 and Figure 8. To improve its readability and clarity, some modifications were implemented:

- The location of the reference stations is now symbolized by a large black cross.
- R_{CO} in Figure 7 and 8 was substituted with M_{rBC}.
- Two color scales describing the overpass frequency and M_{rBC} were added.
- A legend now indicates the two source types (wild fires and gas flaring) and the airmasses moving at low level.

Minor comments of Reviewer#3

P1, L4: expand the acronym "NETCARE" here.

Changed.

P1, L10: "factor of 10", not "factor 10"

Changed.

P1, L22: "rBC was affected" not "got affected"

Changed.

P7, L32: "As opposed to aerosol", not "Other than aerosol"

Changed.

P21, L2: There isn't a Section 6, so what should this refer to?

Corrected.

P25, L10: "available ton the Government"?

Corrected.

REFERENCES

Abbatt, J. P. D., Leaitch, W. R., Aliabadi, A. A., Bertram, A. K., Blanchet, J.-P., Boivin-Rioux, A., Bozem, H., Burkart, J., Chang, R. Y. W., Charette, J., Chaubey, J. P., Christensen, R. J., Cirisan, A., Collins, D. B., Croft, B., Dionne, J., Evans, G. J., Fletcher, C. G., Ghahremaninezhad, R., Girard, E., Gong, W., Gosselin, M., Gourdal, M., Hanna, S. J., Hayashida, H., Herber, A. B., Hesaraki, S., Hoor, P., Huang, L., Hussherr, R., Irish, V. E., Keita, S. A., Kodros, J. K., Köllner, F., Kolonjari, F., Kunkel, D., Ladino, L. A., Law, K., Levasseur, M., Libois, Q., Liggio, J., Lizotte, M., Macdonald, K. M., Mahmood, R., Martin, R. V., Mason, R. H., Miller, L. A., Moravek, A., Mortenson, E., Mungall, E. L., Murphy, J. G., Namazi, M., Norman, A.-L., O'Neill, N. T., Pierce, J. R., Russell, L. M., Schneider, J., Schulz, H., Sharma, S., Si, M., Staebler, R. M., Steiner, N. S., Galí, M., Thomas, J. L., Salzen, K. von, Wentzell, J. J. B., Willis, M. D., Wentworth, G. R., Xu, J.-W. and Yakobi-Hancock, J. D.: New insights into aerosol and climate in the Arctic, Atmospheric Chem. Phys. Discuss., 1–60, doi:https://doi.org/10.5194/acp-2018-995, 2018.

AMAP: AMAP Assessment 2015: Black carbon and ozone as Arctic climate forcers. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. vii + 116 pp., [online] Available from: http://www.amap.no/documents/doc/amap-assessment-2015-black-carbon-and-ozone-as-arctic-climate-forcers/1299 (Accessed 7 January 2016), 2015.

Bond, T. C. and Bergstrom, R. W.: Light Absorption by Carbonaceous Particles: An Investigative Review, Aerosol Sci. Technol., 40(1), 27–67, doi:10.1080/02786820500421521, 2006.

Liu, D., Quennehen, B., Darbyshire, E., Allan, J. D., Williams, P. I., Taylor, J. W., Bauguitte, S. J.-B., Flynn, M. J., Lowe, D., Gallagher, M. W., Bower, K. N., Choularton, T. W. and Coe, H.: The importance of Asia as a source of black carbon to the European Arctic during springtime 2013, Atmospheric Chem. Phys., 15(20), 11537–11555, doi:https://doi.org/10.5194/acp-15-11537-2015, 2015.

McMeeking, G. R., Hamburger, T., Liu, D., Flynn, M., Morgan, W. T., Northway, M., Highwood, E. J., Krejci, R., Allan, J. D., Minikin, A. and Coe, H.: Black carbon measurements in the boundary layer over western and northern Europe, Atmos Chem Phys, 10(19), 9393–9414, doi:10.5194/acp-10-9393-2010, 2010.