

Authors: Olga B. Popovicheva et al.

The title of manuscript: Multi-year black carbon observations and modeling close to the largest gas flaring and wildfire regions (Western Siberian Arctic)

General comments:

The Arctic has warmed three times more quickly than the planet as a whole, making it the most sensitive region to climate change. To understand the impacts of BC emissions on the Arctic from source regions, particularly from the Siberian Arctic, the authors present three and a half years measurements of equivalent BC (eBC) concentrations from 2019 to 2022. These measurements are complemented by elemental carbon (EC) data obtained using thermal-optical method, conducted at the recently established station “Island Bely” (IBS). The station is located along the main pathway through which the highest levels of anthropogenic pollution from industrial regions, as well as emissions from Siberian wildfires, enter the Arctic.

Furthermore, the authors evaluate seasonal variations in intensive optical properties and their dependence on wavelength, as indicated by Absorption Angstrom Exponent (AAE). They also estimate the site-specific absorption coefficient (SAC), which serves as the basis for calculating eBC values.

By coupling the FLEXPART Lagrangian particle dispersion model with the latest black carbon (BC) emission inventories for anthropogenic and biomass burning sources, the study investigates the detailed aerosol aging spectrum, source region attribution, and source sector apportionment for the entire period as well as during pollution episodes. This approach effectively integrates observations with model simulations and emission inventories.

However, it is noted that this work appears very similar to a study published in 2022 by the same group (Popovicheva et al., 2022), conducted at the same location with similar observations and using the same model. The conclusions are largely the same, apart from the inclusion of two additional years of observations.

Response: The purpose of this paper was to present an extensive dataset that spans for 3.5 years instead of 1 year in Popovicheva et al. (2022). The large amount of research results collected during all these years helps us to verify previous observations and present new findings for winter/summertime maxima and the impact of the increasing wildfire events in Siberia. In addition, we have a set of new data on light absorption, such as the spectral dependence of light absorption, multi-annual intercomparison analyses between polar stations according with respect to babs(570) characteristics, analyses of 170 samples for OC/EC in order to virify correlations between EC and eBC (this has never done before), revise the eBC_{AET} values and get site-specific mass absorption characteristics that quantify the light absorption for a given location (that is the most important and representative characteristic).

I would recommend publishing the manuscript in Atmospheric Chemistry and Physics (ACP) after the authors address the following aspects:

- Reduce or condense repetitive content that overlaps with the previous paper published in 2022.

Response: We have tried to reduce repetitive content in the new version. However, it should be noted that since we have extended the dataset, some repetition cannot be avoided.

- Emphasize aspects that are being discussed for the first time in this study, such as “BrC and its relationship with the corresponding AAE” and “eBC (derived from SAC) and its comparison with eBCAET”.

Response: We appreciate Reviewer 2 for pointing this out and we have tried to focus on new findings as much as the results allow. Several changes have been performed throughout the manuscript in this aspect.

- It is noted that the eBC values are approximately half of the eBC_{AET} values (Table 1). Could the authors have a further discussion regarding the difference?

Response: We have now added a discussion in section 3.2 at Lines 468-498 (see Manuscript with Track Changes).

- There appears to be a significant discrepancy between the results in Table S3 and Figure 10 b). For example, in July 2020, Figure 10 b) shows that biomass burning (BB) accounts for about 80%, whereas Table S3 indicates only a 7% contribution of BrC during the same period. Conversely, in February 2020, Figure 10 b) shows less than 10% BB, while Table S3 also reports a 7% BrC contribution. Could the authors clarify why these differences occur?

Response: Table S3 presents the contribution of babs from BrC to total babs(370) calculated from the aethalometer data. It relates to BrC contribution at 370 nm.

On the contrary, Figure 10b shows the source contributions to BC estimated by the FLEXPART model. Hence, the results in Table S3 and Figure 10 are not directly related.

Since we use BB emissions (wildfires) from CAMS GFAS (see Methods section), we can relate the modelled BB contribution to observations. Light absorption at 370 nm is considered as a “proxy” for BrC and can be related to BB (fires), when the BB sources are nearby. At IBS, as well as in other Arctic stations, our research indicates aged BBBC and, thus, comparison is not straightforward.

Specific comments:

L27-L28: Is the “92%” an average value of cold seasons over the entire study period? It appears that the 92% mentioned in abstract is not consistent with the 83% stated in the conclusion.

Response: We have tried to clarify (see Lines 28-30 in Manuscript with Track Changes). 83% is the contribution from anthropogenic sources over the entire period (including both cold and warm periods), 92% corresponds to the anthropogenic contribution during the cold season only, over for the 3.5 years of study).

L87-L88: Are the Biomass Burning (BB) results in Figure 10 a) and b) from ground level or from the altitudes (600-800 hPa)? Do the BB results in Figure 10 support the previously work by Qi and Wang, 2019?

Response: The Legend of Figure 10 states that “(a) Timeseries of monthly mean contribution from different emission source types to surface BC concentrations...”. Hence, the answer is that the contribution corresponds to the surface (always below the boundary layer height).

Exactly! Our main findings support those reported by Qi & Wang (2019). We also find that the main source at IBS is fossil fuel combustion (e.g., gas flaring emissions), while wildfires play secondary role. However, here, we do not examine the impact of deposition on snow/ice, like in Qi & Wang (2019) (since we do not have such measurements). The impact of long-range transport from lower latitude Asian sources to the Arctic has been quantified long before by Evangeliou et al (2016) (<https://doi.org/10.5194/acp-16-7587-2016>).

L150: Should this be Fig. S1, instead of Fig.1d ?

Response: Correct! We have change this in Line 192 (see Manuscript with Track Changes).

L159: What are the two inlets with a much higher flow rate (~ 38LPM) than 5LPM?

Response: Information added in Lines 214-216 (see Manuscript with Track Changes).

L170: Please specify eBCAET as “equivalent black carbon concentration by aethalometer”, when mentioning it for the first time.

Response: Corrected in Line 224 (see Manuscript with Track Changes).

L182: Please spell out for “NIR-VIS”

Response: Corrected in Line 236 (see Manuscript with Track Changes).

L240: What is the highest level of the 137 vertical levels?

Response: Approximately 80 km. We have added it in Line 317 (see Manuscript with Track Changes).

How do you couple with the BB burning injection height in the emission inventory?

Response: The BB emissions from CAMS-GFAS come with estimates of the maximum injection altitude of each gridded emission. We run our model backwards releasing particles at the surface of the receptor (this is the IBS site) and calculate footprint emission sensitivities at 0-100, 100-3000 and 3000-8000. We find the indices of the fires occurring at these heights, we calculate modelled concentrations at these heights separately and we sum them afterwards to retrieve the total modelled concentration at the receptor (IBS site).

L276-L277: Please be consistent with the wavelength mentioned (880nm or 800 nm?). Only 880nm is shown on Fig. 2.

Response: We appreciate Reviewer for pointing out this typo. The correct wavelength is 880 nm and has been now corrected everywhere in the manuscript. Please see section 3.1 and elsewhere in Manuscript with Track Changes.

L277: The expression is confusing, i.e., “the mean \pm sigma (median) values”. It is suggested to use “the mean \pm sigma and the median, respectively”.

Response: Corrected as reviewer suggested in Line 356 (see Manuscript with Track Changes).

L285: Please ensure consistency between the text and Table 2.

Response: There is no Table 2 in the manuscript, so we assume that Reviewer means consistency in terms of babs(880) (and not 800 as it was earlier). This has been corrected throughout the manuscript.

L295-L297: The sentence is not well expressed. Please consider re-phrasing it.

Response: We have removed this sentence as unnecessary, since it does not add anything to the manuscript.

L308 & L1083: Please ensure consistency in the wavelength between the text on page 11 and Figure 3(a). The caption for Figure 3(a) should indicate 590 nm. Additionally, please add (a) and (b) to Figure 3.

Response: We have corrected the inconsistency in legend of Figure 3. It now writes 590 nm (see Manuscript with Track Changes).

L378: Is it the cold season, or the warm season over the entire period?

Response: Line 378 writes “... observed for the cold and warm periods with means 44 ± 47 and 19 ± 57 ng m⁻³, respectively.” We have now added “, for the entire study period.” Please see Manuscript with Track Changes.

L385-L388: The numbers in the text differ slightly from those in Table S2. Please ensure consistency between the text and the table.

Response: We have corrected the number in consistency with Table S2 in Lines 540-544 (see Manuscript with Track Changes).

L427: No box-whisker plots are shown in Figure S3, which presents the emissions of BC from CAMS-GFAS. This should be Figure S4.

Response: Corrected in Line 595 (see Manuscript with Track Changes).

L431: It is suggested to add "in percentage" (shown in Table S3) after the "contribution".

Response: Corrected in Line 599 (see Manuscript with Track Changes).

L439: Can the authors explain why eBC_{AET} is higher than eBC by a factor of two?

Response: We have now added this explanation in section 3.2 at Lines 468-498 (see Manuscript with Track Changes). This has been answered in a previous comment.

L498: Should it be Figure 9?

Response: Corrected in Line 683 (see Manuscript with Track Changes).

L520-L521: It is interesting to see a lack of consistency between Figure 10b and Table S3. It appears that a high BB contribution does not result in a relatively high percentage of BrC.

Response: Typically, a high BB contribution results in a high percentage of BrC in atmospheric aerosols. However, the exact relationship depends on several factors. What we know is that BrC is a major component of BB emissions and that BB produces both BC and BrC.

Although high BB contribution generally means more BrC, the actual percentage depends on (a) combustion conditions (flaming combustion – high temperature – results in more BC and less BrC, smoldering – low temperature – results in more BrC and less BC, peat fires – wet biomass – result in higher BrC), (b) fuel type (wood, agricultural waste, and peat produce high BrC emissions, coal burning – anthropogenic – results in more BC and less BrC), and (c) atmospheric aging (BrC undergoes photobleaching – degrades under sunlight – reducing its absorption over time and aged BrC may convert into non-absorbing organic carbon).

L546: It should be ~ 50%, instead of 60%.

Response: The correct number is 57% (see Line 741, Manuscript with Track Changes).

L547: It is suggested to use 72,400 km².

Response: Corrected in Line 741, Manuscript with Track Changes.

L560: what is the unit of “150,000”?

Response: 150,000 “fires” occurred. Corrected now in Line 755 (see Manuscript with Track Changes).

L574: In general, the 'Conclusion' section is too long and should be more concise.

Response: The conclusions section has been shortened and refined as suggested by both reviewers (see Manuscript with Track Changes).

L596: "BrC light absorption coefficient in the UV spectrum showed similar trends as BC, although it exceeded BC by 2.4 times during both cold and warm periods." What is the basis of this statement?

Response: In Conclusions section we aimed to concentrate the basic findings of this work, considering that the paper is relatively long. In this sense, we simply report the findings. We have now changed the title of the section to Summary and conclusions.

L607: Please confirm the numbers. They should be consistent with these in Table S2, where the numbers are $106 \pm 67 \text{ ng m}^{-3}$.

Response: Corrected in Line 806 (see Manuscript with Track Changes). In total, we have tried once again to make text consistent with data shown in Tables.

L669: Please use “MAC”, instead of “MAC” to ensure consistency when writing the initials. Otherwise, it would be confused with the Mass Absorption Coefficient (MAC).

Response: Corrected in Line 939 (see Manuscript with Track Changes).