Response to Anonymous Referee #1

We appreciate the invaluable comments. Our answers to the comments are provided below. The reviewer comments are written in italics.

The manuscript presents BC data, based on absorption measurements from 5 Arctic ship-based campaigns (2016-2020). It is a nice data-set, and considering the scarcity of available data from this remote area, very valuable. I would recommend the paper for publication in ACP, however have some comments that are needed to be addressed before.

Both the AE33 and COSMOS are instruments, that measure absorption coefficients. It is already challenging and makes different corrections necessary to retrieve the correct absorption coefficient from filter-based measurements. The absorption coefficients are then used to estimate the value of the BC mass concentrations. Reading the paper, one could think that the instrument would measure directly the BC mass, or at least it would be a trivial or straight-forward thing to get the BC concentrations correctly from filter-based absorption measurements. Only very few details are given according to the measurement uncertainty and possible corrections and assumptions. I would like to read a more thorough discussion/introduction about it, how well these instruments can be used for direct BC mass measurements. The manuscript from Backmann et al., 2017 (https://doi.org/10.5194/amt-10-5039-2017) deals with the uncertainties of Aeth measurements from the Arctic and proposes a region-specific correction factor. I am not even completely sure, how and if the aethalometer data was corrected, but this would be maybe an option to consider.

Reply> Thank you for the comments. First of all, the model of the Aethalometer which was deployed during the cruise is AE22, not AE33. This mistake is corrected in the revised manuscript. We sincerely apologize for any inconvenience caused by this mistake.

Another major change is that we successfully obtained AE22 data for the 2017 and 2018 cruises, which were not available in the preprint. Therefore, we can compare the two instruments, which led our analysis to focus more on the COSMOS data because of the larger uncertainty in the AE22 data.

We used the manufacturer recommended standard BC mass concentration data from COSMOS and AE22 throughout the analysis. In an effort to give readers more information about the measurements, we modified the text to include the MAC values, the uncertainties, and other specific features of each instrument, and the comparison between the two instruments. With this, paragraph two in Sect. 2 of the preprint has been expanded to three paragraphs in the revised manuscript. That is, the original expression “A continuous soot monitoring system (COSMOS, model 3130, KANOMAX, Japan) and an Aethalometer (model AE33, Magee Scientific Co., USA) were used in the cruises in 2016–2019 and in 2020, respectively, to measure the mass concentrations of BC aerosols. Both instruments use light absorption methods. Therefore, the obtained mass concentration of BC is equivalent black carbon (eBC, Petzold et al., 2013). Nonetheless, BC will be used throughout the manuscript if not specifically mentioned. COSMOS monitors changes in transmittance of 565 nm wavelength LED light across an automatically advancing quartz fiber filter tape. To achieve measurements with high sensitivity and a lower detectable light absorption coefficient, COSMOS uses a double-convex lens and optical bundle pipes to maintain high light intensity and signal data are obtained at 1000 Hz. The data integration time was set to 1 min, which was then averaged to 1 h for further analysis. In addition, its sampling flow rate (0.9 L min^{-1}) and optical unit temperature were actively controlled. The inlet line for COSMOS was heated to 400 °C to effectively volatilize non-refractory aerosol components that were internally mixed with BC. The lowest detection limit of COSMOS at 1 min time resolution is 50 ng m^{-3} and at 1 h time resolution is 1 ng m^{-3} (Ohata et al., 2019). Aethalometer uses the absorption of light at a wavelength of 880 nm by the ambient aerosols collected on a Pallflex Teflon-coated glass fiber (TFE) filter tape to determine the BC concentration. The flow rate was set to 5 L min^{-1}. The data integration time was set to 5 min, which was then averaged to 1 h for further analysis. The default mass absorption cross section value of 7.77 m^2 g^{-1} and internal multiple scattering correction factor of 1.57 were applied (Drinovec et al., 2015). The lower detection limit of Aethalometer at 1 h time resolution is 5 ng m^{-3}. It is noted that the default parameter settings of the aethalometer may cause the obtained BC mass concentrations to be twice the actual values (Laing et al., 2020; Asmi et al., 2021)." has been modified to

"A continuous soot monitoring system (COSMOS, model 3130, KANOMAX, Japan) and an Aethalometer (model AE22, serial number 1057:1010, Magee Scientific Co., USA) were used during the cruises to measure the mass concentrations of BC aerosols. Whereas both instruments use light absorption methods, COSMOS was equipped with a 400 °C heated inlet line. This feature effectively eliminated interference from volatile non-refractory aerosol chemical species internally mixed with BC, ensuring a high accuracy of m_{BC} measurement. This aspect has been critically assessed in previous studies (Ohata et al., 2019; Sinha et al., 2017). Consequently, COSMOS measurements differ from traditional light absorption methods, where the mass concentration of BC is referred to as equivalent BC (eBC, Petzold et al., 2013). Therefore, instead of using eBC, the term BC can be used for COSMOS data in a general sense (Ohata et al., 2019). Henceforth, when comparing data from the two different instruments, we will use m_{eBC} to represent the BC mass concentration measured with the Aethalometer during the 2017, 2018, and 2020 cruises, and m_{BC} (COSMOS) to represent the BC mass concentration measured with COSMOS during the 2016–2019 cruises. Otherwise, BC mass concentration is denoted as m_{BC} for simplicity.

COSMOS monitors changes in transmittance of 565 nm wavelength LED light across an automatically advancing quartz fiber filter tape. To achieve measurements with high sensitivity and a lower detectable light absorption coefficient, COSMOS uses a double-convex lens and optical bundle pipes to maintain high light intensity and signal data are obtained at 1000 Hz. In addition, its sampling flow rate (0.9 L min^{-1}) and optical unit temperature were actively controlled. The measurement interval was set to 1 min, which was then averaged to 1 h for further analysis. The default mass absorption cross section (MAC) of 10 m^2 g^{-1} was applied for the derivation of m_{BC}. The lowest detection limit of COSMOS at 1 min time resolution is 50 ng m^{-3}. On an hourly basis, COSMOS can measure m_{BC} in the range of 1–3000 ng m^{-3} with an average accuracy of ~10 %, as compared with measurements by a single particle soot photometer (SP2) (Moteki and Kondo, 2010); and its sensitivity to the changes in the BC size distributions was less than 10 %, within the typical BC sizes in ambient atmosphere (Ohata et al., 2019). The SP2 is used as a reference instrument in previous studies (e.g., Ohata et al., 2019; Sinha et al., 2017). Further details about the measurement principles of COSMOS can be found in previous studies (Ohata et al., 2019; Kondo et al., 2009).

The Aethalometer uses the absorption of light at a wavelength of 880 nm by ambient aerosols collected on a quartz filter tape to determine the BC concentration. The flow rate was set to 5 L min^{-1} and the accumulation area of the filter is 1.67 cm^2. The filter was set to change every 24 hours to minimize the loading effects. The data integration time was set to 5 min, which was then averaged to 1 h for further analysis. The default manufacturer-provided MAC value of 16.6 m^2 g^{-1} was applied. The lower detection limit of the Aethalometer at 1 h time resolution is 20 ng m^{-3} with the error limit of ±30 ng m^{-3}. Comparison between m_{eBC} and m_{BC} (COSMOS) for cruises in 2017 and 2018, when both data are available, shows that the two data are in high consistency (Pearson correlation coefficient R>0.96) and that m_{eBC} was 1.3–2.5 times m_{BC} (COSMOS) (Fig. S2). Previous studies also show that the default parameter settings of the Aethalometer, as mentioned above, may cause the obtained BC mass concentrations to be 1–3 times the mass measured by SP2, depending on the sources and mixing states of the BC aerosols (Wang et al., 2014; Sharma et al., 2017; Laing et al., 2020). Due to the above reasons, the AE22 data in this study are mainly used as a reference. Hereinafter, for cruises conducted from 2016 to 2019, the analysis primarily relied on COSMOS data. In the case of the 2020 cruise, when only AE22 data was available, AE22 data was utilized for the analysis." (P4, L13 - P5, L21)

Regarding the BC correction for the Arctic region as proposed by Backmann et al., 2017, we would rather use the standard BC concentration recommended by the manufacturer at this stage, since the assumptions for the specific AE22 BC data correction may cause a wider error limit because the measurements covered a quite wide range of latitudes.

If I understood correctly, the AE33 was used in 2020, otherwise the COSMOS was deployed. The wavelength dependence of the AE33 measurement is also often used to differentiate between biomass burning and anthropogenic BC. At least for 2020, you could do this differentiation and compare that to the model results. That would be a nice validation for the model, considering the moderate agreement between the measured and modeled total BC concentration.

Reply> The model of the Aethalometer used in this study is AE22, not AE33. This typo error has been corrected in the revised manuscript. Since the AE22 model has only 2 channels, we decided not to apply source apportionment analysis using AE22 BC data.

Specific comments:

1. ***P2,L9: "can work as CCN" please rephrase, e.g. to can act as***

   Reply> The word "work" has been replaced with "act". (P2, L10)

2. ***P4, L8: Please mention already here, that the specific placement of your inlet was not the only method you have used to get rid of the ship emission influence, and that that the data sorting will be explained later.***

   Reply> The original expression "The air intake was set at the handrail of the front upper deck to avoid ship exhaust pollution." has been modified to "The air intake was set at the handrail of the front upper deck to prevent contamination from ship exhaust pollution. Furthermore, detailed information regarding data filtering techniques to mitigate the impact of ship exhaust will be provided later.". (P4, L9-11)

3. ***P4, L10-22: what mass absorption efficiency is used in the COSMOS instrument?***

   Reply> The mass absorption cross-section used in COSMOS is 10 m^2 g^{-1}. This has been included in the revised manuscript. (P4, L31)

4. ***P4, L23: you state that the BC mass concentration is derived from the AE33 880 nm measurements. What about the other wavelengths? Why not another one? The COSMOS uses 565nm, why not the closest wavelength of the AE33 was used to be more consistent?***

   Reply> The model of the Aethalometer used in this study is AE22, not AE33. This typo error has been corrected in the revised manuscript. The other AE22 channel uses 370 nm, of which values are interfered with by contributions from organic materials and are not suitable for direct comparison with the COCMOS 565 nm BC data.

5. ***P4, L28-29: did you use these "default parameter settings"? And please provide more details about this fact. Please see my general comment about requesting much more details on how BC concentration was derived, and what are your uncertainties!***

Reply> The corrected and modified text on **pages 4 and 5** of the revised manuscript contains the mentioned information. Please see our response to the general comments.

6. ***P5, L13-22: If I understood correctly, your method of discriminating the data which might be influenced by the ship emissions was the following: firstly the discrimination by wind direction, and in the years when the mixing ration of O3 was also measured you used that as an extra criterion. How much more data was considered invalid using the O3 criteria in 2017 and 2018? Which was not already discriminated by the wind direction? Can you estimate how much bias could it cause not having O3 data for the other years, and with that still using some data that might have been influenced by the ship?***

Reply> Using the O_3 criteria, 100 and 173 1-min m_{BC} (COSMOS) data were considered invalid in 2017 and 2018, respectively. They accounted for less than 0.3 % and 0.4 % of the total amounts of valid data in 2017 and 2018, respectively. That is, further scrutiny by O_3 data had little influence on the overall properties of the observed BC. This point has been included in the manuscript as:

"It is noteworthy that the additional scrutiny based on the O_3 criteria had minimal impact on the overall characteristics of the observed BC by COSMOS. This screening process resulted in the exclusion of less than 0.3 % and 0.4 % of the total valid data in 2017 and 2018, respectively." (P6, L12-14)

7. ***Figure 2, panel a: could you make the y axis logarithmic? Or do something else to see the lower values as well?***

Reply> To enhance the visibility of the lower values in Fig. 2a, a zoomed-in view of Fig. 2a with the y-axis maximum set to 80 ng m^{-3} is shown in Fig. S3b of the revised supplementary.

8. ***Figure 3: a logarithmic y axis for the BC concentration would help here a lot too. Caption: "short dashed gray line" it is enough if you write dashed gray line, I was looking for short lines that are dashed and gray :).***

Reply> The y-axis of Fig. 3 has been modified to a logarithmic scale. The word "short" has been removed from the caption. (P15, L3)

9. ***P6, L30-31: This is what you do not see based on Fig2a, and therefore would be a logarithmic axis nice***

   Reply> To enhance the visibility of the lower values in Fig. 2a, a zoomed-in view of Fig. 2a with the y-axis maximum set to 80 ng m^{-3} is shown in Fig. S3b of the revised supplementary.

10. ***P7, L25-26: what was this similar value reported by Taketani et al.?***

   Reply> After careful examination of the data provided by Taketani, we found that the mean m_{BC} reported by Taketani et al. (2022) for the same period is 0.6 (±0.8) ng m^{-3}, much lower than the value of 8.2 (±6.0) ng m^{-3} obtained in our study. Consequently, we have elected to remove this section from the revised manuscript. We apologize for any confusion this may have caused.

11. ***P11, L13: "anthropogenic productive activities producing" strange wording, please change***

   Reply> The expression "anthropogenic productive activities producing" has been modified to "industry activities producing". (P15, L16)

12. ***P11, L14: "may export" please change to "may be exported"***

   Reply> The expression "may export" has been changed to "may be transported". (P15, L17)

13. ***P11, L26-27: if I understood correctly, the AE33 and the COSMOS were not running parallel, and the AE33 measured in 2020 and otherwise the COSMOS. You state that the background periods were defined based on the measurements of the COSMOS. Does this mean that you did not even try to look for background periods in 2020? Why?***

   Reply> Thank you for the comments. First, the model of the Aethalometer used in this study is AE22, not AE33. This typo error has been corrected in the revised manuscript. Second, the definition of background periods has been modified in the revised manuscript. The new definition includes background periods from 2016 to 2020. For details on the new definition, see our response to the next comment.

14. ***P11, L29: your definition of a background period included that the BC mass concentration had to be above 1ng/m3, which is the 1 h detection limit of the instrument. My problem is with this, that with such a criterion you define that your background BC mass concentration has to be above 1 ng/m3. I understand that you are not able to reliably measure such low concentrations, but these values should be considered, otherwise the background concentration that you report is false, and at most you can call it as the upper limit, and the real background concentration could still be well below this value.***

Reply> The definition of the background periods has been modified from "The background periods in the western central Arctic Ocean (>72° N) were defined according to the m_{BC} measured by COSMOS and the 5 day HYSPLIT back trajectories as follows: the 1-min m_{BC} was below the lowest detection limit of 50 ng m^{-3} for continually 2 hours or longer, the 1-h m_{BC} was above the lowest detection limit of 1 ng m^{-3}, and the air masses were from the Arctic Ocean." to

"The background periods in the western central Arctic Ocean (>72° N) were determined according to the following criteria: first, for each hour with effective BC data, all three 5-day HYSPLIT back trajectories initiated at starting heights of 10, 500, and 1000 m originated from the Arctic Ocean. Additionally, all 1- min m_{BC} or 5-min m_{eBC} data within that hour were not removed due to ship exhaust according to data screening criteria described in Sect. 2. The second criterion is to ensure the accuracy of the selected data." (P15, L30-34)

15. ***P11, L33- P12, L2: You state that the background value (that you have determined) being smaller that the overall mean BC concentration measured during a full year in the central Arctic suggests that even in the summer months the central Arctic is influenced by imported BC pollutants. Please compare the background to the mean summer values that were measured during MOSAIC. And if the relation is still the same, then your statement will be correct.***

Reply> Thank you for the comments. Following the revision of the selection criteria of background periods, this part has been removed from the revised manuscript.

16. ***P12, L6-7: "The result should be representative as the data covered the summer seasons of three years." Why? Representative for what? I would just delete this statement, or make it more specific, please.***

Reply> Thank you for the comment. This statement has been deleted from the revised manuscript. We apologize for any confusion this may have caused.

17. ***Section 4.4: you use the GEOS-Chem simulated biomass burning BC to total BC ratio to evaluate the high BC episodes as well. Table 2 shows in some cases of the episodes, enormous difference between the measured and modelled BC concentration (e.g. E1 20 vs 1 ng/m3) exits. In such cases, if the modelled vs. the measured BC concentration are many factors away from each other, I do not think that you can use the modelled biomass burning to total BC ratio to assume anything about the real measurement. Even for 2 out of the 3 selected episodes, the modelled BC mass is quite far away from the measured. Please comment on this.***

Reply> Despite significant differences between measured and modeled BC concentrations in certain episodes, we contend that using GEOS-Chem model simulation results to estimate the biomass burning to total BC ratio during the selected episode periods is justified for two main reasons. Firstly, the model, capable of reproducing 44% of the temporal spatial variation of the overall observed BC (Sect. 4.3), indicates the ubiquitous dominance of biomass burning BC North of 65 °N, where all episodes were identified (Fig. 3). Secondly, uncertainty analysis, involving shifting the episode period back or forward by 18 hours while maintaining its length, revealed changes of no more than 10 % in the modeled biomass burning to total BC ratio for most episodes, except for E2 and E6, as illustrated in the figure below.

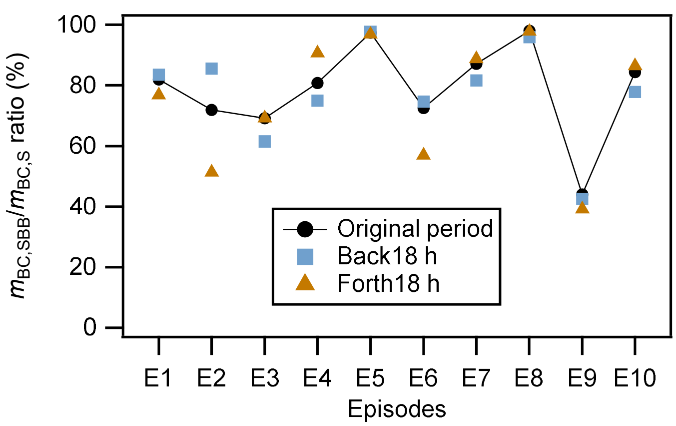

Figure R1-1: The biomass burning to total BC ratio for each episode (line and circle marker) estimated from the GEOS-Chem model simulation results. Also shown in the figure are the ratios derived by moving each episode period back (square marker) or forward (triangle marker) by 18 hours.

This point has been included in the revised manuscript as: "Note that despite substantial normalized mean biases in model simulations compared to observed m_{BC} for these episodes, ranging from −95 % to 178 %, we consider it reasonable to estimate the contribution of biomass burning to the total BC based on the model results. This is attributed to the pervasive dominance of biomass burning BC north of 65 °N, where all episodes were identified (Fig. 3). The estimate is further supported by the uncertainty analysis, involving shifting the episode period back or forward by 18 hours while maintaining its length, which revealed changes of no more than 10 % in the modeled biomass burning to total BC ratio for most episodes." (P18, L8-14)

Moreover, E3, E8, and E10 were selected for source and transport analyses because their temporal and spatial variations were well reproduced by the GEOS-Chem model. The analysis results were supported by the HYSPLIT back trajectory model. This point is better clarified in the revised manuscript by modifying the original expression "Furthermore, the temporal and spatial variations of E3, E8, and E10 were well reproduced by GEOS-Chem model (Fig. 3). Therefore, in the following, the sources of BC during E3, E8, and E10 are elaborated based on GEOS-Chem model as well as HYSPLIT back trajectory model." to

"Additionally, the temporal and spatial variations of E3, E8, and E10 were well reproduced by GEOS-Chem model, showing nearly simultaneous peaks in observed and model data during these episodes (Fig. 3). Therefore, in the following sections, the sources and transports of BC during E3, E8, and E10 are elaborated based on GEOS-Chem model, with findings further corroborated by the HYSPLIT back trajectory model.". (P18, L14-18)

18. ***P13, L11-15: Question regarding to the definition of the high BC episodes? Did you also use a geographic restriction? Only taking periods when the vessel´s latitude was higher than a certain value? Because looking at Figure 3 it looks like that. I do see periods, that look long enough and have high BC concentrations but were not considered as an episode at low latitudes.***

Reply> Yes, we do restrict the episode selection to being north of 65° N. Therefore, the original expression "To characterize the sources of the high concentrations of BC in the Arctic Ocean and the marginal seas," has been modified to "To characterize the sources of the high concentrations of BC in the Arctic Ocean and the marginal seas (north of 65° N),". (P17, L29-31)

19. ***P13, L13-14 and Table2: you state that the definition of the episodes included that the mean of the 1h BC concentration has to be above 20 ng/m3. How can it be that the overall observed mean BC is below 20 ng/m3 for E1 (first row of Table 2)?***

Reply> We apologize for the mistake. We have modified the expression "the mean of valid 1-h m_{BC} during the defined periods was greater than 20 ng m^{-3}." to "the mean of valid 1-h m_{BC} during the defined periods was not less than 20 ng m^{-3}.". (P17, L32-33)

In addition, we have rounded all m_{BC} values not less than 10 ng m^{-3}, including those in Table 2, to integers.

20. ***Figure 4: Please change the color for the ship positions, it is very hard to see it. And the wind arrows are also hard to see. (also for all other similar figures)***

Reply> Figure 4 and other similar figures have been modified to clearly show ship positions and wind arrows.

21. ***Figure S5: You cannot see the BCbb sources, the dots are so small. Anyway: not only for S5 but also for Figure 4, S6, why don't you show a zoomed in picture of the region which you are talking about, one could see everything much better on the plots.***

    Reply> Figures S9 (S5 in the preprint), S10 (S6 in the preprint), and 4 have been modified to show a zoomed in view of the region related to Episode 3. Besides, Figure S9 as well as Figures S11 and S14 (S7 and S10 in the preprint) are modified to clearly show the BCbb sources by coarsening the longitudinal and latitudinal dimensions by a factor of 5.

22. ***Figure 5: same as for Figure 4, actually do it please for all similar figures.***

    Reply> Figures 4 and 5 and other similar figures have been modified to clearly show ship positions and wind arrows.

23. ***P16, L20: "CO2 and O3 were not or even slightly decreased" I see there quite a decrease. Maybe use other wording.***

    Reply> When compared with CO and CH_4, the decreases in CO_2 and O_3 do not show significant differences from their temporal changes before and after episode 8. Therefore, we will retain the original statement. (P21, L21)

24. ***P16, L23-27: I only see a decrease of CO2 when the bb plume was present. All the possible explanations would rather explain a constantly low CO2 concentration, am I not right?***

    Reply> While we agree that CO_2 uptake by high latitude vegetation may explain the constantly low observed CO_2 concentrations, we still think that this uptake might have contributed to the slight decrease in CO_2 concentrations observed at a location distant from the continent. This consideration is in addition to acknowledging the possible influence of smoldering combustion conditions on that minor decrease.

    Accordingly, we retain the original statement, but modify "The decrease" to "The slight decrease". (P21, L24)

25. ***P17, L5-6: why was the background BC not subtracted from mBC/ΔCO?***

    Reply> To ensure that there were sufficient data to characterize the spatiotemporal changes in $m_{BC}/\Delta CO$, the background of m_{BC} was not subtracted. Actually, after subtracting the background m_{BC} of 2.1 ng m^{-3} (Table 1), 53 % of the $m_{BC}/\Delta CO$ data are lost.

Therefore, the original expression “note that in order to obtain more $m_{BC}/\Delta CO$ data points, the background of m_{BC} was not subtracted” has been modified to “note that in order to ensure that there were sufficient data to characterize the spatiotemporal changes in $m_{BC}/\Delta CO$, the background of m_{BC} was not subtracted”. (P22, L6-8)

26. P23, L2: “at higher than low latitude regions” ???

Reply> It has been modified to “at higher latitude regions”. (P29, L5)