Response to Anonymous Referee #2

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

Review of the manuscript titled 'Measurement report: Shipborne observations of black carbon aerosols in the western Arctic Ocean during summer and autumn 2016–2020: boreal fire impacts'

The manuscript titled 'Shipborne Observations of Black Carbon Aerosols in the Western Arctic Ocean During Summer and Autumn 2016–2020: Boreal Fire Impacts' authored by Deng et al., presents a comprehensive analysis of black carbon levels in the Arctic Ocean during the summer and autumn seasons. Furthermore, it conducts an in-depth examination of the emission sources contributing to elevated black carbon episodes using simulations from a global chemistry-transport model. This manuscript contributes a significant and valuable dataset to the Arctic region, which is often limited in available data. After addressing the major comments outlined, I highly recommend this manuscript for publication in ACP (Atmospheric Chemistry and Physics).

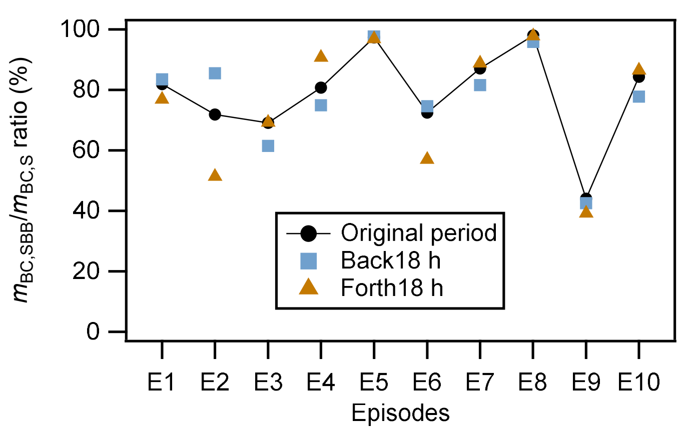
The manuscript details the analysis of 10 specific episodes of black carbon (BC), yet it overlooks the explanation of additional episodes present in the dataset. There's a notable lack of comprehensive explanations for the identification of these episodes and the criteria utilized in the selection process for these events. Additionally, the manuscript employs a model with a notably coarse resolution for simulations, which might introduce biases into the results, particularly when applied to Arctic conditions. The authors must provide a more detailed account of the uncertainties inherent in the model and highlight potential biases when interpreting the simulated values within the manuscript. Addressing these concerns will significantly strengthen the manuscript's scientific rigor and ensure a more robust interpretation of the findings.

Reply> We appreciate the reviewer's valuable comments. We apologize for the incomplete explanation of the selection process for high BC episodes in the preprint. In the preprint, the criterion of 10 ng m^{-3} represents three times the background m_{BC} level determined in Sect. 4.2 of the preprint. Furthermore, considering that longer episodes and higher BC mass concentrations would offer better representation, we refined the selection criteria to ultimately identify approximately 10 episodes. In the revised manuscript, although the estimated background m_{BC} level changed due to the revision of the background period definition, we retained this definition but modified the presentation.

The original expression regarding the definition of high BC episodes "To characterize the sources of the high concentrations of BC in the Arctic Ocean and the marginal seas, high BC episodes were defined as periods when the 1-h m_{BC} was continually greater than 10 ng m^{-3} for 18 h or longer and the mean of valid 1-h m_{BC} during the defined periods was greater than 20 ng m^{-3}. In total, 10 high BC mass concentration episodes were identified (Figs. 1, 3, S1, and S3, Table 2)." 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), we identified periods when the 1-h m_{BC} exceeded 10 ng m^{-3}. From these periods, we further selected those lasting 18 h or longer and the mean of valid 1-h m_{BC} during the selected period was not less than 20 ng m^{-3}. This process allowed us to identify and refine 10 high BC episodes (Figs. 1, 3, S1, and S6, Table 3).". (P17, L29-P18, L1)

As for the other high BC episodes than E3, E8, and E10 that have been explained in detail in the manuscript, their temporal and spatial variation failed to be well reproduced by GEOS-Chem model. From this point of view, it is inappropriate to interpret their origins based on the model result. However, due to the ubiquitous dominance of biomass burning BC in the Arctic Ocean region where those episodes occurred, the biomass burning to total BC ratio was estimated based on the model results in the manuscript. This 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, except for E2 and E6. This is illustrated in the figure below.



This point raised in the preprint is supplemented in the revised manuscript as follows: "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)

Although coarse horizontal resolution may cause larger uncertainty in model simulation results, other mechanisms including transport, deposition, and emissions may play more important roles in determining the uncertainty range of the model. In fact, a multi-model study by Whaley et al. (2022) indicates that models with higher resolution didn't produce more accurate simulations of BC mass. Therefore, we will not re-run the model with a higher resolution.

Uncertainty analysis of the simulations is presented in Sect. 4.3 of the revised manuscript.

Line 20 (Page-1): I would suggest specifying the particular seasons under consideration here. Failing to do so might lead to a misconception that the findings represent an annual average. Clarifying the specific seasons, such as summer and autumn, within the analysis would prevent any misinterpretation regarding the temporal scope of the data and offer a more accurate representation of the findings.

Reply> The expression "in 2019" has been modified to "by the 2019 cruise". And the whole sentence has been modified from "The average m_{BC} in the surface air over the Arctic Ocean (72–80° N) observed in 2019 was over 70 ng m^{-3}, which was substantially higher than in other years (approximately 10 ng m^{-3})." to "The average m_{BC} in the surface air over the Arctic Ocean (72–80° N) observed by the 2019 cruise exceeded 70 ng m^{-3}, which was substantially higher than cruises in other years (approximately 10 ng m^{-3}).".(P1, L19-21)

Line 27 (Page-1): what does this mean " with some near-surface and others in the mid-troposphere"?

Reply> The original sentence "The model analysis indicated that most episodes were attributed to the airmasses transported from boreal fires to the Arctic Ocean, with some near-surface and others in the mid-troposphere." has been modified to "The model analysis indicated that most episodes were attributed to BC containing airmasses transported from boreal fire regions to the Arctic Ocean, with some transport occurring near-surface and others in the mid-troposphere.". (P1, L26-28)

Line 28-30 (Page-1): The abstract appears to contain repetitive sentences. I suggest that the authors revise the abstract to convey essential information more concisely. Condensing the content while retaining crucial details will enhance the abstract's clarity and effectiveness in summarizing the study's key findings and contributions.

Reply> Thank you for your valuable comment. Upon review, we have maintained the majority of the abstract content and made modest alterations. (Page 1)

Line 2 (Page-2): AMAP, 2021b ? Where is 'a'?

Reply> The reference “AMAP, 2021b” has been changed “AMAP, 2021a”. (P2, L4)

Line 7 (Page-2): can include? Rephrase the sentence.

Reply> The word “can” has been changed to “may”. (P2, L7)

Line 13(Page-2): Add relevant citation

Reply> A citation to “AMAP, 2011: The Impact of Black Carbon on Arctic Climate.” has been added to the revised manuscript. (P2, L13)

Line 18 (Page-2): I would recommend citing Gogoi et al., 2021 (Long-term changes in aerosol radiative properties over Ny-Ålesund: Results from Indian scientific expeditions to the Arctic) for the continuous long term monitoring of aerosols in the Arctic. Further, I would recommend comparing the values in this study with Gogoi et al., 2021.

Reply> Thank you for the suggestion. In addition to the cited works and the Gogoi et al. (2021) recommended by the reviewer, there are other papers describing continuous long-term monitoring of BC aerosols in the Arctic that are worthy of being acknowledged. Therefore, instead of including Gogoi et al. (2021) into the reference, we have modified the expression from "(Stohl et al., 2013; Schmale et al., 2022)" to "(e.g., Stohl et al., 2013; Schmale et al., 2022)" to provide examples. (P2, L18-19)

Moreover, due to the significant spatial separation between our study's measurements and those reported by Gogoi et al. (2021), we have chosen not to directly compare the values obtained in our study with theirs.

Line 32 (Page-2): What do authors want to convey here? It is obvious that the BC concentration reduces when the observed location is far from the source region.

Reply> The phrase “source region” has been modified to “continent”. In addition, the whole sentence “They also revealed important characteristics of the spatial distribution of BC in the Arctic Ocean and its marginal seas such as the BC concentration decreases with the growing distance from the source region (Xie et al., 2007; Sakerin et al., 2015, 2021).” has been modified to “They also revealed important characteristics of the spatial distribution of BC in the Arctic Ocean, demonstrating that BC concentration diminishes in the northern direction and decreases as distance from the continent increases (Xie et al., 2007; Sakerin et al., 2015, 2021).”. (P2, L31-P3, L1).

Line 29-32 (Page-3): Rewrite the sentence.

Reply> The original sentence "BC monitors based on light absorption theory were operated during the 2016–2020 summer and autumn expeditions from the North Pacific Ocean to the Arctic Ocean, encompassing the western Arctic Ocean and part of the east Siberian Sea, and back to the North Pacific Ocean to measure the BC mass concentration (m_{BC})." has been modified to "to enhance comprehension of the distribution and sources of BC in the Arctic, the mass concentration of BC (m_{BC}) was monitored across five round-trip expeditions conducted between the North Pacific Ocean and the Arctic Ocean during the summer and early autumn of 2016–2020." (P3, L30-32)

Line 28 (Page 4): What does the 'default parameter settings' mean here? Please explain. Also, explain more about the loading effect corrections and other corrections applied to the data. I would also recommend the authors to explain the uncertainties in the measurements. COSMOS measures at 565 nm, and Aethalometer measurements were at 880 nm. How did the authors compare these measurements at two distinct wavelengths?

Reply> The model of the Aethalometer which was deployed during the cruise is AE22, not AE33. This mistake has been corrected in the revised manuscript. Details including the default parameter settings, uncertainties, and a comparison between the two instruments have been added to **paragraphs 2-4, Sect. 2** of the revised manuscript.

As for details on loading effect corrections and other corrections for the measurements, they are beyond the scope of this paper and will not be included.

Line 13 (Page 5): To avoid the influence of ship exhausts, the authors have used 1- or 5-min data records that occurred when the 1-min wind direction and speed relative to the ship's course within ±60° of the bow and >3 m s−1. Why is it done so for these specific values? What are the reasons for choosing these specific limits?

Reply> We determined the data screening criteria by experimenting with various criteria as well as referring to previous studies, such as Taketani et al. (2016). Other criteria we tested include requiring the 1-min wind direction and speed relative to the ship's course to be within ±70° of the bow and >2 m s^{-1}, respectively. Ultimately, we selected wind direction within ±60° of the bow and a wind speed >3 m s^{-1}. This choice effectively screened out data associated with simultaneous significant decreases in O_3 and increases in BC observed in the open North Pacific Ocean (approximately north of 42° N) and the Arctic Ocean.

Regarding the BC mass concentrations obtained south of 42° N during the 2017 and 2018 cruises, we subjected them to additional scrutiny. Whenever there was a simultaneous decrease in O_3 and an increase in BC, we considered both 1-minute BC and O_3 data as invalid. This additional scrutiny resulted in the exclusion of only 0.12 % and 0.17 % of the observed data from the 2017 and 2018 cruises, respectively. This information has been added to the revised 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)

Line 26 (Page 5): Since the horizontal resolution of GEOS-chem was 2° × 2.5°, how is it valid over the Arctic regions due to the higher grid resolution?

Reply> Although coarse horizontal resolution may cause larger uncertainty in model simulation results, other mechanisms including transport, deposition, and emissions may play more important roles in determining the uncertainty range of the model. In fact, a multi-model study by Whaley et al. (2022) indicates that models with higher resolution didn't produce more accurate simulations of BC mass. Therefore, we will not re-run the model with a higher resolution.

Line 25 (Page 7): Authors have compared the measurements of BC using an Aethalometer and SP2 here. I would not recommend the direct comparison of the measurements with SP2 since the values are highly dependent on the BC size distribution. SP2 has lower and upper bounding limits for the measured BC distributions. This will result in a biased comparison.

Reply> First, the comparison was between COSMOS (not Aethalometer) and SP2. According to Ohata et al., 2019 (Accuracy of black carbon measurements by a filter-based absorption photometer with a heated inlet), the COSMOS can measure m_{BC} in the range 1–3000 ng m^{-3} with ~10 % accuracy as compared with SP2. Furthermore, despite the size-dependence observed in the comparison between COSMOS and SP2, the estimated sensitivity of the COSMOS m_{BC} values, with a 1 h time resolution, to changes in the BC size distributions was less than 10%, falling within the typical natural variabilities of BC size distributions. Therefore, we will maintain our comparison between COSMOS and SP2. Please note that the accuracy of the comparison between COSMOS and SP2 measurements has been addressed in the preprint. In the revised manuscript, this point is addressed even clearer as follows:

"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)." (P5, L1-5)

Figure 1: I would recommend modifying the color bar to a different color option (either jet or something else). It is difficult to identify the variability using these colors.

Reply> The color table has been revised to WarmCold.

Line 15 (Page 9): I would recommend modifying the plots with Lower whisker – 1st percentile, upper whisker – 99th percentile.

Reply> Due to the considerably high values of the 99th percentile of the BC data, we have chosen to retain the 9th percentile as the lower whisker and the 91st percentile as the upper whisker. Additionally, we have included a supplementary figure (Fig. S3) that presents the entire range of observed data, along with a focused view specifically showcasing data within the range of not exceeding 80 ng m^{-3}.

Figure 3: I would recommend changing the left y-axis to a logarithmic scale

Reply> The left y-axis of Fig. 3 has been modified to a logarithmic scale.

Line 6 (Page 11): The derived background concentrations in this study are misleading. The criteria used here need to be proven.

Reply> The definition of background periods have 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)

Line 13 (Page 11): Rewrite the sentence.

Reply> The original expression "Those activities include anthropogenic productive activities producing large amount of air pollutants in low latitude regions that may export to the Arctic through long-range transport (Ikeda et al., 2017; Zhu et al., 2020)," has been modified to "Those activities include industry activities producing large amounts of air pollutants in lower latitude regions that may be transported to the Arctic through long-range transport (Ikeda et al., 2017; Zhu et al., 2020),". (P15, L15-17)

Line 27 (Page 11): Why the authors have chosen 5 days airmass trajectory for this study? I would suggest to explain the reason here.

Reply> The 5-day duration was chosen because this time is long enough to be able to indicate the possible source regions of high BC episodes as well as short enough to ensure the accuracy of the trajectories. This is mentioned in the revised manuscript: "The selection of a 5-day duration allows for identifying potential source regions of high BC episodes (Sect. 4.4) while ensuring trajectory accuracy (Backman et al., 2021)." (P7, L5-7)

Line 10 (Page 12): I would suggest that the authors compile the various comparisons made regarding black carbon concentrations observed in their study with those from other relevant studies in the Arctic region. This compilation could be organized into a table format detailing concentrations, seasons, instruments used, and corresponding references, etc. This tabulated presentation would provide a comprehensive and accessible comparison, aiding readers in understanding the context and variability of BC concentrations in the Arctic across different studies.

Reply> Thank you for the valuable comments. In the revised manuscript, we have incorporated a table (Table 2) containing BC mass concentrations derived from shipborne observations reported in previous studies. Based on this information, we have revised the comparison between this study and previous studies from:

"The BC mass concentration measured in the western central Arctic Ocean is comparable to those in previous studies. In this study, the median and mean (± 1 standard deviation) m_{BC} in August 2020 were 3.4 and 14 (±35) ng m^{-3}, respectively; the values are close to those measured in the central Arctic Ocean during the same period using a similar AE33 aethalometer, where the median and mean (± 1 standard deviation) values were 6.5 and 10 (±22) ng m^{-3}, respectively (Boyer et al., 2023). In addition, on 8–9 September 2016, when the *Araon* was cruising in the Arctic region similar to the observation region of Taketani et al. (2022) (i.e., approximately 70–74° N and 170–153° W), the mean (± 1 standard deviation) values of m_{BC} was 8.2 (±6.0) ng m^{-3}, which is consistent with that presented in Taketani et al. (2022, Fig. 2a) during the same period using a single particle soot photometer (SP2) (Moteki and Kondo, 2010). However, the overall mean m_{BC} observed in the Arctic Ocean of this study (10 ng m^{-3}, Table 1) is 10 times higher than that reported by Taketani et al. (2022). The large difference was likely caused by the spatial and temporal difference between the measurements in the two studies. The cruise routes in this study covered part of the East Siberian Sea region, whereas that in Taketani et al. (2022) was within the Chukchi and Beaufort Sea regions; and the cruise in this study occurred mainly in August whereas that in Taketani et al. (2022) mainly in September. Resultingly, different airmasses containing different BC concentrations could have been observed by this study and Taketani et al. (2022). Therefore, caution on the temporal and spatial ranges should be taken when comparing the mass concentrations of BC observed in the Arctic Ocean. This also indicates the necessity to further study the spatial-temporal variations of BC in the Arctic Ocean. It is noted that the COSMOS can measure the BC mass concentration in the Arctic with ~10 % accuracy as compared with SP2 in 1 h time resolution (Ohata et al., 2019), therefore the instrument difference shouldn't have influenced the comparison between this study and Taketani et al. (2022) largely." to

"The BC mass concentration measured in the western central Arctic Ocean is comparable to some of those in previous shipborne observation studies (Table 2), most of which adopted aethalometer methods, except for those conducted by Taketani et al. (2016, 2021). In this study, the median and mean (± 1 standard deviation) m_{eBC} measured with the AE22 in August 2020 were 3.4 and 14 (±35) ng m^{-3}, respectively; the values are close to those measured in the central Arctic Ocean during the same period using a AE33 aethalometer, where the median and mean (± 1 standard deviation) values were 6.5 and 10 (±22) ng m^{-3}, respectively (Boyer et al., 2023). In addition, the mean (± 1 standard deviation) m_{BC} (COSMOS) in August and early September 2016 was 10 (±11) ng m^{-3}, aligning with the m_{eBC} value of 23 (±55) ng m^{-3} obtained in late July and August 2016 by Ding et al. (2018), considering the relative uncertainty factor of 1–3 for the Aethalometer as discussed in Sect. 2. However, this m_{BC} (COSMOS) value is 10 times higher than that reported by Taketani et al. (2022). The large difference was likely caused by the spatial and temporal difference between the measurements in the two studies. The cruise routes in this study covered part of the East Siberian Sea region, whereas that in Taketani et al. (2022) was within the Chukchi and Beaufort Sea regions; and the cruise in this study occurred mainly in August whereas that in Taketani et al. (2022) mainly in September. Resultingly, different airmasses containing different BC concentrations could have been observed by this study and Taketani et al. (2022). Therefore, caution on the temporal and spatial ranges should be taken when comparing the mass concentrations of BC observed in the Arctic Ocean. It is noted that the COSMOS can measure the BC mass concentration in the Arctic with ~10 % accuracy compared with SP2 in 1 h time resolution (Ohata et al., 2019), as mentioned in Sect. 2, therefore the instrument difference should not have influenced the comparison between this study and Taketani et al. (2022) largely. Furthermore, the m_{BC} (COSMOS) measured in this study is lower than most of the m_{eBC} observed in the Eurasian Arctic Seas, except for that observed in the Laptev Sea in 2018 (Pankratova et al., 2020). This is also likely caused by differences in air mass resources." (P8, L25-P9, L14)

Line 17 (Page 12): I would recommend the authors show R2 values than R throughout the manuscript.

Reply> We present the Pearson correlation coefficient, denoted as R. Hence, we will consistently use R instead of showing R^2.

Table 2: I would recommend showing the region for these episodes in this table.

Reply> The regions for these episodes have been compiled as longitudinal and latitudinal ranges in Table 3.

Line 10 (Page 15): I would recommend the authors to compare the median values of BC for all these episodes since the mean will not represent the actual variabilities.

Reply> The median and mean values for the 10 selected episodes are displayed in Table R2-1. With the exception of Episode 8, the differences between the median and mean values for each episode are not significant. Moreover, we think that the mean values better represent the magnitude of the increment of each episode from the background. Consequently, we will continue to utilize mean values in the manuscript to characterize these episodes.

Table R2-1: The median and mean values (unit: ng m^{-3}) for the 10 selected episodes.

Episodes	E1	E2	E3	E4	E5	E6	E7	E8	E9	E10
Median	21	33	38	20	25	23	22	23	24	28
Mean	20	34	44	25	25	26	32	55	25	29

Figure 6: I would recommend changing the y-axis to a logarithmic scale here for the BC mass concentration.

Reply> The y-axis for the BC mass concentration has been modified to a logarithmic scale.

Figure S1: I recommend changing the color options for the color bar. It is difficult to understand.

Reply> The color table has been revised to WarmCold.

Figure S2: I would recommend changing the axis to a logarithmic scale.

Reply> Changing the axis of a box plot figure to a logarithmic scale proves to be challenging. As an alternative, we include a zoomed-in view of the figure as Fig. S5 in the revised supplementary.

Figure S5, S7, and S10: These figure needs to be revised with detailed color contrasts.

Reply> The color table of these figures has been updated. These figures are Figs. S9, S11, and S14 in the revised supplementary.

Figure S14: Recommend to modify the color bar

Reply> The color table has been revised to WarmCold. This figure is Fig. S18 in the revised supplementary.

Figure S15: I would recommend showing all the episodes in this plot rather than removing the other episodes.

Reply> Back trajectories of all the episodes are included in the revised figure, labeled as Fig. S19 in the revised supplementary.