

Response to Anonymous Referee #3

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

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

Summary

Yange Deng¹ and co-authors present a really valuable data set of mass concentrations of eBC within the Arctic Ocean, a region with sparse data coverage. The authors outline through the use of in situ shipborne measurements and a global chemistry transport model that the proportion of eBC mass arising from biomass burning increases with the latitude at which observations are performed.

I recommend this paper after the following corrections are made.

Major comments:

Method section I suggest

A lack of any description of how HYSPLIT was utilised i.e. which settings were chosen, whether it was a single or ensemble run, the initialised altitude, why 5-days was chosen, whether or not air masses below the mixed-layer were selected for (assume not given the altitude plots), which metrological fields were utilised. Please add more information about how HYSPLIT was utilised.

Reply> Single runs of HYSPLIT were calculated with a time step of 1 hour, a starting height of 500 m above model ground level, and a duration of 5 days. The 5-day duration was chosen because this time is long enough to be able to indicate the possible source regions of observed high BC episodes as well as short enough to ensure the accuracy of the trajectories. Air masses below the mixed layer were included because air masses from boreal fires may transport through the mixed layer to the ship position. The NCEP's GDAS data was utilized to provide meteorological fields. Furthermore, in the revised manuscript, back trajectories initiated at starting heights of 10 and 1000 m above model ground level were calculated to assist in the selection of Arctic Ocean background periods.

The original expression “Furthermore, the NOAA Air Resources Laboratory Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT; Stein et al., 2015) was also applied to assist the interpretation of the BC sources.” has been modified to

“Furthermore, backward trajectories were generated using the NOAA Air Resources Laboratory Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT; Stein et al., 2015) to aid in interpreting the sources of the observed BC and identifying background periods in the Western Arctic Ocean. These trajectories were calculated with a 1-hour time step, initiated at the ship positions with starting heights of 10, 500, and 1000 m above model ground level, and extended for 5 days. 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). The meteorological data used for HYSPLIT was the NCEP’s GDAS data, featuring a horizontal resolution of $1^\circ \times 1^\circ$ and 24 pressure levels extending from the ground to 20 hPa in the vertical direction.”. (P7, L1-9)

Please include the amount of data which is removed when doing the cleaning of the data. Also, include the amount of data which is below the limit of detection. What proportion of the measured observations is left after the various cleaning criteria are applied to the data set e.g. detection limits, averaging thresholds, and ship contamination.

Reply> We mainly cleaned data that may have been contaminated by ship exhaust. After the cleaning process, all other data, including values below the limit of detection, were presented as recorded (e.g., Figs. 1b-1f, 2a, 3, and 6a). Moreover, Fig. S6 illustrates the BC mass concentration before the removal of data influenced by ship exhausts. The quantity of data removed due to potential ship contamination was included in the original manuscript, corresponding to **Lines 10-12 on Page 6** of the revised manuscript.

Specific information regarding data below the limit of detection post the exclusion of potentially ship-contaminated data is provided in the current manuscript: “Within the hourly BC mass concentration data, 5–13 % of COSMOS data and 63–71 % of Aethalometer data fall below their respective detection limits.”. (P6, L16-18)

More justification is required to support why a particle MAC is applied to the absorption coefficient measurements as opposed to another i.e. a more site-specific MAC. The paper could simply report the absorption coefficient instead?

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. The AE22 defines BC from 880 nm absorption with the manufacturer provided MAC, sigma, value of $16.6 \text{ m}^2 \text{ g}^{-1}$. COSMOS also uses the default instrument MAC, value of $10 \text{ m}^2 \text{ g}^{-1}$. Since the measurements were done over a quite wide latitudinal range, we decided to use manufacturer recommended standard MAC rather than site specific values. Paragraphs 2 to 4 of Sect. 2 in the revised manuscript gives more information about the two instruments including the MAC applied.

Furthermore, we insist to report m_{BC} instead of aerosol absorption coefficient because the main instrument we used for the measurements is COSMOS with a heated inlet, whose accuracy in measuring m_{BC} has been critically assessed by previous studies (Ohata et al., 2019; Sinha et al., 2017). For a detailed explanation, please see our response to the reviewer's first specific comment on pages 5 and 6.

Results and discussion section

Elevated values:

The authors highlight 10 events but the underlying reason for why the events are highlighted in the first place is not so clear, this maybe due to a lack of stated research questions.

Also, the explanation for why the criteria was used to defined the episodes was not explained i.e. 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.

Reply> We selected the 10 high BC episodes to characterize the sources of the high concentrations of BC observed during the cruises. This point has been explained in the original manuscript, corresponding to **Line 30 on Page 17** of the revised manuscript.

The criterion of 10 ng m⁻³ represents three times the background m_{BC} determined in Sect. 4.2 of the original manuscript. 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. 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)

Was there also a latitudinal criterion imposed on the event selection to study ‘Arctic Ocean and the marginal seas’ as all events are above 65 degrees norther, and either in the ‘North of 72° N group’ or ‘between 52 and 72° N’ group.

Reply> Yes, we specifically restrict the episode selection to locations 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)

However, it is worth noting that the limitation of north of 65° N does not align with the grouping of latitudes used to illustrate the temporal and spatial variability of BC mass concentrations in Section 4.1. This information is included in the revised manuscript as: “Note that the grouping mentioned here does not comply with the latitudinal constraints (i.e., north of 65° N) used to select high BC episodes in Sect. 4.4.”. (P7, L25-27)

Background:

It is not clearly explained what the authors are trying to achieve by presenting ‘background’ values. Do they mean values that are unaffected by anthropogenic emissions? Also, the criteria has no justification and seems arbitrary.

Reply> We apologize for the misleading in our previous definition of the background periods. The background in the context of this manuscript refers to the preindustrial atmospheric conditions. Whereas finding a situation identical to preindustrial atmospheric conditions is challenging, examining periods in the Arctic Ocean unaffected by regional transport could offer insights into the preindustrial background conditions. This assumes that the impact of natural terrestrial activities, such as wildfires, on BC in the preindustrial Arctic Ocean atmosphere was negligible. This information has been included in the manuscript as:

“While finding a situation entirely identical to the preindustrial atmosphere is challenging due to the pervasive influence of anthropogenic activities on even natural events like wildfires (McCarty et al., 2021), examining periods in the Arctic Ocean unaffected by regional transport could offer insights into the preindustrial atmospheric situations. This assumes that the impact of natural terrestrial activities, such as wildfires, on BC in the preindustrial Arctic Ocean atmosphere was likely negligible, recognizing the inherent uncertainties in making such historical assessments.”. (P15, L9-14)

The definition of 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 ($>72^\circ$ 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)

Supplement:

The supplement is of course important to explain interesting but not essential results. In this specific work, the supplementary is 18 pages long with 15 figures, are all necessary.

Figures S5 & S7: can the mass concentration of eBC not be grouped based on the metres AGL of the collocated back trajectories.

Reply> For Episode 3, corresponding to Fig. S9 in the revised supplementary, the average m_{BC} associated with back trajectories originating above 1.5 km AGL (49 ng m^{-3}) is higher than the average m_{BC} associated with back trajectories originating below 1.5 km AGL (39 ng m^{-3}). For Episode 8, corresponding to Fig. S11 in the revised supplementary, the m_{BC} cannot be well grouped based on the meters AGL, possibly due to the limited number of valid data collected during Episode 8.

We have decided not to include the above information in the revised manuscript because we think it contributes little to the interpretation of m_{BC} transport.

Figure S15: There is a mention of the fact that backward air mass trajectories (back trajectories) are initialised at 500 m above the ship.

Reply> More detailed information on the back trajectories has been included in the revised manuscript as mentioned in our reply to the reviewer’s first major comment.

Specific comments:

The title is a bit misleading. The manuscript present absorption data, not BC data or eBC data. Justify why you made this decision.

Reply> This is because the main instrument we used for the measurements was COSMOS with a heated inlet, whose accuracy in measuring m_{BC} has been critically assessed by previous studies (Ohata et al., 2019; Sinha et al., 2017). We apologize that we haven’t stated this clearly in the preprint. We apologize also for missing any consistency check between measurements by the two different instruments. These two points are included in the revised manuscript as follows:

“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).” (P4, L15-21)

“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.” (P5, L12-21)

Title: ‘impacts of boreal fires’

Reply> The original expression “boreal fire impacts” has been modified to “impact of boreal fires”.

Abstract:

L3: observations of BC ? L14?

Reply> The original expression “BC” has been modified to “BC aerosols”. (P1, L14)

L15-17: Say the actual number of research expeditions

Reply> The total number of research expeditions is disclosed in the final paragraph of the “Introduction” section in the revised manuscript. (P3, L31) This information is not included in the abstract as it does not constitute a key element.

L20: replace ‘over’ with ‘greater than’

Reply> The original expression “was over” has been changed to “exceeded”. (P1, L20)

L22: perhaps due to ‘that’ more frequent wildfires ‘had occurred’ in the Arctic... or perhaps due to more frequent wildfires ‘occurring’ ...

Reply> The word “occurred” has been changed to “occurring”. (P1, L22)

L23: biomass burning contributed the most to the observed BC in ...

Reply> The original expression “composed the largest contribution” has been modified to “contributed most”. (P1, L23)

L23: by mass

Reply> The expression “to the observed BC in the” has been modified to “to the observed BC by mass in the”. (P1, L23)

L26: change to transported from regions with boreal fires

Reply> The original expression “transported from boreal fires to the Arctic Ocean,” has been modified to “transported from boreal fire regions to the Arctic Ocean,”. (P1, L26-27)

Introduction:

L2: ‘climate warming rate’ be more specific e.g. average surface temperature

Reply> The original expression “The climate warming rate in the Arctic is more than three times of the global average,” has been modified to “The annual average surface temperature increase in the Arctic is more than three times the global average increase,”. (P2, L2-3)

L3: ‘decline in Arctic sea ice’ be more specific, what is the parameter? Volume? Extent? What is the season you are referring to? Annual? summertime?

Reply> The original expression “resulting in a rapid decline of Arctic sea ice and extreme cold events,” has been modified to “resulting in a rapid decline of Arctic sea ice extent in all months, a decrease in extreme cold events,”. (P2, L3)

L5: ‘SLCFs’ to SCLF

Reply> We think “SLCFs” is more appropriate. (P2, L6)

L6: ‘has’ to have

Reply> The word “has” has been changed to “have”. (P2, L6)

L6: Can you not refer to the more updated AMAP reports?

Reply> Yes, the “AMAP Assessment 2021: Impacts of Short-lived Climate Forcers on Arctic Climate, Air Quality, and Human Health” has been added to the reference list as “AMAP, 2021b”. (P2, L7).

L6: Arctic aerosol chemical

Reply> The phrase “Aerosol chemical composition” has been changed to “Arctic aerosol chemical composition”. (P2, L7)

L7: American spelling: ‘sulphate’ to sulfate

Reply> The word “sulphate” has been changed to “sulfate”. (P2, L8)

L7: ‘sea-salt’ to sea-spray aerosol

Reply> We think “sea-salt” is more appropriate because the subject of the sentence is “Arctic aerosol chemical composition”. (P2, L8)

L9: causes direct and/or semi-direct or can cause direct and semi-direct climate forcing

Reply> The expression “direct/semi-direct” has been modified to “direct and/or semi-direct”. (P2, L9-10)

L9: it is not just BC acting as cloud condensation nuclei (CCN) – it makes it seem like this by the way you single BC out.

Reply> The original sentence “Particularly, BC aerosols in the Arctic atmosphere can absorb solar radiation directly which causes direct/semi-direct climate forcing and work as cloud condensation nuclei (CCN) which causes indirect climate forcing (McFarquhar et al., 2011).” has been modified to “Particularly, BC aerosols in the Arctic atmosphere can absorb solar radiation directly which causes direct and/or semi-direct climate forcing (AMAP, 2011). Besides, BC aerosols can also act as cloud condensation nuclei (CCN) which causes indirect climate forcing (AMAP, 2011; McFarquhar et al., 2011).”. (P2, L8-11)

L11: change ‘radiation’ to radiation budget and ‘besides’ to leading to an acceleration in the melting of snow and ice

Reply> The original sentence “When deposited onto snow/ice surface, BC can also affect the radiation due to reduction of the surface albedo; besides, it can accelerate the snow/ice melting due to its light absorption ability.” has been modified to “When deposited onto snow/ice surface, BC can also affect the radiation budget due to reduction of the surface albedo, leading to an acceleration in the melting of snow and ice (AMAP, 2011).”. (P2, L11-13)

L15: remove the word ‘warranted’

Reply> The word “warranted” has been changed to “critical”. (P2, L16).

L16: ‘in observatories’ to at ground-based Arctic observatories

Reply> The phrase “in observatories” has been modified to “at ground-based Arctic observatories”. (P2, L17)

L17: ‘Barrow/Utqiagvik’ choose one or write formerly called Barrow for example

Reply> The expression “Barrow/Utqiagvik” has been modified to “Utqiagvik”. (P2, L18)

L18. ‘whereas those data’ to These long-term data sets provided...

Reply> The expression “those data” has been modified to “these long-term datasets”. (P2, L19)

L22: ‘Airborne observations, especially aircraft-based ones, earth-surface conditions’ – not completely understood what is meant by this. Please rephrase.

Reply> The original sentence “Airborne observations, especially aircraft-based ones, are less constrained from earth-surface conditions and allow the vertical profiles of BC in different seasons to be evaluated (e.g., Schulz et al., 2019; Ohata et al., 2021a; Jurányi et al., 2023).” has been modified to “Airborne observations have illustrated the vertical distributions of BC above the Arctic Ocean surface (e.g., Schulz et al., 2019; Ohata et al., 2021a; Jurányi et al., 2023).”. (P2, L22-24)

L22: Why ‘different seasons’ makes it seem like the reason for airborne observations are to expand on the number of seasons in which measurements can be performed

Reply> The original sentence “Airborne observations, especially aircraft-based ones, are less constrained from earth-surface conditions and allow the vertical profiles of BC in different seasons to be evaluated (e.g., Schulz et al., 2019; Ohata et al., 2021a; Jurányi et al., 2023).” has been modified to “Airborne observations have illustrated the vertical distributions of BC above the Arctic Ocean surface (e.g., Schulz et al., 2019; Ohata et al., 2021a; Jurányi et al., 2023).”. (P2, L22-24)

L23: ‘Shipborne observations allow for in situ measurements in the remote Arctic Ocean especially in summer and autumn when the Arctic sea ice is at the minimum’ – Shipborne observations allow for measurements in Arctic Ocean at all times of the year, it is just that in summer and autumn these measurements are easier to perform as the Arctic Ocean is more accessible.

Reply> The original sentence “Shipborne observations allow for in situ measurements in the remote Arctic Ocean especially in summer and autumn when the Arctic sea ice is at the minimum” has been modified to “Meanwhile, shipborne observations have facilitated in situ measurements in the remote

Arctic Ocean, especially in summer and autumn when the Arctic sea ice is at the minimum, making access to the Arctic Ocean easier”. (P2, L24-26)

L28: ‘over’ to ‘in’

Reply> The word “over” has been changed to “in”. (P2, L29)

L29: remove the word ‘preliminary’

Reply> The original expression “These shipborne studies have provided preliminary results of BC mass concentrations for” has been modified to “These shipborne studies have provided BC mass concentration results used for”. (P2, L30)

L30: Arctic Ocean?

Reply> The phrase “Arctic Seas” has been modified to “Arctic Ocean”. (P2, L31)

L30-33: Break up sentence too long

Reply> The original 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)

L32: ‘decreases with the growing distance from the source region’ – remove ‘the growing’ and replace with ‘increasing’

Reply> The expression “with the growing distance from the source region” has been modified to “as distance from the continent increases”. (P2, L33)

Page3:

L1: Mention the Arctic Haze phenomenon or refer to

Reply> The original expression “with high values in winter and low values in summer.” has been modified to “with high values in winter and spring – the Arctic Haze season (Barrie, 1986), and low values in summer and early autumn.”. (P3, L2-3)

L1: Relatively ‘large’ what BC mass concentrations

Reply> By “relatively large” we mean that the seasonal change during MOZAiC observation is larger than that at ground-based observatories. We are sorry for the unclear expression. For clarity, the phrase “relatively large” has been modified to “the changes are larger”. (P3, L2)

L2: ‘those’ to ‘these studies were limited’

Reply> The expression “those studies limited” has been modified to “these studies were limited”. (P3, L4)

L4-5: the parts of the seas which are close to land or are you simply mentioning that these seas are close to land (if so not really necessary)

Reply> The phrase “close to land” has been deleted. (P3, L6)

L9-13: mention the importance of performing measurements in regions with sparse coverage

Reply> This information is included in the revised manuscript by adding “, where data coverage is sparse,” after “especially in the western central Arctic Ocean and East Siberian Sea”. (P3, L14)

L16: Remove ‘whereas models have been improving in the past two decades’

Reply> “Whereas models have been improving in the past two decades” have been deleted. (P3, L18)

L18: ‘The main obstacles include poor understanding in’ replace in with of

Reply> The word “in” has been replaced with “of”. (P3, L19)

L24: ‘in the context of climate change, the likelihood of extreme Arctic fire weather will increase.’ – what do you mean by ‘Arctic fire weather’ this term is unknown to me.

Reply> The expression “extreme Arctic fire weather” has been modified to “extreme fire weather in the Arctic”. (P3, L26)

L25: the impact on BC emissions

Reply> We think that “the impact of BC emissions ... on ...” is appropriate, so we have retained it without alternations. (P3, L26-27)

L27: ‘is’ replace with ‘are’ as you mention ‘studies’

Reply> The word “is” has been changed to “are”. (P3, L29)

L29: ‘BC monitors based on light absorption theory’ rephrase’

Reply> This information has been removed from the revised manuscript. (P3, L30)

L1-4: No aims of the study are listed but the results are described

Reply> The first sentence of the paragraph “In this study, 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

“In this study, 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)

Shipborne observations:

L8: Makes it sound like this is the only thing that was done to clean the data from the ship exhaust, however, later on you mention other steps.

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)

L11: Would be nice to mention clear how many expeditions were carried out in total even though it is clear from seeing the results and assuming that one cruise was carried out each year.

Reply> This information has been added to the last paragraph of the Introduction section. (P3, L31)

L13: Mention the concept of the mass absorption cross-section (MAC) here as this is an important concept

Reply> The MAC values used for the two instruments have been included in the revised manuscript as follows:

For COSMOS, “The default mass absorption cross section (MAC) of $10 \text{ m}^2 \text{ g}^{-1}$ was applied for the derivation of m_{BC} .” (P4, L31-32)

For AE22, “The default manufacturer-provided MAC value of $16.6 \text{ m}^2 \text{ g}^{-1}$ was applied.” (P5, L11)

L14: Why is it referred to as ‘BC’ when what you are measuring is eBC?

Reply> We apologize for the misleading explanation in the preprint. In the revised manuscript, a detailed explanation regarding this is added as follows:

“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 (m_{eBC} , Petzold et al., 2013). Therefore, instead of using m_{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.” (P4, L15-25)

In addition, a comparison between the two instruments, combined with previous studies led us to decide to use Aethalometer data mainly as a reference. This point is included in the revised manuscript as follows: “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.” (P5, L12-19)

L18: ‘data integration time’ what is this term and what does it refer to? Not completely clear.

Reply> The phrase “data integration time” has been changed to “measurement interval”. (P4, L30)

L23: Why was the wavelength 880nm chosen? Why not the wavelength most similar to 565nm, can you justify your reasoning?

Reply> The 2nd AE22 channel uses 370 nm, which is found to respond with great sensitivity to aromatic organic species, and thus is not suitable to compare directly with the COCMOS 565 nm BC data.

L26: Is this MAC value site-specific i.e. for the Arctic? Is there a more relevant MAC value?

Reply> The manufacturer-provided MAC value for AE22 at 880 nm is $16.6 \text{ m}^2 \text{ g}^{-1}$, and this value has been incorporated into the revised manuscript. We opted for the use of this single value over applying a correction method (e.g., Backmann et al., 2017) due to the wide range of latitudes covered in the measurements conducted in this study.

L28: default parameter settings such as ...

Reply> The default parameter settings mainly refer to the default manufacturer-provided MAC value of $16.6 \text{ m}^2 \text{ g}^{-1}$ of AE22. The original expression “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

“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).” (P5, L15-18)

L28: Aethalometer

Reply> The expression “aethalometer” has been modified to “Aethalometer”. (P5, L16)

Page 5:

L6: ‘lump’ replace with lamp

Reply> The word “lump” has been replaced with “lamp”. (P5, L30)

L21-22: why impose this criterion of 40 minutes of valid data records. Does that mean that at least 40/5 = 8 5-min values or 40 1-minute values are needed.

Reply> We use standards that require 40 minutes of valid data recording to ensure that each hour’s data accurately represents that corresponding hour. Yes, at least 8 5-min values or 40 1-min values are required to calculate hourly values. (P6, L15-16)

Model simulations:

L31: When was the data accessed? Why were small fires not included?

Reply> The inclusion of small fires in the model was unintentionally omitted in the manuscript. The original expression “The Global Fire Emissions Database (GFED v4.1)” has been revised to “The Global Fire Emissions Database with small fires (GFED v4.1s).” (P6, L22-23) We apologize for any confusion.

Page 6:

L5: Expand on the criteria you used for the HYSPLIT runs. Defend and state the number of hours you ran the model for? What was the initialised height? What was the temporal resolution for the runs? How did you track the movement of the ship using the model?

Reply> We didn't use the model to track the movement of the ship. Other responses regarding this comment have been addressed in our response to the reviewer's first major comment.

Line 18-20: unclear what is achieved by the groupings and they are not used so much afterwards:

South of 52°N: (in the North Pacific Ocean),

North of 72° N: (mainly in the Canada Basin and the east part of the East Siberian Sea, which are noted as western central Arctic Ocean in the following sections of this study),

between 52 and 72° N: (mainly in the Bering, Chukchi, and Beaufort Seas).

Reply> The groupings are used to characterize the spatial-temporal variations of m_{BC} and the contribution of biomass burning to the observed BC along the cruise route. The main findings are a decrease in m_{BC} with increasing latitude (Paragraph 2, Sect. 4.1) and an increase in the contribution of biomass burning to the observed BC as latitude increases (Paragraph 1, Sect. 4.4).

Please note that the grouping mentioned here does not comply with the latitudinal constraints (i.e., north of 65° N) used to select high BC episodes in the Arctic. This information has been added to the second paragraph of Sect. 4.1 as follows: "Note that the grouping mentioned here does not comply with the latitudinal constraints (i.e., north of 65° N) used to select high BC episodes in Sect. 4.4." (P7, L25-27)

Page 7-8:

L4: remove 'pre-existing'

Reply> The phrase "pre-existing" has been removed. (P8, L11)

L34-page8, L1: 'necessity to further study the spatial-temporal variations of BC in the Arctic Ocean' move to discussion

Reply> The sentence "This also indicates the necessity to further study the spatial-temporal variations of BC in the Arctic Ocean." has been removed from the paragraph.

L3: 'shouldn't' to should not

Reply> The expression "shouldn't" has been modified to "should not". (P9, L11)

L7: 'Barrow' to 'Utqiagvik'

Reply> The word "Barrow" has been changed to "Utqiagvik". (P10, L3)

L10: Is it possible to know whether BB reach further north or that the ratio increases because the anthropogenic influence decreases at the latitude increases?

Reply> Yes, it is possible by comparing the spatial distribution of anthropogenic BC and biomass burning BC based on the model result as illustrated in Sect. 4.4 in this study. In the revised manuscript, this information is more clearly represented by adding references to previous studies: “This aligns with previous model studies indicating that during summer, the transport efficiency of low latitude anthropogenic BC to the Arctic was low, and biomass burning BC contributed more than 63 % to the surface BC in the Arctic (Ikeda et al., 2017; Zhu et al., 2020).” (P17, L25-28)

Page 9:

Figure 2: The smaller values are not very clear. Seems as though values are cut-off – represent or detail in the caption the values that you cut-off. The font size for the y-axis can be increased. Remove ‘Year/month/day’ not needed. You could separate the plot based on latitudes and focus on the events which are all above 65 degrees north. Not 100% clear what time resolution is presented here.

Reply> Figure 2 has been revised to include individual data in panel (a). A statement “All data presented here is at 1 h time resolution and the data influenced by ship exhaust has been removed.” has been added to the caption. The label for the bottom axis has been removed. The full-scale panel (a) and a zoomed-in view of panel (a) with the y-axis maximum set to 80 ng m⁻³ are now shown in Fig. S3.

4.2 Background BC concentration in the western central Arctic Ocean

L9: remove ‘pristine region’ not scientific or perhaps rephrase to distant from sources of anthropogenic pollutants

Reply> We apologize for the misleading information in the preprint. The original expression “Ideally, the Arctic Ocean is one of the pristine regions in the world and background concentrations of primary air pollutants such as black carbon there should be close to zero. However, zero background black carbon may not be true to the Arctic marine boundary layer atmosphere due to the influence of” has been modified to

“While finding a situation entirely identical to the preindustrial atmosphere is challenging due to the pervasive influence of anthropogenic activities on even natural events like wildfires (McCarty et al., 2021), examining periods in the Arctic Ocean unaffected by regional transport could offer insights into the preindustrial atmospheric situations. This assumes that the impact of natural terrestrial activities, such as wildfires, on BC in the preindustrial Arctic Ocean atmosphere was likely negligible, recognizing the inherent uncertainties in making such historical assessments.” (P15, L9-14)

L10-13: 'should be close to zero' why? What does 'close to zero' mean? Use the literature to suggest typical values as opposed to some arbitrary 'close to zero' statement which is quite meaningless. Who claims that the marine Arctic boundary layer should have a 'zero background'? BC has been transported up to the Arctic prior to pre-industrial times (McConnell et al., 2007). McConnell et al., (2007) present historical BC from Greenland ice-cores showing non-zero values.

Reply> We apologize for the misleading information in the preprint. The original expression “Ideally, the Arctic Ocean is one of the pristine regions in the world and background concentrations of primary air pollutants such as black carbon there should be close to zero. However, zero background black carbon may not be true to the Arctic marine boundary layer atmosphere due to the influence of” has been modified to

“While finding a situation entirely identical to the preindustrial atmosphere is challenging due to the pervasive influence of anthropogenic activities on even natural events like wildfires (McCarty et al., 2021), examining periods in the Arctic Ocean unaffected by regional transport could offer insights into the preindustrial atmospheric situations. This assumes that the impact of natural terrestrial activities, such as wildfires, on BC in the preindustrial Arctic Ocean atmosphere was likely negligible, recognizing the inherent uncertainties in making such historical assessments.” (P15, L9-14)

L13: 'anthropogenic productive' replace with 'industry'

Reply> The phrase “anthropogenic productive” has been replaced by “industry”. (P15, L16)

L14: lower latitude

Reply> The phrase “low latitude” has been modified to “lower latitude”. (P15, L16)

L14: 'export' replace with transport

Reply> The word “export” has been replaced by “be transported”. (P15, L17)

L15: how do commercial fisheries significantly impact Arctic BC?

Reply> The phrase “commercial fisheries and” has been removed. (P15, L19)

L17: rephrase the sentence 'coastal region along the Arctic climate warming' doesn't make sense

Reply> The original expression “along the Arctic climate warming” has been modified to “driven by the warming Arctic climate”. (P15, L19)

L18: which allows for the transport...

Reply> The expression “which allows transport...” has been modified to “which allows for the transport...”. (P15, L21)

L19: Separate sentence/start a new sentence: ‘and the stable atmospheric conditions’

Reply> The original sentence “In winter and early spring, the buildup of anthropogenic pollutions, due to the expansion of the polar dome, which allows transport of anthropogenic pollutants from continental regions further south, and the stable atmospheric conditions, can lead to monthly mean m_{BC} of as high as more than 100 ng m^{-3} (e.g., Boyer et al., 2023).” has been modified to “In winter and early spring, the buildup of terrestrial anthropogenic and natural pollutants occurs due to the expansion of the polar dome, which allows for the transport of pollutants from continental regions further south. This buildup, combined with stable atmospheric conditions, can result in monthly mean m_{BC} levels exceeding 100 ng m^{-3} (e.g., Boyer et al., 2023).”. (P15, L20-23)

L20: replace with higher

Reply> The expression “of as high as more than” has been changed to “levels exceeding”. (P15, L23)

L21: surface layer atmosphere

Reply> The original expression “Arctic Ocean surface atmosphere” has been modified to “Arctic Ocean surface layer atmosphere”. (P15, L25)

L24-25: Why are summer and early autumn months most suitable to evaluate background levels? What are you trying to achieve? By background do you mean pre-industrial – the background will have a seasonality as described by the Arctic Haze phenomenon.

Reply> We appreciate your precious questions and comments. We apologize for the misleading in our previous definition of the background periods.

Yes, the background in this context refers to the preindustrial Arctic Ocean conditions, which could have seasonality due to changes in atmospheric transport through the year. In the revised manuscript, we modify the definition of the background period to be a period in the Arctic Ocean that is not affected by regional transport, which is more likely to occur during summer and early autumn. Please see Sect. 4.2 for details.

L26: The definition of ‘background periods’ needs to be properly explained it is not clear why the criteria you mentioned have been applied. What is it you want to achieve with this notion of background (perhaps unaffected by anthropogenic emissions?). Background Arctic Ocean conditions perhaps is better wording. The definition relies on values below the detection limit ‘below the lowest

detection limit' there is it valid? You could additional work out how often the measurements exceed these background values.

Reply> We apologize for the misleading in our previous definition of background period. The background we intended to define was the background Arctic Ocean conditions which was not influenced by regional transport of air pollutants from terrestrial regions. This situation was more likely to occur in summer than winter months and may represent the preindustrial summer conditions in the Arctic Ocean. Therefore, we have revised the definition of Arctic Ocean background periods from “The background periods in the western central Arctic Ocean ($>72^\circ$ 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 ($>72^\circ$ 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)

L28: remove 'continually'

Reply> The entire sentence was deleted from the revised manuscript.

Page 12:

L2: replace 'imported' with long-range transport

Reply> The comparison between estimated background m_{BC} and central Arctic Ocean m_{BC} has been deleted from the revised manuscript.

L3-4: 'might be twice the actual values because the default' this is repeated

Reply> The comparison between estimated background m_{BC} and central Arctic Ocean m_{eBC} has been deleted from the revised manuscript.

L7: There are decreasing trends in the Arctic and this has been well reported and hence is representative of this time period e.g. Hirdman et al., 2010, Sharma et al., 2004,

Reply> Although previous studies reported a decreasing trend in winter eBC, the annual trend in summer eBC remains uncertain (e.g., Sharma et al., 2004; Schmale et al., 2022). Could the reviewer be more specific with the comment? Thank you so much.

L9: Unsure how the background is calculated.

Reply> We apologize that we haven't stated this point clearly. The original expression "The overall mean of background m_{BC} calculated from m_{BC} at 1 h time resolution for 53 hours was $3.3 (\pm 1.5) \text{ ng m}^{-3}$ (Table 1)." has been modified to "The overall mean of background m_{BC} , calculated from COSMOS m_{BC} at 1 h time resolution over 52 hours, was $7.5 (\pm 8.5) \text{ ng m}^{-3}$." (P16, L10-11)

L17: Why not R2?

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

Page 13, 4.4 Sources of High BC episodes:

In 4.4. Sources of High BC episodes, the phrasing is not consistent, these episodes are referred to as 'high BC episodes', 'elevated BC mass concentration periods' and 'high BC mass concentration episodes.'

Winiger et al., 2016 might be a good study to read and reference.

Reply> We appreciate the paper the reviewer recommends.

In the revised manuscript, these episodes are uniformly referred to as high BC episodes. Accordingly, the following revisions are made in the revised manuscript:

"elevated-BC episodes" → "high BC episodes". (P1, L24)

"high BC mass concentration episodes" → "high BC episodes". (P17, L33)

"elevated BC mass concentration episodes" → "high BC episodes". (P29, L22)

L12 – 15: Why are the elevated values defined as such what is the reasoning? i.e. why the criteria '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} . Why 18 h? Why 10 ng m^{-3} , why 20 ng m^{-3} ? What is the motivation?

Reply> 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, we have 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)

L21-22: ‘Note that in addition to these 10 episodes, high BC was also observed in the Arctic at other times, such as on 14-18 August 2020’ – is this sentence needed?

Reply> This sentence has been removed from the revised manuscript.

L25: support what you mean by ‘well reproduced’?

Reply> What we wanted to say here is that both observed and model data showed peaks during these episodes. Therefore, the original expression “Furthermore, the temporal and spatial variations of E3, E8, and E10 were well reproduced by GEOS-Chem model (Fig. 3).” has been complemented as “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).” (P18, L14-16)

Page 14:

L6: replaced ‘occurred’ with was measured

Reply> The expression “occurred” has been modified to “was measured”. (P19, L6; P20, L11; and P26, L12)

L11: Remove ‘Contour plots’

Reply> The expression “Contour plots in Fig.” has been changed to “Figure”. (P19, L11; and P21, L7)

Page 16:

L3: rephrase ‘... burning BC and surface winds before to after Episode 8’

Reply> The expression “... burning BC and surface winds before to after Episode 8” has been modified to “... burning BC and winds before, during, and after Episode 8”. (P21, L4)

5 Summary and conclusions:

Page 23:

L2 ‘higher than low latitude’ – what does this mean?

Reply> The original sentence “Relatively low m_{BC} were observed at higher than low latitude regions.” has been modified to “Relatively low levels of m_{BC} were observed at higher latitude regions.”. (P29, L5-6)

L8-10: Repeated definition

Reply> This part has been deleted from the revised manuscript.

L24: Arctic BC mass

Reply> The expression “observed Arctic BC” has been modified to “observed Arctic BC mass”. (P29, L32)