Answer to referee #2

The manuscript titled "Drivers controlling black carbon temporal variability in the Arctic lower troposphere" by Gilardoni et al. investigates the seasonality and meteorological influences on black carbon (BC) concentration in the Svalbard region through a combination of modeling and observational measurements. It is found that wet scavenging plays a large role in modulating seasonal variability and that circulation, from a boundary layer to synoptic scale, impacts the shorter-term BC variability. The paper has significant potential to increase understanding on Arctic BC concentration and its controlling factors.

Overall, the manuscript is well-written, with the knowledge gap and scientific objective of the manuscript clearly laid out. The paper flows well and is organized in a way that distinctly addresses each objective. However, there are several areas throughout the paper that are unclear or lack necessary supporting information. These issues should be addressed to improve the clarity and strengthen the claims of the manuscript. Following minor revisions, I recommend publication.

General Comments

The manuscript is strongly based on the idea that there are two periods (cold and warm seasons) with different responses in each period. The data were separated into these two chosen periods, November- April and May-October, before any analysis or underlying trends were observed. What is the basis for the selection of the month range for each period? There is little discussion in the manuscript that gives support and explanation for the reason why the data were separated in this way. Is this cold season of November-April and warm season of May-October similarly used to subset data in this region in previous publications? If so, please include references and brief discussion in the introduction or methods. If not, was this based on analysis of measurements? For example, if it is based on average temperature (or some other variable) and there are clear differences between the two periods, then it would be useful to include a discussion (perhaps in the methods or supplement) on how and why these two periods are distinguished. I understand that the goal is to investigate seasonal variability, but why was it chosen to separate the data into two periods rather than say four? The manuscript would benefit from further clarification and support on this subject.

We would like to thank the referee for this comments and we agree that the separation of the study period in two seasons is explained with limited details in the original manuscript. The identification of a warm and a cold period is based on previously published analysis of eBC variability at Zeppelin (Eleftheriadis et al., 2009, Stathopoulos et al. 2021), at about 1 km from the Gruvebadet Atmospheric Laboratory (GAL). eBC showed significantly different source regions during the two periods, defined as cold season from November to April and warm season from May to October. Furthermore, Stathopoulos et al. (2021), showed that large scale circulation patterns that impact the pollutant transport from lower latitudes (NAO, OA, and SCAN) are characterized by opposite behaviors during this two periods of the year.

Although eBC during transition months might not be well captured using a simplified seasonality composed by only two periods, introducing a larger number of seasons would have led to smaller seasonal datasets with limited representativeness.

The "Generalized additive Model" paragraph in the section method was modified as follows:

> "We built two different GAMs to describe eBC concentration observed during the cold (November - April) and the warm (May - October) periods, assuming that different mechanisms

might control pollution variability. This assumption is corroborated by the fact that eBC observed at Zeppelin (at about 1 km from GAL) is characterized by significantly different source regions during the warm and cold season, as defined above (Eleftheriadis et al. 2009; Stathopoulos et al. 2021). Furthermore, Stathopoulos et al. (2021), highlights that large scale circulation patterns that impact the pollutant transport from lower latitudes (NAO, OA, and SCAN) shows opposite behaviors during these two periods of the year."

Specific Comments

Line 37: This paragraph appears to be contradictory and the key point is unclear. The first sentence states that overestimation of BC scavenging may cause BC model underestimation. The following two sentences agree with this first statement. However, the last statement suggests the opposite by stating that models tended to underestimate rather than overestimate BC scavenging. Is this sentence supposed to say that models underestimate *BC* in agreement with the first sentence or underestimate *BC scavenging* which opposes the first sentence? If this last sentence is supposed to contradict the previous sentences, then it should be placed in another paragraph with further discussion on the opposing point. Alternatively, with more emphasis that there are contrasting results in the literature they can be placed in the same paragraph. Please clarify on the key point of this paragraph.

We thank the referee for pointing out the ambiguity of this section and we removed the last sentence, which is misleading in this context. We also added a reference specific to BC scavenging modeling parametrization at global scale and in the Arctic, citing the paper from Lund et al., 2018 and Lund et al., 2017.

"The overestimation of BC scavenging in polar regions, where ice-clouds are dominating, has been proposed as one of the factors responsible for BC model underestimation. Browse et al. (2012) enhanced the model ability to describe BC Arctic seasonality optimising the in-cloud and below cloud scavenging scheme. Zhou et al. (2012) improved the agreement between modelled and observed BC deposition by reducing scavenging in ice and in mixed-phase clouds, but still failed in reproducing the atmospheric concentrations. ~~Furthermore, recent studies indicate that BC atmospheric lifetime is shorter than previously expected (Samset et al. 2014, Lund et al. 2018, Wang et al. 2014, Matsui et al. 2018), indicating that models tended to underestimate rather than overestimate BC scavenging (Lund et al. 2018).~~ Lund et al. (2018) observed that reducing the ice-cloud scavenging significantly increased the BC surface concentration in the Arctic, but declined model performance at lower latitudes, highlighting the need of a deeper understanding of processes and properties controlling BC scavenging (Lund et al., 2017)."

Line 236: The second paragraph of Section 3.2 on the BC MAC reported in literature and the determination of the value used in this paper does not seem vital to this section or the main manuscript. By moving this discussion to the supplement, it would aid in flow and readability of the manuscript and better highlight only the necessary key points of the results. Additionally, there are several literature values listed throughout this paragraph which makes it hard remember each in order to place the 10.2 m^2g^{-1} in context of the literature. It could be beneficial to summarize all values in a figure. This way, it would be easier to visualize where the 10.2 m^2g^{-1} used in this manuscript falls in comparison to previous literature.

We thank the referee for the suggestion. Accordingly, we modified the first paragraph of section 3.2 as follows:

"eBC was then derived from the absorption coefficient time series at 660 nm, assuming a constant Mass Absorption Cross section (MAC) equal to 10.2 m^2g^{-1}, in agreement with the MAC calculated by Ohata et al. (2021) with instrument techniques similar the to ones employed in this study (See section S1 and Table S1)."

We then moved the comparison of MAC values reported in the second paragraph to the supplementary material, where we summarized the MAC values in a Table (Table S1).

Table S1. MAC values reported from previous studies in the Arctic region.

MAC at 550nm	Notes	Reference
7.5 m^2g^{-1}	Freshly emitted BC	Bond and Bengstrom, 2006
8.8 – 10.5 m^2g^{-1}	Arctic	Zanatta et al. 2016
9.8 m^2g^{-1}	Svalbard (spring)	Zanatta et al. 2018
5 – 9 m^2g^{-1}	Alert (3 year data)	Sharma et al., 2017
10.8 – 15.1 m^2g^{-1}	Arctic	Ohata et al. 2021

References have been added to the supplementary material.

Line 272: "the largest difference was observed in July 2020, when eBC concentration was...". It is unclear whether "eBC concentration" here is referring to the mean or median value. I assume it is the mean value, but it would be useful to specify.

The sentence has been rephrased as follows:

"During the warm period, the largest difference was observed in July 2020, when the mean eBC concentration was higher compared to the same months of the remaining analyzed years."

Table 2: Do none of the variables in the table have statistical significance greater than 99%? In the caption it is stated that this is marked with two asterisks (**), but (**) never appears in the table. Please remove this description if it is unused, and/or verify that none of the variables mistakenly have one (*) or three (***) asterisks instead of two (**).

We thanks the referee for pointing out this inconsistency. We modified the Table and Table caption as reported below:

Table 2. As in Table 1, but for the warm season. The p-values are indicative of each variable statistical significance (** corresponds to significance larger than 99.9% and * larger than 95%).

Warm season	Dev. Explained	p-value
Julian Day (Jul)	0.13	$<2\ 10^{-16}$ **
Day of the Year (DOY)	0.22	$<2\ 10^{-16}$ **
Temperature (Temp)	0.32	$<2\ 10^{-16}$ **
Relative Humidity (RH)	0.36	$7.21\ 10^{-5}$ **
Radiation (Rad)	0.40	$4.59\ 10^{-2}$ *
BLH	0.43	$3.45\ 10^{-5}$ **
AO	0.46	$1.38\ 10^{-2}$ *

Line 380: It is hard to tell from Figure S7 that colder temperatures corresponded to airmasses that spend more time over the Arctic Ocean and Greenland coasts. There is hardly noticeable difference between Figure S7b and S7d. I suggest reproducing this figure by plotting a contour map of the difference of Figures S7b and S7d. This would clearly show the locations of greatest difference and perhaps more strongly support this claim. Otherwise, I suggest removing this statement.

We agree with the referee that the difference between panel b and d is difficult to capture. We modified Figure S7 adding an additional panel showing the difference between the residence probability maps to support the statement that colder temperatures corresponded to airmasses that spend more time over the Arctic Ocean and Greenland coasts.

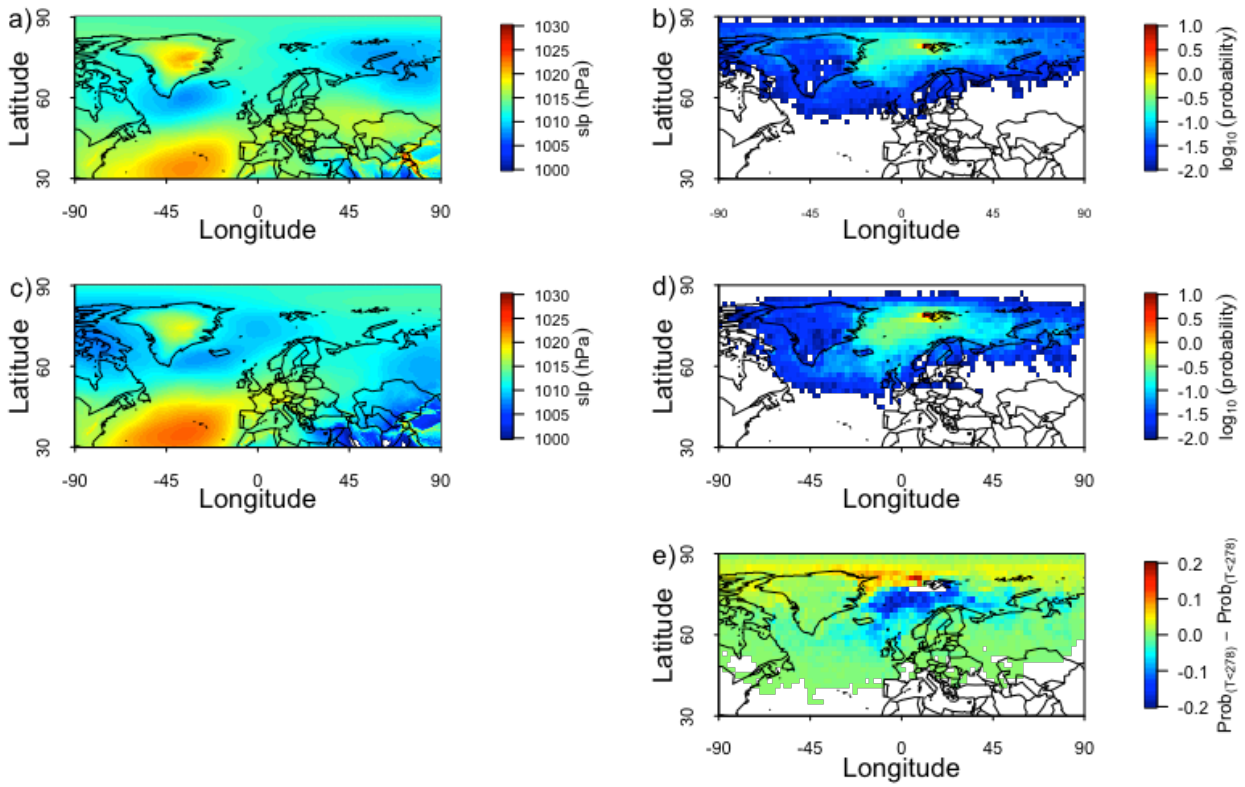

Figure S7. Average sea level pressure maps and residence time probability maps when the temperature at GAL was lower (panel a and c) and higher (panel b and d) than 278 K during the warm season; panel e shows the probability difference map between colder and warmer days (panel b – panel d). Residence time probability maps are based on 7-day back-trajectories. The threshold of 278 K was defined based on the temperature impact on eBC concentration reported in Fig. 7c.

Line 395: This paragraph is lacking support for the reason why eBC increases with increasing radiation. Is the statement "Low-level clouds are usually associated with rain and drizzle, with the later [sic] one not well captured by cumulative daily precipitation measurements" based on previous literature or based on measurements analyzed in this study? Please include supporting references for this statement and/or add further discussion of the analysis that led to this statement.

Cumulative precipitation daily data were derived from hourly precipitation values. Hourly precipitation measurements are usually affected by large error when drizzle and light-precipitation dominates, due to the small precipitation rates (<0.5 mm h^{-1}) (Nystuen et al., 1999). This references were added to the manuscript and the sentence was corrected as follows:

"Low-level clouds are usually associated with rain and drizzle, with the latter one not well captured by cumulative hourly ~~daily~~ precipitation measurements (Nystuen et al. 1999)."

Figure S6: It is hard to visualize how the winds are changing (which is a relevant point discussed in the manuscript) with a different axis range in each plot. Please use the same fixed axis range for all plots to be able to compare and contrast the plots with each other more easily.

We modified the scale of the wind rose plot in order to use the same range for all the panels.

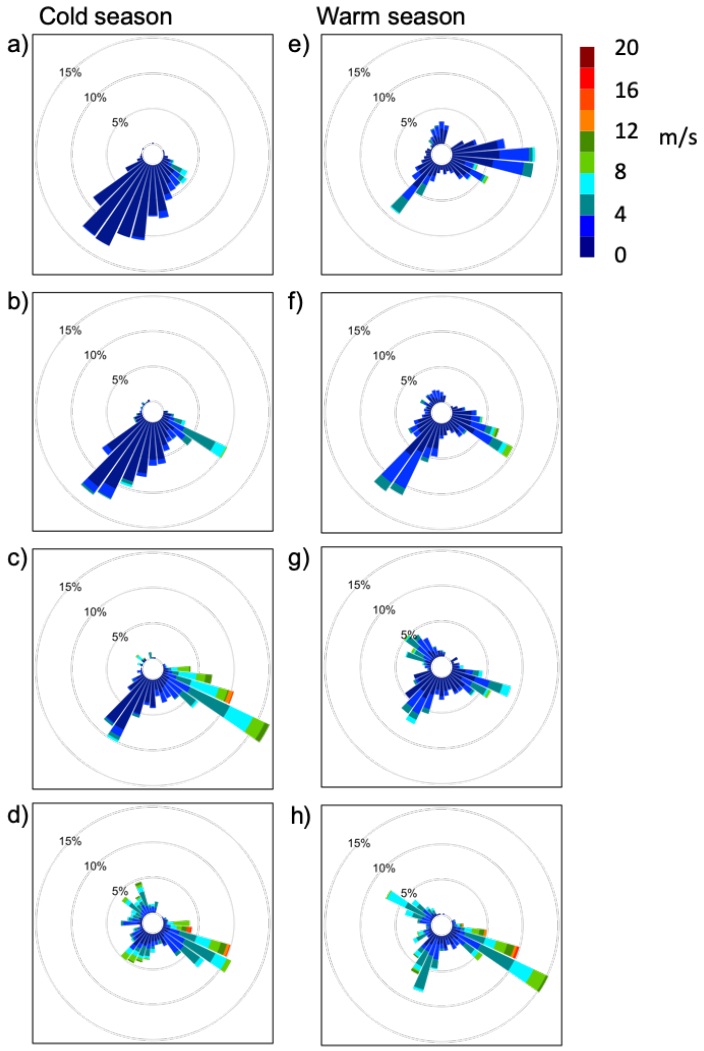

Figure S6. Wind roses describing main wind pattern at GAL during the cold (a-d) and the warm season(e-h) when blh was below 100 m (a and e), between 100 and 200 m (b and f), between 200 and 400 m (c and g), and between 400 and 600 m (d and h).

Figure S8: Are the vertical lines extending to the 25th and 75th percentiles or standard deviation? Is the thick line the median or mean? Please clarify in the caption/description.

Figure S8 caption was modified to specify the meaning of the continuous lines and the vertical lines:

"Figure S8. Change of specific humidity (panel a) and pressure (panel b) along back-trajectories, for air masses arriving at GAL when RH was higher (orange) and lower (blue) than 70%. Continuous lines indicate mean values, while vertical lines correspond to standard deviation."

Several small grammatical issues are listed below, please address them for clarity and ease of reading:

- Line 38: Add “the” so that it reads “optimizing the in-cloud and...”

Corrected

- Line 52: The word “challenging” appears in the wrong place in the sentence. It should read “Both these factors make the quantification of biomass burning impact on the Arctic lower troposphere challenging” or “Both these factors make it challenging to quantify the biomass burning impact on the Arctic lower troposphere”.

The sentence was modified according to the suggestion

- Line 119: Replace “to” with “with”.

“To” was replaced.

- Line 296: Change “increased” to “increase” (or remove “of”).

The sentence was corrected

- Line 298: “investigates” should be “investigate”.

The verb was corrected

- Line 316: “pressire” should be “pressure”.

The word pressure was corrected

- Line 325: This sentence is unclear. It seems it should read as “air masses reaching Svalbard spent most of the time over the ocean”, or “air masses reached Svalbard after spending most of the time over the ocean”.

The sentence was corrected adding the word “after”

- Line 339: Missing “Å”. Should be “Ny- Ålesund”.

The name of the village was corrected

- Line 357: This sentence should be either plural or singular (not both). It should read as either “indicates a larger interannual difference” or “indicates larger interannual differences”.

The sentence was corrected in the plural form

- Line 374: Change “increased” to “increase”.

The word was changed

- Line 396: Add “than” so that it reads “to more than 100...”.

The word “than” was added

- Line 400: Change “later” to “latter”.

The spelling was corrected.