

Referee #1

Black text: Referee comments by Anonymous Referee #1

Blue text: Our replies

Red text: Modified parts of the manuscript

* Line number is the number in the tracked manuscript.

The manuscript by Kinase et al. (Long-term observations of black carbon and carbon monoxide in the Poker Flat Research Range, central Alaska, with a focus on forest wildfire emissions) reports ~ 4 years of continuous monitoring data of black carbon (BC) and carbon monoxide (CO). The observation data were simulated by the FLEXPART-WRF model. The data were analyzed by focusing on high-concentration events that were likely induced by biomass burning. The authors suggest that the emission ratio of BC may depend on fire radiation power (FRP). The topic is within the scope of the journal. The organization and writing quality of the manuscript can be improved. I suggest all the authors of the manuscript carefully read through it again to improve both the general organization and expressions of individual contents. I have the following comments.

→Thank you for your kind review and important comments. All the comments were helpful for us in improving our manuscript. Please see our answers to the specific comments below.

Major comment

I believe that Figure 6 shows the most important message of the manuscript. Although there might be a relationship between BC/delta_CO and FRP, the data are highly scattered ($r = 0.44$). At the current moment, it is very difficult for me to evaluate if such a trend exists. Comparison with some other data may help better understand the data. For instance, BC/delta_CO data from laboratory experiments (smoldering, flaming) can be compared with the present dataset. If the existing dataset from the laboratory supports the idea, the conclusion can be strengthened.

→Thank you for your important comment. There are several studies that have tested the relationship between the BC/ Δ CO ratio and the modified combustion efficiency value (MCE) to characterise the BC/ Δ CO ratio as a function of combustion intensity. For example, Pan et al. (2017) measured BC, CO, and CO₂ in small-scale combustion

experiments. In their experiment, dry and wet wheat straw samples and dry rapeseed plant samples were burned, and the time evolution of the BC/ Δ CO ratio and MCE were observed. They reported that BC is mostly produced during the flaming process, and the evolution of the BC/ Δ CO ratio which depends on the combustion stage could be confirmed ($13.9 \pm 10.1 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ for MCE larger than 0.95 cases, and less than $7.1 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ for MCE smaller than 0.96 cases). Although these BC/ Δ CO ratios are larger than our observed BC/ Δ CO ratio, differences in fuels might be a possible reason. Selimovic et al. (2018) also burned some types of fuels, including coniferous trees, in a large indoor combustion facility and measured BC, CO, and CO₂ with various other chemical species. They reported a high BC/ Δ CO ratio ($13.8 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ on average) and a low BC/ Δ CO ratio ($4.7 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ on average) in the conditions of flaming dominated and smouldering dominated, respectively, in the same range as our observed values. Moreover, (Chakrabarty et al., 2016) tested Alaskan peat and Siberian peat in the combustion chamber under smouldering conditions, and low BC/ Δ CO ratios ($1.2\text{--}2.6 \text{ ng m}^{-3} \text{ ppbv}^{-1}$) were reported.

The positive relationship between the BC/ Δ CO ratio and MCE is also observed in the field measurements (Kondo et al., 2011b; Selimovic et al., 2019). Although MCE and FRP are different parameters, both parameters indicate combustion conditions and have a strong correlation (Wiggins et al., 2020). Therefore, our observed evolution of BC/ Δ CO ratios with increasing FRP can be a reasonable result. We modified our manuscripts as shown below.

“lines 328–340: For example, Pan et al. (2017) measured BC, CO, and CO₂ from biomass burning in small-scale combustion experiments. In their experiment, dry and wet wheat straw samples and dry rapeseed plant samples were burned, and the time evolution of BC/ Δ CO ratio and MCE were observed. They reported that BC is mostly produced during the flaming process, and the evolution of the BC/ Δ CO ratio which depends on the combustion stage could be confirmed ($13.9 \pm 10.1 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ for MCE larger than 0.95 cases, and less than $7.1 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ for MCE smaller than 0.96 cases). Although these BC/ Δ CO ratios are larger than our observed BC/ Δ CO ratio, differences in fuels might be a possible reason. Selimovic et al. (2018) also burned some types of fuels, including coniferous trees, in a large indoor combustion facility and measured BC, CO, and CO₂ with various other chemical species. They reported a high BC/ Δ CO ratio ($13.8 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ on average) and a low BC/ Δ CO ratio ($4.7 \text{ ng m}^{-3} \text{ ppbv}^{-1}$ on average) in the condition of flaming-dominated and smouldering-dominated, respectively, in the same range as our observed values. Moreover, Chakrabarty et al. (2016) tested Alaskan peat and Siberian peat in the combustion chamber under a smouldering conditions, and low BC/ Δ CO ratios ($1.2\text{--}2.6 \text{ ng m}^{-3} \text{ ppbv}^{-1}$) were reported. The positive relationship between the BC/ Δ CO ratio and

MCE is also observed in the field measurements (Kondo et al., 2011b; Selimovic et al., 2019)....”

Other comments

Line 17

The observation period needs to be clearly stated, including the end of the observation.

→Modified.

“line 17: from April 2016 to December 2020.”

Line 18

It is better to show some statistics (e.g., 10th and 90th tile values), rather than only showing the median.

→Modified

“lines 17–19: The medians, 10th, and 90th percentile ranges of the hourly BC mass concentration and CO mixing ratio throughout the observation period were 13, 2.9, and 56 ng m⁻³ and 124.7, 98.7, and 148.3 ppb, respectively.”

Introduction

There are too many usages of 'significant.' Reserve the usage of such an adjective only for really meaningful cases (e.g., demonstrating statistical significance).

→We checked ‘significant’ throughout our manuscript and changed the wording or deleted where necessary.

Line 88

The reviewer is not familiar with the observation site. Based on the description in section 2.1, it seems that the observation site is located in a forest. Can the influence of forest

canopy on particle deposition be ignored when the sampling height is 5.5 m above the ground level?

→Observatory is located on a mountain hill, with non-tall (~2m) sparse black spruce forest. Therefore, deposition effects can be ignored. We added a sentence shown below.

“ Line 79–80: Note, that the effects of deposition by trees and canopies can be ignored because the laboratory is located on a mountain hill, with non-tall (~2m) sparse black spruce forest.”

Line 89

Are there any reasons why the authors used the PM_{1.0} cyclone? Most BC data are collected using PM_{2.5} inlets. The usage of PM_{1.0} cyclone makes comparison with literature data to be difficult.

→As pointed out by this comment, the PM_{2.5} cyclone has been used in many studies. However, also the PM_{1.0} cyclone has been use in as many other BC studies (Kondo et al., 2011a; Mori et al., 2020; Selimovic et al., 2018; Vakkari et al., 2018). The mass median diameter and count median diameter of BC particles are between 120nm and 160nm and between 50nm and 80 nm, respectively, most of BC particles are submicron particles (Bond et al., 2013). Therefore, particle loss through the PM_{1.0} cyclone can be ignored for BC concentration measurement and our result can be compared with previous studies that used a PM_{2.5} inlet. Coarse particles are known to interfere with the BC measurement by filter-based optical absorption technique (Bond et al., 1999). To minimize the interferences by these particles and to achieve high accuracy of BC measurement, it is better to use PM_{1.0} cyclones. We added a sentence showing below.

“lines 95–96: Note, as most BC particles are smaller than 1 μm, BC loss through the PM_{1.0} cyclone can be ignored.”

line 113-115

How well are these parameters constrained by previous studies? It would be helpful to have some references so that readers can find the original papers for these numbers.

→Parameters for the BC removal process used in this study were chosen from (Grythe et al., 2017), which conducted a sensitivity test for the combination of BC removal parameters

for BC removal over Barrow, Alert, and Zeppelin (cf. Table 4 of their paper). We have conducted similar sensitivity tests with our observations and found that this combination showed the lowest RMSE with the observations. We added an explanation below.

“lines 124–125: which estimated by Grythe et al. (2017) as the best parameters over several Arctic regions, i.e., Barrow, Alert, and Zeppelin”

Line 130

What kind of back trajectory model was employed? Are back trajectory calculations for ground surface trustable for the duration of 20 days?

→FLEXPART-WRF version 3.3 (Brioude et al., 2013) and WRF version 4.4 (Skamarock et al., 2019) were employed in this research. The meteorological field near the surface was recalculated using WRF with increased vertical layers near the surface (around 14 layers within 0–2km from the surface) from ERA5 pressure-level data (around 9 layers in 1000–800hPa), and the vertical mixing within the PBL was diagnosed using the meteorological field of WRF. For the advection, hourly-averaged mass-weighted winds, which were calculated in WRF, were used to conserve the total mass. Although we conducted 20-day backward calculations, most simulated particles reached PFRR within approximately 10 days (Figure 4(c)). We therefore believe that our simulation is trustworthy. We added an explanation below.

“lines 127–128: , and most simulated particles reached PFRR within approximately 10 days (Figure 4(c))”

Line 166

Concentrations of most aerosol species are different from one place to another place. The description is not very informative. It would be better to describe 1) how the concentration level of the present data is compared with previous studies, and 2) what kind of geographical differences might have induced the difference.

→Thank you for your important suggestion. The median hourly BC concentration at PFRR was 13 ng m^{-3} and that at Utqiagvik was 12 ng m^{-3} , almost the same level. Relatively high BC concentrations were observed at Utqiagvik between January and March, however, BC concentrations at PFRR did not increase in the same seasons. This difference may be attributed to the BC accumulation in the polar dome (Quinn et al., 2007; Sharma et al.,

2013). On the other hand, large peaks of BC concentration (up to 5540 ng m⁻³) were sometimes observed at PFRR, however, these peaks were not observed at Utqiagvik. This difference is possibly caused by the topological separation by the Brooks mountain range. We modified our manuscript as below.

“lines 174–183: Observed median BC mass concentrations were the same level as previous reports at Utqiagvik (Barrow) (12 ng m⁻³), which showed BC mass concentration over the long term using the same instrument (BCM3130) employed in this study (Sinha et al., 2017; Mori et al., 2020). Abrupt peaks (up to 5540 ng m⁻³) were occasionally observed during summer at PFRR, but these peaks were not observed at Utqiagvik. On the other hand, increases in BC mass concentrations were reported in Utqiagvik between January and March, while not in PFRR. These different variations may be attributed to the topological separation by the Brooks mountain range and to the polar dome structure (Quinn et al., 2007; Sharma et al., 2013).”

Line 172

The comparison of the CO data should belong to the method section.

→We moved comparisons with aircraft observations provided by NOAA to the method section to support the accuracy of our CO observations. A part of the comparison with previous studies remained in the result section.

(moved)

“ line 108–111: To validate our CO observations, we compared our observed CO mixing ratio with aircraft observations (less than 500 m AGL above the PFRR) provided by the NOAA Global Monitoring Laboratory (<https://doi.org/10.15138/39HR-9N34>; accessed on 2 November 2023) (Figure S1), confirming a good agreement between these two observation results.”

Line 186

Figure S3 is not well organized. It may be better to separate the panel.

→Modified.

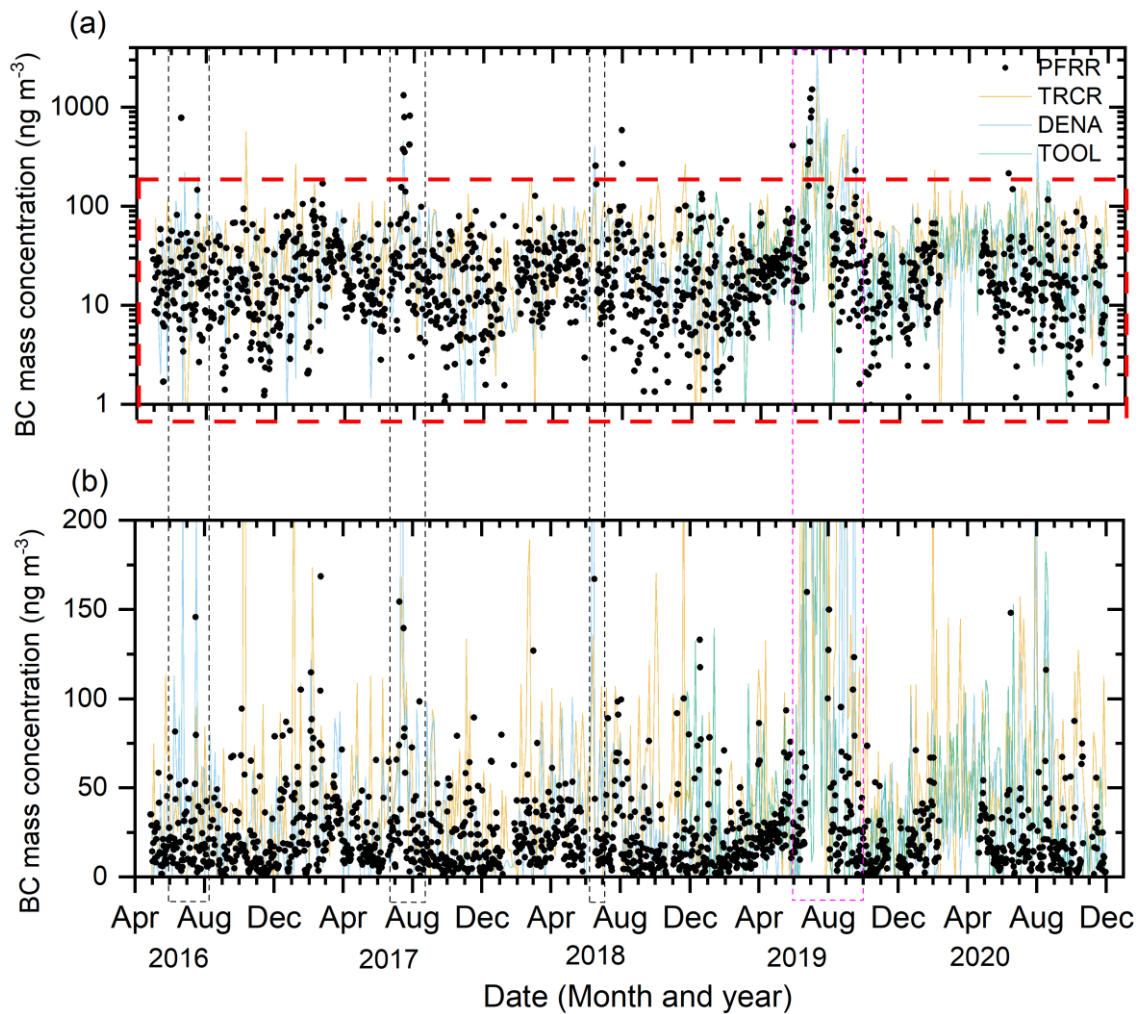


Figure S3. ...To show the variation of BC mass concentrations in the low concentration level, an expanded plot for low BC mass concentrations ranging between 0 and 200 ng m⁻³ was shown in (b) by a linear scale.”

Line 202

Why was the observation suitable for obtaining the data that were influenced by wildfires?

→Because PFRR is the only site located in the central interior of Alaska while other BC observation sites are located on the edge or outside of interior Alaska. Most forest wildfires

occur in the interior of Alaska (Figure 1). Therefore, PFRR is surrounded by forest wildfire occurring regions, differently from the other BC observation sites (Figure 1), resulting in higher BC concentration peaks than other sites. We modified our manuscript as below.

“lines 220–222: because PFRR is the only BC-measuring site located in the central interior of Alaska and is surrounded by forest wildfire occurring regions while other BC observation sites are located on the edge or outside of interior Alaska.”

Line 254

Are there any typical criteria to separate smoldering and flaming by FRP?

→No typical criteria. As we explained in the reply to the major comment, there are some studies that investigated the relationship between the BC/ Δ CO ratio and MCE, however, the relationship between the BC/ Δ CO ratio and FRP has never been introduced. Therefore, our result (Figure 6) for the evolution of the BC/ Δ CO ratio with increases in FRP will be the first report.

L290

Back trajectories only suggest the origin of air masses. It does not indicate anything about the occurrence of wildfires.

→We analyzed as if the observed air mass was affected or not by the forest wildfire by using back trajectory analysis coupled with the FRP satellite observation (section 2.4). So we added ‘with FRP (hereafter, we simply use ‘back trajectory’)’ to Line 315.

“lines 318–321: We selected 406 hourly cases between June and September from the data selected in Section 3.4 as high BC cases from forest wildfires and chose 184 cases of hourly BC observations results affected by near forest wildfires detected in Alaska and western Canada by back trajectory analysis with FRP (hereafter, we simply use ‘back trajectory’)”

“lines 407: Finally, we tracked air mass origin for 4 days using the HYSPLIT model with FRP satellite observations...”

L298

I am not sure if the correlation can be considered as being robust.

→We deleted ‘robust’ and ‘clear’ from this sentence.

“line 341: ... , we report a positive correlation...”

“line 344: This relationship should be taken...”

L303

The selection of the window is somewhat arbitrary. How does the original data (without classification by the window) look like? Why do the authors think that such a window is needed? These points will need to be clearly described.

→Generally, air mass location estimated by the trajectory analysis can not be pinpointed, having some spreads. The original VIIRS observation has a very fine resolution (375 m), and this resolution will be too fine to detect the actual areas of forest fire. Therefore, we defined an FRP detection window for this analysis.

We used GDAS1 meteorological data for back trajectory analysis which has a $1^\circ \times 1^\circ$ spatial resolution. Based on this constraint, we set our initial windows as $\pm 0.5^\circ$ for latitude and longitude. However, PFRR is in a high latitude and the geometrical length of latitude is approximately 2 times longer than that of longitude. For this reason, we defined latitudinal width as $\pm 0.25^\circ$ finally. This will take into account fires within an area of $\sim 25 \times 25 \text{ km}^2$. Although we tested some finer spatial and time resolution cases, a similar positive trend was considered, a similar positive trend was confirmed. We modified our manuscript as below.

“lines 349–352: Based on the spatial resolution of GDAS1 ($1^\circ \times 1^\circ$), we set our initial windows as $\pm 0.5^\circ$ for latitude and longitude. However, PFRR is in a high latitude and the geometrical length of latitude is approximately 2 times longer than that of longitude. For this reason, we defined latitudinal width as $\pm 0.25^\circ$ finally. Although we tested finer window size cases, a similar positive trend was confirmed. “

(references used in this reply)

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between fire behavior and aerosol and gas emissions, *Geophys. Res. Lett.*, 47,
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Referee#2

Black text: Referee comments by Anonymous Referee #2

Blue text: Our replies

Red text: Modified parts of the manuscript

* Line number is the number in the tracked manuscript.

Kinase et al., 2023 "Long-term observations of black carbon and carbon monoxide in the Poker Flat Research Range, central Alaska, with a focus on forest wildfire emissions" presents long-term observations of BC mass concentrations and CO mixing ratios in the Poker Flat Research Range, which is located in Central Alaska. The authors use the FLEXPART-WRF model to estimate contributions from black carbon source regions and sectors and the HYSPLIT model to trace airmasses that originate from forest wildfires. This study shows a good correlation between the BC/ Δ CO ratio and the fire radiative power (FRP) and concludes that the FRP should be used to improve emission inventories. This work provides very useful datasets and novel ideas. The methods and analysis support well the conclusion of this study. The text is well-written although in some parts the structure needs to be carefully revised. I have some minor comments for the authors to consider, presented below:

→Thank you for your kind review and important comments. All the comments were helpful for us in improving our manuscript. Please see our answers to the specific comments below.

Abstract: Although the measurements of this study were taken in Alaska, the high BC mass concentrations were originated from wildfires in both Alaska and Western Canada (section 3.5). Therefore, I would suggest discussing in the abstract about wildfires at high latitudes rather than just in interior Alaska. I would also suggest to mention already in the abstract the whole period of sampling at the PFRR.

→Thank you for your important comment. We extended the source regions from only Alaska to boreal forests in Alaska and Canada. We also added a whole period of observations as below.

“line 17: from April 2016 to December 2020.”

“line 22: a contribution of boreal forest wildfires in Alaska and western Canada”

“lines 395–396: The estimated BC source sectors and regions were biomass burning from Russia, Alaska, and sometimes Canada...”

Section 2.2: Some additional information about COSMOS is needed here. At least it should be added that this is a filter-based absorption technique.

→Modified.

“lines 91–93: The measurement technique of BCM3130 is based on filter-based optical absorption, thus other light-absorbing particles and scattering particles can be a source of interferences on BC measurement (Bond et al., 1999; Kondo et al., 2009).”

Lines 163-164: Unclear sentence; please, rephrase.

→Thank you for your important comment. We discussed for the differences in BC concentration at PFRR and Utqiagvik again and modified to be more specific. Please check.

“lines 174–183: The median hourly BC mass concentration and 10th and 90th percentile values throughout the observation period were 13, 3, and 56 ng m⁻³, respectively. No clear increase in annual median BC mass concentration was observed (Table 1). Observed median BC mass concentrations were the same level as previous reports at Utqiagvik (Barrow) (12 ng m⁻³), which showed BC mass concentration over the long term using the same instrument (BCM3130) employed in this study (Sinha et al., 2017; Mori et al., 2020). Abrupt peaks (up to 5540 ng m⁻³) were occasionally observed during summer at PFRR, but these peaks were not observed at Utqiagvik. On the other hand, increases in BC mass concentrations were reported in Utqiagvik between January and March, while not in PFRR. These different variations may be attributed to the topological separation by the Brooks mountain range and to the polar dome structure (Quinn et al., 2007; Sharma et al., 2013).”

Lines 201-202: Why are the black carbon emissions better captured in central Alaska? Is Alaska the only source region of the measured black carbon? A discussion on the topography of the sites and meteorological conditions is needed to support this statement.

→As shown in Figure 1, most boreal forest wildfires occurred in the interior of Alaska and western Canada, and PFRR is located in the centre of these forest wildfire-occurring areas, i.e., PFRR is surrounded by strong BC emission sources, while other observation sites compared in this study are located on the edge or outside of forest wildfire occurring regions.

As reviewer #2 mentioned and we showed in Section 3.3, other boreal forests, i.e., Russia and Canada, can also affect the BC concentration at PFRR. However, as shown in Figure 5, most large peaks (exceeding $1 \mu\text{g m}^{-3}$) were associated with Alaskan forest wildfires. Also, BC from the Russian forest wildfires can affect BC concentration at PFRR, however, BC concentration did not exceed $0.1 \mu\text{g m}^{-3}$ in these cases, likely due to the deposition and diffusion during their long-range transport. BC from Canadian forest wildfires also affected PFRR BC concentration, however, it was a minor case. We added the sentence below.

“lines 220–222: because PFRR is the only BC-measuring site located in the central interior of Alaska and is surrounded by forest wildfire occurring regions while other BC observation sites are located on the edge or outside of interior Alaska.”

“lines 285–287: BC from forest wildfires that occurred in western Canada also affected the BC concentration at PFRR (Figure S6), but to a lesser frequency. Russia was also estimated as an effective BC source region (Figure 3), but BC concentration did not exceed $0.1 \mu\text{g m}^{-3}$ in most cases (Figure 5).”

Lines 219-223: Figure 2 shows also that emissions in Canada contribute to BC concentrations at the PFRR during the warm season. How important is this contribution? I would suggest discussing this in this section since you will link later the high BC concentrations with the forest wildfires in Canada.

→Fractions of BC from Canada in warm seasons were 1.0–21% in the 10–90 percentile, relatively lower than that of Alaska and Russia. However, as pointed out by this comment, sometimes BC plumes from western Canada were captured at PFRR during BC high

concentration periods. Therefore, we added this information to the manuscript as shown below.

“line 241: and sometimes Canada (1.0–21% in the 10–90 percentile)”

“lines 285–286: BC from forest wildfires that occurred in western Canada also affected the BC concentration at PFRR (Figure S6) but to a lesser frequency.

”

Lines 251-256: I would suggest moving this text somewhere in the next section because it is not relevant to section 3.4.

→Modified. These sentences were moved to Lines 312–318.

“lines 312–318: In the previous section, we showed that most high BC mass concentration cases were related to forest wildfires in Alaska. Increases in biomass-burning-derived BC/ Δ CO ratios with combustion efficiency were suggested from an observational study on boreal forest wildfires (Kondo et al., 2011b) and from laboratory-scale burning experiments of crop residues (Pan et al., 2017); however, in-depth studies examining variabilities in BC/ Δ CO ratios based on long-term, near-forest observations have not been conducted. To consider the possibility that combustion conditions (flaming and smouldering) primarily control the BC/ Δ CO ratio, we are going to investigate the relationship between the BC/ Δ CO ratio and forest wildfire intensity in this section. We selected ...”

Line 259: Can you support the statement here with the back trajectory analysis?

→We added example cases of footprints when Alaskan and Canadian forest wildfires affected the BC concentration at PFRR in Figures S5 and S6 in our supporting information.

“

Examples of PES footprints and contributions of BC from biomass burning emissions at 12:00 on 27 June 2019.

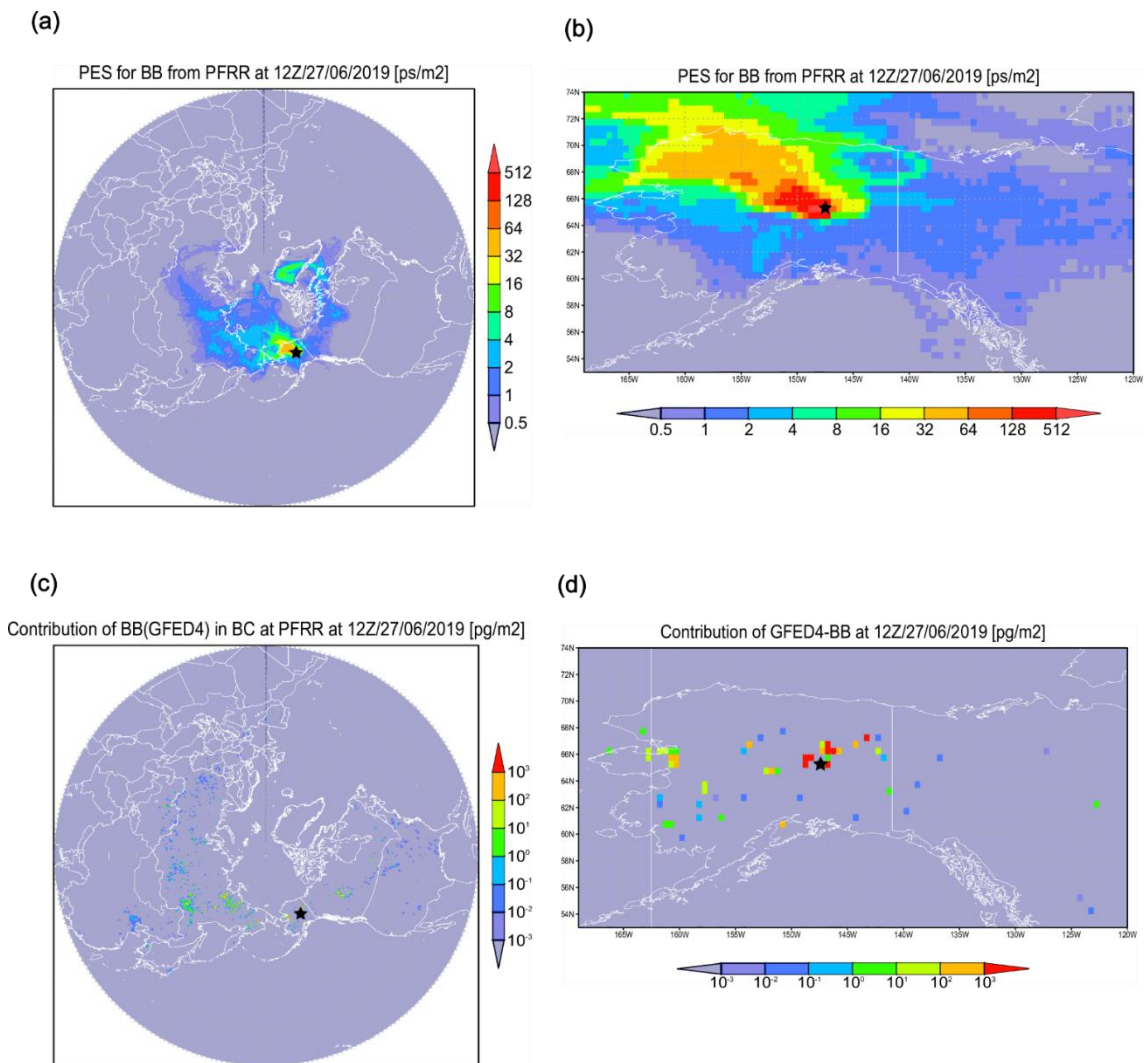


Figure S5. Examples of (a) a PES footprint on a global scale, (b) that focused on Alaska and Western Canada, (c) a BC emission contribution from biomass burning on a global scale, and (d) that focused on Alaska and Western Canada at 12:00 on 27 June 2019. In this case, high BC concentration (1611 ng m^{-3} on hourly average) was observed at PFRR and most BC came from forest wildfires around PFRR. Star marks indicate the PFRR location.

Examples of PES footprints and contributions of BC from biomass burning emissions at 00:00 on 12 July 2017.

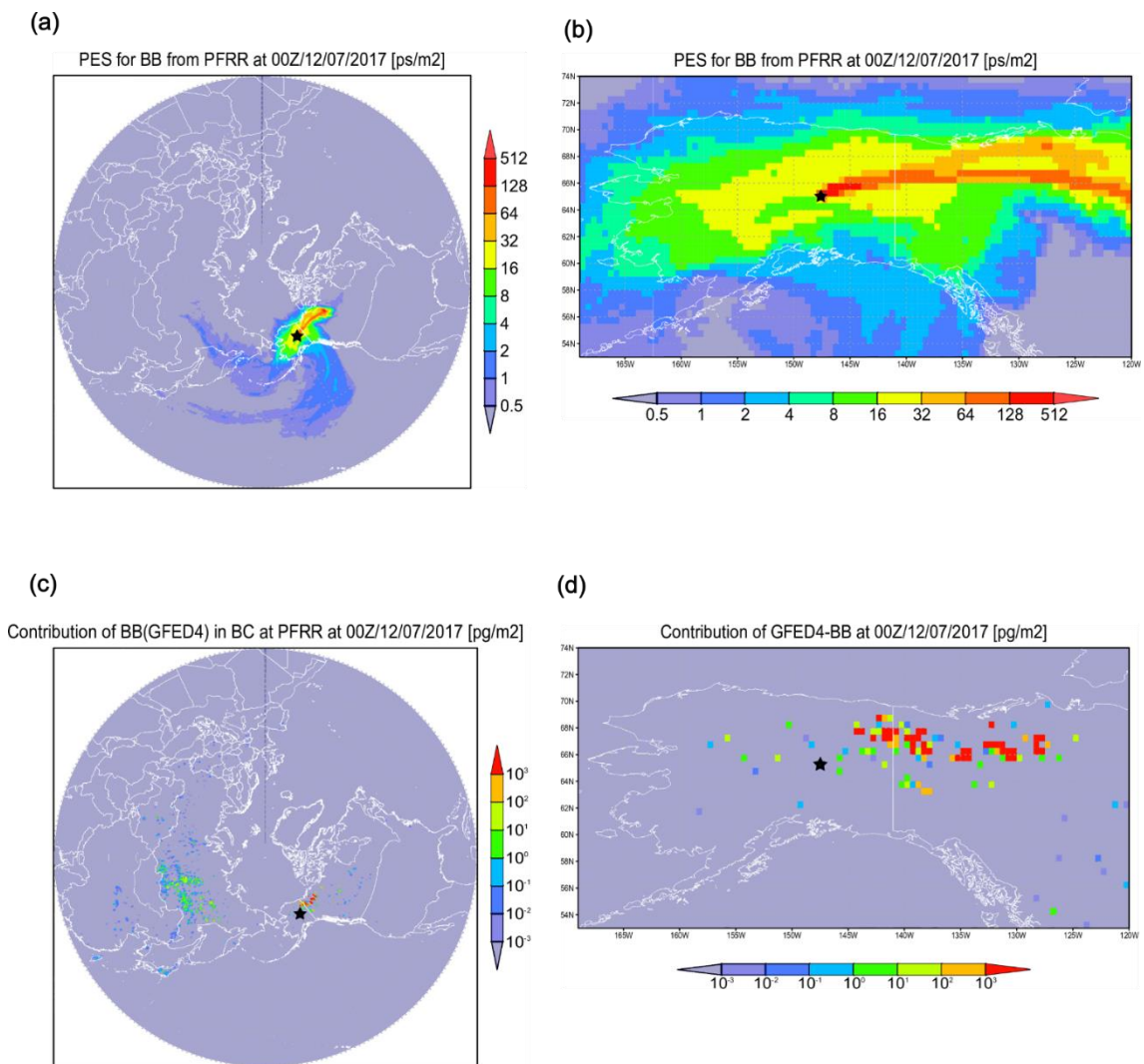


Figure S6. Same as Figure S5 but at 00:00 on 12 July 2017. In this case, high BC concentration (204 ng m^{-3} on hourly average) was observed at PFRR and most BC came from forest wildfires in western Canada and partly Alaska. Star marks indicate the PFRR location.

lines 278–280: The medians of the contributions of biomass burning and the mean age of BC estimated by the FLEXPART-WRF simulation in these high BC mass concentration cases were higher and shorter (95.5% and 2.6 days) than those in other periods (7.6% and 6.9 days) (Figure 4(b) and (c)), indicating a strong contribution of BC from neighbouring forest wildfires (Figures S5).”

Line 261: East Asia is a separate category in Fig. 5. The text should be corrected.

→Our explanation in the sentence was wrong, the text in Figure 5 is correct. We modified our explanation in the manuscript.

“line 281–282: 6 categories based on Figure 3(b), Central Asia and Europe are included in Others”

Figure 4b. Typo in the label (x-axis).

→Modified.

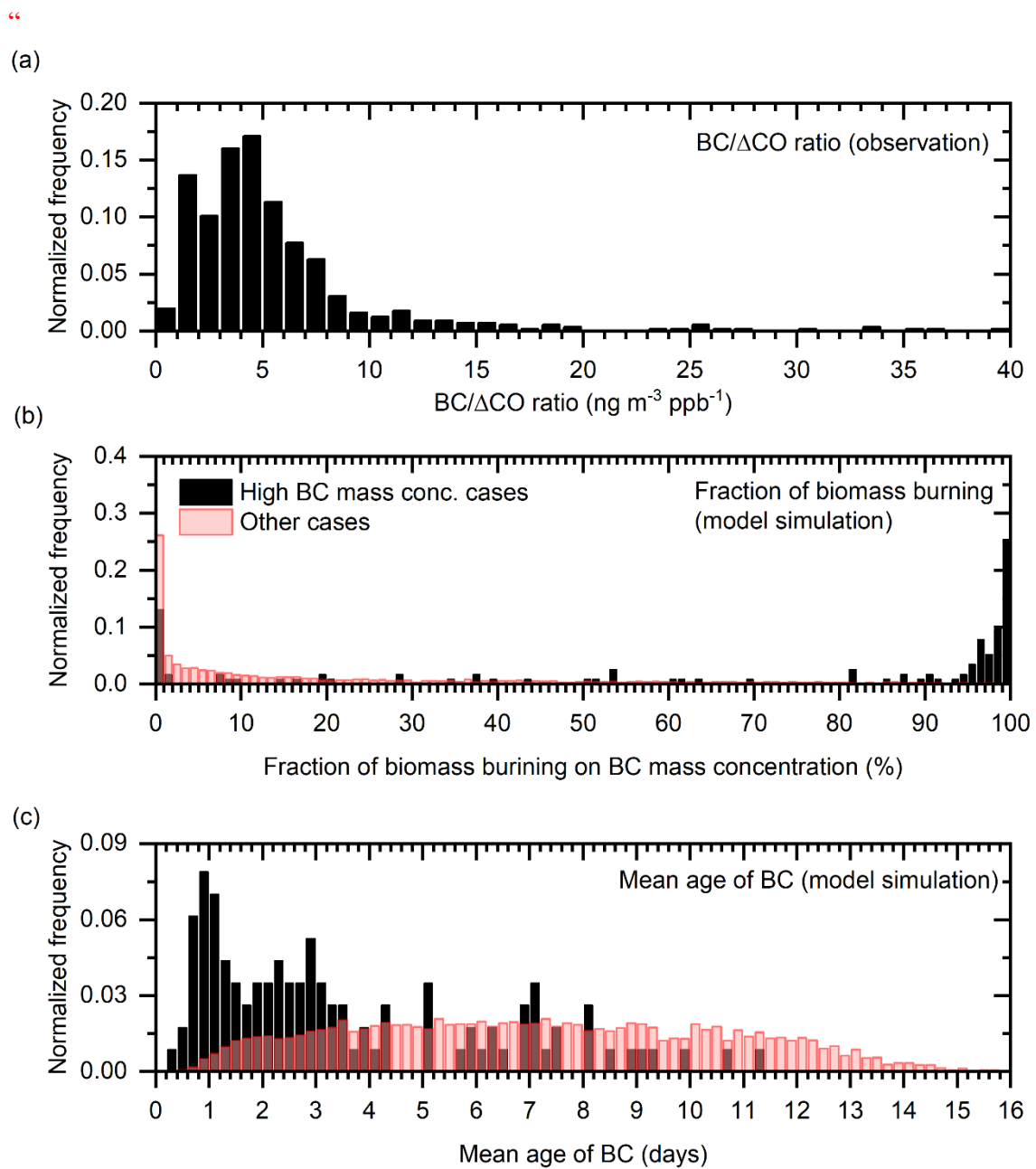
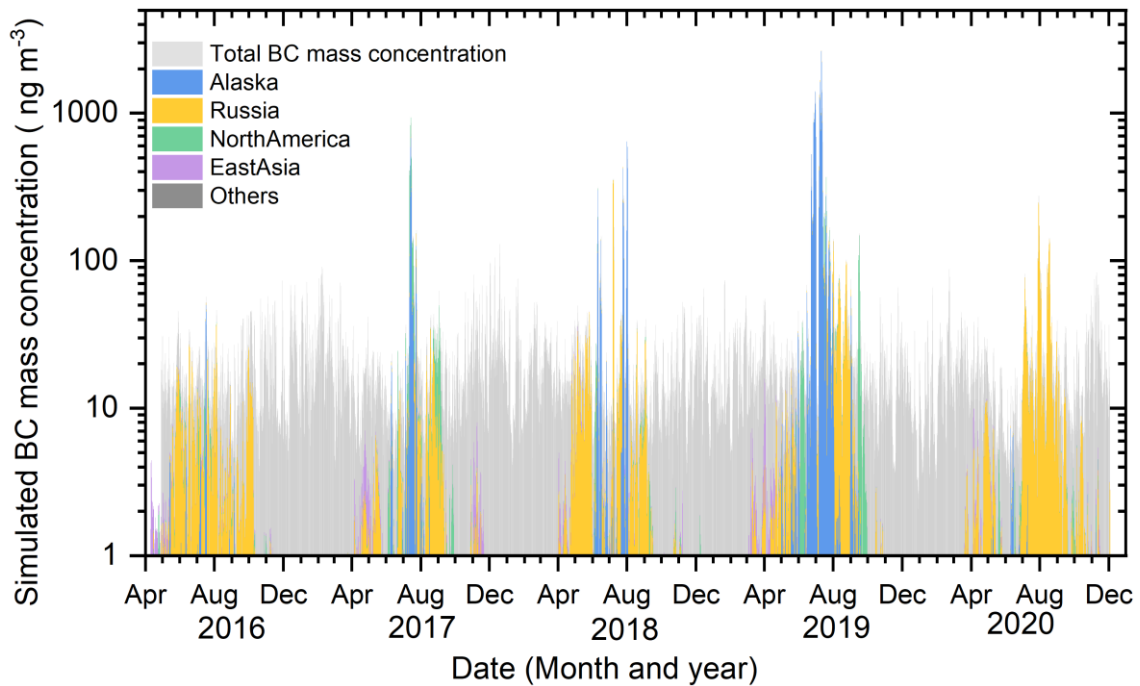


Figure 5: I would suggest the x-axis to be the same as in Figure 2 for easier comparison.

→Modified. In addition to this comment, the colour profile has been changed to the same as in Figure 2.

“



»

Lines 291-292: Unclear sentence; please, rephrase.

→There was too much information in that sentence and it probably made it confusing and unclear. Therefore, we deleted redundant parts as below.

“lines 322–323: We found a positive correlation ($r = 0.44$, $p < 0.0001$, $n = 184$) between the BC/ Δ CO ratio and Σ FRP/point values (Figure 6).”

Line 311: There is a typo.

→Modified.

“line 357: BC/ Δ CO”