Response to RC2

We thank Referee 2 for the very helpful and valuable comments. We will take all the comments into consideration and revise our manuscript. Our responses to the Referee's comments are shown below. The Referee's comments and our replies are numbered and shown in blue and black, respectively.

The manuscript entitled

High-resolution analyses of concentrations and sizes of black carbon particles deposited on northwest Greenland over the past 350 years - Part 2: Seasonal and temporal trends in black carbon originated from fossil fuel combustion and biomass burning

by Goto-Azuma et al.,

presents data of concentrations and size distributions of refractory black carbon (rBC) particles from an ice core in the Arctic based on a novel CFA system equipped with a more efficient nebulisation system used to fed a wide-range SP2 system, used for the rBC determination. Overall the system provide an unprecedented temporal resolution and extend the size range of rBC determination.

RC2-1 The manuscript (labelled Part2) discusses trends and sources of BC particles, presents a back trajectory analysis, the identification of potential sources (biomass burning and fossil fuel combustion) and an estimation of potential albedo reductions. The results presented in this manuscript rely strongly on the technical details presented in Part 1 paper. I recommend Part2 to be finalised after the acceptance of Part1.

AC2-1 We will revise the Part 1 manuscript first and would like to leave the decision to the Editor.

Major points

RC2-2 The assumptions used to obtain the monthly resolution looks artificial because has been obtained by assuming an equally spaced monthly time step. This imply the assumption of a constant monthly dry and wet deposition rate which is not the case in particular considering the 350 years time span investigated. Furthermore it is not clear how much the signal is blurred by the dispersion in the CFA system. Indeed, the data have a clear and well resolved seasonal signal, which is already

a very nice results in my opinion. The sections of the manuscript where the monthly analysis is presented should be revised.

AC2-2 We acknowledge that the term "month" in our manuscript does not correspond to a real calendar month, as wet and dry deposition do not occur evenly throughout a year. The use of the word "monthly" may have been confusing. As we explained in our reply (AC1-1) to Referee 1's comment RC1-1, we defined the mid-winter and mid-summer of each year, then divided the periods from mid-winter to mid-summer and mid-summer to mid-winter into six equal sections each. In the manuscript, we referred to each of these sections as a "month." However, there is inherent uncertainty in this definition. The discrepancy between our artificial months and actual calendar months could be one or two months. We will revise the manuscript to clearly state that the "months" defined in this study are not real calendar months and to acknowledge the associated uncertainty.

The signal dispersion is discussed in the companion manuscript (Part 1 of our study). Therefore, we did not explain it in this manuscript. However, to address Referee 2's concern, we will include a brief explanation in our revised manuscript, as we mentioned in our reply (AC1-3) to Referee 1's comment RC1-3.

RC2-3 Back Trajectories analysis - The 1958-2015 timeframe was used for BT calculations. This period is not necessarily representative of the entire 350 years covered by the ice core. Also, monthly precipitation data have been used to weight BT's which is highly uncertain. Finally, dry deposition processes have been neglected in this analysis. Overall this part of the work is weak.

AC2-3 We agree that the 1958-2015 timeframe used for back trajectory calculations is not necessarily representative of the entire 350-year period. However, we are constrained by the availability of reanalysis data, which begins in 1958. Nagatsuka et al. (CP, 2021), based on the back trajectory studies for the SIGMA-D site, have shown that the contributions from different regions remained relatively constant during 1958-2013. Thus, we can discuss the source regions of BC for this period based on our back trajectory calculations. For earlier periods, we can only hypothesize the back trajectories by assuming similar atmospheric circulations to those of the 1958-2015 period. In our revised manuscript, we will clarify this limitation and acknowledge the significant uncertainties for the period prior to the 1950s.

We did not use monthly precipitation data; instead, we used daily precipitation data, which has much lower uncertainties. For back trajectory calculations, we assumed wet deposition. As stated in our reply (AC1-2) to Referee 1's comment (RC1-2), it is reasonable to assume that the contribution of

dry deposition is minor. However, we have performed new calculations of back trajectories without weighing for daily precipitation. In our revised manuscript, we will present these new results and include a brief discussion on wet and dry deposition.

Minor points:

RC2-4: 1-rBC (refractory BC) should be used in place of BC everywhere in the paper.

AC2-4 We will replace "BC" with "rBC" in our revised manuscript.

RC2-5: 2-line 18 - "roles" is vague. "processes" ?

AC2-5 We will change the word in our revised manuscript.

RC2-6: 3- line 40 - "Changes in". --> "Increase of"

AC2-6 We will change the word in our revised manuscript.

RC2-7: 4- line 177 - 179 - Na+ was used for annual layer counting a part cases when this was not possible because of the low signal. Therefore the authors recurred to the standard 18O isotope signal. Using Na+ is interesting but why 18O was not used if the signal is more clear? A brief explanation should be given.

AC2-7 Due to the diffusion of water isotopes, seasonal peaks of $\delta^{18}O$ are often less pronounced compared to those of Na, as illustrated by the $\delta^{18}O$ peak around 16.5 m in Fig.2. Consequently, we primarily relied on Na for annual layer counting. Nonetheless, we also examined the $\delta^{18}O$ data and confirmed that both $\delta^{18}O$ and Na yield the same dating results. We will add a brief explanation on annual layer counting in our revised manuscript.

RC2-8: 5- Figure 6 - the meaning of the x axis is not clear. Also, what happen to the IC contribution at the D4 site for low x values?

AC2-8 We apologize for the missing X-axis title. The X-axis denotes days. We will add the X axis title to Figure 6. At the D4 site, the contribution of IC is large in the initial few days, likely due to its proximity to Greenland. IC is the only land region near Greenland that can serve as a BC source region. Despite its small area, the contributions from IC in the initial few days are much larger than those from other land regions. However, if we include oceanic regions along with land regions, the contribution from IC decreases substantially.

RC2-9: 6- Figure 8 the normalisation procedure should be briefly described

AC2-9 In our revised manuscript, we will provide a brief explanation of how we normalized the size distributions.

RC2-10: 7- line 284 - why a Gaussian distribution is assumed? usually a log normal distribution is more appropriate.

AC2-10 We made an error in our manuscript. In our study, we assumed a log-normal distribution. We will correct this mistake in our revised manuscript.

RC2-11: 8-lines 285 -301 how the use of a single MMD or single mBC parameter to describe the entire BC size distribution can be consistent with two very different BC sources ?

AC2-11 This is an important and intriguing comment. The all-season size distribution patterns of BC particles across different decades were similar (Fig. 8), showing distributions close to the log-normal distribution, with additional large particles beyond the log-normal curves. However, both MMD and mBC increased during periods when the contribution of fossil fuel combustion BC particles was high. To better differentiate between fossil fuel combustion and biomass burning BC, we will present decadal means of summer and winter mean mass size distributions. We tentatively label this new figure as Fig. 8-2. Both summer and winter patterns are close to log-normal distributions, though MMDs were larger in winter during periods with significant contributions from fossil fuel

combustion BC, suggesting that fossil fuel combustion BC particles were larger than those from biomass burning. As Referee 2 commented, using only single MMDs or single mBCs is insufficient to describe the entire BC size distributions. We will include the new figure (Fig. 8-2) and explain this in the revised version of our manuscript.

RC2-12: 9- Figure 10 - What I see in these plots are yearly BC mass concentration trends. Not monthly BC concentration trends.

AC2-12 In Fig. 10, we plotted monthly mean BC mass concentrations (though they might not represent true monthly mean concentrations due to uncertainties in monthly dating), not annual mean concentrations. The figure caption is correct.

RC2-13: 10-line 351-353. the BT analysis was inconclusive and should not mentioned further

AC2-13 We agree that our back trajectory analysis was inconclusive in explaining anthropogenic BC mass concentration trends. Furthermore, there are uncertainties in back trajectories for the period prior to 1958. Despite these uncertainties, we believe it is still worthwhile to speculate on the sources of biomass burning BC, assuming that the back trajectories for the period prior to 1958 were not significantly different from those in the 1958-2015 period. In the revised version of our manuscript, we will clearly state that we can only speculate on the sources and that we cannot draw firm conclusions.

RC2-14: 11- line 366 - how long the high peaks have really lasted? It is possible to catch strong events shorter than 1 season if "the data for a few months after large BC concentration peaks could have been affected" (as affirmed by the authors at line 206-207).

AC2-14 The wording "the data for a few months after large BC concentration peaks could have been affected" might have been misleading. Most of the high peaks lasted for only a month or two (with “month” not being a real calendar month, as “month” is not a real month). We will revise this sentence to avoid any confusion.