Response to RC1

We thank Referee 1 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.

RC1-1 This manuscript "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" submitted by Goto-Azuma et al., provided monthly resolved 350-year records of concentrations and size distributions of black carbon (BC) particles from an ice core that was drilled in the northwest Greenland Ice Sheet. The authors discussed sources of BC particles originated from biomass burning and fossil fuel combustion based on backward trajectory analyses, and estimated the potential albedo reductions. The main advantage of this work concerns with the extremely high resolution records from the updated CFA system that was consisted of single-particle soot photometer and a high-efficiency nebulizer. <u>As a result, the annual layer of the SIGMA-D ice core can be reasonably divided into 12 months, which provides a chance to decipher the monthly variations of the ice core BC particles that have been impossible before. However, these monthly-averages depend on assumptions, such as evenly month distribution of precipitation, that can be hardly met. Therefore, uncertainties due to the assumptions should be examined carefully before a solid conclusion can be reached.</u>

AC1-1 We have identified the mid-winter and mid-summer points of each year and divided the period from mid-summer to mid-winter to mid-summer into six equal sections. Similarly, the period from mid-summer to mid-winter was also divided into six equal sections. In the manuscript, we referred to each of these sections as a "month." However, we acknowledge that these sections do not correspond to real calendar months, as precipitation does not occur evenly throughout the year. The "months" defined in this manuscript are artificial. Nonetheless, we think that dividing each year into 12 sections provides valuable insights into the seasonality of BC and its temporal changes. 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 uncertainties.

Other minor comments:

RC1-2 *Lines* 196-197: *Please give pieces of evidence that contribution of dry deposition can be ignored.*

AC1-2 When we computed back trajectories, we assumed that the contribution of wet deposition was greater than that of dry deposition. It is difficult to estimate the wet and dry deposition ratio directly since there are no observations at the SIGMA-D site, as at most of the sites in the Arctic. Instead, we computed the terminal velocity (V) of BC particles falling onto the SIGMA-D site using the equation $V=2\rho r^2 g/9\zeta$ assuming spherical BC particles. Here ρ , r, g, and ζ denote the density of BC particles, the radius of BC particles, the acceleration of gravity, and the viscosity coefficient of the atmosphere, respectively. We used the values 1800 kg m⁻³ and 1 μ m for ρ and 2r (diameter), respectively. Assuming an atmospheric temperature of -40°C, ζ was calculated to be 1.5×10^{-5} N s m^{-2} . With these values, the terminal velocity was estimated to be 6 m day⁻¹. Given that the BC particles fall from 500 m above the ice sheet surface at the SIGMA-D site, it would take approximately 100 days for the BC particles to reach the ice sheet surface, indicating a very small dry deposition velocity at the SIGMA-D site. A study using the GEOS-Chem global chemical transport model (Breider et al., JGR, 2014) also indicated that the annual mean fraction of dry deposition in the Arctic was only 11 %. Furthermore, Sinha et al. (JGR, 2018) showed that the dry deposition was a small contributor (less than the uncertainties of the measurements, which were about 20%) to the total BC deposition at Ny-Ålesund, Svalbard, where the total water equivalent snowfall amount during September-April was similar to the annual accumulation amount at the SIGMA-D site. Thus, it is reasonable to assume that the contribution of dry deposition is small. We will add these statements when we revise our manuscript.

We also conducted additional back trajectory analyses without weighting the air mass probability by local daily precipitation. The probability distribution of air masses (Fig. A2) indicates that, throughout the year, back trajectories for the SIGMA-D and D4 sites originate from wider regions without weighting, with the difference being more pronounced in winter than in summer. Fig. 6_new compares the results with and without weighting. Whether weighted by daily precipitation or not, the contribution from North America is the largest at both SIGMA-D and D4 sites throughout the year. Without weighting, the contribution from Russia increases at both sites throughout the year, with a more significant increase at the SIGMA-D site, especially in winter. Without weighting, the contribution from Europe decreases in winter and increases in summer at both sites (at SIGMA-D after six days in winter). The slight difference in temporal patterns of mass concentrations of anthropogenic BC between the two sites might reflect the different contributions from Russia in winter. We will replace Fig. 6 with Fig. 6_new and add Fig. A2 in the revised manuscript. Our interpretation of the ice core data remains unchanged if there is a minor contribution from dry deposition.



Fig. A2: Probability distributions of air masses at (left) the SIGMA-D site and (right) the D4 site calculated without weighing with the local daily precipitation: (a) averages of all seasons, (b) averages of winter months (December-February), and (c) averages of summer months (May-July)



Fig. 6_new: Temporal variability in contribution of air masses arriving at (left) the SIGMA-D site and (right) the D4 site from four regions: (a) averages of 12 months, (b) averages of winter months (December-February), and (c) averages of summer months (May-July). Right-hand axes indicate contributions from NA, and left-hand axes indicate contributions from the other regions. Solid and dotted curves denote results with and without weighing with the daily local precipitation, respectively.

RC1-3 Section 3.2. Is there possibility that corresponding of the seasonal Na and BC peaks (Fig. 10) can be disturbed by signal dispersion in the CFA system?

AC1-3 Figure 10 shows Na and BC concentrations at ~ 1/12 year resolution, which corresponds to ~ 22 mm resolution. As demonstrated in the companion paper (Part 1 of our study), the dispersion lengths are ~35 and ~39 mm for Na and BC, respectively. Consequently, the signal dispersion might slightly reduce the heights of the seasonal peaks. Due to the slightly asymmetrical shape of the Na and BC peaks (as discussed in the companion paper), their positions might change slightly in the CFA system. However, these change in peak positions would be minimal and would not alter the relative positions of the seasonal Na and BC peaks.

RC1-4 Figure 3: The extremely high peak around 1710 needs to be explained.

AC1-4 The extremely high BC peak in 1710 occurred in summer and was accompanied by a very high NH₄⁺ peak. Similarly, other very high BC peaks in 1711 and 1712 occurred in summer and were accompanied by high NH₄⁺ peaks. These observations suggest that these BC peaks likely originated from large forest fires. As suggested by Referee 1 commented, we will add a few sentences highlighting these peaks in the revised version of our manuscript.

RC1-5 *Line 257: "GrIS"* \rightarrow *"GrIS"*

AC1-5 We will correct this typo.

RC1-6 *Figure 6: Please indicate the meaning of the X axis.*

AC1-6 We apologize for the missing X-axis title. The X-axis represents days. We will add the X-axis title in the revised version.

RC1-7 *Line 291: "Of the two size parameters, mBC is easier to calculate than MMD; hence, it can be used to investigate changes with high temporal resolution". This is not a good reason to choose mBC.*

AC1-7 We fully agree. This does not sound scientific. We will revise this sentence.

RC1-8 Lines 300-301: An objective statistical method should be applied to reach the results.

AC1-8 To address Referee comment RC1-8, we performed breakpoint analyses (Muggeo, Statis. Med., 2003). The results suggest that the increases of BC mass concentrations, mBC and MMD began around the 1850s, 1870s and 1810s, respectively. Although these dates are slightly different from those stated in our manuscript, the increases in mBC and MMD still occurred earlier than the increase in concentrations, as we originally indicated. In the revised manuscript, we will incorporate the results of the breakpoint analyses.

RC1-9 Figure 10: The time range of Figure 10C is not 1915-2003.

AC1-9 We apologize for this oversight. We will correct it promptly.