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

General comments

RC1-1 The pre-print manuscript "Technical note: High-resolution analyses of concentrations and sizes of black carbon particles deposited on northwest Greenland over the past 350 years – Part 1. Continuous flow analysis of the SIGMA-D ice core using a Wide-Range Single-Particle Soot Photometer and a high-efficiency nebulizer " by Kumiko Goto-Azuma et al. describes and examines an instrumental coupling of a CFA feeding a Marin-5 nebulizer and a wide range SP2 to analyze the black carbon profile (size spectrum, number and mass) along a Greenland ice core. This article is the first part of the study, which will be supplemented by a companion paper devoted to the results of the analyses.

The article describes in great detail the analytical system used and provides a serious analysis of its performance. In several paragraphs, the authors compare their system with other existing CFA/Nebulizer/SP2 coupling, attempting to demonstrate that the latter underestimate BC masses for reasons of reduced or unstable nebulizer efficiency, or the size spectrum truncated by the classic SP2. To my knowledge, this comparison reflects parameters that are not considered by the authors, which brings into question some of their conclusions. Further investigation would be required to finalize this work.

AC1-1 Many thanks for the general comments. We will consider all the parameters pointed out by Referee 1 in the specific comments.

Specific comments

RC1-2 Lines 76-78 : While the original SP2 allows incandescence measurements of BC particles with diameters of between 70 and 850-900nm (40-450nm according to DMT), the new SP2-XR model on the market is, according to DMT, suitable for incandescence measurements of BC particles with larger diameters ranging from 50 to 800nm. To date, I know of no research team working on snow and ice samples with this new SP2-XR, but it is used for atmospheric measurements.

You will also note the difference in size range between the data proposed by DMT and that of (Mori et al, 2016) for the original SP2 indicating a factor of 2 in measurable sizes. A specific study as done by (Mori et al, 2016) with an SP2-XR would suggest that the SP2-XR would be able to cover a wider range up to >1.5µm without modification. Unfortunately, this study is not available.

AC1-2 Thank you very much for the important comments on off-the-shelf SP2s provided by DMT. There was confusion about the measurable size ranges for different versions of off-the-shelf SP2s and the SP2 with the modifications made by Moteki et al. (2010). The standard SP2 referred to by Mori et al. (2016) meant a version with the modifications made by Moteki et al. (2010), which is not the off-the-shelf SP2 or SP2-XR. We mistakenly compared Moteki et al.'s (2010) version of SP2 with the Wide-Range SP2 developed by Mori et al. (2016). We will correct the mistake and compare the Wide-Range SP2/Marin 5 setup with the off-the-shelf classic SP2 (measurable size range presented in the brochure was 70-500 nm) / U5000AT setup and the off-the-shelf SP2/APEX-Q setup.

We are not convinced by the operator manual of the SP2-XR that it can cover a wider size range up to >1.5 µm. If the study is not available (has not been published), we are unable to refer to this study in the manuscript.

RC1-3 Line 117-118 : Can you explain how you obtained this depth resolution value of 0.3±0.1mm ? Is it only the resolution related to the laser positioning sensor or the resolution once the water is analyzed by the different online instruments ? Is this value defined solely by the fluidics of the CFA system (line, valves, debubbler, …) ?

AC1-3 The depth resolution mentioned is that of the laser positioning sensor, not the resolution of the water analysis performed by different online instruments. We apologize for any confusion caused by the text. We will revise it to ensure clarity. The depth resolution of the laser positioning sensor has been defined and published by Dallmayr et al. (2016). Therefore, we will not explain its definition again.

RC1-4 Line 135 : This is not the focus of this article but can you clarify why your Picarro calibration is only done after the CFA session and not also before the measurements? Have you observed sufficient stability of the instrument if it is running all the time?

AC1-4 After receiving this comment, we checked the log data of the CFA sessions again. Although we currently calibrate our Picarro before and after a CFA session, we calibrated it only after a CFA session for the SIGMA-D, most likely to save time. Since we ran Milli-Q water before and after every CFA session, we could confirm that our Picarro was stable enough during the 4-5 hours of each CFA session. Figure 5 indicates that the stability of the Picarro was sufficient for our purposes.

In addition to confirming the Picarro's stability, we found an error in the description of our Picarro. Although we wrote that we had used the Picarro L2130-i, we had also used the L2120-i when the L2130-i was out of order. We will correct the description of the Picarro.

RC1-5 Line 151 : This comment concerns the supply of melt water to the nebulizer and then to the SP2. For accurate measurement of BC, the water flow must be controlled as it is proportional to the BC measurement. Peristaltic pumps are not the most stable over time and the flow rate varies according to wear on the tygon tube. What's more, this type of pump does not produce a stable flow, but a pulsed flow, as can be seen on an APEX/SP2 setup. Do you have any clarification on these points, and don't you think it's necessary to add a precise flow measurement before introduction into the nebulizer, using a Sensirion micro flow sensor for example ?

AC1-5 Before and after each CFA session, we measured the flow rate. After each CFA session, the flow rate usually decreased slightly (~5%), likely due to wear of the tube. Just before the next CFA session, the flow rate of the peristaltic pump was adjusted. In this way, we maintained an almost constant flow rate with a variability of less than 5%. Strictly speaking, peristaltic pumps produce pulsed flow, as Referee 1 commented. However, the peristaltic pump was run at ~7.50 rpm, which was high enough that the pulses were not observed in our BC data. We will add this information to the manuscript.

RC1-6 Line 166 + : While the internal mass and size calibration of the SP2 is relatively stable with time if the instrument is not moved, experience has shown that the nebulizer's efficiency is less so. Your method consists of measuring once this efficiency using PSL particles and Aquablack according to the size range. However, whether feeding an ICPMS or, in this case, an SP2, regular calibration of the nebulizer is necessary and this is generally done by analyzing a range of calibration solutions of known concentration on a daily frequency. How can you demonstrate that the Marin-5 model is more stable than the other nebulizers used for this type of experiment ?

AC1-6 We have not tested the stability of APEX-Q. Hence, it is difficult to compare the stability of APEX-Q with that of Marin-5. The repeated measurements of the nebulizer efficiency of Marin-5 (please see Fig. S1) show almost the same nebulizer efficiencies over time, though there is some variability in the nebulizer efficiency data shown in Fig. 2(a). For BC diameters < 2 µm, the error was ±8 %, which does not significantly affect the BC data. Therefore, we used the same nebulizer efficiency values. We also frequently checked the SP2/nebulizer system using PSL and confirmed the stability of the system. We will replace Fig. 2(a) with Fig. S1, or add Fig. S1 as supplementary material, and include a few lines in the text to explain the stability of Marin-5 over time. We have also compared the efficiencies of APEX-Q and Marin-5 nebulizers. We will add this comparison to the manuscript.

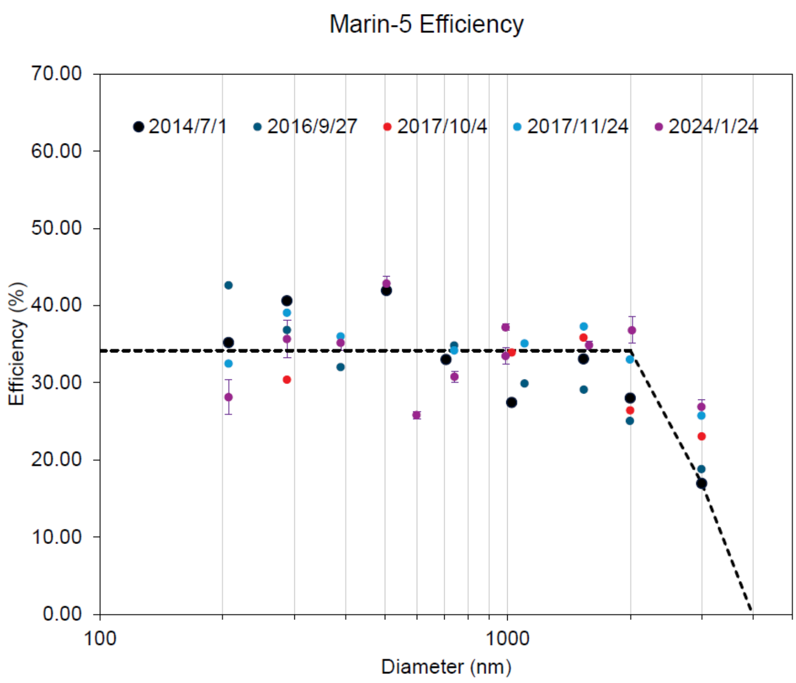

Fig. S1

RC1-7 In addition to these technical comments, can you provide more information about the post-processing of SP2 data in order to obtain BC mass and size profiles? The SP2 data files are relatively heavy, so some users try to extract them directly from the DMT software, but others turn to the PSI ToolKit.

AC1-7 We used the "Standard SP2 Software" and the "Probe Analysis Package for Igor (PAPI)", both provided by DMT, to acquire and process the incandescent signal in binary data and convert it to text format. Then we used our original code to calculate the mass and size of BC particles. We will add this information to the text. We did not use the PSI ToolKit.

RC1-7 Line 243 : Figure 2 does not convince me about the stability of the nebulizer, whatever the flow rate or particle size range. For 0.384mL.min-1, for example, for the <2µm section, the efficiency varies from around 27% to 42% (Fig. 2a).

AC1-8 We determined the nebulizer efficiency to be 34.2% ± 8.0% for BC particles < 2 µm. As Referee 1 commented, the nebulizer efficiency does vary between 27% and 42%. Therefore, when we estimated the total error in BC data, we took this variation into account following Mori et al. (2016). However, as described in AC1-6 and Fig. S1, the nebulizer efficiency did not change over long periods.

RC1-9 Line 250 + : Signal dispersion. Dispersion tests are carried out using two solutions with different characteristics (in BC, ionic charge and isotopic composition) injected alternately through a valve under the melting head and then circulated to the analytical instruments. This is a good method, but a step is missing to estimate the impact of the melting head on this dispersion. Several parameters are not taken into account. 1) Even if the stratigraphy in the ice samples were perfectly horizontal, mixing would occur between the samples in the center of the ice stick and those on the outside of the inner ring (13mm?) mixed up to the port of the CFA line, 2) the ice strata are not always horizontal in the stick.

AC1-9 We injected the solutions near the center hole of the melt heads (i.e., from above the melt head), not through a valve under the melt head. Since this was not clear in the manuscript, we will revise Line 204. Mixing occurs between the samples in the center of the ice stick and those on the outside of the inner wall (26 x 26 mm square-shaped melt head as described by Bigler et al. (2011)). However, due to the very short distance and very small dead volume within the melt heads, the mixing that occurs within the melt heads is negligibly small compared to the mixing that occurs in other parts of the CFA system, such as the debubbler, valves, conductivity cells, tubing, and nebulizer.

If the stratigraphy in the ice samples is not horizontal, it does not affect the resolution of the CFA system, although it affects the temporal resolution of the ice core data. To evaluate the signal dispersion in the CFA system, we do not think that the stratigraphy in the ice samples matters. Nevertheless, the stratigraphy of the SIGMA-D core was nearly horizontal, allowing minimal mixing of ice from different ages.

RC1-10 If we take into account only the interesting results of your method, this provides the basic parameters on the dispersion of the CFA and the analytical instruments. I'm quite surprised to see that the dispersion lengths (L1 and L2 average) are fairly similar between the instruments. It is known that the large dispersion in the Picarro is linked to a long cavity flush time, but this should be much shorter for the SP2 and ICPMS (to my knowledge closer to 10mm on other configurations). In addition, some studies have used these dispersion parameters to simulate a non-dispersed signal.

AC1-10 The length of the tubing between the melt head and the Picarro was much shorter than the lengths of the tubing for SP2 and ICP-MS. This would explain why the dispersion lengths for the Picarro are similar to those for the SP2 and ICP-MS.

RC1-11 L261 : Yes of course the resolution of your CFA is better than these dispersion values, you may indeed observe a signal at a higher frequency, but the values observed will be reduced by this dispersion.

AC1-11 We completely agree with the comment. Therefore, as we wrote in the manuscript, the monthly mean values might have been affected by the preceding months.

RC1-12 L268 + : Minimal loss of BC. That's a great information for all BC measured by CFA that should be reproduced elsewhere!

AC1-12 Thank you for the comment. We were relieved when we saw this result.

RC1-13 L295 + : BC profile. A rolling average over 10mm is indeed necessary to smooth out the technical characteristics of the CFA, and for an initial assessment of the data. Unfortunately, Figure 6 does not allow this work to be properly appreciated, as it is too crowded. Consideration could be given to adding an enlarged extract of the profile over a short period of a few years in order to appreciate any seasonal variation in the BC, which would be an added value to the use of the CFA and its high resolution. You can save the full profile for future publications.

AC1-13 Apologies for Fig. 6 being too busy. We will add an enlarged extract of the profile or replace it with the current Fig. 6 as suggested by Referee 1.

RC1-14 Lines 315 + : This brings us to the crux of the article, which proposes to demonstrate that configurations other than Nebulizer Marin-5 and WR-SP2 underestimate BC mass concentrations by XX%. It's not just the instrument and the measurement that come into play, but also the data processing. The low size limit of traditional SP2s is well known, which is also why DMT now offers an SP2-XR. Just because there are no measurements taken on sizes above 650 or 850nm does not mean that this part of the size spectrum is not considered. As shown in Figure 7, a Normalized dM/dlogD fit can be used to calculate the total mass (lognormal fit size distribution). This fit does not necessarily require measurements above 650 or 850nm to be correct if most of the peak is covered. To the best of my knowledge, but you can get in touch with the main users, to overcome the problem of the size spectrum being truncated at the top, classic SP2 users use the PSI ToolKit, which proposes the use of this fit in order to extract correct mass values. This last point should change the hasty conclusions of this manuscript.

For users of the U5000T nebulizer, on the other hand, there is a real problem of underestimation coming from instable nebulizer's efficenty.

AC1-14 We appreciate these very important comments. Although the U5000AT ultrasonic nebulizer has recently been replaced by the APEX-Q nebulizer, many previous ice-core BC studies, including most of those in Greenland, used the U5000AT nebulizer. To investigate spatial variability within Greenland and the Arctic, we need to compare our new data with the valuable BC data previously obtained with the U5000AT nebulizer. Thus, we think it is important to evaluate the degree of underestimation of the BC data obtained with the classic SP2/U5000AT nebulizer setup. When we compare the two setups, we will cut off the BC particles > 500 nm (not 650 or 850 nm as written in the current version of our manuscript) because the measurable size range presented in the brochure of a classic SP2 was 70-500 nm.

We also think it is important to compare the Wide-Range SP2/Marin-5 setup with the classic SP2/APEX-Q setup, which is used in more recent ice-core BC studies. We agree that the total mass concentration of BC can be more accurately estimated assuming a lognormal size distribution if most of the peak is covered. We will investigate recent studies using the classic SP2/APEX-Q setup and revise this manuscript accordingly. However, we would like to point out that the size distribution of BC in snow or ice cores does not always follow a lognormal distribution. Bimodal size distributions with second peaks > 500 nm have been reported by Mori et al. (2019) and Kinase et al. (2020). Unless the size distribution is actually measured with a Wide-Range SP2, we cannot ensure that the size distribution follows a lognormal distribution and that the total mass can be accurately calculated assuming the lognormal distribution. We will revise the manuscript to include these issues.

Technical corrections

As a non-native English speaker, I will not be making any technical corrections to this manuscript.