Response to RC2

We thank Referee 2 for the very valuable and helpful comments. We would like to revise the manuscript, taking all the comments into consideration. 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.

RC2

RC2-1 This manuscript by Goto-Azuma et al. describes a continuous flow ice core analysis system with parameters used for the analysis of the SIGMA-D core from NW Greenland. This manuscript aims to describe the NIPR CFA system (including SP2, ICPMS, Picarro, etc.), conduct a detailed assessment of continuous ice-core BC analysis with the Marin 5 and wide range SP2 system, and introduce the analysis and dating of the SIGMA-D ice core. My overall impression of this manuscript is that while the methods presented here underpin some very interesting BC data from the SIGMA-D core (which are presented in a companion paper), it does not have sufficient novelty or focus to stand as a separate manuscript. I would suggest the authors revisit the purpose of this manuscript and reframe it with a more central goal as I think most of what is included would be more appropriate for the methods section of the science-focused manuscript. Hopefully my suggestions below are useful. I do think the resulting datasets (discussed in the companion paper) are very interesting and appear to be quite robust, but reiterate that I do not think this methods manuscript holds up very well on its own in its current form.

AC2-1 Although the BC measurement technique using the Wide-Range SP2 and the Marin-5 nebulizer has already been reported by Mori et al. (2016), this manuscript presents the first attempt to apply this method to a CFA system, allowing continuous and high-resolution measurements of the size distribution as well as concentrations of BC particles in ice cores. We believe it is important to describe such a combined system and assess its performance. Furthermore, to utilize and maximize the valuable data obtained by pioneering work such as that of McConnell et al. (2007), which used a classic SP2 and ultrasonic nebulizer U5000AT, we need to estimate the degree of underestimation for such a classic system. However, some important information and details are missing from the current manuscript, as pointed out by Referees 1 and 2. If we make revisions following all the Referees' comments and add the necessary technical information, we believe that a revised manuscript could have sufficient novelty on its own.

As for the general description of the entire CFA system and the units other than the BC unit, we could simplify or move some parts to the Appendix or Supplementary Material, as suggested by Referee 3. It is not new to use SP2, ICP-MS, Picarro, etc., with a melting system. Nevertheless, our CFA system enabled simultaneous analyses of many parameters in one laboratory, which we believe is unique. Although multi-parameter CFA analyses of ice cores have been previously conducted during CFA campaigns using different measurement units, those units were usually brought to a CFA laboratory by multiple laboratories and used only during the campaign. To our knowledge, CFA systems used in such campaigns using different units have been rarely reported. We think it is worth introducing at least briefly the general features of the CFA system built at the National Institute of Polar Research, which covers a wide range of analyses and consistently analyzes all the parameters. We also think it is worth briefly assessing the performance of the different units in the CFA system.

RC2-2 First, the title and abstract indicate the main goal of this manuscript is to present the application of the wide-range SP2 + Marin 5 for continuous ice core analysis. The SP2+nebulizer system has been used for continuous analysis in a number of ice core labs and the details of this specific system for BC measurements in liquid water have been presented previously (Mori at al., 2016), so the assessment of the modified SP2 and Marin 5 nebulizer system is not particularly novel. Most of the other methods presented here (e.g. ice core CFA SP2, Picarro, and ICPMS analysis) are also well-established, with the exception of the BC particle size measurements, and therefore are more appropriate for a methods section of a science-oriented paper in my opinion.

AC2-2 We believe that the assessment of the modified SP2/Marin 5 nebulizer system attached to a melting system and the evaluation of underestimation by previous BC measurements have sufficient novelty for the following two reasons. First, to our knowledge, the dispersion of BC particles, potential losses of BC particles, and resulting changes in BC size distribution in a CFA system have never been assessed. Second, the degree of underestimation of BC mass concentration for the classic SP2/nebulizer system has not been quantitatively evaluated. We would like to emphasize these points in our revised manuscript. Including these results in the methods section of a science-oriented manuscript (Part 2 of our study) would obscure the purpose of the science-oriented manuscript and would be distracting. Moreover, it would be too lengthy. We believe that the important information derived from the Part 1 manuscript should be highlighted; hence, it is not appropriate for Supplementary Material. Therefore, we would like to separate the methods paper (Part 1 of our study) from the science-oriented paper (Part 2 of our study), subject to approval by the Referees and

Editor. We admit that the current manuscript may give the impression that it lacks sufficient novelty. We plan to add more technical information and make revisions to address Referee 2's concerns.

RC2-3 The measurement of BC size distributions throughout the core, though, is quite novel and exciting, but this manuscript lacks detail or justification for this specific measurement. There have not been published long-term reconstructions of BC particles size from Arctic ice cores, largely because as the authors correctly state, "obtaining accurate estimation of the size distribution of BC particles on a routine basis is not easy" (line 67). I agree, and a major reason why is because it is extremely difficult to maintain a stable SP2 response/calibration and nebulizer efficiency throughout an ice core CFA campaign. However, the manuscript did not justify how the authors have overcome these challenges to apply this method to ice core CFA analysis, where it is crucial to demonstrate stability and reproducibility of the method to ensure consistent measurements over weeks and/or months of ongoing analysis. I think more detail is warranted on how the authors ensure a coherent BC size dataset throughout the SIGMA-D analysis, which likely spanned a few months given the stated analysis rate of 6-7 m on one to two analysis days per week. Was the SP2-Marin5 system stable throughout an analysis day, week, month, etc.? Were replicate ice sections analyzed with good agreement? What protocols were used or standards run to ensure a consistent dataset? How was SP2 data processed? Investigating these questions will require presenting some timeseries BC size distribution data, which is omitted entirely in this manuscript despite its emphasis in the title and abstract. Only BC mass and number concentration timeseries are shown and even then, the figures are too small to evaluate the timeseries data.

AC2-3 We agree with these comments, which were also stated by Referee 1. We apologize for not sufficiently describing the stability of the SP2/Marin 5 nebulizer system. We will demonstrate the stability and reproducibility of our method to ensure consistent measurements over a long period of time, as noted in our reply (AC1-6) to Referee 1. Additionally, the stability of the system was ensured by repeated measurements of the same samples over several months or a couple of years, as reported by Mori et al. (2019). We will also briefly explain how we processed the data, as written in our reply to Referee 1 (AC1-7). Unfortunately, we could not analyze replicate ice sections for BC due to the limited amount of the SIGMA-D core. Instead, we believe that the results of the BC loss test presented in Fig. 4 ensure the reproducibility of our method. Again, we apologize for the overly busy Figures 6 and 8. We will add enlarged extracts of the profiles or replace the current figures with them. We will also consider presenting more of the BC size data in a way that does not overlap with the Part 2 manuscript of this study.

RC2-4 Other aspects of the BC dataset that would be valuable to assess would be the Marin 5's performance against the Apex Q, which is more prevalent now for ice/snow analysis than the Cetac U5000AT and also has much better nebulization efficiency for large particles (Wendl et al., 2014). While the Cetac was originally the nebulizer of choice for the SP2 ice core method when it was first introduced (McConnell et al., 2007), I don't think the Cetac should be the benchmark for the underestimation of BC concentration for a 'standard' ice core method anymore since many groups have moved away from it (largely because of its efficiency and stability issues). Lastly, it should be made explicit that many of the findings related to BC concentration underestimation in ice cores presented here apply primarily to Arctic and alpine ice core sites. The choice of nebulizer (Cetac U500AT vs Apex Q, at least) does not seem to impact BC concentrations for Antarctic ice cores sites as much given the much smaller particles and lower BC concs observed at those sites (Arienzo et al., 2016, JGR, Supplemental Fig 1).

AC2-4 We appreciate the constructive suggestion to compare the performances of the Marin-5 and APEX-Q nebulizers. We did compare the efficiencies of both nebulizers for the size range between ~200 and ~700 nm at a flow rate of 0.384 mL/min (Fig. S2). We will add Fig. S2 in the text or as supplementary material. Fig. S2 shows that the efficiency of the Marin-5 is slightly higher than that

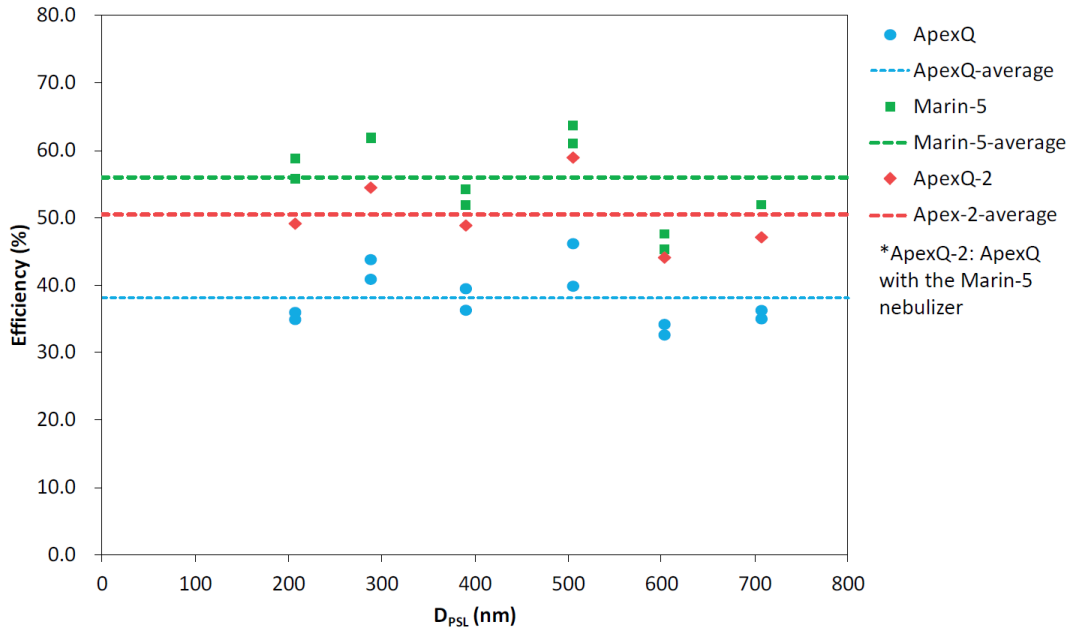

Fig. S2

of the APEX-Q. However, we could not perform stability tests for the APEX-Q or analyze the nebulizer efficiency for larger BC particles because we did not have an APEX-Q in our institute. We could borrow it from a distributor only for a short period.

We could not find Arienzo et al., 2016 in JGR. Do you mean Arienzo et al., 2017, JGR? If you mean Arienzo et al., 2017, JGR, Supplemental Fig. 1 presents concentrations only. We could not find any size distribution data. Could you please provide more information on the paper so that we can download it?

We believe that many of the findings related to BC concentration underestimation in ice cores apply not only to Arctic and alpine ice core sites, but also to Antarctic sites. The size distributions of BC in the surface snow of Eastern Antarctica reported by Kinase et al. (2020, JGR) indicated that the mass ratios of BC particles > 500 nm were large, although the concentrations were very low.

RC2-5 Other sections of the manuscript, including the description of the complete CFA setup with the new addition of the ICPMS and preliminary dating of the SIGMA-D ice core, seemed extraneous and distracting to me from the more exciting BC size distribution idea. As I mentioned previously, I think those sections are more appropriate for the methods section of the science focused manuscript as they are largely established methods. Additionally, the dating section did not include enough detail to be compelling (for example the dating section only showed ~3 m of annual layer counting and did not show the tritium ties or volcanic synchronization).

AC2-5 As we wrote earlier in our reply (AC-2-1), we will simplify or move some parts to supplementary material so that the manuscript does not appear extraneous or distracting. However, we would like to separate Part 1 and Part 2 as explained earlier. Since the dating method, including tritium and sulfate data, has already been published elsewhere (Nagatsuka et al., 2021), we did not repeat it in this manuscript. We will consider moving the dating section to the Part 2 manuscript.

RC2-6 In short, this Part 1 manuscript, which is framed as a BC methods paper by the title and abstract, does not have sufficient novelty or detail to stand alone in its current form. In my opinion, it is better suited to be included as a methods section for the scientific paper unless the manuscript is refocused around the novel BC size distribution method.

AC2-6 As we wrote earlier, we would like to make revisions to the manuscript, emphasizing the novel method for size distribution measurements, to ensure it has sufficient novelty.

Other comments

RC2-7 Line 117: 0.3 +/- 0.1 mm depth resolution seems incorrect- are the units right?

AC2-7 Yes, the units are correct. As mentioned in our reply to Referee 1 (AC1-3), this depth resolution is that of the laser positioning sensor, not the resolution once the water is analyzed by the different online instruments. We apologize for the confusing text. We will revise it to avoid any confusion. The depth resolution of the laser positioning sensor has been defined and published by Dallmayr et al. (2016).

RC2-8 Line 288-289: If the dating section stays, it would be worth including a figure showing volcanic synchronization. What is meant by 'made adjustment' prior to 1783? What exactly was adjusted?

AC2-8 As we mentioned earlier (AC2-5), the results of the volcanic synchronization have been presented in a previous paper by Nagatsuka et al. (2021). Therefore, we do not want to repeat it in this manuscript. However, if requested by the Referees or Editor, we can add a figure showing volcanic synchronization. In any case, we will indicate which volcanic peaks were used to refine the previous dating results. Having said this, we will also consider moving the entire dating section to the Part 2 manuscript of our study if it is deemed more appropriate.

RC2-9 Lines 298-300: Are the sporadic peaks attributed to large particles reproducible? What do the BC size distributions look like for those depths? It would be interesting to understand if any meaningful interpretations can be drawn from them. If they are just filtered out of the data and considered noise, then what is there any advantage of using the wide-range SP2 over a standard one?

AC2-9 Thank you very much for the important comments. Many of the sporadic peaks are attributed to large particles, which are not always reproducible. However, large BC peaks are often found in other ice cores from Greenland and in the SIGMA-D NH_4^+ record. Such peaks are likely due to large boreal forest fires, as presented in the Part 2 manuscript of our study. At these peaks, BC sizes are larger. When we revise the manuscript, we will show size distribution data for such BC peaks.

Although many of the sporadic peaks are filtered out of the data, even 10 mm averages of the raw data show high concentration peaks. The data averaged over 10 mm (Fig. 8) show a difference in mass concentrations for different upper limits of the measurable BC size. However, Figure 8 is too busy, as commented by Referees 1 and 2, and this feature cannot be seen very well. We will present an enlarged extract of the profile to show this feature more clearly. By improving Figure 8, we hope the advantage of using the Wide-Range SP2 over a standard one will be clearer. Figure 9 also shows the advantage of using the Wide-Range SP2 over a standard one.

RC2-10 Lines 329-331: While the CFA system is capable of measuring water isotopes, ICPMS, microparticles, and methane, I don't think that is demonstrated in this manuscript and distracts from the BC focus.

AC2-10 We will revise the manuscript as stated earlier to address this comment.

RC2-11 Does the paper address relevant scientific questions within the scope of ACP?

Yes, ice core BC size distribution and concentration measurements are within the scope of ACP.

RC2-12 Does the paper present novel concepts, ideas, tools, or data?

The BC size distribution method is novel, but the other aspects of the manuscript (dating, ice core CFA analysis) not so much.

AC2-12 To address this comment, we will revise the manuscript as stated earlier.

RC2-13 Are substantial conclusions reached?

No. I do not think the manuscript reaches substantial conclusions, as the novel aspect of the manuscript (the BC particle mass method) is not well described, and the results/conclusions of the SIGMA-D analysis are discussed in a companion paper.

AC2-13 To address this comment, we will revise the manuscript as stated earlier.

RC2-14 Are the scientific methods and assumptions valid and clearly outlined? Are the results sufficient to support the interpretations and conclusions? Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

No. More information needed on the stability and reproducibility of the BC size distribution method over the course of the ice core analysis. The BC size distribution records are not presented in this manuscript making it difficult to assess the method.

AC2-14 We will add more information on the stability and reproducibility of the BC size distribution method, as stated earlier. We will also consider presenting the size distribution data.

RC2-15 Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

Yes

RC2-16 Does the title clearly reflect the contents of the paper? Does the abstract provide a concise and complete summary? Is the overall presentation well structured and clear?

Somewhat. The title and abstract focus on BC measurements, but the manuscript also includes sections about the full NIPR CFA system and SIGMA-D ice core dating that I found distracting.

AC2-16 We will restructure the distracting parts of the manuscript.

RC-17 Is the language fluent and precise? Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

Yes

RC-18 Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

Yes. Described in comments above.

AC2-18 We would like to keep this manuscript separate from Part 2 manuscript of our study for the reasons stated above.

RC2-19 Are the number and quality of references appropriate?

Yes, though lacks citations to more recently published Arctic BC records.

AC2-19 We will add more recently published Arctic BC records to the references.