

## **Response to the comments of referee #2 (RC C677):**

First, we would like to thank the anonymous referee very much for the comments and suggestions. Below the original comments -shown in italics- answered point-by-point, which will be taken into account in the revised manuscript accordingly.

*It would be very helpful if the experimental section contained some description of the snow and ice core samples used.*

We inserted the following in the manuscript:

### **2.4.3 Snow and ice samples**

We used several snow and ice samples to monitor the stability of the setup and to test the optimal method on real samples. The so-called internal snow standards were prepared from fresh snow from Blewett Pass, WA, USA, and from Ewigschneefeld, Switzerland. A large amount of fresh snow was melted and kept in a glass bottle in the refrigerator ( $\sim 5^{\circ}\text{C}$ ). The ice core samples used for testing the optimal method include two core segments from an ice core from Tsambagarav glacier, Mongolian Altai, drilled at 4130 m asl in 2009, and two core segments from Lomonosovfonna, Svalbard, Norway, drilled at 1202 m asl in 2009. Those two ice cores are thought to represent the extremes in terms of mineral dust content with the Mongolian core being highly influenced by dust from the nearby deserts and the Svalbard core being remote from any large dust source. The dust content was estimated based on the average calcium concentration in the core segments.

*Page 5 lines 15-17: It would be helpful here to include more information on how the SP2 was calibrated for larger particle sizes since this is not as straightforward as calibration at smaller sizes is. It would also be helpful to report the precise range over which the SP2 calibration was done.*

Page 3079, line 2-5: We inserted the following in the manuscript:

“The calibration of the SP2 was conducted up to a mass of 70 fg BC. The measured calibration curve was linearly extrapolated to cover the full dynamic range of the incandescence detector (up to  $\sim 500$  fg, which corresponds to a BC mass equivalent diameter of  $\sim 810$  nm). Moteki and Kondo (2010) showed that the SP2 calibration curves can deviate from linearity for larger BC mass, depending on the effective density of the particles. In this study, no deviation from linearity was observed up to a BC mass of 70 fg BC and the doubly charged particles indicated that this still holds with little uncertainty up to 140 fg BC. Sensitivity analyses with using an empirical power law calibration curve for the BC mass range above 100 fg, in a similar manner as applied by Schwarz et al. (2012), indicated deviations from the linear calibration approach well below the general calibration uncertainty of the SP2. This confirms that choosing a linear calibration curve is appropriate for this study. However, this would not necessarily hold, when a substantial fraction of the BC mass would be detected at BC mass equivalent diameters above one micrometer.”

*Page 7 lines 29-30: "Repeated measurements of the same sample varied within 15% standard deviation of the mean . . ." This meaning here is not clear, did you mean varied within 15% of one standard deviation or something else?*

Page 3081, lines 25-26: We changed the sentence in the manuscript as follows:

"The standard deviation of repeated measurements of the same sample fell within 15% of the mean, indicating the setup is stable."

*Page 8 lines 22-23: "It may be necessary to choose different SP2 calibration for the aqueous BC standard and e.g. an ice core sample." This meaning of this sentence is unclear.*

Page 3082, lines 23-24: This sentence means that dependent on the BC-type in standard and sample that might differ, it might be necessary to use different SP2 calibrations for standard and sample. So that for each standard and sample to suitable SP2 calibration is used according to the BC-type contained.

We changed the sentence in the manuscript as follows:

"The BC-type contained in the aqueous BC standard may differ from that e.g. in an ice core sample. Thus it may be necessary to apply different SP2 calibration curves for the analysis of the BC standard and the ice core samples, so that the most suitable SP2 calibration is used for either BC-type."

A more detailed discussion about the choice of calibration curves is provided in the supplement (S.4 and S.5), as already noted in the discussion version of the manuscript.

*Page 10 lines 15-17: "This restricts the choice of calibration materials to BC-types for which the SP2 sensitivity is known, whereas it is not necessary that the SP2 sensitivity is equal for the sample and the standard." This is a somewhat unclear. The supplemental material gives a nice explanation of how the SP2 sensitivity to each of the standard and sample can introduce uncertainty into the measurement, but the explanation here in the manuscript is somewhat confusing. Do you mean that ideally one would choose a calibration material for which the SP2 sensitivity is known, but that it is not necessary for the sensitivity to the sample and standard be the same (although this may add some uncertainty to the measurement)?*

Page 3084-3085, lines 22-23, lines 1-2: We modified the text in the manuscript as follows:

"This restricts the choice of internal calibration standards to BC-types for which the SP2 sensitivity is known ( $\Rightarrow f_{\text{bias}}^* = 1$ ). The factor  $f_{\text{bias}}$  only becomes unity if the sensitivity of the SP2 to the BC-type in the sample is known, therefore potentially leaving some uncertainty. Note that it does not matter whether the SP2's sensitivity is equal or different for the standard and sample nor does it have any influence on the resulting uncertainty of the method, if appropriate calibration curves are chosen for the analysis of both measurements. The reason for this is that, in this approach, the measurement of the standard is solely used to quantify the efficiency of the nebulizer, while it is not used to quantify the sensitivity of the SP2."

Note, the measurement of the standard could in principle also be used to determine the overall

efficiency/sensitivity of the whole nebulizer-SP2 unit. In this case, the condition to minimize the measurement uncertainties would be that the SP2 is equally sensitive to the BC type in the standard and the sample under investigation. However, such an approach is different from the approach described in this part of the manuscript. A more detailed discussion about this topic, including for example the treatment of non-BC components in standard BC materials, is provided in the supplementary material.

*Supplemental Material:*

*Page 24 line 5: D on the bottom right-hand side of the equation should be DBC*

*Page 24 line 12: DBC on the right-hand side of the equation should be DPSL*

*Page 24 line 14: The first instance of DBC on the right-hand side of the equation should be DPSL*

We changed this in the manuscript accordingly.

**Additional correction:**

*Page 3082, line 19: "...commonly done with a mass-selected Fullerene Soot..."*

We changed this in the manuscript as follows:

*"...commonly done with mass-selected Fullerene Soot..."*