Author response to:
Review of "Assessing recent measurement techniques for quantifying black carbon concentration in snow" by J. P. Schwarz et al.

Anonymous Referee #2

Author response italicized.

General comments:
This paper presents evaluations of the two recent methods for the measurement of BC concentration in snow. One utilizes the Single Particle Soot Photometer (SP2) and the other does the Integrating Sphere/Integrating Sandwich Spectrophotometer (ISSW). For the SP2 method, authors found that the efficiencies of the nebulizers used in this study and previous studies are strongly dependent on the size of the particles in liquid samples. Authors also indicated that BC particles in snow tend to be larger than typically observed in the atmosphere, which brings about larger uncertainty of the SP2 calibration (and possibly for ISSW). These findings are very important considering recent increasing usage of the SP2 for measurement of BC in water samples. However, discussion on the estimate of the measurement uncertainty is not clear as detailed below. They should be fully explained before this paper is considered for publication.

> *The authors appreciate Anonymous Reviewer #2's assessment of the value of our manuscript and his/her suggestions for improving clarity.*

Major comments:
1. The descriptions of the critical parameters of the nebulizer are unclear, especially for the paragraph on page 3777, Line 7-18.

a) Here only a relative efficiency is shown (Figure 1). The procedure to derive absolute efficiency should be provided, including mathematical formulations. The use of stopping distance and effective density should be related to the efficiency in a more understandable way.

> *We have rewritten the discussion of the procedure to derive absolute efficiency, expanding the explanation and reshaping it to simplify understanding. We now include mathematical formulae. To relate the efficiency in a more understandable way to stopping distance and effective density, we have adopted Reviewer #2's suggestion, b), below.*

b) The efficiency for BC should be shown in the same way as for Figure 1. After all, what matters is the efficiency of BC. Page 3778 L5-26. The authors assume that the nebulizer efficiency depends on particle stopping distance, and convert the PSL-based efficiency into the efficiency for BC. In that case, the figure of relative transmission efficiency for BC as a function of BC diameter (VED) should be shown, corresponding to Fig. 1 (the efficiency for PSL as a function of PSL diameter).

We have modified Figure 1 to include the BC-relevant efficiency curve, as suggested.

How different are the size-dependent efficiencies between for PSL diameter and for BC diameter? Moreover, the authors adopted the effective density of BC of 0.8 g cm-3 by averaging six BC materials in Moteki and Kondo (2010), but the value of fullerene soot (the same laboratory standard of this manuscript) in Moteki and Kondo (2010) is 0.5 g cm-3 and that of ambient BC in Tokyo is 0.3 g cm-3. The uncertainty of the assumed effective density of BC would lead to the uncertainties of the estimated size-dependant nebulizer efficiency for BC and determined mass concentrations of BC in snow. These uncertainties derived from the assumption of the effective density of BC should be explained.

In addition to showing the sensitivity to the density assumption between 0.8 and 0.9 g/cc density, we have added the following sentences to clarify that the 0.8/0.9 g/cc estimates for BC effective density in the snow are conservative, and explicitly stated that the connection between the uncertainty in the density with that in the impact of the nebulizer efficiency size dependence, as follows:

> For BC, we adopted a value for the effective density of 0.8 g cm^{-3}, which is the average density measured between 400—900 nm mobility diameter for six BC materials in Moteki and Kondo~(2011); these materials did not include the glassy carbon spheres from that study, which are clearly not related to BC structure in the ambient, and have much higher density, but did include presumably fresh BC measured in Tokyo with an effective density of only 0.3 g cm^{-3}. Such a low effective density is likely not appropriate for processed BC such as would be found in snow, and would therefore result in very short calculated stopping distances and thus underestimate the losses of larger BC. Therefore, We believe the 0.8 g cm^{-3} density estimate is reasonably conservative; although it is clear that uncertainty in the actual effective density of the BC translates into uncertainty in the nebulizer losses.

Page 3777 L7-11, in relation to a) and b). If the size-dependence of the nebulizer efficiency is due to the aerodynamic properties of the PSLs with the different diameters after the droplets evaporation in the nebulizer, the description "this efficiency was of order 1 ug-liquid cm-3 for 505nm PSLs" (L11) is somewhat confusing because "how much liquid the nebulizer aerosolized into a given volume of air" (in L9) may not depend on the size of the PSLs. Possible reasons for the size-dependence of the nebulizer efficiency should be mentioned clearly, and the efficiency of extracting PSL particles from liquid to air (i.e. to the SP2) should be quantified or explained in an appropriate way (e.g. the number fraction of the extracted particles to the total nebulizer-induced particles).

We have clarified in the text that the absolute efficiency at a given size does not represent an actual measure of the quantity of liquid being mixed into air, to avoid confusion between the absolute nebulization efficiency differing at different particle *size, with the concept of a fixed amount of fluid being mixed in the air.*

We also expand the existing discussion of aerosol losses to include the fact that the loss term of PSLs in transport to the SP2 laser in our laboratory conditions was small compared to the losses occurring in the nebulizer.

Finally, we make clear that we are using an empirical approach to evaluating nebulization efficiency impacts and directly determining the efficiency of extracting PSL particles from liquid to the SP2.

2. Biases to the ISSW measurements are discussed, including non-BC light absorbing and purely scattering particulate in water. In this paper, the shift in the BC size in snow is discussed. However, the effect of this shift on ISSW measurement is not discussed. It should be estimated.

In the time that this manuscript was under review, we performed additional analyses, and wrote an additional manuscript, describing the implications of the BC size distribution on MAC (which we alluded to as a possibility in this paper). However, given the possible impact of filter effects in the ISSW, we are not able to estimate the possible impacts on the ISSW, beyond the existing discussion.

3. This paper is aimed to assess the two measurement techniques. However, no inter-comparisons using the snow samples collected by the field campaigns are shown. This inter-comparison provides overall uncertainties of the two techniques and therefore should be included.

As discussed in the text, the real snow samples were not stored correctly (after initial measurements were made with the ISSW, but before the SP2 was used), and thus do not provide a meaningful comparison. We included information about the BC concentration changes in the storage bottle as a helpful side note for other experimentalists concerned with this issue.

4. Page 3785 L9-18. The authors reports that the most contaminated dust in samples is equivalent to 15 ng-BC/g-H2O. What is the approximate BC-equivalent size of these dust particles? Is it possible that some large BC particles which the authors observed in Fig. 2 are artifacts due to the contaminated dust?

We have expanded the discussion of the identification of the dust, including the following facts: 1) the BC-equivalent size of the dust was predominantly below 500 nm VED; and 2) that the color-temperature of the large particles measured by the SP2 were consistent with BC composition, indicating that dust is not the source of the size distribution shift shown in Figure 2. We also expanded discussion of SP2 setup so the reader will easily understand the gain settings used to achieve these results.

Page P3785 L9-18. The offset due to the contaminated dust (15 ng-BC/g-H2O) looks "small" (in L9 and P3787 L18) when considering the higher BC concentration (up to about 1200 ng/gr) seen in Fig. 3. However, BC concentrations in many snow samples in previous studies are lower than 100 ng/g, and much lower than this for some arctic or mountain site snow samples. The lower the BC concentration in snow is, the larger the uncertainties due to dust will be. The offset indicates that previous studies using a SP2 for measurement of BC in snow and ice may overestimate BC concentration for highly dust-contaminated samples. I would suggest mentioning these points.

We have added the suggested discussion.

In relation to this point, the comparison shown in Figure 3 is made for the BC size ranges far exceeding the concentrations encountered in natural environment. The figure and discussion should focus on the BC range below 200 ng/g.

The figure and discussion focuses on an appropriate range of BC concentration in snow: Ming et al., 2008 (citation below) indicate loadings up to 980 ng/g in the natural environment.

Ming et al. (2008): Black Carbon (BC) in the snow of glaciers in west China and its potential effects on albedos, Atmospheric Research, ***92,*** *114-123.*

Minor Comments: # P3773 L4-5. Some papers are not included in the reference list.

We have ensured consistency between citations and references.

P3776 L9-10. What is the volume flow rate of the peristaltic pump? How much total sample volume is required for the analysis of typical snow?

We have added the answers to these questions in the paper: the flow rate was 0.005 $cm^3\ s^{-1}$*, and the total snowmelt volume required for a BC concentration measurement with consistency check in very clean snow was ~ 4 gr.*

P3778 L11. Moteki and Kondo (2011) should be Moteki and Kondo (2010)
Corrected

#P3779 L26. sonewhat should be somewhat

Corrected.

#P3783 L19-24. The range of BC concentrations for laboratory experiments also should be mentioned here.

Added.

#P3789 L21-26 The order of the references is incorrect.

Corrected.

Figure 3. The unit of the X-axis should be ng g-1, to be consistent with the text.

Modified.