

Interactive comment on “Development of an improved two-sphere integration technique for quantifying black carbon concentrations in the atmosphere and seasonal snow” by Xin Wang and Xueying Zhang

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We are very grateful for the referee's critical comments and suggestions. The followings are our point-by-point responses to the comments. Our responses start with “R:”.

General comments The authors report the development of a two-sphere integration spectrophotometer for quantitative measurement of the mass concentration of ambient BC (black carbon) and BC in the snow. As BC is a major light-absorbing aerosol,

C1

which will accelerate the snow melt after its deposition onto the surface of snow or ice, thus plays a key role in regional and global climate change. The reported instrument provides an important and useful method for measuring BC in snow. By using the developed instrument, the authors investigated the spatial distribution of BC light absorption in surface snow across northern China during Jan. to Feb. 2014. This section is interesting, which may provide a better constrain of BC simulation in the earth system. I have the following comments to improve the manuscript.

Specific comments Abstract, line 41 and line 42: BC absorption contributed 68.5%-95.9% of total light absorption in the atmosphere and 52.3%-93.3% in seasonal snow over northern China. In my experience, the values for atmosphere air are a bit too high. The wavelength needs to be specified here. Or are they mass concentration contribution?

Response: Sorry for the misleading. We agreed with the reviewer that the wavelength should be given. In previous version, the absorption contribution of BC to total light absorption was at a wavelength of 600 nm. Compared with previous studies, we have decided to update all of the datasets in this study at the most common wavelength of 550 nm. For this issue, the absorption contribution of BC to total light absorption at 550 nm was lower than that of 600 nm in the revised manuscript. The result is also consistent with the previous study by Wang et al. (2013, their Figure 11).

References: Wang, X., Doherty, S.J., and Huang, J.P.: Black carbon and other light-absorbing impurities in snow across Northern China, *J. Geophys. Res.-Atmos.*, 118, 1471-1492, doi: 10.1029/2012JD018291, 2013.

Page 4, line 101, what is the reason that causes 60% error for snow measurement with SP2 method? More discussion will better show the clear advantage of the developed instrument in this paper over the current available methods. Also in line 106, “biases remain”, what are the biases sources? I suggest to give a table to list the uncertainties of each instrument for both ambient and snow measurement.

C2

Response: There are two major issues in leading the large uncertainty. Based on the investigation by Schwarz et al. (2012), the relative transmission efficiencies of polystyrene latex sphere concentration standards (PSLs) in liquid to the SP2 after aerosolization remarkably reduce to 20% due to larger diameter of BC particles (> 500 nm, their Fig. 1). Therefore, the larger diameter of BC (>500 nm) is hardly captured by SP2 instrument with a Collison-type nebulizer. Moreover, the mixing status of BC in snow is more complicated than the standard fullerene soot in the laboratory and the typical BC in the atmosphere. As a summary, they found that the SP2 instrument can be used to measure BC mass concentration in snow with substantially larger uncertainty (60 %) than for atmospheric sampling (<30 %).

References: Schwarz, J. P., Doherty, S. J., Li, F., Ruggiero, S. T., Tanner, C. E., Perring, A. E., Gao, R. S., and Fahey, D. W.: Assessing Single Particle Soot Photometer and Integrating Sphere/Integrating Sandwich Spectrophotometer measurement techniques for quantifying black carbon concentration in snow, *Atmos. Meas. Tech.*, 5, 2581-2592, 10.5194/amt-5-2581-2012, 2012.

Page 5, line 132, is there possible for the loss of light absorption organic aerosol?

Response: We agreed with the reviewer that there is possible to lose the light absorption due to organic carbon. Owing to BC in snow is often hydrophobic, long time melting could lose more BC to the container walls instead of collected on the filter (Ogren et al., 1983). In order to minimize the loss of BC and OC mass, we quickly melted the snow samples in a microwave within a very short time. Therefore, the loss of insoluble organic aerosols is very limited, and can be neglectable. At present, this method is widely performed for snow melting procedure. (Doherty et al., 2010,2014; Wang et al.,2013).

References: Ogren, J. A., Charlson, R. J., and Groblicki, P. J.: Determination of elemental carbon in rainwater, *Anal. Chem.*, 55, 1569–1572,1983. Doherty, S.J., Warren, S.G., Grenfell, T.C., Clarke, A.D., and Brandt, R.E.: Light-absorbing impurities in Arc-

C3

tic snow, *Atmos. Chem. Phys.*, 10, 11647-11680, doi: 10.5194/acp-10-11647-2010, 2010. Doherty, S.J., Dang, C., Hegg, D.A., Zhang, R.D., and Warren, S.G.: Black carbon and other light-absorbing particles in snow of central North America, *J. Geophys. Res.-Atmos.*, 119, 12807-12831, doi: 10.1002/2014JD022350, 2014. Wang, X., Doherty, S.J., and Huang, J.P.: Black carbon and other light-absorbing impurities in snow across Northern China, *J. Geophys. Res.-Atmos.*, 118, 1471-1492, doi: 10.1029/2012JD018291, 2013.

Page 7, the spectrum information and an example for the data processing for this part are encouraged to be shown in the supporting information. The wavelength should be specified? Did the authors make an average over the selected range?

Response: We feel sorry for the misleading. We have carefully unified all of the relative spectrum at a given wavelength of 550 nm and replotted the relative Figures in the revised manuscript.

Page 9, line 234, there is a typo. Figure 2 should be Figure 3. How to determine the filter loading?

Response: Corrected.

Page 10, Fig. 4 was lost in the text.

Response: We have corrected this mistake and added the relative description of Figure 4 in Page 11, Line 303.

The results got from TSI method were smaller than that with the two-step method for the snow samples over northeast China (Fig. 6), but the results for Lanzhou were the opposite (Fig. 7). Some more discussion is encouraged to explain the underestimated and overestimated of these two methods?

Response: We feel sorry for the misleading. However, there seems no discrepancy for Figure 6 and Figure 7. As seen in Figure 5, it clearly shows that the loss of BC is in the range of 12-20% due to 1- μ m quartz fiber filters. Therefore, if we scaled the BC

C4

mass in the atmosphere as 120% based on the thermal optical method, the results of BC concentration in the atmosphere should be much similar as Figure 6. Therefore, we concluded that the thermal-optical method underestimates BC concentrations by 35%–45% than that measured by TSI technique.

Page 12, line 322, is this method related to size distribution?

Response: This technique on estimating BC light absorption is only based on the integrated light absorption in the wavelength of 400-750 nm due to all insoluble LAPs on the Nuclepore filters. Therefore, this method is nothing relevant with the snow grain size. However, there is still a need to provide the information of the snow grain size during the snow field campaign, which is the key parameter in determining the snow albedo, and pretty useful in validating the radiative transfer models.

Page 34, Fig. 3, the Y axis should be S/S_0 , not $-\ln(S/S_0)$. The authors gave the fit equation in the figure: $y = a \cdot \exp(-b \cdot x) + c$.

Response: We have corrected this mistake, and replotted Fig. 3. Therefore, the Y axis is still given as the relative attenuation (A_{tn}), " $A_{tn} = -\ln[S/S_0]$ ".

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