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# Influence of the melting temperature on the measurement of the mass concentration and size distribution of black carbon in snow

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Abstract. The influence of temperature and time of snow sample melting on the measurement of mass concentration and size distribution of black carbon (BC) in snow was evaluated experimentally. In the experiments, fresh (Shirouma) and aged (Hakusan) snow samples were melted at different temperatures or at different time lengths, and the BC mass concentration and size distribution in the melted snow samples were measured using a nebulizer and a single-particle soot photometer (SP2). In the experiment where melting temperature was varied, the BC mass concentration in the liquid decreased at a melting temperature of 70 °C. This decrease was 8.0% for the Shirouma sample and 46.4% for the Hakusan sample and depended on BC particle size, with a significant decrease found at BC diameters less than 350 nm. A similar decrease in BC mass concentration was found when the Hakusan snow sample that had been melted at 5 °C was heated to 70 °C. The experiment in which melting time was varied indicated that BC mass concentration in the liquid did not change for the Shirouma sample but decreased significantly with a longer melting time for the Hakusan sample (38.6%). These results indicate that melting of snow samples at high temperatures or over long time periods can significantly affect the measurement of BC mass and its size distribution, especially for aged snow samples.

# 1 Introduction

Black carbon (BC), commonly referred to as soot, strongly absorbs solar radiation in the atmosphere, leading to significant climate effects. The deposition of BC onto snowpacks/ice packs also leads to positive radiative forcing because it can significantly reduce snow/ice albedo (Bond et al., 2013; Warren and Wiscombe, 1980; Wiscombe and Warren, 1980). The snow/ice albedo effect of BC, together with other processes (water supply, grains, etc.), also accelerates the melting of snow and ice (Aoki et al., 2011; Brandt et al., 2011). Therefore, numerous studies have been conducted to measure BC concentration in snow/ice and estimate its influence on the climate via radiative forcing, surface albedo, and so on.

In the latest report by the Intergovernmental Panel on Climate Change (IPCC, 2013), the global radiative forcing of BC in snow since 1875 was estimated to be  $0.04 \text{ Wm}^{-2}$ , with a 90% uncertainty range between +0.02 and  $+0.09 \text{ Wm}^{-2}$ . Bond et al. (2013) also provided a central estimate of  $0.04 \text{ Wm}^{-2}$ , between +0.01 and  $+0.09 \text{ Wm}^{-2}$ . Such work shows that there remains a large uncertainty in the estimation of the climate effects of BC on snow/ice; therefore, more studies are necessary on BC in snow, including the improvement of its measurement techniques.

The mass concentration of BC in snow/ice is commonly measured using one of the following three methods. The first utilizes light absorption by the BC. In this method, a snow sample is melted and filtered. Diffuse light is irradiated to the filter to measure its light transmission, and the mass of BC retained on the filter is derived from the decrement of the transmission (Clarke and Noone, 1985; Grenfell et al., 2011; Warren and Clarke, 1990). The second method utilizes the thermal optical technique. In this method, elemental carbon (EC), rather than BC, is measured. The snow sample is melted and filtered with a quartz fibre filter. The EC retained on the filter is thermally converted to CO2 gas, and the mass of EC is derived by measuring the CO<sub>2</sub> concentration (Aoki et al., 2011; Chow et al., 2001, 2007; Jenk et al., 2006; Lavancy et al., 1999). The third method utilizes the single-particle soot photometer (SP2). In this method, snow samples are melted and aerosolized using a nebulizer. The mass and size of each aerosolized BC particle are measured with the SP2 instrument (Lim et al., 2014; McConnell et al., 2007; Ohata et al., 2013; Schwarz et al., 2012, 2013). This method has the advantage that it provides not only the total BC mass but also the size distribution of BC particles in the snow. The BC size distribution is very important because it significantly affects the mass absorption cross section of BC in snow (Schwarz et al., 2013). There are several factors affecting the uncertainty of measured BC mass concentration by using these methods, and several studies have been performed to reduce such uncertainty and increase precision and reliability. For example, Mori et al. (2016), Ohata et al. (2011, 2013), Schwarz et al. (2012), and Wendl et al. (2014) estimated uncertainties in the SP2 method by evaluating the size-dependent efficiency of certain nebulizers, the effect of agitation of a liquid BC sample, calibration techniques, etc. Although the melting of snow/ice samples is a common procedure in the three methods above, uncertainties due to this procedure have not yet been fully examined. Many researchers use a microwave oven or hot water bath for melting snow/ice samples faster, to reduce BC loss onto the inner walls of sample containers as well as variation in the BC size distribution (e.g., Brandt et al., 2011; Doherty et al., 2010; Schwarz et al., 2012; Warren and Clarke, 1990). However, some studies suggest that the heating condition for melting snow/ice samples may significantly affect the BC mass concentration and its size distribution in the melted water, leading to possible uncertainty in the measurement of BC mass in snow/ice. Schwarz et al. (2012), Lim et al. (2014), and Wendl et al. (2014) indicated that freezing and melting cycles significantly affect BC mass concentration and size distribution in water, and that it is possible that BC size distribution can provide information about the thermal history of the snow. Lim et al. (2014) evaluated sample treatment procedures in the SP2 method, including the melting procedure, freezing/melting cycle, and surface area to volume ratio of the sample containers. They compared two snow melting procedures, melting at room temperature and melting in a warm bath (30 °C), but did not find a significant difference in the measured BC concentration between the two procedures. They suggested that faster melting of snow samples is preferable and that the melting temperature does

not significantly affect the measurement. However, considering that higher temperatures have been adopted for the faster melting of snow/ice samples using a microwave oven in several studies, the influence of temperature should be evaluated for a broad temperature range. It is possible that the history of snow/ice samples, such as various chemical species deposited on snow or the cycles of partial melting and refreezing, affects the influence of the melting conditions on the measurement.

In this study, experiments to measure BC mass and size distribution in fresh and aged snow samples melted at various temperatures and over various lengths of time were conducted with the SP2 instrument (Droplet Measurement Technology, Boulder, Colorado, USA). By comparing the results for the different temperatures and the time lengths, their influence on BC measurement in snow/ice are evaluated.

# 2 Experiment

## 2.1 Snow samples

The snow samples used in this study were obtained from two locations: Hakusan (36°17' N, 136°62' E) and Shirouma (36°76' N, 137°88' E), Japan. Sampling was conducted on 13 and 21 March 2013, respectively. The Shirouma sample was fresh, powder snow, sampled within 6h of the snowfall. The Hakusan sample consisted of aged, granular snow that was sampled approximately 2 weeks after the snowfall. This sample probably experienced partial melting (by sunlight heating or rainfall) and refreezing; therefore, it was likely more inhomogenized. After the sampling, these samples were stored in polypropylene (PP) containers at temperatures below -30 °C. In order to make the snow samples more uniform, they were stirred well with a mixer in a low temperature laboratory (room temperature of -30 °C) at the National Institute of Polar Research. After stirring, samples were divided and melted. All containers and bottles used in this study had already been cleaned with ethanol and ultrapure water.

## 2.2 Melting process

In this study, two experiments were performed to evaluate the influence of melting temperature and melting time length. For the experiment to evaluate the former influence (melting temperature experiment), each (Hakusan or Shirouma) snow sample was stirred in a 500 cm<sup>3</sup> bottle and then divided into nine 30 cm<sup>3</sup> glass bottles for melting. Snow samples in each three of these nine bottles were melted in a water bath at one of three temperatures: 5, 20, and 70 °C. In order to control the melting temperature accurately, we did not use a microwave oven. Inhomogeneity in each snow sample was estimated using the range of measurement results for the three bottled samples melted at the same temperature. In comparison to the melting temperature experiment, the experiment to evaluate the influence of melting time length (melting time experiment) used a larger amount of snow (sampled across a wider area at Shirouma or to a larger depth at Hakusan). Therefore, the BC mass concentration in each snow sample was not necessarily identical across the two experiments. In the melting time experiment, each snow sample was stirred in a 6000 cm<sup>3</sup> metallic can and then divided into three 30 cm<sup>3</sup> glass bottles and one 500 cm<sup>3</sup> glass bottle. Snow samples in these bottles were melted in a refrigerator at a temperature of approximately 1 °C. Differences in bottle volume affected melting time length: it took 2-3 h to melt snow samples in the 30 cm<sup>3</sup> bottles and more than 6 h for the 500 cm<sup>3</sup> bottles. A low temperature (1 °C) was adopted because the melting temperature experiment had previously shown that a lower melting temperature is preferable for snow BC measurement. Moreover, a lower melting temperature allowed for a broader range of melting time lengths.

In both experiments, each bottle or container was closed during melting and storage, and after the melting, samples were not transferred to other containers, so as to minimize the risk of contamination. All liquid samples were sonicated for 15 min and shaken just before measurement.

#### 2.3 Measurement of the BC mass concentration

The BC mass concentration and size distribution in the melted samples were measured with an experiment system nearly identical to that used by Ohata et al. (2011, 2013) and Mori et al. (2016). Details of the SP2 instrument used have been described elsewhere (Moteki and Kondo, 2007, 2010; Mori et al., 2016; Stephen et al., 2003). The liquid of a melted snow sample was transferred to a concentric pneumatic nebulizer (Marin-5, Cetac Technologies, Inc., Omaha, Nebraska, USA) with a peristatic pump (REGRO Analog, ISMATEC SA., Feldeggstrasse, Glattbrugg, Switzerland) and aerosolized using dry, filtered air at a flow rate of  $15.23 \text{ cm}^3 \text{ s}^{-1}$ . The sample droplets in the airflow were evaporated during passage through a heater tube at 140 °C, and the water vapor in the airflow was removed during passage through a chiller at 3 °C. The BC and other nonvolatile particles remaining in the airflow were introduced to the SP2 to measure the mass and size of each BC particle. Mori et al. (2016) calibrated the entire experimental system using simulated BC material. They also determined that the detection efficiency of 200 to 2000 nm BC in the liquid was consistently around 50%.

## 3 Results

## 3.1 Influence of melting temperature

Figure 1a and b show the median total mass concentration of BC in the melted Hakusan and Shirouma snow samples, respectively. Error bars show the range between minimum and maximum. The total mass concentrations of BC in the Haku-



**Figure 1.** Total mass concentration of BC in (**a**) Hakusan and (**b**) Shirouma snow samples melted at 5, 20, and 70 °C.

san snow sample melted at temperatures of 5, 20, and 70 °C were 60.7 (55.2-69.8), 63.6 (48.1-67.2), and 32.5 (31.8-42.9)  $\mu$ g L<sup>-1</sup>, respectively; in the Shirouma snow sample, they were 24.7 (23.3-29.2), 24.6 (24.2-27.3) and 22.8 (22.6-23.8)  $\mu$ g L<sup>-1</sup>, respectively. To estimate the drift of the experimental system, repeat measurements of selected samples were conducted after all samples had been measured. The average difference in BC mass concentration between the first and second measurements was -2.1 % (-0.4 to -5.1 %). For both samples, the BC mass concentration values do not show a significant difference between the melting temperatures of 5 and 20 °C; however, the BC mass concentration was systematically smaller at the 70 °C melting temperature. In the case of the Hakusan sample, the difference between 70 and 5/20 °C is significant, exceeding random errors from measurement and sample inhomogeneity.

Figure 2a shows the median mass size distribution of BC in the Hakusan snow samples melted at temperatures of 5, 20, and 70 °C; Fig. 2b shows the same but for the Shirouma snow sample. For the Hakusan sample, the mass concentration of BC particles with diameters less than 400 nm is significantly smaller in the snow melted at 70 °C than that at the other temperatures. For the Shirouma sample, the BC mass size distributions for the three melting temperatures are similar to one other; although, the values at the higher melting temperature are smaller for BC particles of less than 350 nm diameter. These results show that the BC mass concentration in melted snow decreases at higher (70 °C) melting temperatures, especially for BC particles smaller than 350 nm. Figure 3 shows the ratio of the median BC mass in the samples melted at 70 °C to those melted at 5 °C, as a function of BC size. Error bars show the minimum and maximum ratios. This figure shows that the ratio is lower for smaller particle sizes. In addition, considering that the Hakusan sample was aged and had much more BC, the difference between the Shi-



**Figure 2.** Mass size distribution of BC in (**a**) Hakusan and (**b**) Shirouma snow samples melted at 5, 20, and 70 °C, where M and Dp denote the mass and diameter of a BC particle, respectively.



Figure 3. Size distribution of the ratio of the BC mass concentration in the snow samples melted at  $70 \,^{\circ}$ C to those at  $5 \,^{\circ}$ C.

rouma and Hakusan samples suggests that the ratio decrease may depend on the age of the snow (and/or other reasons such as pollution).

The influence of high temperature (or heating) was also evaluated by a further experiment. We heated the liquid of the Shirouma snow sample melted at 5 to 70 °C and measured the mass size distribution of the BC. Figure 4 shows the size distribution of the BC mass ratio before and after the heating. The error bars show the reproducibility of the measurement. The ratio decreased significantly with BC diameters below 300 nm and the decrease was more evident below 150 nm, indicating that reduction of the mass concentration of smaller BC particles can occur at higher temperature (70 °C) not only during snowmelt but also after melting.



**Figure 4.** Size distribution of the ratio of the BC mass concentration in the Shirouma snow sample melted at 5 °C and heated at 70 °C to that before heating.

#### 3.2 Influence of melting time

Figure 5a and b show the median total mass concentration of BC in the melted Hakusan and Shirouma snow samples, respectively, comparing values for the samples melted in the 30 and  $500 \,\mathrm{cm}^3$  bottles. Error bars for the  $30 \,\mathrm{cm}^3$  bottles show the range between the minimum and maximum. Because only one 500 cm<sup>3</sup> bottle was measured for each snow sample, the random error range was estimated by assuming that the sample inhomogeneity was the same as that for the three 30 cm<sup>3</sup> bottle samples. Concerning repeatability, the Hakusan 500 cm<sup>3</sup> bottle sample was measured a second time after other sample measurements, and the difference in BC mass concentration between the first and second measurements was 7.8%. The time required for melting was approximately 2 h in the 30 cm<sup>3</sup> bottles and exceeded 6h in the 500 cm<sup>3</sup> bottles. The total mass concentrations of BC in the Hakusan snow sample melted in the 30 cm<sup>3</sup> bottles and 500 cm<sup>3</sup> bottles were 37.2 (33.9–30.1)  $\mu$ g L<sup>-1</sup> and 22.9 (20.8–24.0)  $\mu$ g L<sup>-1</sup>, respectively. This difference is significant, exceeding the random error range. On the contrary, in the Shirouma snow sample, the values were 16.3 (14.3-18.2)  $\mu$ g L<sup>-1</sup> and 15.1 (13.2–16.8)  $\mu$ g L<sup>-1</sup>, respectively. This difference is negligibly small.

Figure 6a shows the median of the mass size distribution of BC in the Hakusan snow samples melted in the  $30 \text{ cm}^3$ bottles and the  $500 \text{ cm}^3$  bottle. Figure 6b shows the same but for the Shirouma snow samples. Error bars were calculated as above. For both the Hakusan and Shirouma samples, the BC mass concentration of smaller BC particles in the  $500 \text{ cm}^3$  bottle sample was less than those in the  $30 \text{ cm}^3$  bottle samples. However, the difference in the Shirouma samples was not significant considering the error range. On the other hand, the difference in the Hakusan samples was significantly smaller at diameters less than 850 nm in the  $500 \text{ cm}^3$  bottle. Figure 7 shows the size distribution of the median of the BC



Figure 5. Total mass concentration of BC in (a) Hakusan and (b) Shirouma snow samples melted in  $30 \text{ cm}^3$  bottles and a  $500 \text{ cm}^3$  bottle.



Figure 6. Size distribution of the BC mass concentration in (a) Hakusan and (b) Shirouma snow samples melted in  $30 \text{ cm}^3$  bottles and a  $500 \text{ cm}^3$  bottle.

mass ratio of the 500 cm<sup>3</sup> bottle sample to the 30 cm<sup>3</sup> bottle samples. Error bars show the minimum and maximum ratios. The ratio is systematically smaller than unity at BC diameters between 100 and 350 nm for the Shirouma sample; however, this was not significant. The ratio is significantly smaller than unity at BC diameters less than 850 nm for the Hakusan sample; however, there is no size dependence. These results suggest that reduction of the mass concentration for BC particles at diameters from 70 to 850 nm may occur during the melting process, especially for aged and/or more polluted snow. The dependence of this reduction on BC diameter was not evident unlike that in the melting temperature experiment.



Figure 7. Size distribution of the ratio of the BC mass concentration in the snow sample melted in the  $500 \text{ cm}^3$  bottle to that melted in the  $30 \text{ cm}^3$  bottles.

It is possible that this decrease in BC mass during snowmelt continues after melting is complete. We stored the melted Hakusan and Shirouma samples in the 500 cm<sup>3</sup> bottles (capped using teflon packing) in a refrigerator at 1°C for 4 days after the melting. The BC mass was measured repeatedly, at time intervals of about 24 h, to evaluate the influence of storage time. Before each measurement, the sample bottles were sonicated for 15 min and shaken just before the measurement. No significant difference in total mass and size distribution was found during storage, indicating that significant BC mass reduction does not occur in the liquid phase during post-melting storage at low temperature. This result is consistent with recent reports. For example, Lim et al. (2014) and Schwartz et al. (2013) showed that the size distribution of BC mass in melted snow samples is stable over 24 h. Wendl et al. (2014) showed that there is no significant BC loss over 18 days. Ohata et al. (2011) showed that the BC mass concentration in water solution did not significantly change over 50 days. It is suggested that BC mass reduction in our melting experiment occurred during the coexistence of the solid and liquid phases.

### 4 Conclusions

To enable a more accurate measurement of the amount of BC in snow, we evaluated the influence of temperature and time during the procedure for melting snow samples. Using various temperatures (melting temperature experiment) and time lengths (melting time experiment), we melted snow samples and measured BC mass concentration and size distribution in the melted samples using the SP2 method. Fresh (Shirouma) and aged (Hakusan) snow samples were used in these experiments, and each snow sample was stirred well before the melting process to reduce its inhomogeneity. To reduce the influence of sedimentation and adsorption on the bottle walls, we sonicated and shook all samples immediately prior to the measurements.

In the melting temperature experiment, snow samples were divided and melted at temperatures of 5, 20, and 70 °C in a water bath. For both Hakusan and Shirouma snow samples, the measured values of total BC mass concentration in the liquid melted at 70 °C were smaller than those in each liquid melted at 20 and 5 °C. The difference between 70 and 5 °C in the Hakusan sample was larger (46.4 %) than that in the Shirouma sample (8.0%), and this exceeded the random error range. This systematic decrease in the mass concentration at a melting temperature of 70 °C occurred for BC particles of less than 350 nm diameter. A similar decrease in the BC mass concentration was also found when the snow sample liquid melted at 5 °C was heated to 70 °C. A similar decrease in the BC mass was also found in samples stored in liquid form that were heated to 70 °C, indicating that the BC concentration decrease occurred not only during melting but also during liquid storage. When the liquid was stored at a room temperature or lower, no significant decrease was detected.

The effect of melting time was also tested using the Hakusan and Shirouma snow samples. Subsamples of approximately  $30 \text{ cm}^3$  took about 2 h to melt at 1 °C, whereas samples of approximately  $500 \text{ cm}^3$  took more than 6 h to melt. In the case of the Shirouma snow sample, measured values of the BC mass concentration of liquid melted in  $30 \text{ cm}^3$  and  $500 \text{ cm}^3$  bottles were nearly the same. In contrast, for the Hakusan snow sample, this value was significantly smaller in liquid melted in the  $500 \text{ cm}^3$  bottle (about 38.6 %) than that in the  $30 \text{ cm}^3$  bottles. The reduction rate in the  $500 \text{ cm}^3$  bottle was nearly constant at BC particle diameters of less than 850 nm. Significant BC mass reduction was not found in the liquid phase during storage at low temperature after melting, suggesting that the decrease occurred during the coexistence of the solid and liquid phases.

These experimental results in this study show that to reduce uncertainty in the measurement of BC mass in snow, samples should be melted at low temperature over a short time. The melting temperature experiment showed that it is possible that heating to a high temperature causes a significant reduction in BC both during the melting process and in the liquid phase. The melting time experiment showed that a slow reduction of BC concentration can occur even at low temperature during the coexistence of the solid and liquid phases. This suggests that it is inappropriate to melt a large volume snow sample all at once, as done in the lightabsorption and thermal-optical methods, because it would need much time for melting the whole sample. These results also suggest that the temperature history of the snowpack may influence BC mass and size distribution. We identified these influences using only two snow samples, suggesting that greater influences may occur in other snow samples. More systematic studies are necessary to quantify these influences as a function of temperature and snow condition to make the temperature and time conditions preferable for melting snow samples clear.

In this study, the mechanisms of BC decrease during snowmelt were not examined. The BC mass decrease may have been caused by the adsorption of BC onto the inner walls of the bottles in which the snow samples were melting. It is also possible that the agglomeration of smaller BC particles into supermicron BC, which was not measured here, caused the apparent decrease in BC mass (especially considering that the decrease of BC mass was more significant at smaller BC diameters in the melting temperature experiment). It should be noted that the size dependence of the BC decrease was different in the melting time and melting temperature experiments, suggesting that multiple mechanisms contribute to the BC decrease during snowmelt. Because BC reduction at 1 °C was found only during snowmelt, this may suggest that it occurs at the interface between the solid and liquid phases. It also should be noted that a significant decrease in BC in the melted snow occurred in the Hakusan snow samples, both in the melting temperature and melting time experiments. This indicates that the influence of melting temperature and melting time length on BC mass would be more significant in aged snow. The BC mass concentration in the Hakusan sample was much higher than that in the Shirouma sample. If the BC loss amount was the same in both samples, the influence would be larger in the Shirouma sample; however, the experimental results showed the opposite. We suspect that contaminating chemical substances may have played a role in this BC decrease. Schwarz et al. (2012) showed that the mixing of nitric acid into liquid samples causes a reduction in the mass fraction of supermicron BC particles. Further, Ohata et al. (2011) showed that the influence of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and Suwannee River fulvic acid (SRFA) was found when concentration of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or SRFA was considerably higher than their typical values in Tokyo. We measured the ion concentrations using ion chromatography (IC-2010, Tosoh Co., Ltd., Tokyo, Japan) in both snow samples. Concentrations of  $SO_4^{2-}$ ,  $NO_3^-$ ,  $Na^+$ , and  $Cl^-$  were significantly higher in the Hakusan sample than those in the Shirouma sample. The concentrations were, respectively, 0.85, 0.59, 0.74, and 0.80 ppm in the Hakusan sample and 0.15, 0.12, 0.47, and 0.29 ppm in the Shirouma sample. The level of such pollutants in snow may have contributed to the BC decrease with temperature. Other impurities, for example, dust in the snow, might also play a role via adsorption onto the dust surface. However, the influence of impurities is not yet fully understood; the influence of organic/inorganic substances as well as dust in the snow should be studied in more detail.

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