Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2015-324-AC1, 2016

Atmospheric Measurement Techniques Discussions

Interactive comment on "Influence of the melting temperature on the measurement of the mass concentration and size distribution of black carbon in snow" *by* T. Kinase et al.

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Received and published: 13 March 2016

The authors thank the reviewer for his/her comments helpful and useful for improving our manuscript. Replies to each comment are shown as the followings:

1.) Section 1, pg 3 paragraph starting on line 6 notes that previous studies have melted snow samples by heating in a microwave and by heating in a warm-water bath. The stated goal of the study is to test whether these approaches affect the size distribution and mass of BC in the melted snow sample. However, the study only tests for the effect of using different temperature water baths. There are no measurements of snow samples melted in a microwave oven. Tests would need to be done to see whether it matters, for example, if the snow is microwaved just long enough to melt the snow or

long enough to actually warm the snow melt water much above freezing. As no such studies are done, the authors should be clear that the results presented only apply to samples heated in a warm bath. This point should be made in the conclusions.

(Ans.1)In this study, we used water bath in order to specify the melting temperature because it is difficult to control and specify the temperature when we use a microwave oven. Prior to the experiments shown in this manuscript, as a trial, we measured and compared the BC mass concentrations in two parts of the Shirouma and Hakusan samples: ones had been melted at the room temperature and the others had been melted with a microwave oven. The BC mass concentrations in the samples melted with the microwave oven were significantly reduced. Thus we think the melting using a microwave oven heat the snow sample to high temperature enough to influence BC measurement. To show them, we have modified our manuscript.

2.) pg. 4, lines 26-27: "Inhomogeneity in each snow sample was estimated with the standard deviation of measurement results for these three bottled samples melted at a same temperature." Then again on pg 7, lines 7-8, it is noted that three samples are used to determine error bars. An n of 3 is not sufficient to calculate a standard deviation. An alternative possibility: Instead of showing error bars in the figures using standard deviations for 3 samples that the relevant figures simply show all three values as, e.g., dots. Similarly, on pg 6, line 23 and in Figure 5, it is not clear if the error bars are again standard deviations of n=3 tests. If so, again, I think these should not be presented as standard deviations but instead show all three data points, as well as the mean. If it's not from n=3 tests, what is it?

(Ans.2)We agree the reviewer's comment, and have modified the text and figures to adopt median with range between minimum and maximum values to the representative value and variability instead of the average and standard deviation.

3.) pg. 5, lines 15-17: snow melt-water samples were aerosolized with "a concentric pneumatic nebulizer (Marin-5, Cetac Technologies Inc., Omaha, Nebraska, USA), with a peristatic pump (REGRO Analog, ISMATEC SA., Feldeggstrasse, Glattbrugg, Switzerland)". Schwarz et al. (2012) and Lim et al. (2014) have demonstrated variable efficiencies for getting BC into the SP2 from liquid samples using different nebulizers. Was the efficiency of the system used here tested/quantified? If there is poor efficiency at larger sizes this could affect the conclusions about the change in total BC mass with heating temperature/rate. This is an important point that must be addressed, or at least acknowledged as a source of uncertainty in the study.

(Ans.3)As pointed out by the reviewer, it is possible that a poor efficiency may affect the result. Our experimental system was nearly identical with that in Mori et al. (2106), who estimated the extraction efficiency of a Marin-5 nebulizer to be mostly 50% in the diameter range of 200–2000 nm, and that the efficiency was stable. This value is much higher than other nebulizers, such as a U-5000AT and a collision-type nebulizer (Ohata et al., 2012, 2013; Schwartz et al., 2012). Although a APEX-Q nebulizer has higher efficiencies up to 72 % in the diameter range of 150–600 nm (Lim et al., 2014) the efficiency of this nebulizer depends on the diameter between 100 and 1000 nm (Wendl et al., 2014). Therefore, we adopted the Marin-5 nebulizer in this study, and we did intercomparison between our system and that in Mori et al. to show the efficiency of two systems agreed within their random error range. We have modified our manuscript to show these.

4.) Figure 1 & Table 1: I don't think Figure 1 is really needed. In the context of this study what is important is that the samples were of new and aged snow. No statement is made about how the geographic location of the samples might affect the study, so I would delete this map and just give the lat/lon of the sample locations for the interested reader. This is currently done in Table 1. The information contained in Table 1 is mostly also in the text. I think this information should be provided either in a table or in the text, but not both, given the brevity of this study/paper. My suggestion would be to delete both Figure 1 and Table 1, and simply include the relevant information in the text.

(Ans.4)Following the reviewer's comment, and have modified the manuscript.

5.) It is not at all clear what the SO4, NO3 and other chemical analyses add to this study. They do not provide any information regarding whether or why the BC concentrations and size distributions are affected by the heating temperature or melt time. All reference to these analyses should be removed from the paper.

(Ans.5)As pointed out in this reviewer's comment, the ion measurement results were not used in the interpretation of the experimental results. We have modified the manuscript to remove information on ion analysis from the snow sample section. However, some studies showed possibilities that ion components may influence the BC measurement and BC size distribution, and the ion measurement showed that amounts of some ions were clearly different in the two samples. We also modified the manuscript to show the possibility that the impurities in snow may be significant in the conclusion section.

Technical corrections: Thanking these reviewer's technical comments, we have modified the manuscript.

6.) Section 1: References to Bond et al. (2012) need to be corrected to Bond et al. (2013). This correction also needs to be made in the References list. 7.) pg. 2, lines 15-16: Bond et al. (2013) also provided a central estimate of 0.04 W/m2, not just a min/max. 8.) pg. 4-5, Section 2.2: Multiple references to "grass bottles" need to be corrected to "glass bottles" 9.) pg 6, lines 14-16: "Figure 4 shows the size distributions of the 30 BC mass ratio of the 70 _C melting sample to the 5 _C melting sample, indicating that the ratio systematically decreases with the decrease of the BC particle diameter." Suggest rewording to: "Figure 4 shows the ratio of BC mass in the samples heated to 70degC to those heated to 5degC, as a function of BC size. This shows that the ratio is lower for smaller particle sizes". 12.) pg. 8, lines 28-30: "In the melting time experiment, the Hakusan and Shirouma snow samples in the 30 cm3 bottles were melted for about 2 hours, and those in the 500 cm3 bottles were melted for more than 6 hours." This wording, and the discussion that follows, implies that this study was about the bottle size, not the amount of time it takes to melt smaller vs. larger snow samples. This sentence should be reworded, e.g. to: “The effect of melting time was also tested using the Hakusan and Shirouma snow samples. Sub-samples of each of approximately 30 cm3 took about 2 hours to melt at 1deg C, whereas samples of approximately 500 cm3 took about 6 hours to melt.” 10.) pg. 6, lines 18-20: “: : :considering that the Hakusan sample was more aged and that it contained more pollutants such as SO42- and NO3- in comparison with the Shirouma sample.” The Hakusan sample didn’t only have higher SO4 an NO3 concentrations – it also has more than double the BC concentrations, as shown in Figure 3. Why not just state this directly?

(Ans.6)As pointed out in this reviewer’s comment, higher BC concentration indicated the Hakusan snow sample was more polluted than the Shirouma sample. We have modified the manuscript to show it, and that the difference in the BC measurement influences between the two samples could not explain only by the BC amount itself.

11.) pg. 8, lines 25-27: “These results indicate that the decrease by the heating to high temperature can occur not only during the snow melting but also during the storage in the liquid phase.” This statement needs to be modified: the decrease in mass was not for samples that were simply stored in liquid form, but that were heated to 70deg C (which is very warm, and so not a temperature samples would encounter simply by being stored at e.g. room temperature).

(Ans.7)The experiment that the heating the liquid to 70C indicate not only the influence of high temperature which the liquid would not encounter by storage at room temperature, but also that the BC decrease and size distribution modification under a high temperature could occur in the liquid phase. We have modified the manuscript for indicating them more clearly.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2015-324, 2016.

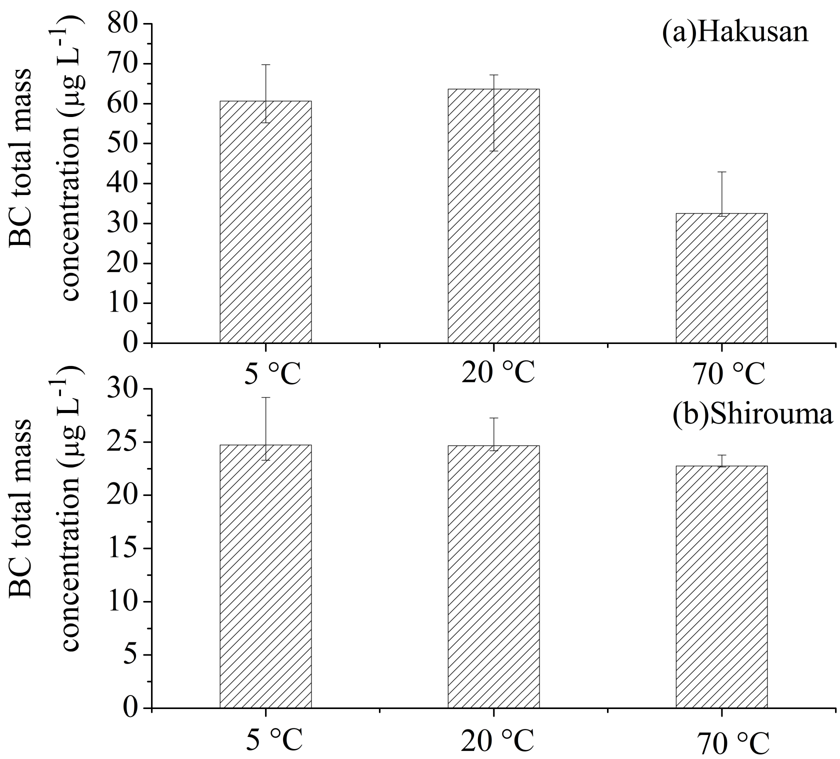
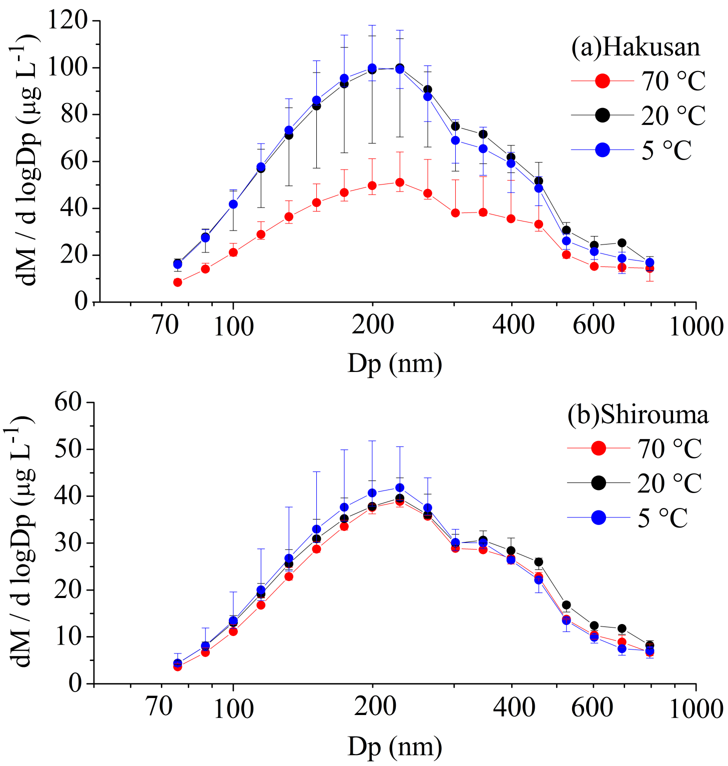
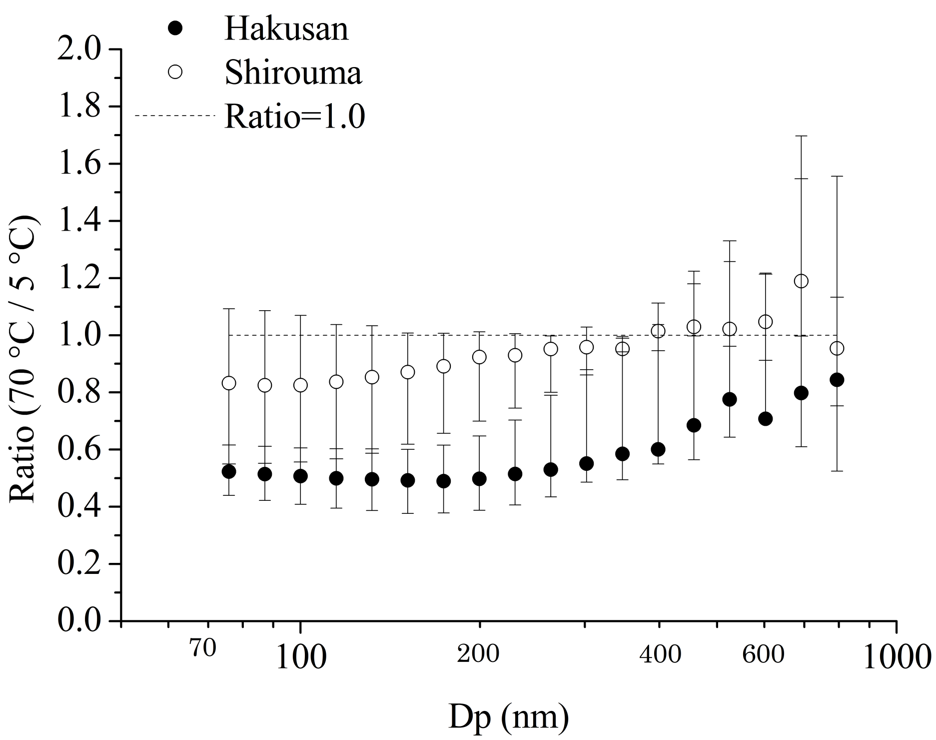


Fig. 1.



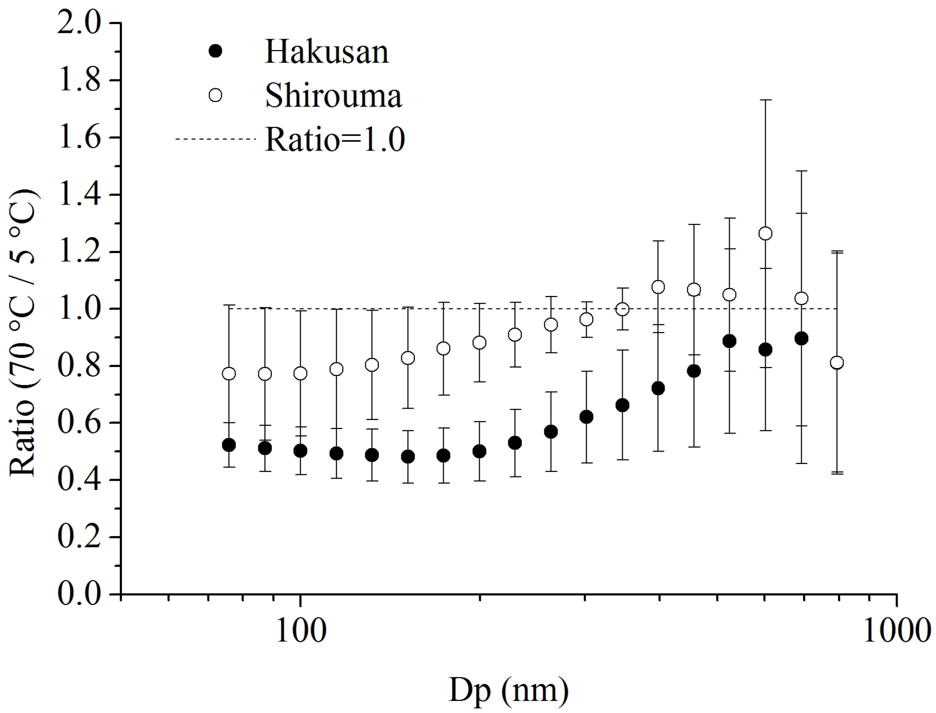
(a)Hakusan
70 °C
20 °C
5 °C
dM / d logDp (μg L-1)
Dp (nm)
(b)Shirouma
70 °C
20 °C
5 °C
dM / d logDp (μg L-1)
Dp (nm)

Fig. 2.



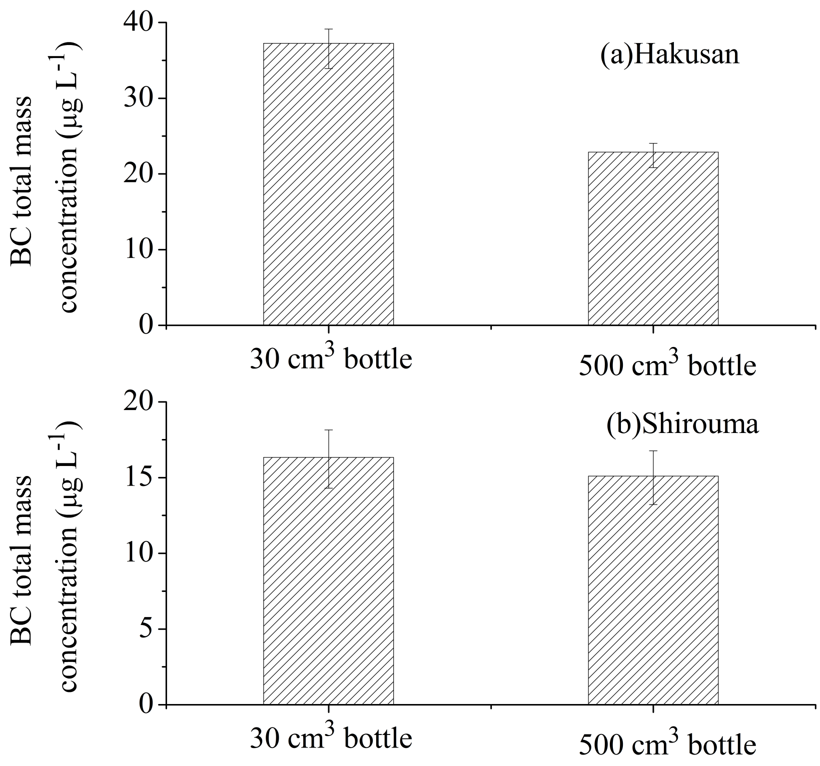
Hakusan
Shirouma
Ratio=1.0
Ratio (70 °C / 5 °C)
Dp (nm)

Fig. 3.



2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Ratio (70 °C / 5 °C)
Hakusan
Shirouma
Ratio=1.0
100
1000
Dp (nm)

Fig. 4.



40
30
20
10
0
BC total mass concentration (µg L-1)
(a)Hakusan
30 cm3 bottle
500 cm3 bottle
20
15
10
5
0
BC total mass concentration (µg L-1)
(b)Shirouma
30 cm3 bottle
500 cm3 bottle

Fig. 5.

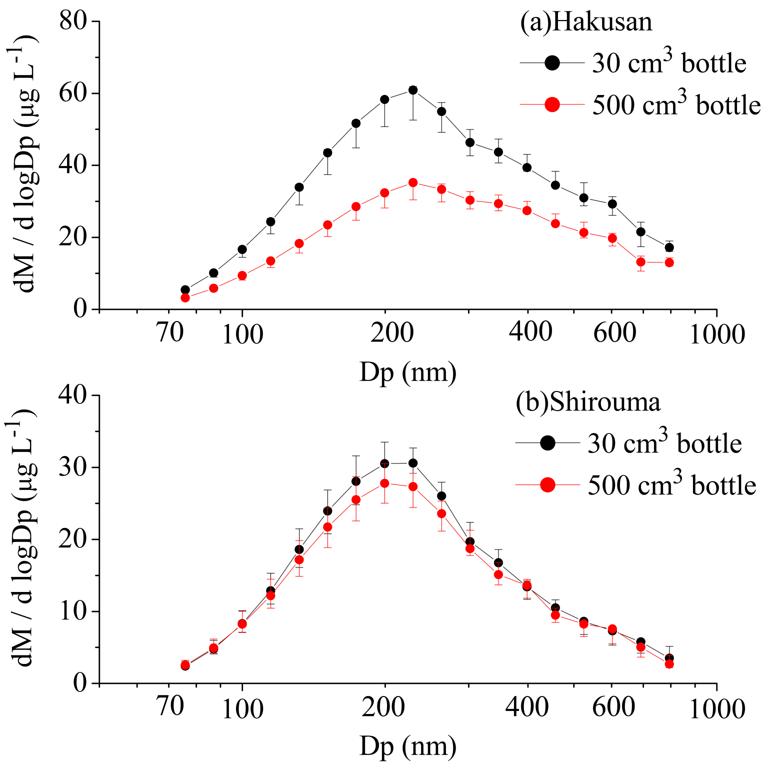

(a)Hakusan
30 cm³ bottle
500 cm³ bottle
dM / d logDp (μg L⁻¹)
Dp (nm)
(b)Shirouma
30 cm³ bottle
500 cm³ bottle
dM / d logDp (μg L⁻¹)
Dp (nm)

Fig. 6.

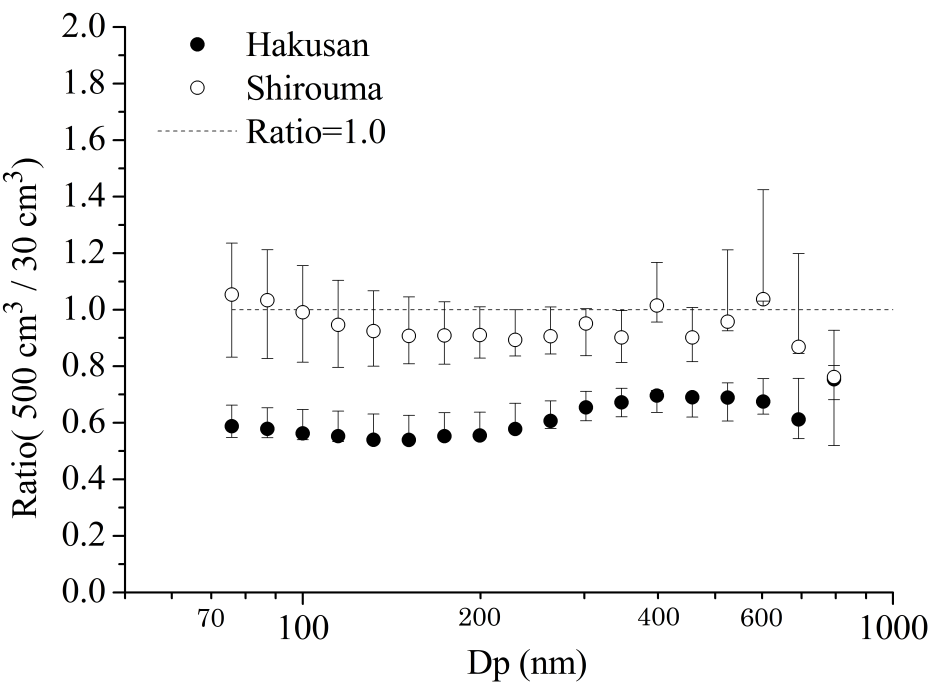

Hakusan
Shirouma
Ratio=1.0
Ratio(500 cm³ / 30 cm³)
Dp (nm)

Fig. 7.