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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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The authors thank the detailed comments useful for improving our manuscript. Replies to each comment are shown as the followings:

(1) What standard material has been used for calibration of the results? The choice of standard material has been discussed in depth previously (Wendl et al., AMT, 2014). This study definitely should be cited in the manuscript since standardization and calibration is an important issue when analyzing BC in snow and ice. I am aware that the description of standard used in the study discussed here can be found in the literature cited (method section) but I do not think this is sufficient. Other related and important questions are: How was the calibration performed? Once a day, weekly: :

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Discussion paper

:? Most importantly, do the discussed effects also show up in standard samples (e.g. did you analyze standard solutions which were treated similar to the individual sets discussed for comparison/as a reference)? How can the observed differences/similarities between standard and sample behavior be explained?

(Ans.1)The experiment system used in this study was nearly identical with that in Mori et al. (2016) who conducted experiments to measure system efficiencies to evaluate absolute sensitivity using model BC particles. We had done the intercomparison of our system with that in Mori et al, (2016). before our study, and found the efficiency and sensitivity of the two systems agreed within their random error range, indicating that sensitivity of our system was also nearly identical with that in Mori et al. (2016). It should be noted that results in this study depends on only relative values, not on the absolute values. Therefore, stability of the system sensitivity is much more significant in this study, rather than absolute calibration. A temporal drift of the system sensitivity during the experiments had been examined by measuring some samples twice (time interval was 1 to 24 hours) in each experiment. The difference of the first and second measurement results was 2.1(0.4 to 5.1)% for 30 cm3 bottle samples, and it was 7.8% for a 500 cm3 sample, indicating that the temporal drift of the system sensitivity was less than or comparable with random errors. To show this result, we have modified the manuscript.

(2) It is unclear if the containers containing the snow samples were kept closed during melting. I assume they were, but still it should be described clearly in the manuscript. This is important since if they were open, effects due to evaporation cannot be excluded. Evaporation would likely result in an increase in concentration but might not be detected because being masked by other, larger effects having an opposite direction (i.e. resulting in decreased concentration) which then might be underestimated as a consequence.

(Ans.2)In order to avoid the contamination and evaporation effect as pointed out with this comment, we sealed containers with bottle cap during the melting, the storage and

the sonication. To show this, we have modified the manuscript.

(3) It remains unclear if once the snow samples were melted they were sonicated in the containers used for melting or if aliquots were first taken, transferred to new containers and sonicated afterwards. If the latter is the case, any wall effects happening in the containers used to melt the samples will not be considered even though they are likely significant (decrease in the observed concentrations, likely size dependent) and would allow a different interpretation of the derived results. This should be clarified and also be discussed if necessary based on the procedure used. It should also be described if the samples were stirred prior/during sonication and/or analysis. This is of particular importance to know for the sample in the 500 cm3 bottle where sedimentation might have a significant effect resulting in the difference observed compared to the 30 cm3 containers.

(Ans.3)After aliquots of the snow samples were taken in a bottle, the melting, the storage, the sonication and the BC analysis of the sample were conducted in the same bottle without a sample transfer to avoid loss or contamination. All sample bottles including 500 cm3 bottles were stirred by shaking them following the sonication just before the analysis. To show these clearly, we have modified the manuscript. In addition, we analyzed 500cm3 samples several time during the 4 days storage (shown in section 3.2), and no significant decrease was found during the storage. These showed that sedimentation effect was insignificant for our experiments.

(4) Further, what was the time passing between sonication and analysis of samples? Was it similar for all samples? Again this information is relevant because wall effects happening between sonication and analysis can result in the observation of decreased concentrations. If there is a large deviation in the time passed between time of sonication and analysis for the individual sample such wall effects might contribute significantly to any observed decrease in concentrations. Has this been tested? This should be clarified and discussed.

(Ans.4)The time passing between the sonication followed by the bottle shake and the analysis of the sample fluctuated between 1 and 10 minutes. We have not examined this time passing effect. However, as already written, some samples measured twice showed the difference of the two measurements, where the above time passing were different, was less than or comparable with random errors. Thus the influence of the time passing less than 10 minutes was less than or included in the random error.

I am fully aware that the author's might have omitted this degree of detailed information because some of it seems rather trivial. Nevertheless, I strongly believe it needs to be addressed carefully for the reasons pointed out. More general and not considering the above, the fact that only two different samples were used for the investigations seems limiting to reach sound conclusions. Are the samples studied here representative? Their concentration is not so different after all. The question remains if the investigated effects are also significant for samples with a much different BC particle size distribution or much lower/higher concentrations? In fact, even for the results presented here, the significance of the described effects may be questioned regarding the uncertainties (see e.g. Fig. 2) and the fact that the number of samples in each set is rather low (n = 3 for 30 cm3 containers and n = 1 for 500 cm3). Is this also the reason why there is not given a clear recommendation what temperature and melting time should preferentially be used in order to obtain the most reliable results?

(Ans.5)We think that it is not very significant that the samples used in this study were representative. (It is not reasonable to think that only two samples represent all snow samples.) Our experiments, which detected the influence of the melting temperature and time adopting only the two samples, strongly suggest that similar influence could occur in more various snow samples. The difference in the two samples indicated that these influences depends on the snow conditions, and this dependence makes difficult to determine clear, preferable conditions for the melting snow sample, as pointed out by this comment. The study enable us notice of significance of the lower melting temperature and shorter melting time for accurate measurement of BC in snow, and

recommends us to test that the adopted melting temperature and time do not affect the measurement for the snow sample. To show this, we have modified the manuscript.

Other remarks: p.7, line 28 – p.8, line 6: Were the stored samples sonicated once again prior to each subsequent analysis? If this is not the case and still no effect of storage time was observed these results would be very different from what has been described in Wendl et al. (2014). Please comment/discuss.

p.2, line 23 ff.: If soot is measured with a thermal optical technique, it is referred to as EC (elemental carbon), not BC. The authors should also cite some of the pioneer work of such measurements in snow and ice: Lavanchy, V.M.H., Gäggeler, H.W., Schotterer, U., Schwikowski, M. and Baltensperger, U. (1999). Historical record of carbonaceous particle concentrations from a European high-alpine glacier (Colle Gnifetti, Switzerland). Journal of Geophysical Research 104: doi: 10.1029/1999JD900408. issn: 0148-0227. Jenk, T. M., Szidat, S., Schwikowski, M., Gaeggeler, H. W., Bruetsch, S., Wacker, L., Synal, H. A., and Saurer, M.: Radiocarbon analysis in an Alpine ice core: record of anthropogenic and biogenic contributions to carbonaceous aerosols in the past (1650- 1940), Atmospheric Chemistry and Physics, 6, 5381-5390, 2006.

p.2, line 28 ff.: To be complete and because it discusses a lot of similar and complementary issues to the ones addressed in this paper, Wendl et al. which was actually published in the same Journal as this study(!) needs to be cited and should also be discussed (see above): Wendl, I. A., Menking, J. A., Färber, R., Gysel, M., Kaspari, S. D., Laborde, M. J. G., and Schwikowski, M.: Optimized Method for Black Carbon Analysis in Ice and Snow Using the Single Particle Soot Photometer, Atmos. Meas. Tech., 7, 2667–2681, 2014, doi:10.5194/amt-7-2667-2014.

Thanking these comments, we have modified the manuscript to quote these references.

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