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Interactive Comment

Interactive comment on "Measuring the specific surface area of snow with X-ray tomography and gas adsorption: comparison and implications for surface smoothness" *by* M. Kerbrat et al.

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This paper compares 2 methods to measure the specific surface area (SSA) of snow : methane adsorption at 77 K and X-ray tomography. Since SSA is a crucial physical variable for porous media and since snow SSA is delicate to measure, a paper that compares 2 potentially accurate techniques is welcome and timely. This study appears to have been conducted carefully by an experienced team and leads to conclusions that are in general reasonable and well justified. I would like to see this paper published in ACP but I recommend significant, although perhaps not major, changes on essentially 2 points before publication.

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1- Since the purpose of the paper is the comparison of 2 methods, it is essential that the accuracy of both methods be evaluated in detail. My feeling is that insufficient efforts have been devoted to this aspect. Regarding the adsorption method, Legagneux et al. (2002) and Domine et al. (2007) have discussed measurement errors in great detail and the authors should remind briefly the main issues raised in those papers. I am also a bit surprised that the authors did not test the reproducibility of this method using their apparatus, as done by Legagneux et al. (2002). It would be reasonable to perform extra measurements to address this point. This is not an unreasonable request, as all that needs to be done is repeat the measurement on a given snow sample, which can probably easily be obtained near Davos now or in the very near future. The authors should also discuss in more detail the impact of the correction for the adsorption of CH4 on the container walls, as a function of the SSA of the sample, as done by Domine et al. (2007). Regarding the tomography method, I am surprised that the error is essentially the standard deviation of a series of measurements. From my understanding, deriving SSA from tomography images is not always trivial, and the algorithms used all produce a certain error that need to be estimated in more detail. The authors may in particular read Flin et al. (2005) IEEE Trans. Image Process., 14, 585. Another issue with the tomography method is that one measurement takes 3 hours at -15°C, during which fresh snow will evolve, especially if a series of measurements are made. It must be stressed that this is an extra reason why tomography may not be suitable for fresh snow, at least with the machine used. In summary for this point, I recommend producing a more detailed analysis of sources of errors, and of reporting those in tables and figures.

2- The authors conclude that both method yield similar results, within 3%, and this is a very interesting conclusion. Since the spatial resolution of the tomographic method is 30 μm, the authors conclude that snow surfaces are smooth at the 30 μm scale and they present some theoretical developments to explain that. They also conclude that the tomographic method is accurate for snow with SSA$<$700 cm2/g, and this is most snows since high SSA snows are very rare. I believe some of these statements are not totally accurate and may be misleading for the ACP reader who is not a snow special-

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ist. First of all, there are many scanning electron microscopy (SEM) studies of snow crystals that show that structures smaller that 30 μm are common. The authors cite Domine et al. (2003) but they should also cite the nice work of Wergin's group, e.g. in Scanning (2002)24, 247 and (2003) 25, 121. Micrographs by both groups do show structures $<$30μm, almost always present in fresh snow, but also not rarely present on metamorphosed snow. They may also refer to optical pictures, for example Fig 5 of Cabanes et al. (2002) Atmos. Environm., 36, 2767. Second, I am not convinced by the usefulness of their theoretical developments (section 4.2 and appendix). The underlying assumption is that the snow evolved under isothermal conditions, but this is rare for surface snow and the physics of snow evolution will then be fundamentally different. For example, Domine et al. (2003) study SSA and micromorphological changes of a recent snowfall evolving naturally under a temperature gradient. The snow does not evolve at all according to a scenario similar to the Fig 5 of Kerbrat et al. On the contrary, under a high T gradient, the growth mechanism are not ruled by Kelvin's law at all and Fig 3 of Domine et al. clearly show that structures thinner than 30 μm are then formed, in opposition to the hypothesis by Kerbrat et al. I Think that section 4.2 and the appendix are really superfluous. If anything, the author can very quickly examine the case where a sphere or a cylinder (see Fig 5 a of Cabanes et al. 2002) sublimates in a PH2O determined by flat surfaces under isothermal conditions, and go on to explain that in nature, things will often be quite different. Third, I think that stating that snows with high SSA are rare is not correct. The latest compilation of Domine et al. (2007) shows that 31 out of 64 fresh snow samples sampled at T$>$0°C (almost 50% !!) have a SSA $>$700 cm2/g. Thus, the tomography method will be inapplicable to at least half of the fresh snow samples, and this limitation must be clearly mentioned. If all the data of Domine et al. are considered, then 37 out of 345 samples have a SSA$>$700cm2/g, i.e. 11%, far from negligible. For many purposes, such as atmospheric chemistry and albedo, surface snow layers are the most important. In the Alpine areas (stressed by the authors), snowfalls are frequent (does not it snow dozens of time a year around Davos ??) so that surface snow will have a SSA$>$700 cm2/g a significant fraction of the time.

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Therefore, I think it would be fair to acknowledge that the tomographic method may underestimate (at least with the instrument used) snow SSA for a significant fraction of snow samples found in Alpine areas. This of course does not mean that the method is not valuable, but its limitations must be clearly stated to the reader. In summary for this point, I recommend acknowledging that snow samples with structures $<30\mu$m are in fact frequent, and that such structures can actually form during metamorphism. As a consequence, tomography may underestimate SSA in a significant number of cases. The theoretical developments "explaining" snow surface smoothness is somewhat disconnected from reality and should be significantly shortened or, better, deleted, and a brief discussion of natural conditions would be in order if the section is kept (in a much shortened form).

Minor points

Abstract Change abstract. It is often not true that natural snow is smooth at the 30μm scale. A voxel size of 10μm will not be sufficient to capture all structural features of about half of fresh snow samples. Using methane adsorption, it will be difficult, but not impossible, to measure thin layers. Cabanes et al. (2002) monitored layers <5 mm thick in the arctic by scraping surface snow into the measurement container. It is clear, however, that tomography is better suited to measure the SSA of thin layers <700 cm2/g, and this needs to be mentioned.

p 10288 l 23 : Cite Taillandier et al. (2007) here also.

P 10290 l 2. These diamond dust crystals are moreover probably hollow, bringing their SSA to about 5000-6000 cm2/g. See e.g. Satow (1983) "Observations on the shapes of snow crystals in the summer season in Mizuho plateau, Antarctica" 5th symposium on polar meteorology and glaciology, NIPR, Japan, 103-107.

P 10290, l 4-5. The reason why those very high SSA values are wrong has been detailed in Legagneux et al. (2002). This is certainly because of the formation of ice crystals with very high SSA during an inadequate cooling procedure. Please mention

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this briefly to back up the claim that early values were too high.

P 10290 l 9. SSA values around 20 cm2/g are obtained on aged REFROZEN snows (Domine et al., 2007).

P 10292 l 19-20 Cite Matzl and Schneebeli (2006) here.

P 10296 l 20-21 At this stage, one would have expected a more detailed and thorough evaluation of error sources, either in the experimental section (better) or here. This sentence is not sufficient.

P 10297 l 6-8. Same comment, more details on errors.

P 10300, l 10-12. For the SSA of rounded grains, please use instead the more complete and corrected data of Domine et al. (2007).

P 10301, l 8-11. Mentioning that "only 3% of the 340 measurements gathered in Domine et al. (2007) are greater than 1000 cm2/g" is true but misleading. The interesting threshold for this discussion is 700 cm2/g. It would be fair to say that problems will be encountered with the tomographic method more frequently than 3% of the time, as detailed in my above detailed comment. It would be informative to the reader to know that the resolution of the tomographic method used will not be sufficient for half of the precipitated snow samples. (Incidentally the number of measurements performed by Domine et al. is 345, not 340).

P 10305, section 4.3. Extra practical considerations must be considered. The adsorption technique requires liquid N2, not a simple requirements in many field studies. Does the microtomographic method require good electrical grounding ? If this is the case, then this may be a problem on ice caps, where a good ground is often difficult to obtain. The potential of optical methods has already been stressed by Domine et al. (2006) CRST, 46, 60. It would be fair to mention that the accuracy of the optical method of Matzl and Schneebeli (2006) needs to be established in more detail. Painter et al. (2007) do not measure SSA. They measure "optical grain size" and the relation

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to SSA is not sufficiently well established to equate it to SSA.

P 10305, section 4.4. It is true that adsorption measurements require larger sampling volumes. However, loose snow can be sampled by scraping thin layers into the container. Coring is not always necessary. Thin layer can therefore also be measured.

P 10306, l 24-25. A sweeping statement such as "our measurements prove that the ice surface in Alpine snow is smooth to a scale of 30 μm few hours after precipitation" is exaggerated and must be removed. Precipitating dendritic snow, the most frequent type of falling snow in the Alps, very often has an SSA$>$1000 cm2/g and its SSA will remain $>$700 cm2/g for more than a few hours at most Alpine temperatures.

Fig 4. How about an inset detailing the errors for the low SSA values ?? After all, most of this paper is about comparison, precision and errors.

Following some of the above comments, Fig 5 and Fig 6 can be deleted.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 10287, 2007.