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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurement of the ambient organic aerosol volatility distribution: application during the Finokalia Aerosol Measurement Experiment (FAME-2008)" *by* B. H. Lee et al.

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I have a short comment on the estimation of CE from SMPS-AMS comparisons for the thermally-denuded (heated) aerosols. The authors ignore the potential effect of non-spherical particles in the SMPS volume estimation. As more volatile components evaporate, it is also likely that the particles become less spherical. A small change in the dynamic shape factor (X) of the particles can cause an overestimation of the particle volume calculated from the SMPS number distribution. For example using equation 25 of DeCarlo et al. (2004) we can estimate that a very small change of X from 1.00

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to 1.03 would cause an overestimation of particle volume (calculated from the mobility diameter) of about 10 percent, comparable to the change which is attributed to a change in AMS CE in this paper. Such shape factors are common (e.g. recent papers by Zelenyuk and co-workers), and for comparison soot particles can have X up to 3. Due to this effect one should be careful to remember that the volume estimated from the SMPS is not the true particle volume but rather an "apparent volume." I am not aware of any evidence to rule out a small change of the shape factor of the order of 0.03 upon particle heating by 100C. The topic of possible changes of AMS CE upon heating (first discussed by the TD-AMS publications of Huffman et al., to my knowledge) deserves further study, but a verification of particle sphericity or a measurement of the dynamic shape factor is necessary for a quantitative characterization with an SMPS-AMS method. The authors could have used the beam width probe of Huffman et al. (AST 2005) to probe possible changes in particle shape, and such experimental characterization is recommended for future studies. In the absence of evidence of lack of small changes in X, the conclusions about a quantitative measurement of the change of CE are not warranted. As an unrelated point, I was very surprised to see no mention of the recent paper by Cappa and Jimenez (2010) who also present volatility distributions derived from TD-AMS measurements over a wider range of temperatures than in this study. A comparison of the methods and results of that and this study would seem necessary.

The comment is related to the estimation of the collection efficiency for the thermodenuded particles and the possibility that our results are due to a small change in the shape of the particles. We should first clarify that the algorithm of Kostenidou et al. (2008) used for the calculation of the collection efficiency uses the complete volume size distribution from the SMPS and not just the total volume concentration. As a result it is sensitive to the details of that function (something that can be seen for example in Figures 4 and 5 of the paper) and not just to its integral. If a particle of a given mass is not spherical then not only its volume changes but also its aerodynamic diameter. This results in shifting of the size distribution both horizontally and vertically during the

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matching and our algorithm is quite sensitive to these changes. This issue has been discussed by Kostenidou et al. (2008) in their analysis of the sensitivity of the algorithm to shape changes.

We repeated the analysis for different values of the shape factor including the 1.03. The error score of the algorithm increases significantly if a non-unity value of the shape factor is used. The minimum error corresponds to X=1 and the error at X=1.03 is double that of the X=1 case. Even more importantly, when a value of 1.03 was used the same collection efficiency was calculated for the thermodenuded particles. The estimated density on the other hand increases significantly for X exceeding unity. These results support the robustness of the calculated collection efficiency.

Our consistency checks for the complete number and volume size distributions shown in Figures 4 and 5 are also inconsistent with a significant change in the shape of the particles. Using a non-unity shape factor results in non-negligible change of the agreement of the corresponding distributions.

Finally, for a physical explanation the thermodenuded particles during FAME-08 were approximately 85 percent ammonium sulfate and bisulfate and only 15 percent organic. Given our knowledge of the shape factors of these salts it is difficult to justify a significant deviation from the spherical shape.

So while the concern about a potential change in shape is valid, we believe that it is unlikely that it can explain all the change in the collection efficiency under the conditions of this study. These were high-aged sulfate dominated particles and had no resemblance to fresh soot particles. We have added a paragraph discussing this potential alternative explanation of the observed changes together with a sensitivity study and the corresponding references. We do agree that this is an issue that deserves attention in future studies of more complex aerosol.

A discussion of the Cappa and Jimenez (2010) paper that was published after this paper had been completed has been added to the introduction and also the conclusions

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of the paper.

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