

Response to Reviewer 2

We thank the referee for their comments and suggestions. Our responses to the comments (along with the original comments in italic) are given below

The authors define correlations between HTDMA and AMS data. SOA hygroscopicity strongly correlates with the relative abundance of the ion signal m/z 44. The analysis is applied to ambient measurements at Jungfraujoch and Mexico City and similar results are found. An empirical correlation between, the hygroscopicity parameter, K_{org} and f_{44} is given. The work provides an approximation that could be introduced as a simplification in the parametrization of hygroscopic OA in atmospheric models. The subject matter is relevant and of interest to the larger scientific community. The provided measurements and correlations will add to the existing body of work on atmospheric aerosol and their subsequent influence on particle hygroscopicity. The document is easy to read and the following concerns will clarify points in the paper.

CONCERNS:

The reviewer cautions that the application of the correlation (as published) is very narrow. The authors restrict their data set such that $\epsilon_{org} > 80\%$ for nonhygroscopic org and $>25\%$ for hygroscopic organic (P19319). As presented, the work appears applicable to a global data set on organic aerosol, when in fact it is not. The authors even suggest a multiple linear regression of f_x for a more accurate correlation. This should be emphasized in the abstract.

The restriction $\epsilon_{org} > 80\%$ was necessary because the overall GF (GF_{total}) becomes insensitive to GF_{org} (κ_{org}) at smaller ϵ_{org} , and thus inferring κ_{org} from GF_{total} becomes an ill-posed problem. Although the results shown are only based on data with $\epsilon_{org} > 80\%$. We feel the κ_{org} obtained with this approach is still generally applicable because it allows accurate predictions of GF_{total} also for smaller ϵ_{org} , even if the κ_{org} would depend to some degree on ϵ_{org} .

Why is the chamber RH set to 50%? What is the relevance of this value? Will the effect of humidity change hygroscopicity values for the SOA produced?

50% RH has been chosen as it represents an average value of the atmosphere and for technical reason (we could not operate the chamber to very high RH $>90\%$ where the risk of condensation on the wall is too high). We do not expect large changes of hygroscopicity when the reaction is performed at a different relative humidity. For the aromatics Cocker et al. (Atmos. Environ 35 (2001) 6073–6085) have shown no such effect. In the case of α -pinene we performed a photooxidation experiment where ozonolysis plays only a minor role. Ozonolysis products depend somewhat on the water vapour concentration.

P19314. L17. How do you know the organic precursor was not completely reacted?

A PTRMS was measuring the decay of the different organic precursors. We added on page 19314, L5 the sentence: "The precursors were measured with a proton-transfer-reaction mass spectrometer."

P19314. L24. "tetramethylethylene: : reacts with ozone and produces OH in high yields" Were OH concentrations measured? In addition, "HONO was continuously replenished to keep a semi-constant OH source." How was this verified? Or was there excess HONO in the system?

The yield of OH from TME ozonolysis is known. The OH radical concentration was also determined indirectly from the decay of α -pinene reaction products by PTR-MS. There was about 10 ppb of HONO in the system and HONO concentration was also measured by a LOPAP instrument.

P19318. L9. How do you know that “the aerosol was mostly neutralized with ammonium”? It is not clear to the reader.

This is known by comparing the AMS or filter sample anions and cations. The two references for these two sites have been added at the end of the sentence.

DeCarlo, P. F., Dunlea, E. J., Kimmel, J. R., Aiken, A. C., Sueper, D., Crouse, J., Wennberg, P. O., Emmons, L., Shinozuka, Y., Clarke, A., Zhou, J., Tomlinson, J., Collins, D. R., Knapp, D., Weinheimer, A. J., Montzka, D. D., Campos, T., and Jimenez, J. L.: Fast airborne aerosol size and chemistry measurements above Mexico City and Central Mexico during the MILAGRO campaign, *Atmos. Chem. Phys.*, 8, 4027-4048, doi:10.5194/acp-8-4027-2008, 2008.

Cozic, J., Verheggen, B., Weingartner, E., Crosier, J., Bower, K. N., Flynn, M., Coe, H., Henning, S., Steinbacher, M., Henne, S., Collaud Coen, M., Petzold, A., and Baltensperger, U.: Chemical composition of free tropospheric aerosol for PM1 and coarse mode at the high alpine site Jungfraujoch, *Atmos. Chem. Phys.*, 8, 407–423, 2008, <http://www.atmos-chem-phys.net/8/407/2008/>

P19318. L24. How do you know that the aerosol in Mexico was an external mixture? Is there any other evidence for this (besides GF measurement)?

The HTDMA did measure a distribution of different GFs while selecting one dry size, which is a clear indication of external mixture. There is no other evidence beside this, as the AMS is measuring the aerosol bulk.

Why do the authors present GF data at 0.95% RH?

The data presented in Fig 7 are with the axis at 0.85 and 0.95 aw. These two aw correspond to the two different setup of the HTDMA as described in P 19316, lines 4-8.

P19322 L 28. Define acronym BC.

Black carbon has been added.

Figure 3. Why do data points scatter as the experimental time increases? Does the greater variation in chemical composition correspond to a greater variation in GF? And Kappa? Please discuss

After nucleation the particle concentration decreases over time due to wall losses and becomes rather small after 20 hours. At long experimental times the AMS data have higher uncertainty with lower mass loadings. Since the HTDMA data is a number based measurement, and sufficient numbers of particles were still present at longer times, the uncertainty does not increase as much..

P19313 L13. Replace “and complemented” with “and are complemented”

Done

P19320. L19. Text reads “all smog chamber data” but figure 5 says “all photooxidation experiments”. Which one is true? How will experiments 7 and 8 (ozonolysis) modify the figure and argument?

We thank the reviewer for pointing out this typo. The text and the figure 5 caption are correct. The legend inside the figure 5 has been corrected: "all photooxidation experiment combined" has been replaced by "all chamber data combined"

P19321. L4. How does f_{43} have "a strong correlation with GF"? $f_{43} > 15\%$ of what? R is at most 0.9 for α -pinene but in Figure 5, f_{43} is not shown. Also please define strong correlation for the reader. Is this what is meant by significance? What is this statistical significance parameter? It is not defined. How many standard deviations from the distribution are assumed? Is it 3sigma, 4 sigma, etc?

The unclear text "From a chemical point of view f_{43} also has a strong correlation with GF ($f_{43} > 15\%$) from SOA generated in smog chambers and could be also considered as an additional parameter to improve the correlation." has been replaced by "From a chemical point of view f_{43} ($f_{43} > 15\%$) could also have a correlation with GF from SOA generated in smog chambers and could also be considered as an additional parameter to improve the correlation. However, as shown in Figure 4, f_{43} and GF can be either correlated or anti-correlated depending of the precursor." Additionally, in Figure 5, f_{43} is not shown as the significance alpha is higher than 0.05 (alpha=0.11 and $R=-0.078$).

Fig 4. Which α -pinene experiment is presented? Is it a photo oxidation or ozonolysis? Is there a difference in correlations for the different systems?

In Figure 4 only photo-oxidation experiments are compared and presented. We changed the Figure caption from: "for the smog chamber data with different precursors" to "for the photooxidation experiments with different precursors"