

We are very grateful to the reviewer for the constructive suggestions and for the proposed corrections to improve our paper. Here, all the issues raised had been addressed. Accordingly, the manuscript had been modified.

Detailed comments

Experimental methods

Page 7651; line 1: where do the cloud events take place, on the measurement site or at the mountain top?. If on measurement site, is there an adapted inlet?

Response: The cloud took place at the mountain top. There is no cloud at the measurement site mentioned in this study. We added “Cloud event occurred at Schmücke research station” into the manuscript.

Line 5 to 9 : to me these lines are contradictory : density is calculated using AMS size distributions, but the AMS size distribution are calculated using a supposed aerosol density ?

Response:

The particle density (ρ_p) is calculated by comparison of bulk particle mass concentration (PM1) derived from AMS measurements and total particle volume concentration derived from SMPS measurements. The particle size is not involved into this calculation.

The particle mass size distribution measured by AMS is represented as vacuum aerodynamic diameter. Differently, the mobility particle diameter is taken in the SMPS measurement. In the manuscript, vacuum aerodynamic diameter (D_{pv}) is converted into mobility diameter (D_{pm}) according to:

$$D_{pm} = D_{pv} / \rho_p$$

Here, ρ_p is the estimated particle density. As above-mentioned, this particle density is estimated according to the bulk properties, independent of particle size. Therefore, the method for estimating particle density is not contradictory to the usage of this density to convert vacuum aerodynamic diameter to mobility diameter.

Modifications in the MS:

“Particle density is estimated by comparison of total particle volume concentration calculated from particle number size distribution assuming a spherical particle and bulk particle mass concentration measured with the AMS. The vacuum aerodynamic diameter for AMS measurements was converted to mobility diameter by division of AMS vacuum aerodynamic diameter by the estimated particle density (1600 kg/m^3).”

What was the CE used during the campaign? Is it supposed to be constant whatever the aerosol chemical composition? Since ammonium nitrate losses are discussed in the paper, it would be nice to apply a chemical composition-dependant CE as suggested by Middlebrook et al. 2012 (Middlebrook, A., Bahreini, R., Jimenez, J., and Canagaratna, M. R.: Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, *Aerosol Sci. Technol.*, 46, 258–271, doi:10.1080/02786826.2011.620041, 2012.)

Response:

The AMS data were re-processed using chemical composition-dependent CE. But, we realized that the mass fraction of different chemical species is used only in this study. The mass fraction using constant CE is the same with that using varied CE. Therefore, the data is not necessary to update the dataset in the manuscript.

Page 7650: uncertainty on the CCN measurement and corresponding KCCN? This is important in a closure study.

Response:

A major source of uncertainty in CCN measurement is the derived supersaturation (ss) in CCN counter. The uncertainty for supersaturations $\geq 0.2\%$ with the 95% confidence range is 10% relatively. For $SS < 0.2\%$, the absolute uncertainty is 0.02%. The uncertainty is quantified according to the relationship between ΔT and SS in CCN counter derived from calibrations using $(\text{NH}_4)_2\text{SO}_4$. Here, we assume that the uncertainty of kappa calculated from CCN measurement is around 10%, roughly.

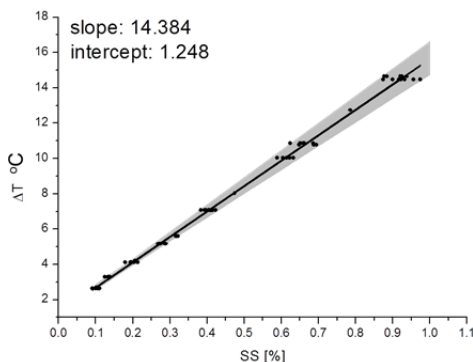


Figure: The relationship between ΔT and supersaturation in CCN counter. The grey area shows the uncertainty in the derived supersaturations (95% confidence range).

Modifications in the MS:

“A major source of uncertainty in CCN measurement is the derived supersaturation (SS) in CCN counter. The relative uncertainty for supersaturations $\geq 0.2\%$ with the 95% confidence range is 10%. For $SS < 0.2\%$, the absolute uncertainty is assumed as the same with $SS = 0.2\%$, i. e., 0.014%. This estimation is made on a basis of the relationship between ΔT and SS in CCNc derived from CCNc calibrations using ammonium sulfate (See supplementary material).”

“According to the uncertainties in CCNc and HTDMA measurements, we roughly estimated that the uncertainty in both κ_{HTDMA} and κ_{CCN} is 10%.”

Results and discussion

Page 7653 lines 18-21: the fraction of hydrophilic/hydrophobic particle may be linked to the fraction of accumulation/aitken particles for a given size.

Response:

Here, the hydrophobic and hydrophilic fraction means the hydrophobic and hydrophilic number fraction in the particle population with the same size. Accumulation and Aitken mode describe the modes in particle number size distribution. Therefore, there is no link between hydrophobic and hydrophilic fraction and accumulation-Aitken mode particles.

Page 7656 line 16: The r^2 obtained on the scatter plots relating $_chem$ and $_HTDMA$ is indeed higher when using size segregated chemical composition than when using bulk chemical composition for 150 nm and 200 nm particles, but it is not the case for 250 nm particles (the slope is also higher than when using bulk chemical composition). Can you comment?

Response:

Firstly, the mass size distribution derived from AMS measurements shows that the peak is around 250 nm in mobility diameter as shown in the following plot. Different from other particle sizes, the bulk chemical composition can well represent the chemical composition of 250 nm particles detected by HTDMA. Compared to bulk chemical composition, using size-resolved chemical composition does not improve the closure very much. Secondly, the data points are 435 for size-resolved chemical composition, while 1639 for bulk chemical composition. Due to the smaller number of data points for scattering plot using size-resolved chemical composition, the slope and r^2 are more sensitive “outlier” than that using bulk one.

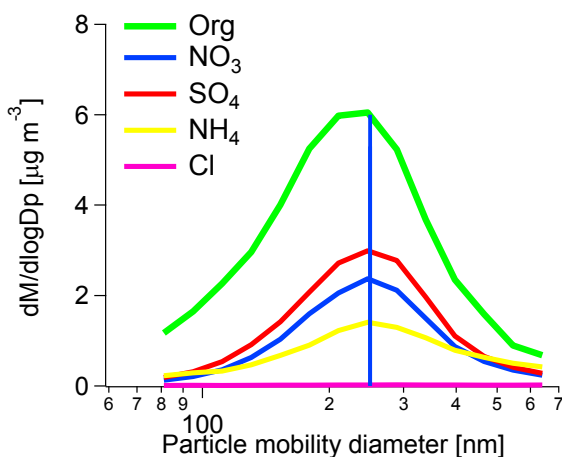


Figure: Particle mass size distribution of different species measured by AMS. 250 nm is marked.

Page 7657: line 10 if the bias between $_chem$ and $_HTDMA$ is higher for high NH_4NO_3 loadings, presumably due to losses in the HTDMA, then the conclusions should state that HTDMA measurement underpredicts the particle hygroscopicity rather than that the AMS/ZSR method overpredicts the particle hygroscopic growth.

Response:

We agree. Due to the evaporation of NH_4NO_3 in the HTDMA system, the growth factor detected by HTDMA is lower than the true hygroscopic growth factor. As a result, the κ_{HTDMA} calculated from growth factor is lower than κ_{chem} predicted by AMS/ZSR method.

Modifications in the MS:

“This observation is consistent with previous studies that also reported that particle hygroscopic growth measured by HTDMA is lower than that predicted AMS/ZSR method when the mass fraction of nitrate was high (Aklilu et al., 2006; Gysel et al., 2007)”

Page 7659, line 1: slopes are improved with the new settings, but not r^2 .

Response:

Yes, due to the number of data point shown in Fig. 8 is much less than that in Fig.5, the goodness-of-fit of linear regression is more sensitive in Fig. 8 to the data “outliers”. This could be the reason why r^2 is not improved.

Page 7659, line 10: It should be made clearer from this point that the authors make the hypothesis that only the more hygroscopic mode of the HTDMA is activated (which is not straightforward). This paragraph is inconsistent with sentences line 10 in the conclusion and line 17 in the abstract: if only the hygroscopic mode is considered, the closure is achieved within 10 %, right ?

Response:

The referee is right. The assumption that only the more hygroscopic particles become activated is not straight forward. However, as we are searching for a possible reason, why we find discrepancies between κ derived from HTDMA and from CCN, we consider this case (more hygroscopic mode particles are activated only) as the upper limit for κ derived from HTDMA measurements. We found that even for this extreme case the κ values derived from CCN measurements are not reached. Therefore other possible explanations are discussed later on.

We clarified this in the text as follows:

"The results are given in Table 3 from which can be seen that for similar sizes, κ_{CCN} values are 37% higher than κ_{HTDMA} values, on average. The upper limit for κ derived from HTDMA measurements is reached by assuming the activation of the more hygroscopic mode only during CCNc measurements. κ values were calculated from the hygroscopic growth factor (κ_{mode} , in Table 3) of hydrophilic mode to compare with κ_{CCN} . The κ_{CCN} values are still higher than κ_{mode} , while the difference between them decreases around 10 %, as can be seen from Table 3."

Page 7659, line 22: I am not sure that the reasons for discrepancies in the closure can be discussed at the level of less than 10 % discrepancy. The level of confidence of each of the measurements added together is higher than this. The authors should provide the uncertainty on the difference between κ_{HTDMA} and κ_{CCN} . Moreover, these effects are discussed for analysing a difference between two average values (calculated over the whole campaign) that show significant variability each. A similar analysis than previously performed when comparing κ_{chem} and κ_{HTDMA} (scatter plot, R^2 and slope) would be more appropriate.

Response:

Yes, we agree. The uncertainties of κ_{CCN} and κ_{HTDMA} will be discussed in the manuscript. “, and the differences between κ_{HTDMA} and κ_{chem} are within 10%” was removed from the abstract. See the response above.

Most of the sampling time, the critical diameters determined with the CCNc were not exactly the same as the dry diameters (D_{dry}) considered by the H-TDMA. The data for critical diameters within the range $D_{dry} \pm 10$ nm were assembled for this comparison, only. Consequently, the number of data points

(pair of κ_{HTDMA} and κ_{CCNc}) is very small. The authors think that a scatter plot and linear regression may not be a better way to represent such comparison.

Conclusion

Page 7661, line 11: The 30% discrepancy between κ_{CCN} and κ_{HTDMA} can be reduced if only the more hygroscopic particles are activated. This effect should be included before surface tension etc. effects are discussed.

Response:

Our statement is unclear. If only hydrophilic mode considered, the CCNc-derived κ is around 30% higher than those determined from HTDMA measurements. The discrepancy is even larger by considering both hydrophilic and hydrophobic modes. We modified the MS:

“Consistency between HTDMA and CCNc-derived κ values was not achieved in our study. If hydrophilic mode in HTDMA measurements is considered only, the CCNc-derived κ is around 30% higher than those determined from HTDMA measurements.”

Technical corrections

Page 7646, line 25 : Under or overprediction instead of Under of Overprediction

Response: It was modified in the manuscript

“would result in an under or overprediction of the hygroscopic growth factor in closure studies”

Page 7649, line 17 size range of SMPS to correct

Response:

The size range was corrected. (10-800 nm)

Page 7656, line 17 ($r^2=0$) to erase

Response: $r^2=0$. was deleted from the texts.

Figure 5: legend should mention size segregated chemical composition instead of bulk chemical composition

Response: We modified the legend of Figure 5.

“Fig. 5: Correlation between κ_{chem} and κ_{HTDMA} for particles with dry $D_p=100, 150, 200,$ and 250 nm, using size-segregated chemical composition”

Page 7659, line 9 to 14: several errors in English language, the whole paper should be checked again;

Response:

We read the manuscript again and improved the English language.

Page 7661 line 7 closure studies

Response: It was corrected.