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This reply is formatted as follows:

Reply to Reviewer #4

Reviewer's comments

Authors' reply

Changes to Text

This work provided comprehensive measurements during HCCT-2010 campaign at the Schmucke mountain site towards to understand the cloud properties. The simultaneous measurement of cloud droplets residues and interstitial aerosols with two AMS highlight the role of cloud processing in enriching aerosol particles by uptake of reactive gas species. This kind of dataset with high quality is rarely obtained and of great value. The analysis of temperature dependence of mass fraction of nitrate and distribution of f44 and f43 shone a light on the uptake/scavenging process of the chemicals, indicating the adverse effect of temperature on the uptake of nitric acid gases and more oxidized organics onto cloud droplets. Moreover, the organic nitrate concentration in cloud droplet residual (CDR) is discussed as well. A positive correlation between organic nitrate in CDR and temperature was suggested. In addition, the authors have added a case study to further compare the effect of activation of nitrate containing particles and uptake of nitric acid gas onto cloud droplets and therefore making the whole article a thorough and logical analysis of the chemical composition evolution between CCN, precursor gases, interstitial aerosol and cloud droplet residual. In general, the conclusion was reasonably supported by the data and analysis. The manuscript was overall well written. I recommend this work can be published after some miner revision.

We thank the reviewer for this positive rating of our manuscript

P.2 line 15-25 The formula of molecule shall be rewritten with subscripts.

Corrected.

P.3 line 30 Please clarify the “similarity” of trajectories. Is it subjective or did you use any objective method?

The similarity was estimated by subjective inspection of the graphs shown in Figure 1

P.5 line 4 Please unify the expression of temperature unit (‘-’ or ‘minus’) according to the ACP writing instructions.

Changed.

P.8 line 1-14 The map of trajectories in figure 1 are somehow changed, making it difficult to understand the exact properties of different trajectories like length of path or the location of polluted regions. Please offer some vertical description like the height of airmass’s center. It may help to understand the uptake of gases if there were no precursor measurements.

We added the pressure history of the air masses to Figure 1. We tried to unify the latitude and longitude scaling of the maps as good as possible, without losing information for those air masses that had traveled only shorter distances during the 96 hours.

P.8 line35 The conclusion drawn here shall be more careful, since if the fraction of nitrate in CDR was elevated, the out-of-cloud aerosol could have an elevated fraction of nitrate as well due to the evaporation or re-partitioning.

> Our data show that in general the nitrate mass fraction is highest in the cloud residuals, higher than in the out-of-cloud-aerosol, and higher than in the interstitial aerosol. It is very likely that the out-of-cloud aerosol is influenced by previous cloud passages and may therefore contain more nitrate than in earlier times, but still our observations show that CDR have the highest nitrate fraction. A depletion of the nitrate fraction in the interstitial particles was not observed. Thus, to our opinion the only explanation for the elevated ammonium nitrate in the CDR remains uptake of gaseous HNO3 and NH3.

P.10 line 18, Considering that the uptake of nitric acid gas, ammonia and organics caused elevation of corresponding CDR compositions, the authors should be more careful to use the term of "xx% of total submicron aerosol mass partitioned in to cloud phase", which is very likely to lead to a misunderstanding that the CDR composition came all from aerosols.

> This is a good point. We reformulated the statement to:
>
> Figure 7 shows that on average 85 % of the total submicron aerosol mass is present in the cloud phase, with a maximum value of 94 % in FCE22.0 and minimum values of about 66 % in FCE13.3. This can be the result of three different processes: activation of the pre-existing aerosol particles acting as CCN, scavenging of interstitial, non-activated aerosol particles, and uptake of gas-phase species by the cloud droplets as it was discussed for nitrate and ammonium above.

P.14 line 15-20 The abundance of a certain chemical composition is not always equal to formation. Only by taking the ratio and the absolute concentration into consideration could tell one if there is formation of the specie. Therefore, the difference here might indicate different mechanisms or different form of organic nitrates.

> We agree with that. Our observations suggest that the abundance of organic nitrate in the particle phase increases with temperature in our data set. We can't conclude from this observation to the formation mechanism. Therefore we reformulated the paragraph to:
>
> This finding suggests that the abundance of organic nitrate in the particle phase increases with temperature. However, recent observations by Lee et al. (2014) showed that formation of organic nitrate is enhanced at lower temperatures. Since also the equilibrium between particle phase and gas phase should be shifted towards the gas phase at higher temperatures, the observed higher amount of organic nitrates in the aerosol phase at higher temperatures cannot be explained by the formation mechanism proposed by Lee et al. (2014).

P.14 line 24, Rephrase “our CDR data do not such a clear trend:”.

> Changed to: "our CDR data do not show such a clear trend"

P.16 line 30, A r2=0.32 could not be described as “significant linear correlation”.

> Significance was tested using both t-test and F-test (using Wavemetric's IGOR function "statslinearregression") with 95% confidence interval. The correlation of both, NO3 and NH4 versus temperature is significant (the regression coefficient is different from zero with 95% confidence).

P.35 Figure 9, There is approximate of O:C on the right axis in the upper panel while it’s absent in the lower panel.

This is on purpose, because the O:C values were derived from the f_{44} value (left scale in the upper panel) using the parameterization given in Aiken et al. (2008) and Canagaratna et al. (2015) (as explained in section 3.2.3), whereas this can not be done using the f_{43} value (left scale in lower panel).

References

Aiken, A. C., Decarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., and Jimenez, J. L.: O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high-resolution time-of-flight aerosol mass spectrometry, Environ. Sci. Technol., 42, 4478-4485, doi: 10.1021/es703009q, 2008.

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Lee, L., Wooldridge, P. J., Gilman, J. B., Warneke, C., de Gouw, J., and Cohen, R. C.: Low temperatures enhance organic nitrate formation: evidence from observations in the 2012 Uintah Basin Winter Ozone Study, Atmos. Chem. Phys., 14, 12441-12454, doi: 10.5194/acp-14-12441-2014, 2014.