

**acp-2016-835**

## **Reply to Reviewer #2**

This reply is formatted as follows:

Reviewer's comments

Authors' reply

**Changes to Text**

Schneider et al. report the results of a field study aimed at studying the uptake of trace gases into clouds during the HCCT-2010 campaign in Thuringia, Germany. For this study two co-located aerosol mass spectrometers (AMS) measured the composition of residues from evaporated cloud droplets (i.e., residual particles) and interstitial aerosols during full cloud and cloud-free events. Aerosol species that form the focus of this manuscript include nitrate, ammonium, black carbon, organic carbon, and various (physical) aerosol and meteorological properties. The data accumulated during this campaign is quite unique among cloud composition studies. Only a handful of AMS studies exist related to the composition of cloud residues. This study appears to be the first to measure interstitial aerosol and cloud droplets simultaneously using the two collocated AMS and their respective aerodynamic cutoff diameter and counterflow virtual impactor inlets.

The authors have collected a wealth of very high quality data from ~14 full cloud events, in addition to non-cloud events for comparison. An extensive analysis of the data has been carried out that provides important insights into trace-gas-cloud and aerosol cloud interactions. Very strong evidence, backed up by theoretical considerations and back trajectories, is provided showing that clouds effectively scavenge gas phase nitric acid, ammonia, volatile organic compounds, and less so black carbon. The data was used to calculate scavenging efficiencies for these gases, which range from 0.6 to 1.0 for nitric acid, ammonia, and VOCs, and 0.1 to 0.5 for black carbon. The methods employed are state-of-the science. The only data missing (due to lack of instrumentation at the campaign) were direct gas phase measurements of HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, NH<sub>3</sub> and VOCs. However, what is missing in gas phase measurements is made up for by the thoroughly and cautiously interpreted AMS data, which leads to high confidence in the conclusions. I support publication in ACP after the few minor comments listed below are addressed.

We thank the reviewer for this very positive rating of our manuscript

Minor Comments:

The title includes the label of "orographic" clouds, but orographic does not appear again in the abstract or main text of the article. I recommend including in the text a description of the cloud type and what measured parameters defined the clouds as orographic during the campaign. Perhaps a discussion related to this could be added to section 3.1?

A description of the cloud types is given in detail in other publications in the special issue on the HCCT campaign, e.g., Tilgner et al. (2014) or Roth et al. (2016). Therefore, we added only a brief description of the cloud to section 3.1.:

**The whole time series of the cloud droplet number distribution measured by the FSSP is given in Figure S4. The 14 FCE are also indicated by the grey bars. These FCE were chosen based on certain criteria as detailed in Tilgner et al. (2014), focusing on connected flow conditions between the upwind, the summit and the downwind station. These conditions went along with stable south-west flow conditions, thus the clouds were mainly of orographic nature, however in certain cases the meteorological analysis revealed that the clouds were not purely**

orographic (FCE1.1, FCE2.1, FCE26.1). For details see Table 5 in Tilgner et al. (2014) and Table 1 in Roth et al. (2016).

Molecular formulae should include subscripts on lines 19-21 on p. 2.

This was corrected

p. 4: Consider spelling out the abbreviations C-ToF-AMS and HR-ToF-AMS where they are first used.

Done.

p. 8, line17: Should read: "...relative to the sum of all species detected by the AMS..."

Changed

p. 8, line 23: replace "forth" with "fourth"

Changed

p. 9, line 25: replace "+" with "-" and visa versa for the charge on nitrate and ammonium, respectively.

This was corrected

p. 9, lines 27-31: The sentence beginning and ending in "If the air...occurs in the cloud phase," is a run-on sentence and is unclear. Please simplify or rephrase to improve clarity.

We changed the sentence to:

If no chemical processing of nitrate and ammonium occurs in the cloud phase and the air returns to the same temperature and relative humidity conditions after the cloud as before the cloud, is it to expect that the overall equilibrium between particle phase  $\text{NH}_4\text{NO}_3$  and gas phase  $\text{NH}_3$  and  $\text{HNO}_3$  after the cloud will be equal to that before the cloud.

p. 14, line 23: Insert "show." Should read: "...our CDR data do not show such a clear trend..."

Corrected

p. 16, line 32: Insert "with." Should read: "...was better correlated with CNDC than..."

Corrected

p. 17, lines 21-23: The sentence beginning and ending with, "At the same temperature...the cloud is established," is unclear. Please rephrase.

We changed the sentence to:

If the aerosol experiences the same temperature and relative humidity after the cloud passage as before the cloud passage, the same equilibrium between particle-phase ammonium nitrate and gas phase nitric acid and ammonia is established.

Figure 9: I found the blue-green diamond symbols difficult to see over the background of green squares. I suggest changing the C-ToF CDR symbols to different colors so they stand out.

We changed the CDR symbols in both panels to black.

Figure 11: I find the different shades of grey difficult to tell apart in some print outs.  
Consider changing colors of the CDR data

We changed the colors of the CDR data to black, purple and red

#### References:

- Roth, A., Schneider, J., Klimach, T., Mertes, S., van Pinxteren, D., Herrmann, H., and Borrmann, S.:  
Aerosol properties, source identification, and cloud processing in orographic clouds  
measured by single particle mass spectrometry on a central European mountain site during  
HCCT-2010, *Atmos. Chem. Phys.*, 16, 505-524, doi: 10.5194/acp-16-505-2016, 2016.
- Tilgner, A., Schöne, L., Bräuer, P., van Pinxteren, D., Hoffmann, E., Spindler, G., Styler, S. A., Mertes,  
S., Birmili, W., Otto, R., Merkel, M., Weinhold, K., Wiedensohler, A., Deneke, H., Schrödner,  
R., Wolke, R., Schneider, J., Haunold, W., Engel, A., Wéber, A., and Herrmann, H.:  
Comprehensive assessment of meteorological conditions and airflow connectivity during  
HCCT-2010, *Atmos. Chem. Phys.*, 14, 9105-9128, doi: 10.5194/acp-14-9105-2014, 2014.