

Interactive comment on “Uptake of nitric acid, ammonia, and organics in orographic clouds: Mass spectrometric analyses of droplet residual and interstitial aerosol particles” by J. Schneider et al.

Anonymous Referee #3

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This study describes aerosol chemical properties measured from both droplet residues and interstitial aerosol particles. The principal aims of this work are to study the cloud processing, including the enrichment of aerosol particles within clouds from the uptake of different gas-phase species. This paper also addresses the role of different chemical species in the activation of cloud droplets. The study is very thorough with an impressive instrumental setup and a large number of statistically relevant cloud events. The paper, figures, and text are well prepared. However, I have some major concerns regarding the experimental approaches used to derive the conclusions made in this manuscript.

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General comments:

This experiment, the HCCT was intended to understand how aerosols are activated into clouds as well as the impact of different cloud processing on cloud properties. As outlined in Tilgner et al., (2014), this study was designed in such a way as to have three well equipped stations before, in, and after the formation of an orographic cloud. I would assume that combining measurements from these three stations would have made this study much more robust, rather than only comparing in and out of cloud residues on the cloud top.

1) One of my major critics is the lack of a clear discussion on the aerosol (CDR and interstitial) physical properties (size and number concentration). These factors play an essential role in the activation of cloud droplets and should not be separated from aerosol chemical properties. Aerosol size distributions should have been taken into account to provide a measure of aerosol activation diameter. It would have been interesting to investigate how this parameter (aerosol activation diameter) varied as a function of chemical composition. It would have been equally interesting to study the sampling efficiency of the CVI inlet through comparison of the total number of CDR particles (CPC/SMPS) with the total number of cloud droplets measured with the FSSP.

a. In section 2.3, the authors state that there are SMPS measurements available, however unless I am mistaken I do not find any other reference to these measurements, either in the manuscript or in the supplementary material.

b. Figure S6 shows OPC size distributions measured behind the CVI and Interstitial inlet. The GRIMM instrument normally provides particle size distribution measurements from 300 nm up to > 10 microns. At 300 nm, all particles are expected to act as CCN. Therefore, these measurements are not useful to observe activation parameters of aerosol particles. I did not find any reference to this figure in the main manuscript.

2) The papers main results are based on the comparison of interstitial aerosol particles and cloud droplet residues. These two “types” of aerosol particles are found in largely

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different size categories, with interstitial aerosol particles generally having diameters < 90 nm and CDR particles having diameters > 90 nm. It has been reported in a large number of studies that the contribution of organic aerosol particles increases as particles size decreases. Equally inorganic nitrates are often measured in larger particle diameters. Can the authors show that the increased organic compounds measured in the interstitial aerosol during cloud events are significant to the cloud event itself and that the concentrations (in the same size class) are different in the NCE?

a. Page 7, Line 35: The authors state that the mass concentration of the interstitial aerosol is lower than that of the CDR. This would be expected since CDR particles are larger in diameter (hence more mass) than the interstitial aerosol.

b. Figure 4. It would be useful to see the significance of the difference between the interstitial and CDR composition, through comparison of the similar size fractions (< 90 nm (INT) and > 90 nm (CDR)) during NCE.

3) Another concern is that the transmission efficiency of the aerodynamic lens used for Aerodyne products sample aerosol particles with “good” efficiency between ~90 nm and ~300 nm (Liu et al., 2007), however below (and above) these limits the transmission efficiency of the instrument decreases rapidly. One needs to take this transmission efficiency into account and also the implications that this may have on the quality of the AMS data at these lower diameters. Baseline errors will likely have an impact at these diameters so it is necessary to take precautions to ensure that measured aerosol compositions and concentrations at these small diameters are real and not just arbitrary noise.

Peter S. K. Liu, Rensheng Deng, Kenneth A. Smith, Leah R. Williams, John T. Jayne, Manjula R. Canagaratna, Kori Moore, Timothy B. Onasch, Douglas R. Worsnop, and Terry Deshler Transmission Efficiency of an Aerodynamic Focusing Lens System: Comparison of Model Calculations and Laboratory Measurements for the Aerodyne Aerosol Mass Spectrometer *Aerosol Science And Technology* Vol. 41 , Iss. 8,2007

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4) A large part of this manuscript is focused on the enrichment of nitrate in aerosol particles after cloud processing. However, these conclusions were made through comparing CDR with NCE before, and in some cases, after the cloud event. If cloud processing was indeed used to result in the enrichment of nitrate in particles, would one not expect to observe higher nitrate in aerosol particles once the cloud event has passed?

5) A constant correction efficiency (CE) of 0.5 was applied to all data. However, there are several periods (shown in Figure 2 a)) where the contribution of nitrate aerosol particles was greater than 25% to the total aerosol mass. In general, within the aerosol mass spectrometry community, it is recommended to apply a composition dependent CE as outlined in the manuscript Middlebrook et al., (2012). Middlebrook, A.N R. Bahreini, J. L. Jimenez, and M. R. Canagaratna (2012) *Aerosol Sci. Tech*, 46:258–271.

6) Cloud events listed in table 1 varied from 3 hrs up to 12 hrs. Air mass trajectories were used to verify that there was no change in air mass properties, however could there be more robust criteria used to classify these cloud events. Could the authors incorporate the FSSP cloud droplet distribution and LWC measurements to evaluate whether the cloud properties changed outside a certain limit. For example Fig. 12 shows the cloud droplet diameters and concentrations changing during the cloud event, this was accompanied also by a change in the LWC. How can the authors ensure that these changes in cloud properties were not accompanied by slight air mass changes, or entrainment of new aerosol types. This might influence the comparison with NCE.

a. Likewise, how long a time period should be compared from the NCE data? It might not be judicious to include data from 24 hours prior to the measurements.

Minor comments: The only mention of orographic clouds is in the title of this manuscript. Although, full details of the experimental design is included in Tilgner et al., some discussion of the importance and how these cloud events were verified as orographic should be included.

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Page 4, section “Analysis instruments” How was the aerosol dried prior to sampling in the interstitial aerosol?

Page 13, Line 33: The authors mention that biogenic emissions could be a source of the higher OA measured at higher temperatures. Is there any evidence of biogenic emissions during these periods? Gas-phase measurements, lower than average BC concentrations, etc.

Page 16, Line 29: What is the significance of these correlations? 99%, 95% and how is the ‘significance’ determined?

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