Response to comments

Manuscript Number: acp-2017-23
Title: In situ chemical measurement of individual cloud residue particles at a mountain site, South China. Qinhao Lin et al.
Received and published: 20 March 2017

Referee #2: J. Schneider

In their manuscript "In situ chemical measurement of individual cloud residue particles at a mountain site, South China", Qinhao Lin and co-workers report on the analysis of single particles from cloud residues using a single particle aerosol mass spectrometer. They observed a high fraction of EC-containing particle in the residuals and detected amines with a high variability. Nitrate was found to be increased in residuals compared to ambient particles, while sulfate showed a dependency on the chemical composition of the residues. The topic of the paper is well suited for ACP, and the data itself are interesting, because single particle measurements of cloud residuals are still sparse. However, the manuscript suffers from many unclear statements and some severe uncertainties regarding the analysis of interstitial particles. I have many points where more information is needed or where I disagree. None of these points alone would be a "major" comment, but the multitude of my remarks and questions suggest to require a major revision and to reconsider the manuscript after my comments listed below have been addressed.

We would like to thank Prof. J. Schneider for his useful comments and recommendations to improve the manuscript. We agree with the comments, and careful revision has been made accordingly, please refer to the following responses for details.

Comments and remarks:
Title: I suggest to change the title to "In situ chemical composition measurement of individual cloud residue particles at a mountain site, South China"

We have changed accordingly. Please refer to Lines 1-2 of the revised manuscript.

Page 4, lines 64 – 68:
More references are needed here to discuss the anthropogenic influence on cloud particles, not just two papers on single particle analysis.

As also suggested by Reviewer 1, We have added references (Stier et al., 2005; Sorooshian et al., 2007b; Lohmann et al., 2007; Rosenfeld et al., 2008; Roth et al., 2016; Seinfeld. et al., 2016; Li et al., 2017) to discuss the anthropogenic influence on cloud particles. Anthropogenic particles can increase number concentration of small cloud droplets, in turn, affect reflectivity and life time of clouds (Rosenfeld et al., 2008; Stier et al., 2005; Lohmann et al., 2007). In-situ cloud chemical measurements show varied chemical composition of cloud droplets at various regions (Sorooshian et al., 2007a; Roth et al., 2016; Li et al., 2017). Although a large number of aerosol/cloud studies over the past 20 years, the uncertainty for evaluating radiative forcing due to aerosol-cloud interactions has not been reduced (Seinfeld. et al., 2016). Please refer to Lines 55-62 of the revised manuscript.

Page 5, line 79: Replace "Nf of sulfate" by "NF of sulfate-containing particles"

We have changed it accordingly. Please refer to Line 74 of the revised manuscript.

Page 5, line 80: Replace "other study" by "other studies"

We have changed it accordingly. Please refer to Line 76 of the revised manuscript.

Page 7, line 122: Was the humidity measured in the evaporation chamber? How do you make sure that all water evaporates?

Relative humidity (RH) was around 30% in the evaporation chamber, thus it can be assumed that the majority of water was evaporated. Please refer to Lines 131-134 of the revised manuscript.

Page 8, lines 147-149: Did you do the size calibration on the mountain top station? What was the ambient pressure during the measurements and during the calibration? Did you check the inlet flow or the pressure inside the aerodynamic lens?

We did the size calibration on the mountain top station. Polystyrene latex spheres (Nanosphere Size Standards, Duke Scientific Corp., Palo Alto) of 0.2-2.0 μm in diameter were used to calibrate the sizes of the detected particles on the mountain top station. The ambient pressure was 830 hPa (826-842 hPa) during the measurements and during the

calibration. The pressure inside the aerodynamic lens maintains about 3 hPa during the measurements and during the calibration. Please refer to Lines 152-155 of the revised manuscript.

Page 8, line 161: The SMPS does not measure the cloud droplet concentration but the cloud residue concentration. Cloud droplets would have to be measured outside in the cloud (by FSSP or similar instrumentation).

We have corrected the mistake. "cloud droplet concentration" was replaced with "cloud residual concentration". Please refer to Lines 165-168 of the revised manuscript.

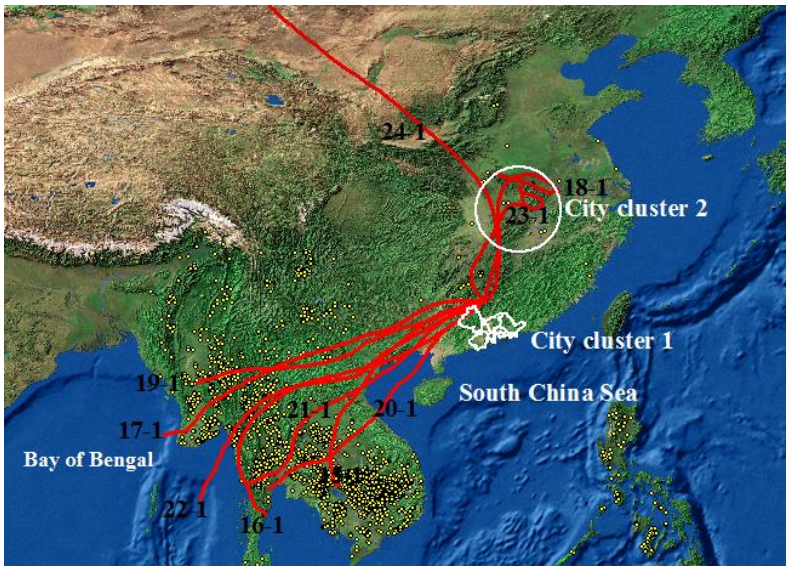
Page 9, line 171: As I will outline in more detail below, I doubt the existence of interstitial particles in this size range.

The period of collecting interstitial particles on 22-23 Jan encountered initial mixing of northerly cloud-free air (dry and cold airstreams) and southwesterly cloudy air (moist airflows). The dry northern air mass might lower supersaturation, only larger particles could be activated. This might result in non-activated particles observed to be above 200 nm here (Mertes et al., 2005; Kleinman et al., 2012; Hammer et al., 2014). To make it more accurate, we prefer to name "non-activated particles", rather than "interstitial particles". We have clarified them. Please refer to Lines 452-461 of the revised manuscript.

Page 9 lines 184-185 and Figure 2: More info on the trajectories is needed: How did the vertical evolution look like? How well is the mountain represented in the model? Is 1800 m the best altitude that represents the mountain site? Please add also the most important megacities to the map to help estimating the influence of anthropogenic emissions.

We have added the vertical evolution of the trajectories. The beginning of southwesterly air masses traversed at lower heights relative to northerly air masses. Please refer to the Figure 2 (b). Heights of the HYSPLIT model in the study region (a spatial resolution of $0.5° \times 0.5°$) was averaged 500 m a.s.l, which was lower than height of the observed site (1,690 m a.s.l). Therefore, a height of 1,800 m a.s.l. (approximately 100 m above the observed site) was used as an endpoint in the model. Continental air masses crossed industrial areas where located in the Yangtze River Mid-Reaches city cluster (Figure 2a). The site was possibly affected by industrial emissions under the influence of continental air masses. Please refer to Lines 189-196 of the revised manuscript.

(a)



(b)

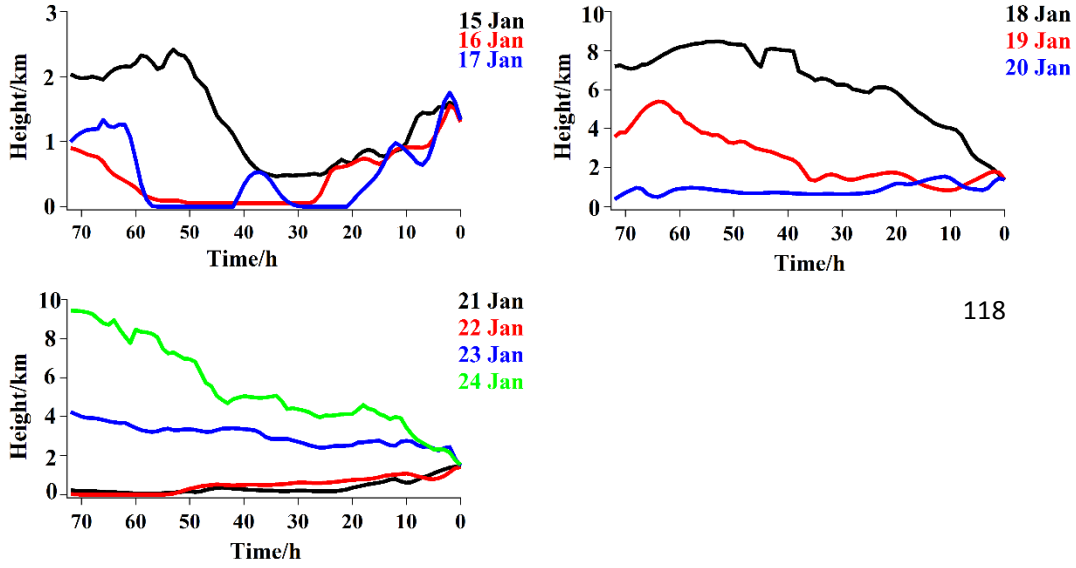


Figure 2: (a) HYSPLIT back trajectories (72 h) for air masses at 1,800 m during the whole sampling period. The white borders and circle refers to the Pearl River Delta (city cluster 1) and Yangtze River Mid-Reaches city cluster (city cluster 2), respectively. The yellow rots represent fire dots during the study periods. The fire dots are available at https://earthdata.nasa.gov/; (b) Heights (above model ground) of the air masses as a function of time.

Page 10, line 207: I suggest moving Figure S3 to the main paper.

Figure S3 has been moved to the main paper, please refer to Figure 4 in the revised manuscript.

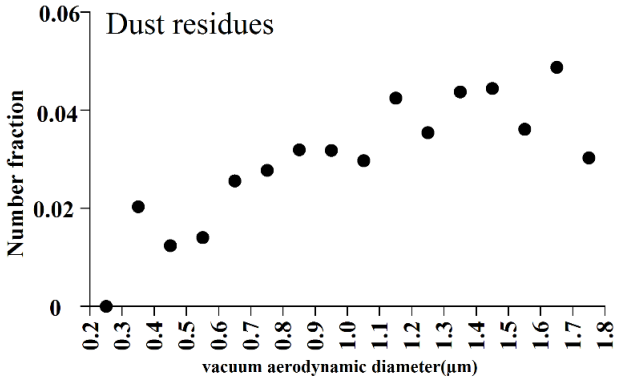
Page 11, lines 221-222: But Roth et al. found a clear enhancement of amines in residues compared to the background aerosol (9% to about 2%).

We have added a comparison Nf of amine-containing particles between cloud residues and background aerosol reported by Roth et al., (2016). Please refer to Lines 265-267 of the revised manuscript.

Page 12, lines 235-237: I agree that dust is found more frequently in the coarse particle size range, but then I would expect to see an increase of the dust fraction in the residues with increasing diameter. This is not seen in Figure S3. Is the identification of dust reliable? What about the Fe-containing particles? They might be dust as well.

We agree with the comment. As a matter of fact, Nf of dust cloud residues generally increased with increasing diameter. Please refer to Lines 276-278 of the revised manuscript.

Approximately 16% of the Fe cloud residues contained Ca peak (m/z 40). Relatively weak Na and K peaks in the Fe particles possibly contributes to anthropogenic sources (Zhang et al., 2014), especially northern air mass across iron/steel industrial activities in Yangtze River Mid-Reaches city clusters (Figure 2). These might suggest that Fe cloud residues was likely to have come from mixed sources. Please refer to Lines 290-294 of the revised manuscript.



Page 12, lines 238-243: What could be the source of these Pb- and Fe-containing particles? See also comment above. Can the Fe-containing particles belong to the dust-type?

As mentioned above, the Fe cloud residues contain Ca peak (m/z 40, 16% by number) and relatively weak Na and K peaks, which possibly contributes to anthropogenic sources (Zhang et al., 2014), especially northern air mass across iron/steel industrial activities in Yangtze River Mid-Reaches city clusters (Figure 2). These might suggest that this particle type likely originated from mixed sources. Please refer to Lines 290-294 of the revised manuscript.

The Pb particles showed its typical ions at m/z 208Pb^+ and internally mixed with K and Cl. Previous studies found that K and Cl internally mixed with Pb particles have a possible origination of waste incineration (Zhang et al., 2009) or iron and steel facility (Tsai et al., 2007). Please refer to Lines 315-318 of the revised manuscript.

Page 12, lines 244-252: If these particles were from sea salt, they should contain chloride ions. That is hard to see in Figure 3. Are these Na-rich particles correlated with air masses coming from the ocean?

Na-rich particles result from varied sources of industrial emissions or sea salt particles and dry lake beds (Moffet et al. 2008). The Nf of the Na-rich cloud residues did not increase from continental (Northerly) air mass to maritime (southwesterly) air mass on 21 Jan (3.3% versus 2.4% by number). However, sea salt ion peak areas (m/z, 81/83$Na_2{}^{35}Cl$/$Na_2{}^{37}Cl$) were enhanced for Na-rich particles origination from maritime air mass relative to continental air mass (3.8 ±l2.4 times). Continental air masses crossed industrial areas where located in the Yangtze River Mid-Reaches city cluster (Figure 2). Industrial emissions was a possible contributor to Na-rich particles under the influence of continental air masses (Wang et al. 2016). This might suggests that the Na-rich particles were contributed by both the industrial emissions and sea salt. Therefore, under the influence of maritime air mass, the signals for sea salt contribution became stronger. Please refer to Lines 299-310 of the revised manuscript.

Page 13, line 259 and Figure 5: How do you distinguish between sulfuric acid and sulfate? Besides, spelling (sulfate, sulphuric acid) should be consistent ("f" or "ph").

Sulfate ion peak at m/z -97 HSO_4^- and sulfuric acid cluster ion peak at m/z -195 $[H(HSO_4)_2^-]$

were given in previous single particle studies (Pratt et al., 2009; Rehbein et al., 2011). "Sulphuric acid" has been replaced by "Sulfuric acid". Please refer to the caption of Figure 5.

Page 13, lines 265-267: What other forms of nitrate do you suggest to be present on the Na-rich and dust residues? What about uptake of nitric acid from the gas-phase by the cloud droplets? How certain is the identification of ammonium? Which peak was used?

The Na-rich and Dust types were mainly composed of alkaline ion peaks (m/z, $23Na^+$,$39K^+$ and $40Ca^+$) in the position mass spectra (Figure 3). This suggests that rather than NH_4NO_3, nitrate might exist in the form of $Ca(NO_3)_2$, $NaNO_3$ or KNO_3 in the dust and Na-rich cloud residues. Please refer to Lines 340-344 of the revised manuscript.

We agree with the comment. We have discussed the contribution of uptake of gas-phase HNO_3 to enhanced nitrate in the cloud residues and cited Schneider et al. (2017). Please refer to Lines 334-335 of the revised manuscript.

Generally, a NH_4^+ ion signal (m/z 18) was used for identification of ammonium in the analysis of single particle mass spectrometry (Pratt et al., 2009). Please refer to Line 338 of the revised manuscript.

Page 13, line 268-272: The stability of ammonium nitrate depends also on the humidity. In the book by Seinfeld and Pandis (2nd edition, Wiley and Sons, 2006, Chapter 10.4.3) it is shown that at 30% RH ammonium nitrate does not exist above 30 C. I would assume that the dry carrier gas in the evaporation section is below 30% RH. Thus, it may well be that NH4NO3 evaporates in your system.

We agree with the comment. We have clarified the artificial effect on ammonium nitrate in the cloud residues. Please refer to Lines 344-346 of the revised manuscript.

Page 13, lines 275-276: The sentences "The presence of abundant sulfate in aged EC cloud residues was considered to be a good CCN species before activation:" needs rephrasing. It is not clear to me what you want to say. Do you mean "aged EC particles mixed with sulfate are good CCN"?

The sentence has been changed to "aged EC particles mixed with sulfate are good CCN".

Please refer to Lines 350 of the revised manuscript.

Page 13, line 279: Ammonium will most likely play a key role in the form of ammonium sulfate or ammonium nitrate. In organic particles, amines may play that role (methylamines). Again: how do you identify ammonium and how do you distinguish between amines and ammonium?

We agree with the comment. Ammonium will most likely play a key role in the form of ammonium sulfate or ammonium nitrate in the OC and aged EC cloud residues (Zhang et al., 2017). Please refer to Lines 353-354 of the revised manuscript.

A NH_4^+ ion signal (m/z 18) was used for identification of ammonium. The existences of m/z 59 $N(CH_3)_3^+$ (trimethylamine,TMA) and related amine ion signals m/z 58$C_2H_5NHCH_2^+$ (diethylamine, DEA) and m/z 86$C_5H_{12}N^+$ (triethylamine, TEA)were used for identification of amines (Angelino et al., 2001). Please refer to Lines 254-257 and 338 of the revised manuscript.

Page 14, line 281 (and Figure 5): Why does oxalate nor correlate with OC?

Classification of the OC particles mainly based on intense organic carbon ion signals (e.g., m/z 27$C_2H_3^+$, 37C_3H^+, 43$C_2H_3O^+$ and 51$C_4H_3^+$). However, majority of oxalate-containing particles internally mixed with the K-rich type. Therefore, oxalate was classified to the K-rich type, probably contributed from biomass burning. Noted that K-rich could contain a large abundant of organics (Pratt et al. 2011), however, the signals of organics were covered by the potassium due to its high sensitive to the laser. Please refer to Lines 371-378 of the revised manuscript.

Page 14, lines 284-285: What do you mean by "enrichment of TMA in amine cloud residuals"? You observe that in 93% of those cloud residuals that are assigned to the "amine" type contain TMA. That is not surprising, more surprising is that it's not 100%. But that's inside the measurement uncertainties, to my opinion.

We have changed "enrichment of TMA in amine cloud residuals" to "presence of TMA in amine cloud residuals". Please refer to Lines 360-361 of the revised manuscript.

Amine family signals m/z 58$C_2H_5NHCH^{2+}$ and m/z 86$C_5H_{12}N^+$ were also selected to

identify amines (Angelino et al., 2001), leading to only 93% of the "amine" residues containing TMA. Particles that exist a peak signal m/z 58$C_2H_5NHCH_2^+$, were found to account for 99% of the Amine residues. Please refer to Lines 254-257 of the revised manuscript.

Page 14, lines 294-295: "This may result in 33% by number to the Amine residues containing oxalate." Please rephrase, not clear what you want to say.

We have rephrased this sentence to "it may facilitate the entrainment of oxalate (33% by number) in the Amine residues. ". Please refer to Lines 370-371 of the revised manuscript.

Page 14, line 298: What does "unscaled" mean? These are absolute particle numbers.

Considering that the SPAMS mainly detected in size range 0.2-2.0 μm and has size-dependent transmission efficiency. Detected particles were not corrected by a SMPS. Therefore, detected particles cannot represent real atmospheric particle level. "unscaled" has been changed to "detected particle counts". Please refer to Line 381 of the revised manuscript.

Page 14, lines 302-303: You say that the air masses change from northerly on 18 Jan to southwesterly on 19 Jan, but the particles remain similar from 17 Jan (around noon) to 20 Jan (noon). On the other hand, the change in particle types is very abrupt from cloud residuals to ambient on Jan 17.

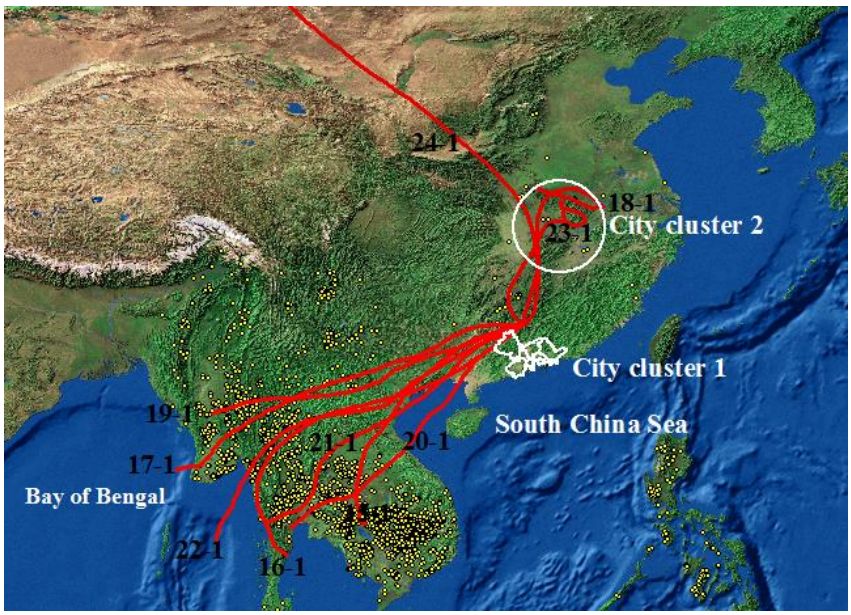
Southwesterly wind flow on 19-20 Jan was too weak (~ 2.75 m s^{-1}) to dilute particles originated from northerly air masses (Figure 1). Additionally, high RH (90%) contour line at height 1,500 m (a.s.l.) gradually moved to north China from 19 to 20 Jan (Figure S5). These might lead to similar residual particle types observed from 19 Jan to 20 Jan, although the site encountered southwesterly cloudy air on 19-20 Jan (Figure 2). Please refer to Lines 400-404 of the revised manuscript.

Ambient RH showed an abrupt decrease from nearly 100% at 10:00 to 85% at 11:00 on 17 Jan (Figure 1). The entrained particles originated from northern air mass might have insufficient supersaturation to activate as cloud droplets. It leads to a very abrupt change in Nf of particle types from cloud residues to ambient particles on Jan 17. Please refer to Lines 382-390 of the revised manuscript.

Page 15, lines 322-325: Do verify the possible transport of biomass burning particles to the site, the vertical history of the trajectories is required.

We have added the vertical evolution of the trajectories. The beginning of trajectories traversed at low heights (about 0-2 km above model ground) of Southeast Asia, where abundant fire dots occurred. Please refer to the Figure 2 (b).

(a)



(b)

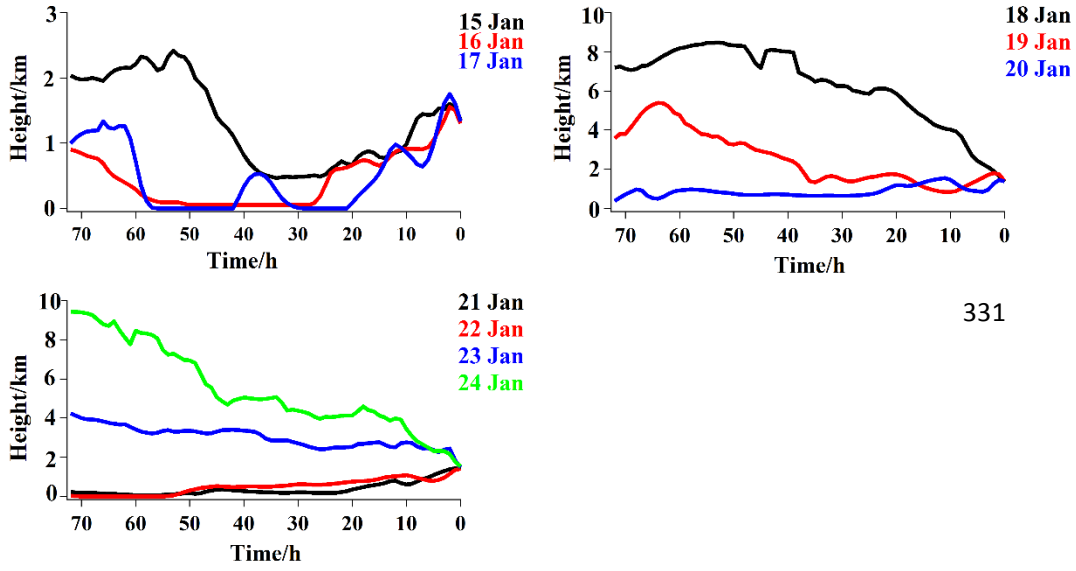


Figure 2: (a) HYSPLIT back trajectories (72 h) for air masses at 1,800 m during the whole sampling period. The white borders and circle refers to the Pearl River Delta (city cluster 1) and Yangtze River Mid-Reaches city clusters (city cluster 2), respectively. The yellow rots represent fire dots during the study periods. The fire dots are available at https://earthdata.nasa.gov/; (b) Heights (above model ground) of the air masses as a function of time.

Page 16, lines 337-229: "Note that after the activation of amine particles, the partitioning of the gas amine on cloud droplets may further contribute to the enhanced Amine cloud residues". That is true, but holds also for other species, as nitrate (HNO3) or water-soluble OC.

We have strengthened the important contribution of uptake of gaseous HNO_3 or water-soluble OC to cloud droplets. Please refer to Lines 334-335, 364-366 and 490-494 of the revised manuscript.

Section 3.5: Here I have a major concern: You report interstitial particles containing sulfate and nitrate in the size range between 200 and 1300 nm. It is very hard to believe (not to say impossible) that such large particles are not activated in a cloud.

Later (page 17, lines 366-368) you write " However, few studies have focused on this issue, in part because interstitial particles show a smaller size than that detected by single-particle mass spectrometry (Roth et al., 2016)." Since the SPAMS has a very similar lower detection size range as the ALABAMA used by my group in Roth et al., 2016), you can not expect that you detect non-activated interstitial particles which should be in the size range below 200 nm.
My suspicion is: The clouds became thinner, and entrainment of cloud-free air has mixed "normal" aerosol particles into the cloud. But such particles cannot be referred to as "interstitial". As long as you don't have cloud microphysics (number and size of cloud droplets) or at least liquid water content (Particle Volume Monitor) available, I would suggest to remove this chapter on interstitial particles.

The period of collecting interstitial particles on 22-23 Jan encountered initial mixing of northerly cloud-free air (dry and cold airstreams) and southwesterly cloudy air (moist airflows). The dry northern air mass might lower supersaturation, only larger particles could be activated. This might result in above 200 nm non-activated particles observed

here (Mertes et al., 2005; Kleinman et al., 2012; Hammer et al., 2014). To make it more accurate, we prefer to name "non-activated particles", rather than "interstitial particles". We have clarified them. Please refer to Lines 453-461 of the revised manuscript.

Page 17, line 358 / Table 1: I would prefer a graph with bars or pie charts. I also strongly recommend showing the SMPS size distributions from residues, ambient and interstitial. That might help to identify the issues with the large interstitial particles.

Table 1 was replaced by pie charts. Please refer to Figure 7 in the revised manuscript. The SMPS size distributions from residues, ambient and interstitial particle were provided in Figure S2.

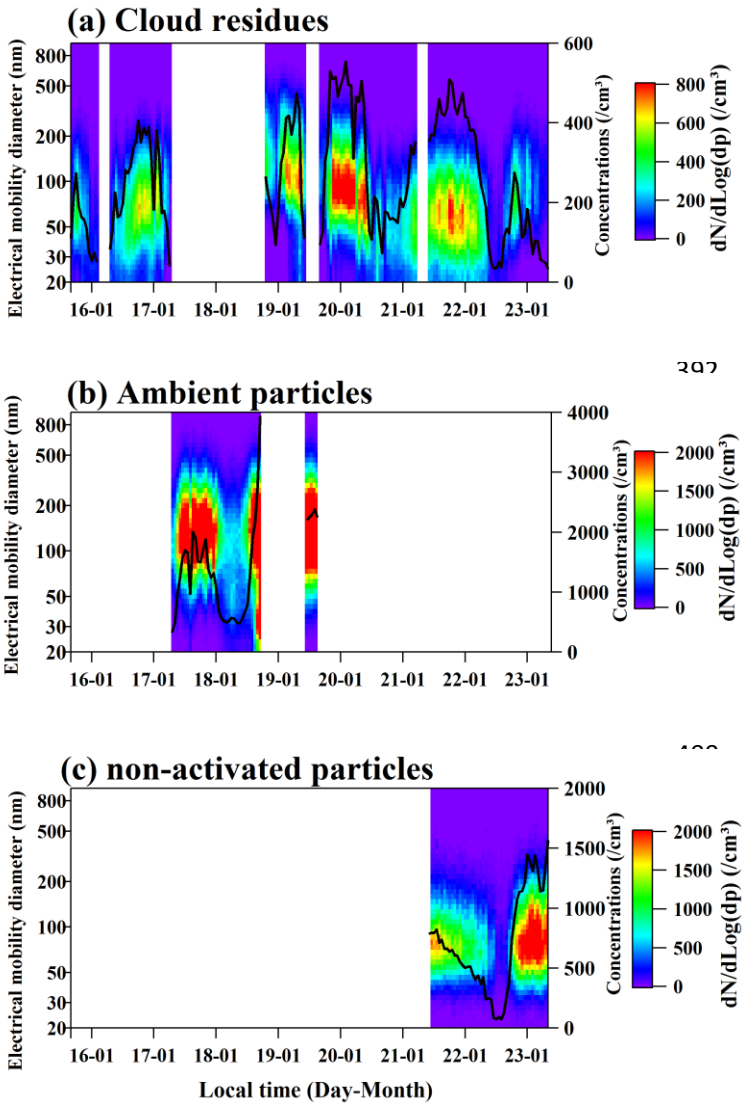


Figure S2: Size distribution (electrical mobility diameter 20-900 nm) of cloud residues (a),

ambient (b) and non-activated (c) particles were measured a scanning mobility particle sizer (SMPS). Black lines represent particles concentrations integrated by the SMPS. The data of cloud residual concentrations was corrected by enrichment factor of 5.25.

Page 18, lines 374-375 / Figure 7 & 8: How are the difference mass spectra of Figure 7 and 8 calculated? Is it ambient - residues and interstitial – residues? Or vice versa? How were the spectra normalized? Please explain.

We have provide the information in the captions of Figures 8 and 9 of the revised manuscript. Please refer to Lines 484-485 of the revised manuscript.

Figures 8: Mass spectral subtraction plot of the average mass spectrum corresponding to cloud residues minus ambient particles. Positive area peaks correspond to higher abundance in cloud residues, whereas negative area peaks show higher intensity in ambient particles.

Figure 9: Mass spectral subtraction plot of the average mass spectrum corresponding to cloud residues particles minus non-activated particles. Positive area peaks correspond to higher abundance in cloud residues, whereas negative area peaks show higher intensity in non-activated particles.

Page 18, lines 376-382: Why not? I drew the same conclusion as Hayden et al. (2008) in my 2017 paper (Schneider et al., 2017, please note the update from ACPD 2016 to ACP 2017). HNO3 uptake may not be the source of the particles but explains the high amount of nitrate found on many particles, also on the Na-rich and dust particles discussed above.

We agree with the comment. We have strengthened the contribution of uptake of gaseous HNO_3 to the enhanced nitrate in the cloud residues. We have update the citation of Schneider et al. (2017) from ACPD 2016 to ACP 2017. Please refer to Lines 334-335 and 490-494 of the revised manuscript.

Page 18, lines 384-386: I agree with that, but wouldn't that support the idea of uptake of HNO3 from the gas phase? If the nitrate content does not play the major role in the activation, but more nitrate is found in the residues, that's an argument for HNO3 uptake.

We agree with the comment. We have strengthened the contribution of uptake of gaseous HNO_3 to the enhanced nitrate in the cloud residues. Please refer to Lines 334-335 and 490-494 of the revised manuscript.

Page 18, lines 387-388: Can intensity simply be compared like this? What about size effects and matrix effects? But again, an explanation how Figures 7 and 8 were calculated might help here.

We agree with the comment. Size and matrix might affect the mass spectra of single particles. Such comparison has been perform in previous single particle studies (Moffet et al., 2008; Pratt et al., 2011). In addition to comparison of certain compound's intensity, its size distribution and number fractions of cloud residues was compared with ambient or non-activated particles, to discuss size effect. Please refer to Lines 487-492 and 504-505 of the revised manuscript.

Figure 8 and 9 show differences in average mass spectra for cloud residues versus ambient particles, as well as cloud residues versus non-activated particles, respectively. Intensity refers to peak area. Please refer to Lines 484-485 of the revised manuscript.

Figures 8: Mass spectral subtraction plot of the average mass spectrum corresponding to cloud residues minus ambient particles. Positive area peaks correspond to higher abundance in cloud residues, whereas negative area peaks show higher intensity in ambient particles.

Figure 9: Mass spectral subtraction plot of the average mass spectrum corresponding to cloud residues particles minus non-activated particles. Positive area peaks correspond to higher abundance in cloud residues, whereas negative area peaks show higher intensity in non-activated particles.

Page 18, lines 391-392: "Compared with interstitial particles, sulfate enhanced in the Fe cloud residues." I think an "is" is missing here.

We have changed "Compared with interstitial particles, sulfate enhanced in the Fe cloud residues." to "Compared with non-activated particles, sulfate was found to enhance in the Fe cloud residues.". Please refer to Lines 503-504 of the revised manuscript.

Page 19, lines 398-399: Better: "The in-cloud process has been reported to be an important pathway: : :"

We have changed accordingly. Please refer to Lines 510-511 of the revised manuscript.

Page 20, lines 421-422: The Jungfraujoch is a station located mostly in the free troposphere and in a remote region, so the biomass burning contribution can be expected to be lower than at other sites.

We agree with the comment. We have discussed less number fraction of biomass burning in the observed cloud residues at the Jungfraujoch station, where located mostly in the free troposphere and in a remote region. Please refer to Lines 251-253 of the revised manuscript.

Figures

Figure 3: Please improve resolution. Labels can't be read upon zooming in.

We have improved Figure 3 resolution. Please refer to the modified Figure 3. Average mass spectrum of Pb, OC and Other types have been moved to the supplemental information (Figure S4)

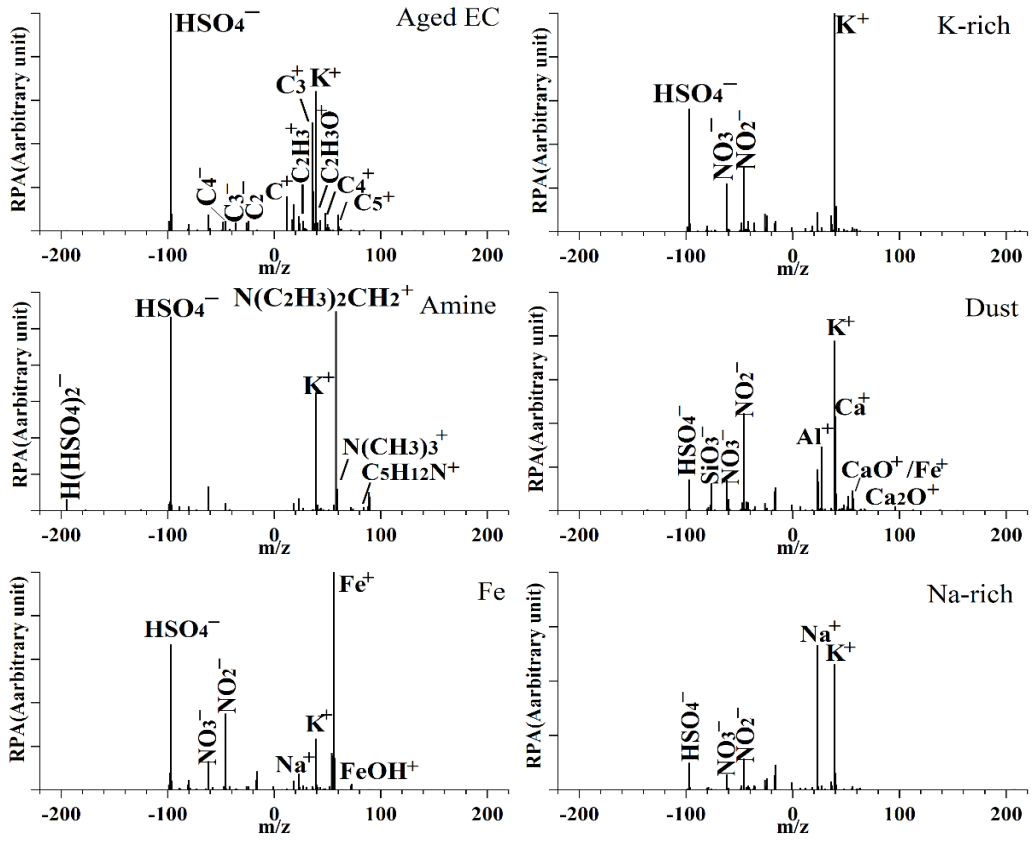


Figure 3: Averaged positive and negative mass spectra for the main 6 particle types (Aged EC, K-rich, Amine, Dust, Fe, Na-rich) of the sampled particles during the whole sampling period. RPA in the vertical axis refers to relative peak area. m/z in the horizontal axis represents mass-to-charge ratio.

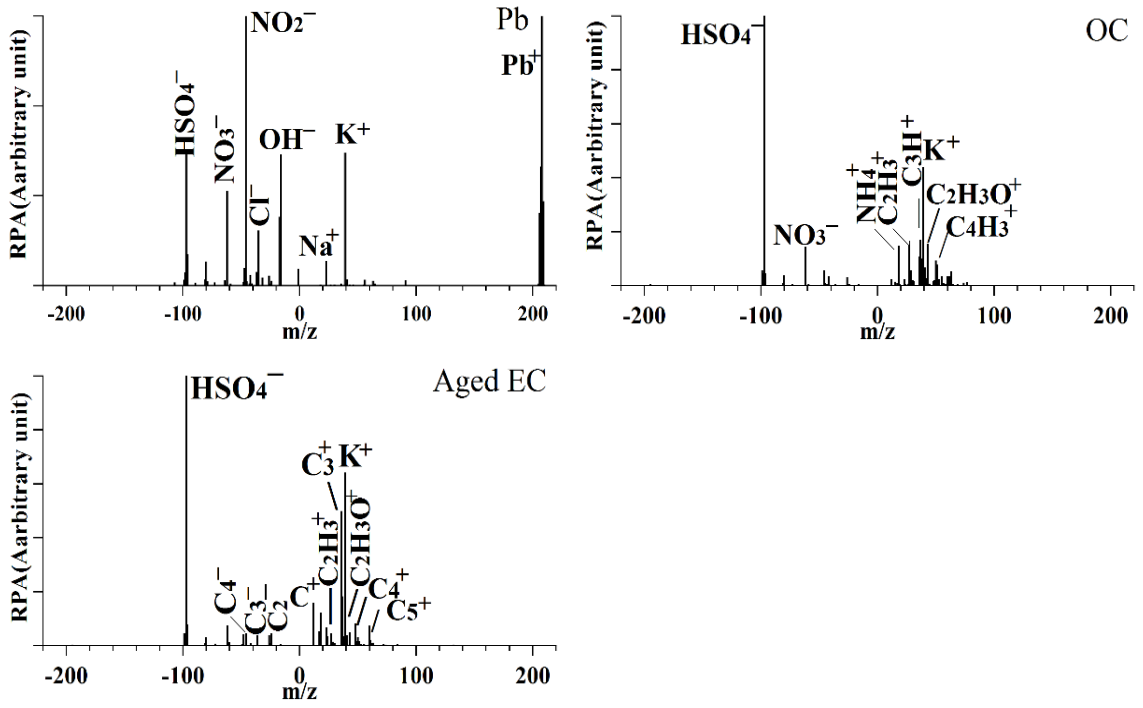


Figure S4: Averaged positive and negative mass spectra for Pb, OC and Other types of the sampled particles during the whole sampling period. RPA in the vertical axis refers to relative peak area. m/z in the horizontal axis represents mass-to-charge ratio.

Figure 6: The ambient particle time series (b) are broader than the corresponding gaps in (a). Please make the Figure broader. You can move the legend with the particle types to above or below the graphs, plus the legend is only needed once.

Ambient and cloud residues were collected at the same hour, which lead to ambient particle time series (b) broader than the corresponding gaps in (a). Figure 6 has been changed accordingly. Please refer to the modified Figure 6.

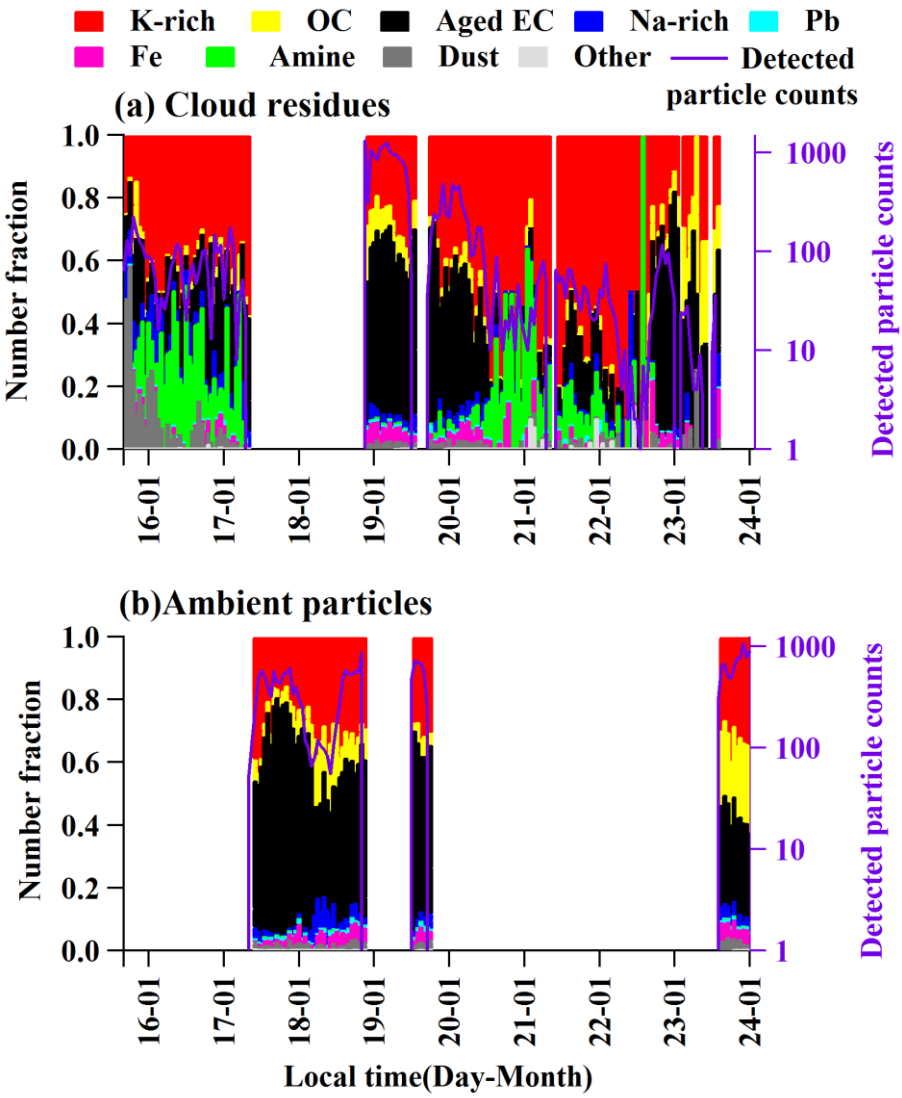


Figure 6: The hourly average variations in the cloud residual and ambient particles during the whole sampling period.

References:

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