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

Anonymous Referee #1

Lin et al describe measurements of the chemistry of cloud droplet residues at a mountain-top site in South China in January 2016. Few measurements of cloud droplet residual chemistry exist, so these are important measurements to help improve our knowledge of cloud formation and properties, which are important for predicting weather and climate. Detailed comments follow.

We would like to thank the reviewer for his/her useful comments and recommendations to improve the manuscript. We agree with the comments, and careful revision has been made according to the suggestions.

It would be helpful for the authors to provide additional information about the cloud events. Please provide, at minimum, ambient temperature during the cloud events to justify the presence of cloud droplets only and no influence from ice crystals would be useful to discuss (related to Line 123); this is shown in Fig 1 and would be useful to refer to earlier to justify the presence of cloud droplets only.

We agree with the comments, and additional information has been added in the revised manuscript as suggested: Ambient temperature on average was 6.9 ℃ (ranging from -7.2 to 11.4 ℃) during the cloud events in this study. Therefore, the clouds here consisted of liquid droplets only. Please refer to Lines 124-126 of the revised manuscript.

It is stated on Line 135 that previous studies found an average cloud droplet size of 10 um at this site, but the distribution is not discussed, nor is the time of year of the previous measurements. Since this work is published in Chinese, these information are not easily obtained by the reader. So, additional discussion would be helpful.

Measurement of drop size spectrum in this region performed during winter of 1999-2001 shows that size of cloud droplets ranged from 4 to 25 μm, with average size of 10 μm and a corresponding liquid water content of 0.11-0.15 g m^{-3} (Deng et al., 2007). Some studies in other locations also showed an average size at ~10 μm (Freud et al.,

2008; Shingler et al., 2012). Therefore, it is reasonable to select a cut size at 8 μm for cloud droplets in the present study. The discussion has been added in section 2 as suggested, Please refer to Lines 126-131 of the revised manuscript.

Is it correct that measurements of cloud droplet size were not completed during this study? It would be helpful to know what fraction of the cloud droplet population was sampled, given the cut size of 8 um. For interpretation of the comparison between the cloud droplet residues and ambient particles, it is important to understand what fraction of the cloud droplets were measured. Previous studies (e.g. Bator & Collett 1997, J. Geophys. Res.) have found that cloud chemistry varies with droplet size. Sampling only the larger cloud droplets may also bias the cloud droplet residue size to larger particles, which is one of the observations. Since the cloud droplet activation process is also size-dependent, it is not possible for the reader to evaluate measurement vs droplet activation size dependencies currently.

It is true that measurements of cloud droplet size were not completed during this study. We agree with the comment. Sampling only the larger cloud droplets may also bias the cloud droplet residue size to larger particles. Previous measurements found that dust, playa salts or sea salt particles are often enriched in larger cloud droplets (~20 μm) (Bator and Collett, 1997; Pratt et al., 2010b). Organic carbon tends to be enriched in small cloud/fog droplets, extending to 4 μm (Herckes et al., 2013). It has been clarified that cloud droplets above 8 μm were sampled by the GCVI. Thus, it partially leads to relatively larger fractions of the Dust and Na-rich cloud residues observed, while the fraction of the OC cloud residues might be underestimated. Please refer to Lines 320-325 of the revised manuscript.

Major Comments:

Lines 20-23, lines 220-221, Figure 4, & numerous other locations: Do these number fractions take into account the size bias in the instrument inlet transmission efficiency, which is clear in Figure S3? It is clear that there are particle size dependencies to the cloud residual chemical composition, particularly for the amine and aged EC particle types, that should be considered when reporting fractions. For example, even on lines 220-221, it is not clear if the authors are reporting 3.8% of the total cloud residues, or 3.8% of the particles measured from 0.7-1.9 um, or 3.8% across each of the size bins from 0.7-1.9 um.

We agree with the comments. The chemical composition of cloud residues is dependent on the particle size (Roth et al., 2016), and the number reported for each particle type

might suffer the bias from size-dependent transmission efficiency (Qin et al., 2006). The relative fraction of cloud residues in 100 nm size interval is presented to minimize the size-dependent transmission efficiency of single particle mass spectrometry (Roth et al., 2016). Similarly, we have provided information on the number fractions of amine and aged EC particle types in cloud residues with size. Nf of the aged EC residues significantly decreased from 54.1% in the size range of 0.2-1.0 μm to 19.2% in the size range of 1.1-1.9 μm. The Amine particles contributed to 3.8% by number of the total cloud residues. Moreover, higher Nf of the Amine residues was detected in size range from 0.7 to 1.9 μm relative to size range from 0.2 to 0.6 μm (16.7% versus 0.4%). Please refer to Lines 20-23, 223-230 and 260-263 of the revised manuscript.

Lines 64-68: Only two other cloud studies are mentioned, or referenced, here, which incorrectly suggests to the reader that the measurement of anthropogenic particles in clouds has only been measured twice. While not all papers need to necessarily be referenced here, it is important to provide a comprehensive view to the reader.

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. Anthropogenic particles can increase number concentration of small cloud droplets, in turn, affect reflectivity and life time of clouds (Stier et al., 2005; Lohmann et al., 2007; Rosenfeld et al., 2008). 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.

Lines 87-89: This list is not comprehensive and is missing many papers. The authors are strongly suggested to conduct a detailed literature search, as comparison of their results with these papers is important.

We agree with the comment, and we have added related references about combined technique of a CVI and AMS or single particle measurement. These studies were mainly conducted in North America including Wyoming (Pratt et al., 2010a), Ohio (Hayden et al., 2008), Oklahoma (Berg et al., 2009), Florida (Cziczo et al., 2004; Twohy et al., 2005), California (Coggon et al., 2014), Europe including Schmücke (Roth et al., 2016; Schneider et al., 2017), Jungfraujoch (Kamphus et al., 2010), Åreskutan

(Drewnick et al., 2007), Scandinavia (Targino et al., 2006), Arctic (Zelenyuk et al., 2010), Central America (Cziczo et al., 2013), West Africa (Matsuki et al., 2010) and Oceans (Twohy et al., 2009; Twohy et al., 2008; Shingler et al., 2012). Please refer to Lines 81-88 of the revised manuscript.

Line 205 & Figure 3: Provide possible ion marker formulae here. Provide a reference for the aged EC type based on comparison to other single-particle mass spectrometry studies (e.g., reference to Moffet and Prather 2009, PNAS). Given the strong K+ signal here, it is likely that both the "Aged EC" and "K-rich" are from biomass burning. Similarly, the OC particles are likely aged biomass burning particles as well, given the strong K+ signal.

Possible ion marker formulae (m/z 27 $C_2H_3^+$, 43 $C_2H_3O^+$) were provided as suggested. We also noted that the aged EC type is similarly observed by other single-particle mass spectrometry studies (e.g., Moffet and Prather 2009, PNAS). We agree with the comment that the "Aged EC", "OC" and "K-rich" might be from biomass burning， as also discussed later. We have discussed the possibility, please refer to Lines 219-222, 232-233 and 314-315 of the revised manuscript.

Lines 209-210: The prevalence of wildfires, shown in Figure 2, suggests that these particles are primarily from biomass burning (see Pratt et al. 2011, ACP for a single particle mass spectrum of fresh biomass burning particles). Pratt et al. (2010, J. Atmos. Sci.) and Hudson et al. (2004, J. Geophys. Res.) discuss the identification of aged biomass burning particles by single-particle mass spectrometry. It seems that the authors can say with greater certainty the source of these particles, and discussion of this would elevate the paper by providing another evaluation of the influence of biomass burning aerosols on cloud activation, which is important and interesting.

We greatly appreciate the comment. Pratt et al (2011) has been cited here to identify the K-rich particles as biomass burning origin. Related references (Hudson et al., 2004; Pratt et al., 2010) were also cited to discuss the identification of aged biomass burning particles and to evaluate the influence of biomass burning aerosols on cloud activation. Majority of the K-rich cloud residues observed here are expected to originate from long-range transportation. Aging process during long-range transportation can increase soluble species (e.g., sulfate, nitrate and oxalate) in the K-rich particles, in turn, improve CCN activity. Please refer to Lines 232-238 and 464-468 of the revised manuscript.

Lines 221-227: It is not clear if Roth et al. observed a decreased fraction of amines in

the clouds compared to the ambient, or if amines just do not influence the site. The presence and behavior of amines would be location and season dependent, so this discussion is not clear and seems to be comparing studies without considering commonalities.

We agree with the comment that the presence and behavior of amines would be location and season dependent. We have made it clear by comparing an Nf of amine-containing aerosol between cloud residues and background aerosol (9% versus 2% by number) in Roth et al. (2016). It indicates a preferential formation of amine in cloud. Aqueous reaction improving the participation of amine has been observed in Guangzhou (Zhang et al., 2012a) and Southern Ontario (Rehbein et al., 2011). Please refer to Lines 263-267 of the revised manuscript.

Lines 244-247: To further understand the source of the Na-rich particles, were they present for both coastal and non-coastal wind directions? Is there a difference in the average mass spectra for these wind directions considering the minor peaks? Are industrial sources located in both directions?

Na-rich particles were resulted from varied sources, including industrial emissions, sea salt or 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, related sea salt ion peak area (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 ± 2.4 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.

Lines 263-267: While ammonium nitrate does not contribute to the dust nitrate observed here, it is not appropriate to generalize this statement to suggest that previous studies in other very different locations did not measure ammonium nitrate.

We agree with the comment. We have revised this sentence to "in this region, ammonium nitrate was not a predominant form of nitrate in the Dust cloud residues", to make it clear. Please refer to Lines 339-340 of the revised manuscript.

Lines 319-329: Was there a mass spectral difference in the K-rich particle type between the N and SW wind directions that would aid in source identification for the two air masses?

A similarity in averaged mass spectrum of the K-rich residues was found for the southwesterly and northerly air masses (Figure S6). Please refer to Lines 419-420 of the revised manuscript.

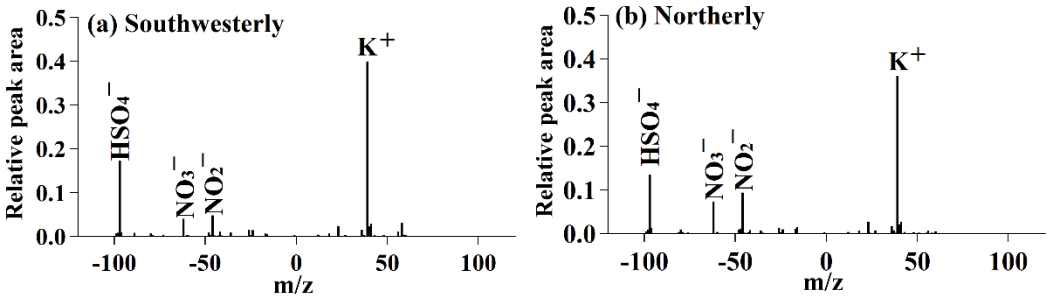

Figure S6: Average mass spectra of K-rich residues for southwesterly (a) and northerly (b) air masses.

Section 3.6: This authors should consider incorporating this discussion into the other sections of the manuscript so that comparisons are made when results are discussed. In addition, the authors should consider differences in atmospheric composition at the various sites. when discussing specific comparisons (i.e. are the contributing sources and magnitudes the same, or may this be a reason for differences? Or, are the seasons the same?). Currently only a general statement on lines 431-433 states that differences are specific to geographic location. As noted above, the literature cited is also not comprehensive, and this section would benefit from additional literature searching.

We agree with the comment. We have incorporated this discussion into the other sections of the manuscript. We have also discussed same or different reason for sources and magnitudes of cloud residues at various sites. We have also cited related literatures (Drewnick et al., 2007; Twohy et al., 2008; Twohy et al., 2009; Matsuki et al., 2010; Kamphus et al., 2010; Pratt et al., 2010b; Zelenyuk et al., 2010; Roth et al., 2016; Bi et al., 2016). Please refer to Lines 241-253, 273-275, 464-471, 489-494 and 505-509 of the revised manuscript.

Figure 4: It would be useful to add comparisons to the ambient and interstitial particles here. In addition, since the cloud residue types changed significantly based on air mass

origin (N vs SW), as stated in Section 3.4, it would be useful to separate out these wind directions and show the fractions of cloud residue, interstitial, and ambient, separated for the two wind directions.

We agree with the the comment. The fractions of the ambient and non-activated particles were provided in Figure 7. The fractions of cloud residue in comparison to ambient particles was performed based on northerly air mass. Please refer to section 3.4 of the revised manuscript.

During the sampling period, the cloud events occurred once the southwesterly air masses were dominant. Therefore, a comparison between cloud residues and ambient particles cannot be addressed under the influence of southwesterly air masses. Please refer to Lines 445-448 of the revised manuscript.

A comparison of cloud residues and non-activated particles has been performed. However, from 22 to 23 Jan during cloud III events, the air mass encountered initial mixing of cloud-free air originated from north and cloudy air originated from southwest. Therefore, a comparison of cloud residues and non-activated particles was not performed for a special wind direction during cloud III events. Please refer to section 3.5 of the revised manuscript.

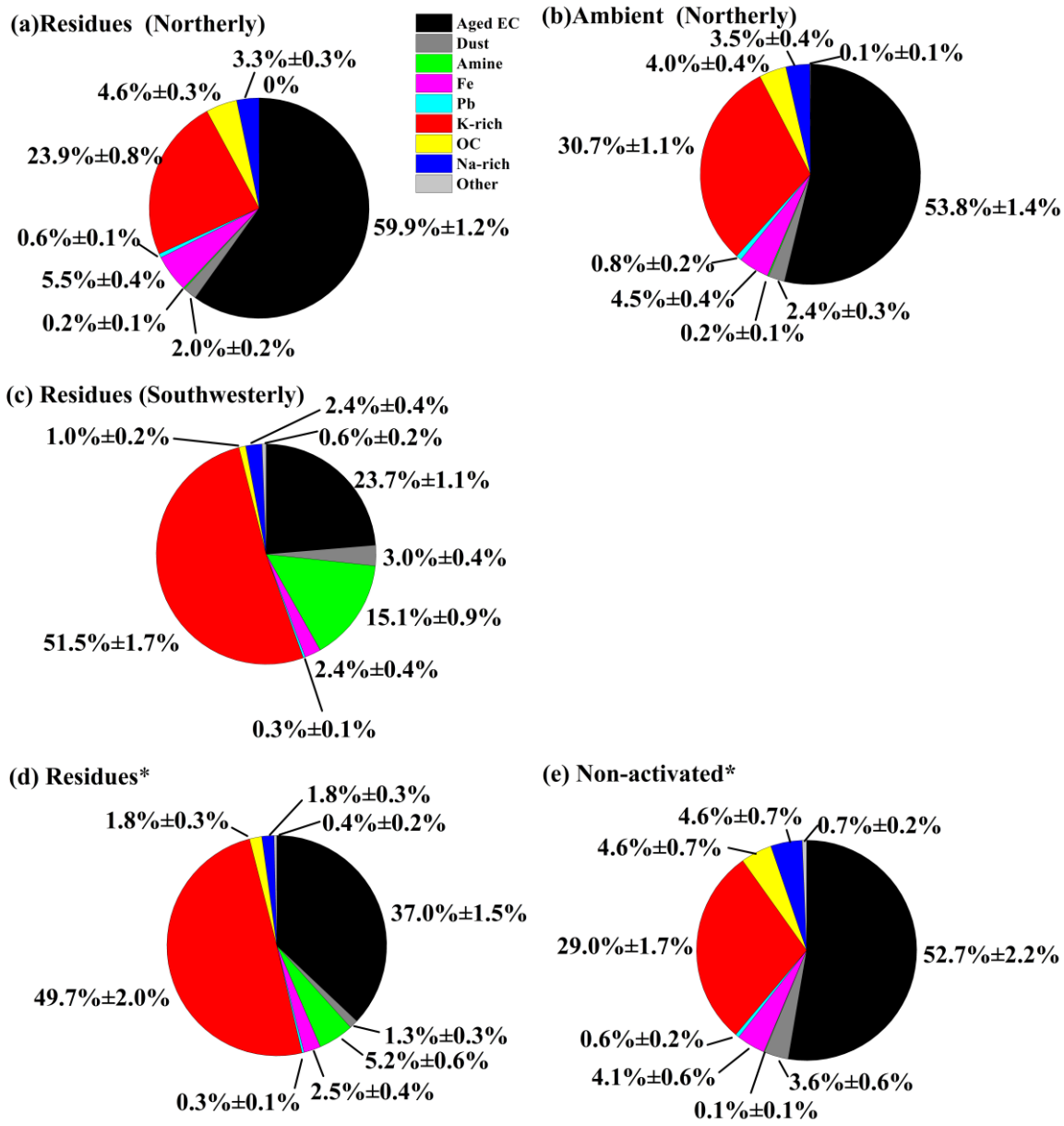

Figure 7: Number fraction of the cloud residues, ambient and non-activated particles. (a) cloud residues during northerly air mass; (b) ambient particle during northerly air mass; (c) cloud residues during southwesterly air mass; (d) cloud residues and (e) non-activated particles were alternately sampled with interval of one hour during the cloud III event; Uncertainties were calculated assuming Poisson statistics for analyzed particles.

Technical Comments:

Lines 24-26, 45, & other locations (search document): Please clarify what is meant by "intensity" in these statements.

Intensity refers to average ion peak area. We have clarified them. Please refer to Lines 24, 43-44 and 486 of the revised manuscript.

Lines 29-30: The phrase "To estimate how atmospheric aerosol particles respond to chemical properties of cloud droplets" is not clear since aerosols determine cloud droplet chemistry, outside of aqueous processing from dissolved trace gases.

The phrase has been changed to " To estimate how atmospheric aerosol particles interact with chemical composition of cloud". Please refer to Line 27 of the revised manuscript.

Lines 41-42: For readers not familiar with the region, it would be helpful to know the suggested source of the amine particles, which the authors are presumably referring to with respect to the wind direction change.

Sources of the amine particles (e.g., ocean and livestock areas) were provided under the influence of southwesterly air masses. Please refer to Lines 40 of the revised manuscript.

Line 44: What does "highly associated" mean in this context? This phrase occurs in other locations in the manuscript as well.

We have modified them. Higher Nfs of nitrate (88-89%) were found in the Dust and Na-rich cloud residues relative to sulfate (41-42%) and ammonium (15-23%). Please refer to Lines 41-43 and 530-532 of the revised manuscript.

Lines 76-80: This sentence incorrectly cites Pratt et al 2010a for cloud droplet residues. The authors should also consider Drewnick et al 2007, J. Atmos. Chem., who did observe lower sulfate mass fractions for droplet residues compared to ambient aerosol. Pratt et al 2010 (J. Geophs. Res.) shows increased mixing with sulfate/nitrate for liquid clouds, compared to ice clouds, as another example.

We have deleted the citation of Pratt et al 2010a and added a citation of Drewnick et al 2007 here. Please refer to Lines 74-75 of the revised manuscript.

A comparison of liquid clouds and ice clouds is beyond the scope of this work, Pratt et

al 2010 (J. Geophs. Res.) was not cited here.

Line 161: It should be clarified that "cloud droplet residue concentrations", not "cloud droplet concentrations" were measured by the SMPS. This is an important distinction.

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

Lines 163-165: Wouldn't hazy days with low visibility be characterized by high, rather than low, PM2.5 concentrations? This is confusing.

Low level of $PM_{2.5}$ (~ 12.7 μg m^{-3}) excludes the influence of hazy days. Please refer to Lines 169-170 of the revised manuscript.

Line 165: Change "rainy" to "rain".

We have changed "rainy" to "rain" accordingly. Please refer to Line 171 of the revised manuscript.

Line 216: It is important to consider the relative enhancement in amine peaks when using a 266 nm laser and that the amines themselves may not comprise the majority of the particle mass. See Pratt et al. 2009, Environ. Sci. Technol.

We have emphasized the effect of 266 nm ionization laser on amine peaks (Pratt et al., 2009). Please refer to Lines 257-259 of the revised manuscript.

Lines 230-232: It should be clarified when discussing previous vs the current study, and if previous studies are being discussed, the season should be noted if there are seasonal variations.

We agree with the comments. We have clarified the specific season in the discussion. At Mt. Tai in northern China, a high concentration of Ca^{2+} in cloud/fog water was mainly attributed to a sandstorm event during spring season (Wang et al., 2011). At Mt. Heng in southern China, abundant crust-related elements (e.g., Al) observed in cloud water is due to Asian dust storms occurring on March−May (Li et al., 2017). Based on backward trajectory, the site in this study was less affected by sandstorm source in northwestern China during cloud events. Local dust emission by anthropogenic-disturbing soils or removing vegetation cover can be excluded as a result of forest

protection. Therefore, a low fraction (2.9% by number) of dust cloud residue is acceptable in the present study. Please refer to Lines 278-287 of the revised manuscript.

Lines 231-237: Some grammar fixes are needed here.

The language has been edited by a native speaker.

Line 279: Please be more specific with the statement "plays a key role in cloud processes". you mean that these particles were preferentially activated? Is there evidence of this?

We have reworded this sentence: "This result also implies that ammonium-containing particles are preferentially activated or enhanced by uptake of gaseous NH_3 to neutralize acidic cloud droplets for the OC and EC types. Please refer to Lines 354-356 of the revised manuscript.

Lines 358-396: The phrasing in these paragraphs should be improved for greater clarity and correct grammar.

These paragraphs have been reworded. Please refer to Lines 463-509 of the revised manuscript.

Line 390: This is an important finding, yet the phrasing "sulfate was observed to diminish" not clear, particularly when considering the following sentence. Please clarify.

We have rephrased and changed to "sulfate intensity was observed to diminish". Please refer to Lines 502-503 of the revised manuscript.

Lines 395-396: What discrepancy? This is not clear.

The discrepancy refers to "the mass or number fraction of sulfate-containing particles in the cloud residues changed between ambient and interstitial (non-activated) particles (Drewnick et al., 2007; Twohy and Anderson, 2008; Schneider et al., 2017)". We have changed "this discrepancy" to "these changes" to make it clear. Please refer to Lines 505-509 of the revised manuscript.

Line 397: Quantitatively, what is "no remarkable change"? The phrase "remarkable

change" is used elsewhere in the manuscript as well, but it isn't defined.

"no remarkable change": We added the data on number factions when the residual particles were compared with ambient or non-activated particles. "remarkable change" has been modified to "remarkable decrease/increase" and added the data on number factions when the residual particles were compared with ambient or non-activated particles. Please refer to Lines 511-513 and 518-519 of the revised manuscript.

Table 2: Not sure what the authors mean by "way" here.

"way" : Cloud residues and non-activated particles were alternately sampled with interval of one hour during the cloud III event. Table 2 has been replaced by pie charts in Figure 7. Please refer to Lines 452-453 of the revised manuscript and the caption of Figure 7.

Figure 2: The lines and numbers on this map are difficult to read. It would be useful to make the text bold perhaps and increase the width of the lines.

We have changed accordingly. Please refer to the modified Figure 2a.

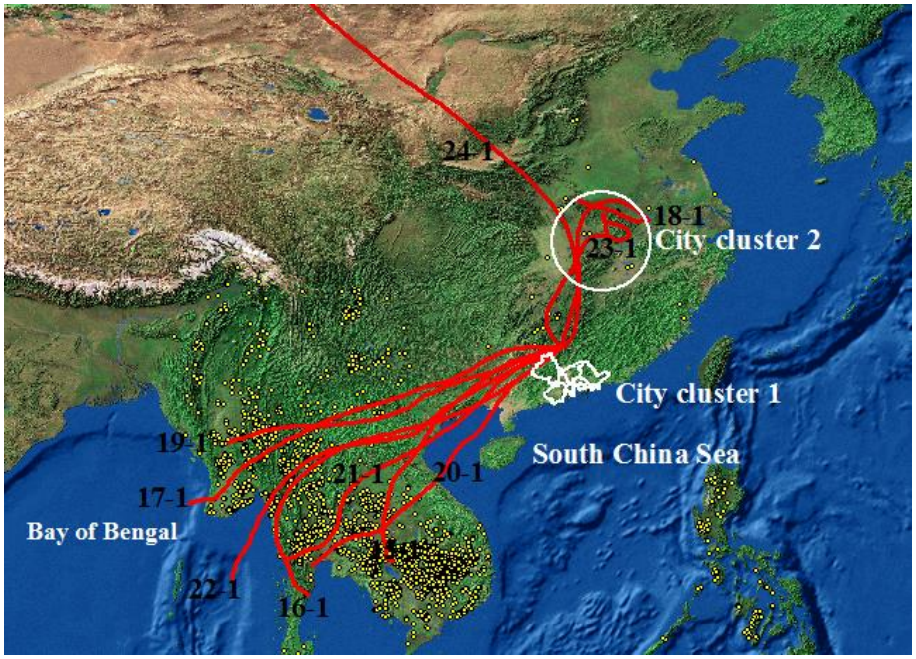

Figure 3: This plot is difficult to read. The authors should consider showing only the most abundant (and discussed) particle types in the main text figure and moving the others (including "Other", which is somewhat meaningless as an average mass spectrum if it is made up of a diverse population of particles) to the supplemental

information.

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)

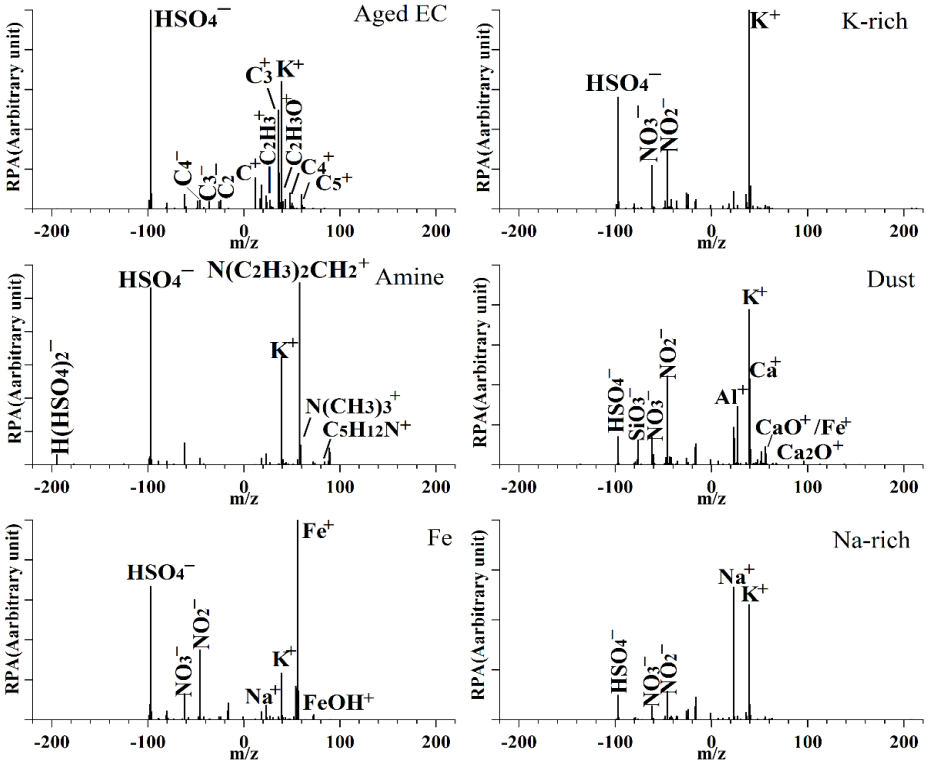

Figure 3: Averaged positive and negative mass spectra for the main 6 particle types (Aged EC, K-rich, Amine, Dust, Fe, Na-rich) of the sampled particles during the whole sampling period. RPA in the vertical axis refers to relative peak area. m/z in the horizontal axis represents mass-to-charge ratio.

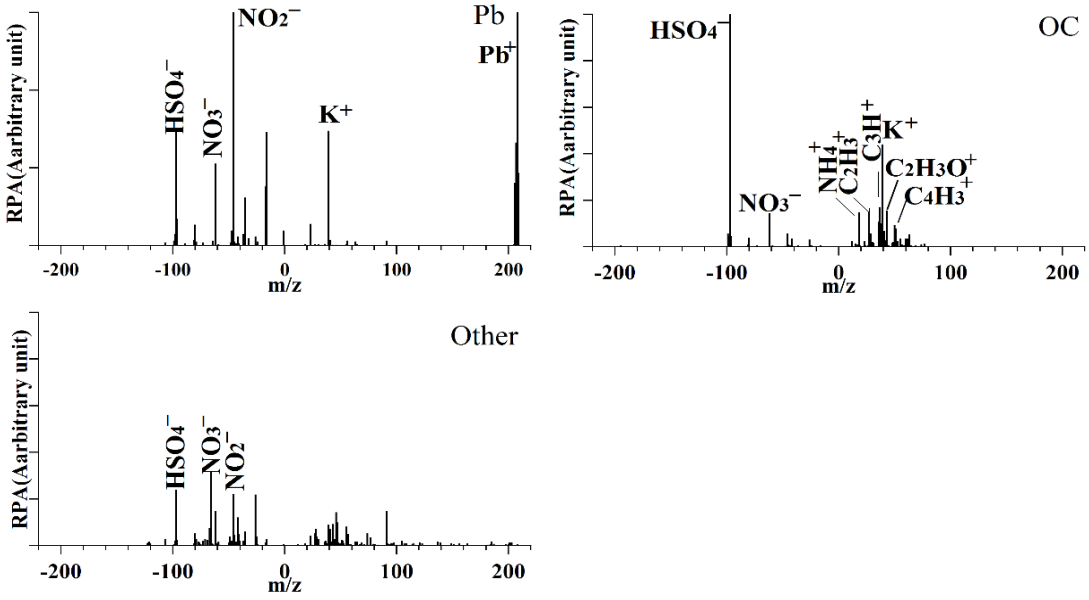

Figure S4: Averaged positive and negative mass spectra for Pb, OC and Other types of the sampled particles during the whole sampling period. RPA in the vertical axis refers to relative peak area. m/z in the horizontal axis represents mass-to-charge ratio.

Figures 7 and 8: Please indicate whether positive peak areas indicate preferentially in the cloud residues and negative indicate preferentially in the ambient/interstitial particles. This is currently not clear in the figure captions.

We have clarified them in the captions of Figures 8 and 9 of the revised manuscript.
Figure 8: Mass spectral subtraction plot of the average mass spectrum corresponding to cloud residues minus ambient particles. Positive peak area corresponds to higher abundance in cloud residues, whereas negative peak area show higher intensity in ambient particles.
Figure 9: Mass spectral subtraction plot of the average mass spectrum corresponding to cloud residues minus non-activated particles. Positive peak area correspond to higher abundance in cloud residues, whereas peak area show higher intensity in non-activated particles.

Figure S4: It is not clear in the maps where RH < and > 90% are located.

We have added contour lines of two relative humidity (50%, 70%) in the Figure S5. Please refer to the modified Figure S5.

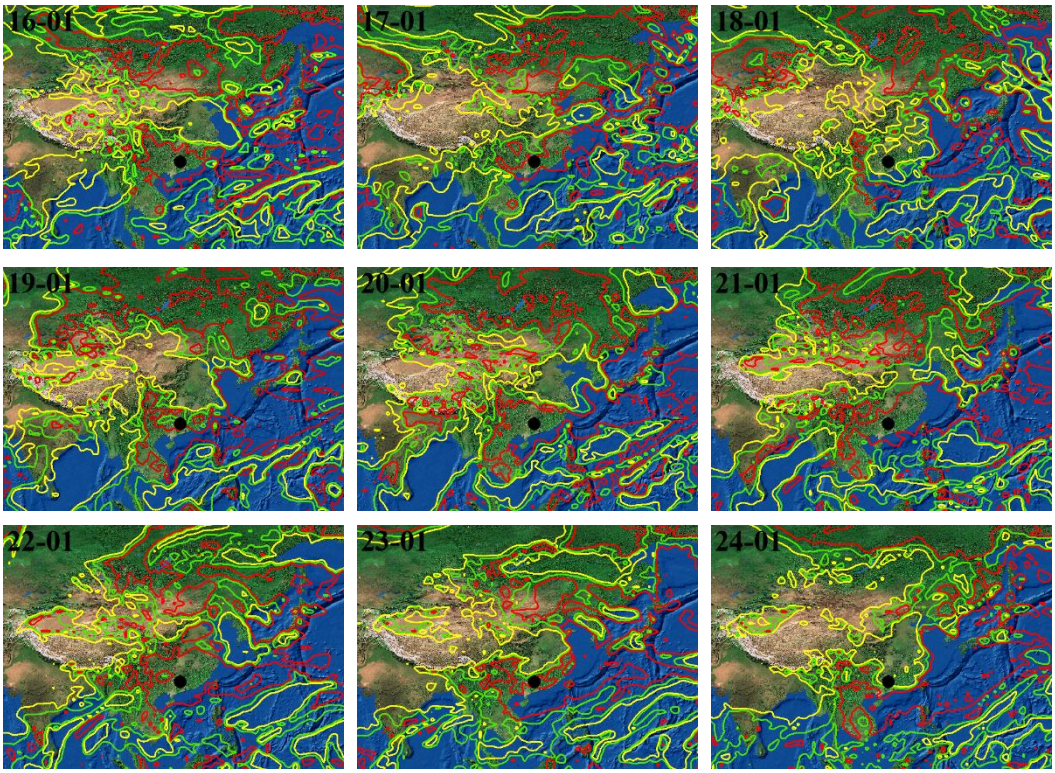

Figure S5: At 850 hPa (about 1,500 m a.s.l), Contour lines (red lines) of relative humidity 90%; Contour lines (green lines) of relative humidity 70%; Contour lines (yellow lines) of relative humidity 50%. Black mark represents the observed site. Data is available at ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/.

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