Response to comments

Reply to the referee #1:

The authors presented single-particle measurements of iron-containing aerosols to analyze the formation of secondary species. The work conducted in this paper may contribute to improve our understanding of effect of iron on the formation of secondary species, although more careful discussions are required for the iron chemistry in aerosol and cloud water. I have some comments and questions to improve this paper.

We would like to thank the referee #1 for his/her useful recommendations to greatly improve our manuscript. All of the comments have been properly addressed in the revised manuscript.

Specific comments

p.9, l.187, Figure S2: Please indicate the upper size of the particles.

Reply: The upper size of particles was 2.0 μm. We have provided the information on the upper size of the particles. Please refer to Lines 214-216 in the revised manuscript and Figure S3 in the revised Supplement.

p.12, l.249: Please present the references and specify the in-cloud chloride formation and irradiation effect.

Reply: Higher NFs of chloride were found in the Fe-containing cloud residues and interstitial particles compared to the cloud-free particles (16 ±1% or 13 ±2% versus 6 ±1%, by number) (Figure 3). The partitioning of volatile gaseous chloride (i.e., HCl) into the aqueous phase was a major contributor to the increased chloride in cloud droplets (Sellegri et al., 2003). On the

other hand, the irradiation effect during daytime would generally result in the reduction of ammonium chloride in the particles due to evaporation at high temperature (Huang et al., 2011). In the case of cloud events, there was no change in NF of chloride in the Fe-containing cloud residues between daytime and nighttime (Figure 4c), suggesting that the amount of in-cloud chloride formation was less affected by irradiation effect. In the case of cloud-free events, no decreased NF of chloride in the Fe-containing cloud-free particles was also observed during daytime relative to the nighttime (Figure 4c), presumably due to the formation of calcium chloride (Sullivan et al., 2007). Please refer to Lines 271-274 and 283-290.

p.12, l.256: Did you measure HCl concentration? What are the sources of HCl in the region?

Reply: The measurement of gaseous HCl has not been conducted in our study. Recently, Fu et al. (2018) reported that the anthropogenic gaseous HCl emission was estimated to be in a range from 20-200 kg/km^2 in southern China, mostly from coal burning, agricultural biomass burning, and waste incineration. The gaseous HCl sources in this region might originate from regional transport of these anthropogenic HCl in southern China. We have clarified them. Please refer to Lines 280-283.

p.15, l.319: The Fenton reaction is a sink of ferric iron. Please consider the redox cycling for the role of the Fenton reaction in the formation of aqueous-phase OH.

Reply: We have added the effect of Fe redox cycling on the formation of aqueous-phase OH through Fenton reaction: "These findings implies that aqueous-phase OH were likely

produced by redox cycling of the Fe-driven Fenton reaction (Hems et al., 2017), and thus enhance oxidation of various precursors to form oxalate during cloud processing." Please refer to Lines 359-361.

p.18., l.378: The photolysis of Fe-oxalate complexes could reduce the net amount of oxalate formation in cloud droplet, even though the Fenton reaction increases the formation rate of aqueous-phase OH (Sorooshian et al., 2013; Lin et al., 2014). Thus, other mechanisms (e.g., strong complexation of iron with organic compounds) are needed to enhance the oxalate concentration in Fe-rich particles. Please consider the effect of Fe chemistry on the net formation of oxalate and discuss the mechanisms to enhance the oxalate concentration in Fe-rich particles.

Reply: We agree with the comment. We have discussed the effect of Fe chemistry on the net formation of oxalate: "Our data showed that aqueous-phase oxidation of the oxalate precursors was an important contributor to the in-cloud formation of oxalate in the Fe-rich particles. The Fe-driven Fenton reaction likely accelerate the conversion of the oxalate precursors to oxalate in the Fe-rich particles during cloud processing. Moreover, the complexation of Fe with oxalate might further increase oxalate content in the Fe-rich cloud residues (Chen and Grassian, 2013), but the photolysis of Fe-oxalate complexes in the Fe-rich cloud residues would lead to the losses of oxalate during daytime." Please refer to Lines 421-428.

p.18., l.384: The formation of stable complexes (e.g., calcium oxalate) is assumed for the

SOA yield to remain entirely in the particulate phase when the water is evaporated. This is different from the suppression of oxalate-promoted Fe dissolution in the model (Ito, 2015), which is based on the laboratory measurements (Chen and Grassian, 2013). At low concentration of oxalate, complexation of oxalate with iron on the surface of the particle is suppressed through formation of Fe-oxalate complexes in solution. Less content of Ca-containing particles suggests low pH in aerosol water. It was proposed that the competition is more significant at low pH. Thus, excess oxalate is needed to enhance Fe dissolution in aerosols. Please consider the effect of the ligand concentration on Fe dissolution in aerosols and discuss the implications.

Reply: We have emphasized the role of oxalate concentration in Fe dissolution in the revised manuscript: "In this work, nearly $45 \pm 1\%$ (by number) of the Fe-rich cloud residues contained only oxalate. The mass concentration of oxalate in the bulk aqueous phase might be overestimated in model simulations. This might suppress Fe dissolution under low concentration of oxalate because of competition of oxalate complexation between dissolved Fe in solution and Fe on the particle surface (Chen and Grassian, 2013)." Please refer to Lines 431-436.

p.19, l.398: Please reconsider this. The Fenton reaction increases the formation rate of aqueous-phase OH, but the net amount of oxalate formation in cloud droplet is reduced by the photolysis of Fe-oxalate complexes. Please also consider the pH upon droplet evaporation when the ammonium species may enhance the SOA yield.

Reply: As suggested by the referee #3, the Sections Atmospheric Implications and

Conclusions have been combined to improve integration of the study results with their impacts. The effect of Fe chemistry on the net formation of oxalate have already been added, as stated above. We have also included the following sentence to mention the contribution of ammonium species to oxalate formation: "Ammonium species possibly enhance oxidation of precursors to form oxalate in the Fe-rich cloud residues through increasing aerosol pH." Please refer to Lines 421-428.

Reply to the referee #3:

Lin et al present an important measurement study of the presence of Fe in individual cloud residual particles, compared to clear air, as well as the presence of secondary species expected to be formed via cloud processing. Given the current interest in Fenton chemistry and importance of considering what fraction of cloud droplets actually contain Fe, this study is quite timely and needed. Overall, the study is well done; however, significant technical corrections and clarifications are needed, as described below.

We would like to thank the referee #3 for his/her useful comments to greatly improve the manuscript. The careful revision has been made according to the suggestions.

Major comments:

As a point of caution in wording, the SPAMS measures the presence of a particular species, but without calibration taking into consideration matrix effects, the mass of that species is not quantified. While much of the paper uses the phrasing of "number fractions", there are many places (e.g. lines 30-31 of abstract, line 394 in conclusions, and many other places) where a reader would expect the authors to be discussing mass concentration, as is the traditional norm of the atmospheric aerosol community. Phrasing should be revised to avoid this potential confusion. For example, on line 32 of the abstract and line 394 of the conclusions, the phrasing "extremely high amounts of sulfate" and "no enhancement in sulfate" is used, but sulfate mass was not measured in this study, rather the number fraction of particles containing sulfate was

measured. This distinction is important, as it impacts the interpretation of results, for example that the authors state "no distinct changes in sulfate during cloud events" (lines 33-34), where the authors did not measure sulfate mass to evaluate this result. The authors need to fix this phrasing (assumption of mass rather than number fraction) and associated conclusions throughout the manuscript. The phrasing such as "number fraction of Fe-containing particles internally mixed with sulfate" would be clearer, for example. Wherever percentages are stated, the phrasing "number fraction", "by number", or similar should be used to avoid confusion. For another example, the authors stated on line 176 "Lithium (m/z 7[Li]+) was found to account for 7% of the Fe-rich type"; I believe the authors mean "7%, by number,", rather than "7%, by mass,", as the typical reader would assume.

Reply: We agree with the comments. The mass concentration of chemical species cannot be quantified by the SPAMS. In order to avoid confusion, the phrasing "number fraction (NF)" or "by number" has been added to state percentages in the modified manuscript. For example, lines 241-243 have been change to "The NFs of sulfate in the Fe-containing cloud residues (93 ± 1%, by number) and interstitial particles (92 ± 4%) were similar to those in the cloud-free particles (96 ± 1%), as shown in Figure 3." Similar statements have been modified throughout the manuscript. Please refer to Lines 30-34, 177-181, 199-202, 205, 211-216, 225-227, 243-245, 256-258, 265-267, 271-273, 276-278, 290-292, 295-296, 299-303, 319-321, 328-331, 333-337, 340-343, 356-357, 369-370, 378-379, 383-389, 391-393, 394-397, and 431-434.

Lines 147-152: In the screening for Fe-containing particles, were particles required to contain m/z 54, to ensure the presence of Fe? This is important and not clear here. It would seem that a range of 56Fe/54Fe is most appropriate, rather than simply a ratio >10, which could mean little m/z 54 present.

Reply: The Fe-containing particles in the current study were identified by the coexistence of peaks at m/z 56 and m/z 54, and their peak area ratio ($^{56}Fe/^{54}Fe$) above 10. The known isotopic ratio of Fe ($^{56}Fe/^{54}Fe = 16$) was used as an indicator for Fe (Beard and Johnson, 1999). The ratio to identify the Fe-containing particles in the atmosphere might be below 16, due to organic peaks at m/z 56 and 54. Considering the varied ratio resulting from the matrix effect of laser desorption/ionization (Allen et al., 2000), Furutani et al. (2011) performed a peak area ratio of m/z 56 to 54 above 3 to identify the Fe-containing particles. However, the average single-particle mass spectra with a ratio of m/z 56 to 54 below 10 can also be obtained in the organic carbon particles (Zhang et al., 2014). Therefore, a high peak area ratio of m/z 56 to 54 above 10 was used in the present study, which had been applied in the previous studies (Zhang et al., 2014; Dall'Osto et al., 2016) to minimize the interference from other species. Please refer to Lines 166-176.

At the beginning of the Results & Discussion, I highly encourage the authors to add a section or several sentences addressing the number fraction of Fe-containing particles out of the total measured for the categories of cloud, interstitial, and clear air. It is

important for those modeling Fenton chemistry to understand the fraction of cloud droplets that contain Fe, and so, I think this information will be very useful to the community.

Reply: We thank the reviewer for this suggestion. A total of 5,682 cloud droplet residues, 395 interstitial particles, and 5,086 cloud-free particles were found to be internally mixed with Fe, representing 3.7 $\pm$ 0.1 % (by number) , 2.6 $\pm$ 0.1% and 3.0 $\pm$ 0.1% of the total measured cloud residues, interstitial particles, and cloud-free particles, respectively. Please refer to Lines 177-181.

Due to matrix effects in LDI, it is not valid to compare peak areas between particle types (e.g. Line 173, Figure S5, Lines 211-213, and other locations). I refer the authors to Hatch et al (2014, Aerosol Sci. Technol.), Gross et al (2000, Analytical Chem.), and Reinard & Johnston (2008, J. Am. Soc. Mass Spectrom.).

Reply: We agree with the comment. The comparisons of peak areas between particle types (e.g., Lines 211-213 and 301-303 as well as Figure S5) have been removed in the modified manuscript.

Lines 209-214 and 259-260: Please check that the percentages being compared are indeed statistically significant. Please report errors in the text as well to provide greater context.

Reply: The errors were calculated assuming Poisson statistics for the analyzed particles in this work. Please refer to Lines 176-177.

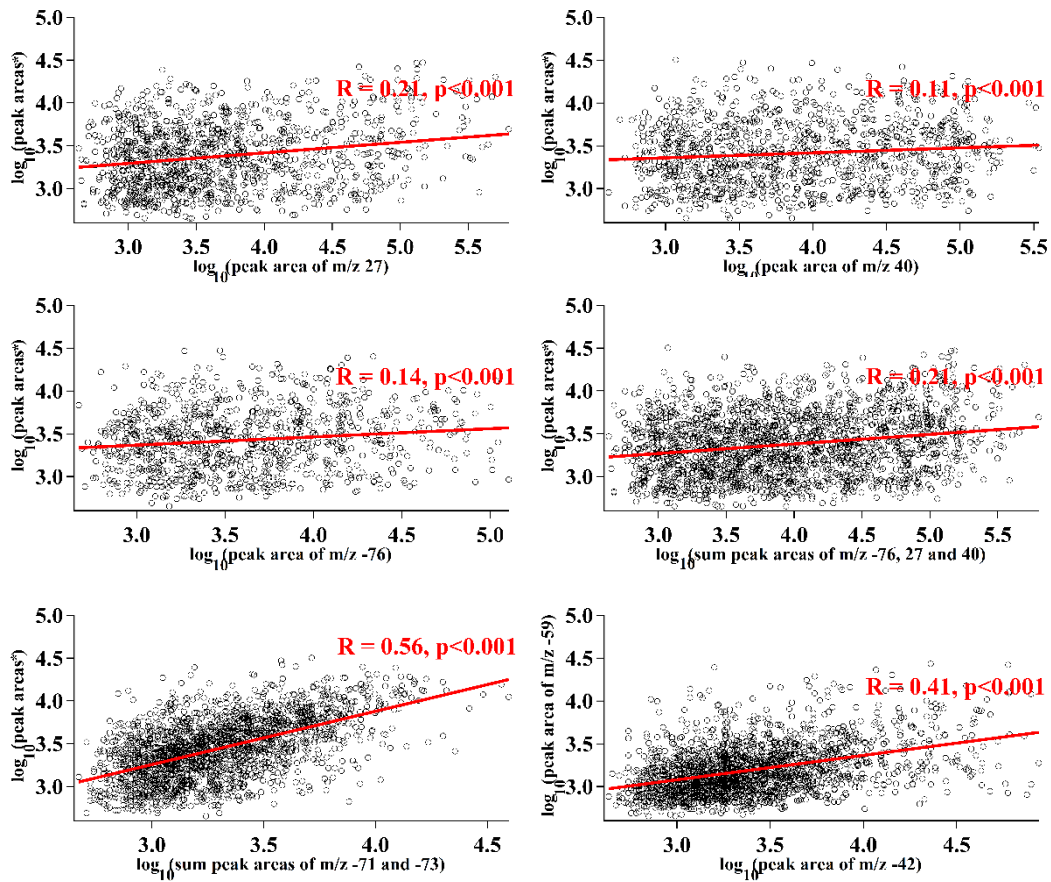
Due to the errors effect on the comparison of percentages, lines 209-214 "The number fractions (NFs) of sulfate in the Fe-containing cloud residues (93%) and interstitial particles (92%) were lower than those in the cloud-free particles (96%), as shown in Figure 3." have been change to " The number fractions (NFs) of sulfate in the Fe-containing cloud residues (93 ±1%, by number) and interstitial particles (92 ±4%) were similar to those in the cloud-free particles (96 ± 1%), as shown in Figure 3." Please refer to Lines 241-243.

Lines 259-256 "The NFs of ammonium in the Fe-containing cloud residues (35%) and interstitial particles (34%) were slightly higher than that in the cloud-free particles (30%) (Figure 3)." have been change to "The NFs of ammonium in the Fe-containing cloud residues (35 ± 1%, by number) and interstitial particles (34 ± 3%) were analogous to those in the cloud-free particles (30 ± 1%) (Figure 3)." Please refer to Lines 299-301.

The errors have also been provided in other places in the modified manuscript. Please refer to Lines 177-181, 199-202, 205, 211-216, 225-227, 243-245, 256-258, 265-267, 271-273, 276-278, 290-292, 295-296, 301-303, 319-321, 328-331, 333-337, 340-343, 356-357, 369-370, 378-379, 383-389, 391-393, 394-397, and 431-434.

Section 3.3: A main result is that Fe-dust particles were more likely to containing oxalate precursors; however, I am concerned about potential ion peak interferences as follows: m/z -59 ($HCNO_2^-$, AlO_2^-), m/z -87 ($AlCO_3^-$), m/z -103 ($AlSiO_3^-$), and m/z -117 ($CaCO_3OH^-$). Either the simultaneous presence of related peaks (e.g. Al^+, Ca^+,

SiO$_3^-$, CNO$^-$) should be investigated, or these ion peaks should not be included when screening for oxalate precursors.

Reply: There is no significant relationship (correlation coefficient = 0.11-0.21) between oxalate precursors (i.e., m/z -59, -87, -103, and -117) and aluminosilicate species (i.e., m/z -76, 27, or 40). A peak of m/z -59 might be the presence of [HCNO$_2$]$^-$, however, the particles including coexistent peaks at m/z -59 and -42 only contribute 3.0 ± 0.1% (by number) to the oxalate precursors that are identified by the presence of peaks at m/z -59, -71, -73, -87, -103, or -117. Higher NF of the oxalate precursors (m/z -59, -87, -103, or -117) in the Fe-containing cloud-free particles during daytime (26±1%,by number) relative to nighttime (20±1%,by number) might suggest not primary source (e.g., aluminosilicate). A positive relationship (correlation coefficient = 0.56) between oxalate precursors (i.e., m/z -59, -87, -103, or -117) and other oxalate precursors (i.e., -71 or -73) is observed. Therefore, we propose that the screening for oxalate precursors using m/z -59, -87, -103, and -117 is less influenced by the interference from aluminosilicate species. Thus, the oxalate precursors in the current study are still identified by the peaks at m/z -59, -71, -73, -87, -103, or -117. Additionally, a peak at m/z -59 might also be produced in the presence of levoglucosan (Bi et al., 2011). We have emphasized the peak at m/z -59 might being from other species in the modified manuscript: "A peak at m/z -59 might also be originated from the presence of [HCNO$_2$]$^-$ or levoglucosan (Bi et al., 2011)." Please refer to Lines 318-319.

Peak areas* in the vertical axis refers to the sum area of peaks at m/z -59, -87, -103, and -117.

Lines 345-346: Since both nitrate and sulfate were present, how can the presence of ammonium nitrate be ruled out?

Reply: Approximately $97 \pm 1\%$ (by number) and $54 \pm 5\%$ of ammonium were found to be internally mixed with sulfate and nitrate in the Fe-EC type respectively, suggesting that ammonium in the Fe-EC cloud residues has a more contribution from ammonium sulfate rather than ammonium nitrate. Please refer to Lines 387-389.

Figure 3: Given the similarities between the cloud residues and interstitial particles, is it possible that the interstitial particles are already cloud processed?

Reply: Previous field measurement observed that median size of cloud/fog droplets was approximately 10 μm at the studied region (Wu et al, 2004). In the current study, the cloud residues and interstitial particles were sampled using the GCVI (aerosol diameter above 8 μm) and cyclone (aerosol diameter below 2.5 μm) inlets, respectively during cloud events. The cloud residues had also been observed to be larger size relative to the interstitial particles (Figure S2). Thus, it was proposed that the interstitial particles had not already processed to become cloud droplet. Please refer to Lines 120-123, 134-138 and Figure S2.

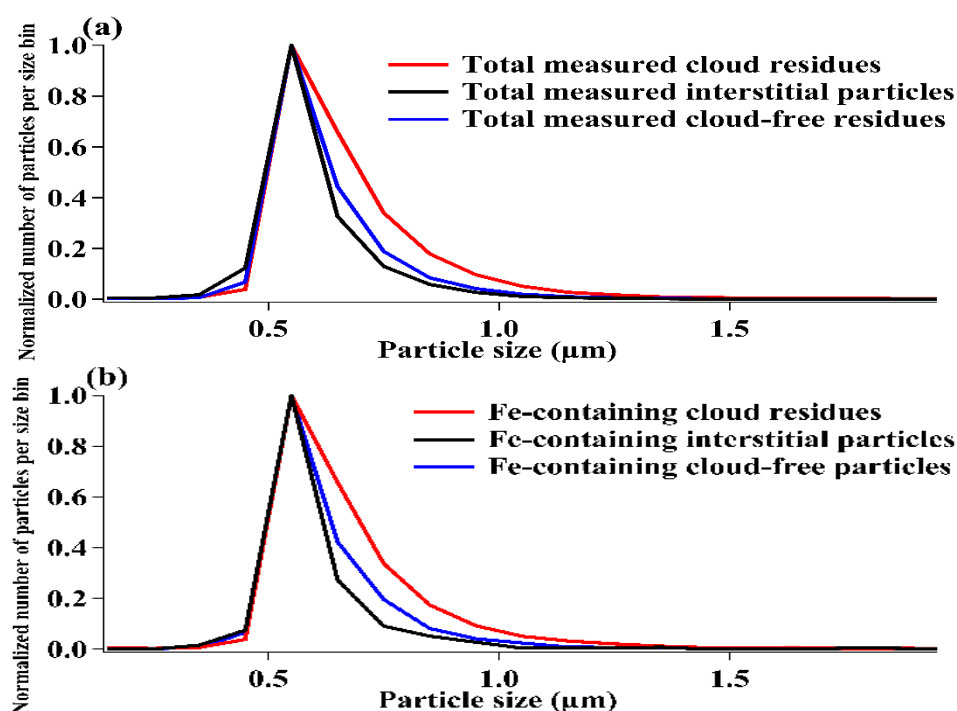



Figure S2. Normalized size distribution of the total measured (a) and Fe-containing (b) cloud residues, interstitial and cloud-free particles.

Technical Comments:

Please clarify the sentence on Lines 38-41.

Reply: The sentence has been change to "Fe-driven Fenton reaction likely increase the formation rate of aqueous-phase OH, improving the conversion of the precursors to oxalate in the Fe-rich cloud residues. During daytime, the decreased NF of oxalate in the Fe-rich cloud residues was supposed to be due to the photolysis of Fe-oxalate complexes." Please refer to Lines 38-42.

Please add a reference to the sentence on Line 47, and provide quantitative and location-based context for "frequently detected". This is surprising to me.

Reply: We thank the reviewer for this suggestion. References and detailed information have been added here "Iron (Fe)-containing particles were frequently detected in the atmosphere, with concentration ranging from 10 ng/m^3 over remote marine environment to 28 μg/m^3 near desert areas (Zhang et al., 2003; Fomba et al., 2013)." Please refer to Lines 48-50.

Line 54: Change "contained" to "can contain".

Reply: "contained" has been changed to "can contain" accordingly. Please refer to Lines 56-57.

Lines 61 & 72: Change "aerosol Fe" to "Fe-containing particles". Watch this phrasing

throughout. It is not correct to say that another chemical species is "in the aerosol Fe".

Reply: We have changed "aerosol Fe" to "Fe-containing particles" in the modified manuscript as suggested. Please refers to Lines 60, 62, 64-67, 69-70, 74, 76, 79, 90-92 and 94.

Lines 75-76: The Sullivan & Prather 2007 paper describes a shipboard study, which would not have been a low RH, as stated here.

Reply: The aerosol liquid water content is smaller by several orders of magnitude than cloud liquid water content (Ervens et al., 2010). "RH" has been changed to "aerosol liquid water content" in order to highlight the role of cloud droplets in the formation of secondary species in the Fe-containing particles. Please refer to Line 80.

Lines 85-87: Vague sentence; please clarify.

Reply: The sentence have been clarified to "Various Fe-containing particle sources (such as dust and anthropogenic sources) might have different effects on the in-cloud formation of secondary species due to their different physicochemical properties (e.g., alkalinity) of these sources (Deguillaume et al., 2005)." Please refer to Lines 90-92.

Lines 89-92: The authors have published several other manuscripts on GCVI-SPAMS measurements at this field site. It would be helpful for the reader for these manuscripts to be described briefly and cited here to provide greater context for the current work.

Reply: We thank the reviewer for this suggestion. We have added our previous studies in the revised manuscript: "We have previously used a ground-based counterflow virtual impactor (GCVI) combined and a real-time single-particle aerosol mass spectrometer (SPAMS) to characterize chemical composition or mixing state of cloud residues (Lin et al., 2017), mixing state and cloud scavenging of the EC-containing particles (Zhang et al., 2017a), and the in-cloud formation of oxalate (Zhang et al., 2017b) at a mountain site, southern China." Please refer to Lines 95-99.

Lines 100-101: Please clarify this sentence and what is meant by "measured area".

Reply: We have revised to "sampling site". Please refer to Line 109.

Lines 121-122: Was a cyclone used? Please state.

Reply: An inlet cyclone with a cut-off aerodynamic diameter of 2.5 μm was used to sample interstitial particles or cloud-free particles during cloudy or cloud-free episodes. We have clarified it. Please refer to Lines 134-136.

Section 2.1: Move the dates of the study (Lines 122-123) to Section 2.1; also move up the sentence on lines 123-124 that gives the time in cloud during the study. What was the cloud type and temperature? State whether these were liquid-only clouds. Refer to Figure S1.

Reply: The Lines 122-123 and 123-124 have been moved to Section 2.1. The ambient temperature varied from 4 to 21 degrees Celsius in this study (Figure S1), indicative

Line 125: How were interstitial particles sampled?

Reply: As described above, an inlet cyclone with a cut-off aerodynamic diameter of 2.5 μm was used to sample interstitial particles during cloudy episodes. Please refers to Line 134-136.

Line 133: Please clarify that the triggers was (I assume) based on the calculated velocity of the particles, rather the intensity of the light scattered, as implied here.

Reply: We agree with the comment. We have changed to "based on the calculated velocity of the particles". Please refer to Line 148.

Line 135: Change "fragments is" to "ions are".

Reply: We have changed to "ions are" accordingly. Please refer to Line 150.

Lines 136-139: Fix grammar throughout sentence.

Reply: The sentence has been changed to "A specific mass-to-charge ratio (m/z) in the positive or negative mass spectrum can correspond to the most probable chemical species dependent on prior field and lab studies." Please refer to Lines 151-152.

Lines 145-146: Change "ranged" to "ranging". Change "bipolar ion mass spectra" to "with the SPAMS".

Reply: We have changed to "ranging" and "with the SPAMS" accordingly. Please refer to Lines 161-162.

Line 147-148: Change sentence starting with "Because the Fe ion peak at m/z 56 may be contaminated: : :" to "Since other species, such as: : :., may also contribute to m/z 56, the natural isotopic composition: : :". The phrasing "may be contaminated" is not correct.

Reply: We have changed to "Since other species, such as $[CaO]^+$, $[KOH]^+$, and $[C_3H_4O]^+$, may also contribute to m/z 56" accordingly. Please refer to Lines 162-163.

Line 168: Change "averaged mass" to "averaged single-particle mass".

Reply: We have changed to "averaged single-particle mass" accordingly. Please refer to Line 194.

Lines 186-189: Please clarify these sentences. The size transmission of the SPAMS needs to be stated in Section 2.2 to provide context for this statement as well.

Reply: These sentences have been change to "However, no clear enhanced NF of the Fe-dust type was detected in particle size range of 1.0-2.0 μm relative to 0.2-1.0 μm (14 ±1% versus 12 ±2%, by number) (Figure S3). The large Fe-dust particles (above 2.0 μm) might have already been deposited during transport. In addition, the Fe-dust particles may have a larger size (above 2.0 μm) that could not be detected by the SPAMS." Please refer to Lines 214-218.

In Section 2.2, we have added a sentence "It should be noted that particles detected by the SPAMS mostly distribute in the size range of d_{va} 0.2-2.0 μm (Li et al., 2011)." Please refer to Lines 156-157.

Line 206: Note that temperature and RH also typically follow diurnal patterns.

Reply: The ambient temperature showed a typically diurnal pattern. However, RH had a high value (nearly 100%) during daytime when the cloud events exited. Please refer to Lines 236-238.

Line 234: Please clarify this sentence. It is not clear what "was compared" means in the context of this sentence.

Reply: The sentence has been change to "The NF of nitrate in the Fe-dust cloud residues was higher than that in the Fe-rich cloud residues (92 ± 3% versus 86 ± 1%, by number)". Please refer to Lines 265-267.

Line 244: Please quantify what is meant by "barely detected".

Reply: Only 0.40 ± 0.01% (by number) of the Fe-containing cloud residues was found to be internally mixed with the sea salt (e.g., m/z 81 $[Na_2{}^{35}Cl]^+$ or 83 $[Na_2{}^{37}Cl]^+$). Please refer to Lines 276-278.

Line 250: Please clarify what is meant by "overwhelm the simultaneous irradiation effect."

Reply: The sentence has been change to "the amount of in-cloud chloride formation was less affected by irradiation effect". Please refer to Lines 287-288.

Lines 261-262 and 267-270: Please clarify these sentences.

Reply: Lines 261-262 have been deleted, because of no clearly increased NF of ammonium in the Fe-rich cloud residues relative to Fe-rich cloud-free particles when considering their errors. Lines 267-270 have been change to "During daytime, the increased NF of ammonium in the Fe-dust cloud-free particles was likely attributed to the enhanced NFs of ammonium nitrate." Please refer to Lines 306-307.

Make sure all figure captions are clear to a reader not familiar with single-particle measurements.

Reply: We have revised the Figure captions to better understanding of the Figures. Please refers to the Figure captions.

While up to the authors' discretion, I suggest combining the Atmospheric Implications and Conclusions sections to improve integration of the study results with their impacts.

Reply: This is a good point. The Atmospheric Implications and Conclusions sections have been combined. Please refer to the Section 4 in the modified manuscript.

Figure 1 caption: Change "averaged mass spectra" to "average single-particle mass

spectra" for clarity.

Reply: We have changed to "average single-particle mass spectra" accordingly. Please refer to Figure 1 caption.

Figure 2 caption: The reference to Pratt et al. is not needed in the caption.

Reply: The reference has been removed.

Figure 5 caption: Should "its precursors" be "the sum of the peak areas of its precursors"?

Reply: We have changed to "the sum of the peak areas of its precursors" accordingly. Please refer to Figure 5 caption.

Figure 6 caption: More information is needed here for the non-SPMS reader to understand the figure.

Reply: Figure 6 caption has been change to "The number fractions of sulfate (m/z -97), nitrate (m/z -46 or -62), chloride (m/z -35 or -37), ammonium (m/z 18), oxalate (m/z -89), oxalate precursors (m/z -59, -71, -73, -87, -103, or -117), Ca (m/z 40), and $[SiO_3]^-$ (m/z -76) in the four Fe-containing cloud residues." Please refer to Figure 6 caption.

Figure S1: What does "alternate sample" mean? What does "used to correct interstitial particles" mean?

Reply: In this study, the GCVI and PM$_{2.5}$ cyclone inlets were alternately applied with an interval of one hour to collect cloud residues and interstitial particles respectively during cloudy events. Thus, air mass, ambient temperature or RH were kept as similar as possible for sampling cloud residues and interstitial particles. Please refer to Figure S1 caption in the revised Supplement.

Figure S4 & S7: Please clarify the captions and include the m/z peaks used in these analyses.

Reply: We have added peaks used in these analyses, such as sulfate (m/z -97), nitrate (m/z -46 or -62), chloride (m/z -35 or -37), ammonium (m/z 18), Ca (m/z 40), oxalate (m/z -89), and oxalate precursors (m/z -59, -71, -73, -87, -103, or -117). Please refer to Figure S5 and S7 captions in the revised Supplement.

Figure S6 caption: Change "of oxalate in the non-Fe" to "oxalate-containing non-Fe".

Reply: We have change to "oxalate-containing non-Fe" accordingly. Please refer to Figure S6 caption in the revised Supplement.

References

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