Responses to anonymous referee #2

General comment:

The manuscript of Kuang et al.: "Contrasting effects of secondary organic aerosol formations on organic aerosol hygroscopicity" shows that the oxidation state of the SOA does not always correlate to their degree of the hygroscopicity, and they propose that the hygroscopicity is controlled by additional factors along with the oxygenation degree of the OA. To show that, the authors use the data set of a field campaign conducted in China (Pearls River Delta) using a suite of online and high-resolution instrumentation. This paper is well written, and I recommend publication after the authors address comments below.

Response: We thank the reviewer for all the valuable comments and suggestions, which promote us to learn from the book *Atmospheric Chemistry and Physics: From Air Pollution to Climate Chang* and provide more solid calculations to improve our narrative.

Major Comments:

Comment: Lines 166-171:   The authors state that they did not perform a calibration at the sampling state. They used an RIE=4 for ammonium taken from the last calibration. This could be a serious issue for ammonium concentration resulting in artifacts (lower of higher ammonium concentration). How well the ammonium concentration correlated with filter measurements (mentioned on line 178) using a RIE=4? Please provide the

Response: Thanks for reviewer's comment. Actually, we have similar worries about the data accuracy of SP-AMS measurements at the very beginning, so we paid a special attention to assure the data quality. Instrument internal calibrations such as m/z calibration and single ion signal scan were performed every day and offline samples were collected daily during the campaign. Aerosol chemical properties measured by parallel instruments were also collected to validate the SP-AMS data. As mentioned in the manuscript, excellent agreement between particle volume concentration derived from SP-AMS and SMPS may support the data accuracy of SP-AMS regarding the total volume (mass) concentration (shown in Figure S5). For better showing the data accuracy, we added Figure S7, S8 and S9 to present the comparisons between the data from SP-AMS and $PM_{2.5}$ monitoring device, as well as GAC, ECOC analyzer, AE33 aethalometer and offline measurements. The good correlation (slope =0.74, R^2 = 0.80) between the total **PM$_1$** mass concentration from SP-AMS (BC included) and **PM$_{2.5}$** mass concentration indicates SP-AMS provided correct total mass concentration. The comparisons of individual components between SP-AMS and external measurements (Figure S8 and S9) also showed good agreements and consequently assure the data quality. Specifically, ammonium (NH_4) mass concentration from SP-AMS was well correlated with offline result (with $PM_{2.5}$ impactor), with a reasonable slope of 0.76 and

a good correlation coefficient (R= 0.83, $R^2 = 0.69$). As reviewer suggested, we used RIE of 3, 3.5 and 4.5 to calculate NH_4 mass concentration as well as its contribution to PM_1. As shown in Table 1, NH_4 mass concentrations varied from 2.99 (RIE=4.5) to 4.48 µg/m^3 (RIE = 3) with tiny difference on mass fractions (8.18%~11.76%, while 9.10% for RIE = 4 was considered in previous analysis). Moreover, comparisons to offline results (Figure 1) also suggest RIE of 4 could be a good choice because applying RIE of 3 and 3.5 might overestimate the NH_4 concentration (slope = 1.01 and 0.86) while using RIE =4.5 might underestimate it (slope = 0.67) considering total PM_1 mass concentration (derived from SP-AMS) took 74% of total $PM_{2.5}$ mass concentration (Figure S5).

Table 1 NH_4 mass concentration and its contribution to PM_1 with different RIE

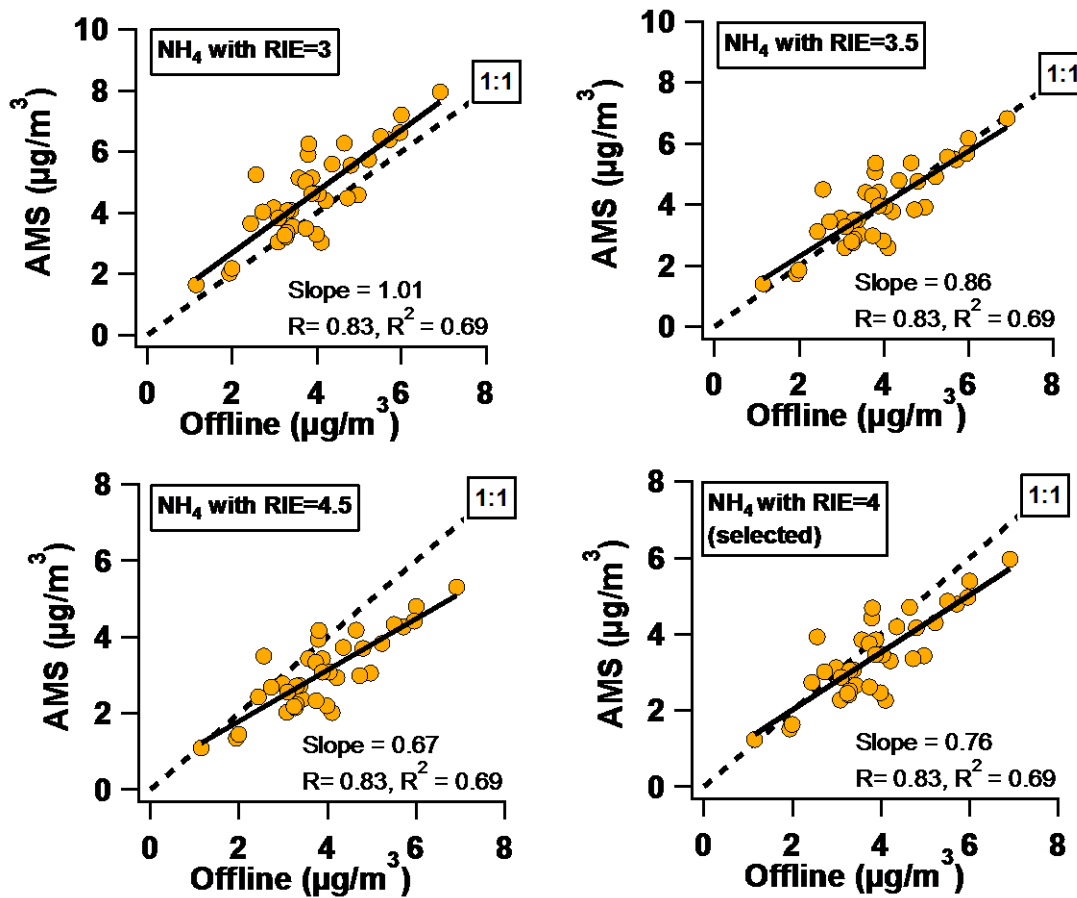| RIE	Average concentration (µg/m^3)	Mass fraction in PM1 (%)
RIE=4	3.39 ± 1.54	9.10%
RIE=3	4.48 ± 2.07	11.76%
RIE=3.5	3.84 ± 1.77	10.26%
RIE=4.5	2.99 ± 1.38	8.18%

Figure 1 Comparisons of NH$_4$ mass concentration between SP-AMS and offline measurements (PM$_{2.5}$) with different RIEs of NH$_4$.

Comment: Lines 235-243, Table 1 and equation 2: How it is possible to have (NH4)HSO4 and (NH4)2SO4 and NH4NO3 at the same time? Ammonia will first neutralize all available sulfate and it will bring the aerosol in the form of (NH4)2SO4. Then, whatever ammonia exists in the atmosphere will react with HNO3 to form NH4NO3. The co-existence of (NH4)HSO4 and NH4NO3 is not compatible according to thermodynamic lows. (Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Seinfeld and Pandis, Wiley, 3rd edition, 2016). The mass balance should be recalculated. After that, please discuss the potential compounds present in the

(NH4)HSO4 should be deleted from equation (2) and (3) and the calculations should be done again. How much the results (kOA) do change with this correction?

Response: Thanks for your comment, we think the reviewer raised a very interesting question. Actually we didn't think of this at all when we were using the ion pairing scheme proposed by Gysel et al. (2007) which have been used in many aerosol hygroscopicity closure or organic aerosol hygroscopicity estimation studies since then.

We have read the book *Atmospheric Chemistry and Physics: From Air Pollution to Climate Chang* and find discussions about the Ammonia-Nitric Acid-Sulfuric Acid-Water system in Chapter 10.4.4, and we also find the figure 10.23 which shows there is no way that both HSO_4^- and NO_3^- can be existed at the same time. The calculation in chapter 10.4.4 follows the way that Ammonia will first neutralize all available sulfate and it will bring the aerosol in the form of $(NH_4)_2SO_4$, and the free part ammonia will then react with HNO_3 to form NH_4NO_3. However, we think this a simplified calculation, and this conclusion does not hold in the real atmosphere (if completely holds, no nitrate should be exist in ammonia poor conditions, which is not the case).

Actually, another reason we never realized this is that we believe that **an equilibrium will always exist between HSO_4^- and SO_4^{2-} under acid conditions through $HSO_4^- \leftrightarrow H^+ + SO_4^{2-}$, and ammonia concentrations under general atmospheric conditions does not raise aerosol pH to exceed pH=7** (Guo et al., 2017), and multiphase buffering mechanism must be considered in considering aerosol pH and ionic form of sulfate (Zheng et al., 2020). If all sulfate is in the form of $(NH_4)_2SO_4$, then

the aerosol should not be acidic. We have initiated a simulation using a thermodynamic model ISORROPIA thus followed both the thermodynamic rule and also the partitioning theory of volatile gases like ammonia and nitric acid. Case 1: total ammonia concentration is 17 ug/m3 (1 umol/m^3), sulfate is 19.2 ug/m3 (0.2 umol/m^3) and nitrate is zero, which means that the ammonia concentration exceeds far the required ammonia for sulfate neutralization, and the RH is 80%, T is 298 K. The simulated aqueous fraction of HSO_4^- in total sulfate is 0.03 umol/m^3 which means that even under this condition, HSO_4^- exists and cannot be neglected. Case 2: change nitrate to 6.3 ug/m3 (0.1 umol/m^3) and other parameters are same with Case 1, thus still ammonia rich condition. For case 2, the simulated HSO_4^- is 0.013 umol/m^3. Case 3: reduce total ammonia concentration is 8.5 ug/m^3 (0.5 umol/m^3), and other parameters remain the same with Case 2. For Case 3, the simulated HSO_4^- will be 0.1 umol/m^3, which means that 50% of sulfate is in the form of HSO_4^-.

These simulation results demonstrate that HSO_4^- , SO_4^{2-} and NO_3^- coexist in the aqueous phase. Due to the lack of gas phase ammonia and nitric acid mass concentration measurements, we didn't perform simulations using thermodynamic model but still use the widely used ion pairing scheme proposed by Gysel et al. (2007), and thus keep same way of estimating κ_{OA} with previous studies for the comparison convenience.

Comment: Lines 293-294: Is this NH4NO3 formation during the night due to the lower temperature during the night? The dissociation constant Kp(T) of the ammonia-nitic acid system (NH3(g) + HNO3 (g) ↔ NH4NO3 (s)) is a function of the temperature and

it is very sensitive to the temperature changes. As the temperature decreases Kp(T) also decreases, and the equilibrium shifts towards the aerosol phase increasing the NH4NO4 mass concentration (Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Seinfeld and Pandis, Wiley, 3rd edition, 2016). It seems that this is the case taking account Figures 2a and 2c. Please discuss.

Response: Thanks for your comment. After reading your comments, we realized that we should be more cautious when discussing the possible nitrate formation mechanism. You mentioned a very good point that the increase of nitrate might be a result of solid and gas phase equilibrium transition due to the temperature decrease. Depending on the RH or the RH history that aerosol particles have been experienced, the aerosol particles might be solid (if crystallized) or aqueous phase (dehydration branch from the morning to the afternoon, metastable) since about 16:00. However, when aerosol particles will be deliquescent is also a puzzle if aerosol particle crystallized in the afternoon due to the complex dependence of deliquescence RH on aerosol mixtures of ammonium sulfate, ammonium nitrate and organic aerosols (Kuang et al., 2016), and the peak RH in the morning was usually lower than 80%, To illustrate the possible mechanisms behind the nitrate increase. We assume the following two states of ammonium nitrate: (1) solid

state; (2) aqueous phase.

For the solid phase case, the $NH_3(g) + HNO_3(g) \leftrightarrow NH_4NO_3(s)$ equilibrium theory is applied (Seinfeld and Pandis, 2016). If we have the initial concentration of gaseous NH_3, then we can guess the initial $HNO_3(g)$ based on the $K_p(T)$ and estimate the nitrate concentration increase due to the decrease $K_p(T)$. The $K_p(T)$ changed from 84 ppb^2 to 24 ppb^2 from 16:30 to 23:30 for the average case with air temperature decrease from 27.7 to 22.7 °C. Figure 2 shows the variations of nitrate mass concentrations under
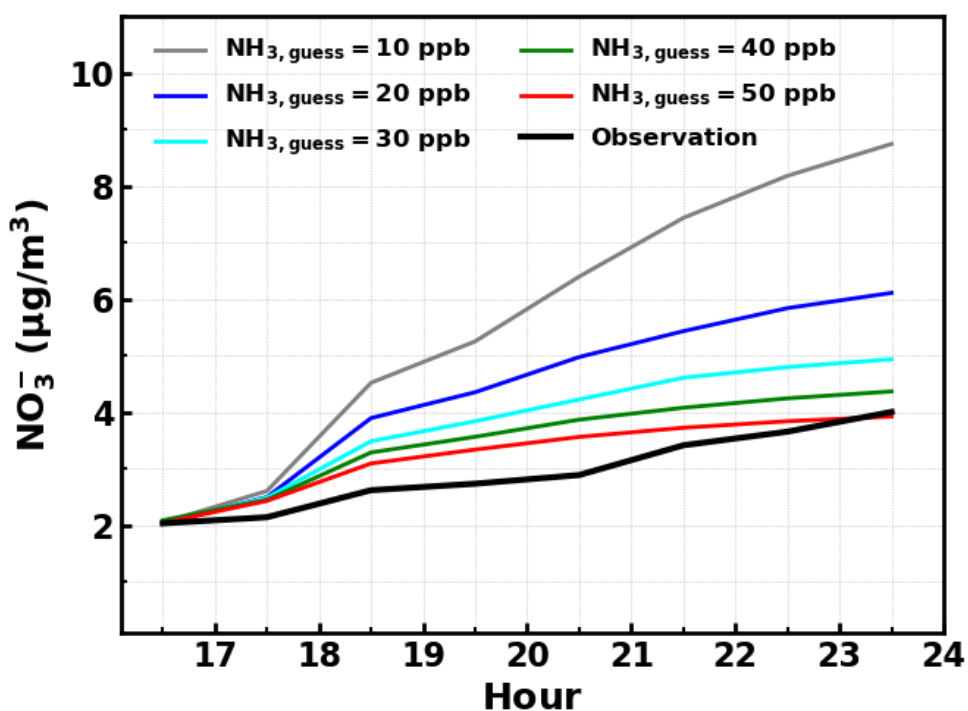

Figure 2. Simulated variations of nitrate under different NH_3 (g) conditions by assuming aerosol particles are aqueous using the ISORROPA thermodynamic model.

different assumed NH_3 (g) concentration conditions by considering variations of $K_p(T)$. The results demonstrate that the temperature induced $K_p(T)$ change is enough for explaining observed nitrate increase, and thus a possible mechanism.

For the aqueous phase case, NH_4NO_3 will be found in the aqueous aerosol phase, and the corresponding dissociation reaction is then $NH_3(g) + HNO_3(g) \leftrightarrow NH_4^+ + NO_3^-$. For this case, both temperature decrease and RH increase play roles in the NH_3 and HNO_3 partitioning. We use a thermodynamic model ISORROPIA (forward, metastable) to simulate the possible magnitude of nitrate increase driven by increased RH and reduced temperature for the average case shown in Fig.2 of the manuscript. We also assume different scenarios of $NH_3(g)$ concentrations and for each $NH_3(g)$ concentration scenario, we estimate the $HNO_3(g)$ concentration with which we can reproduce the observed nitrate concentration at the initial time (16:30), to get a total HNO_3 concentration in gas and aerosol phase, then we can estimate the possible increase of nitrate in aerosol phase due to repartitioning and the results are shown in Fig.3. The results show that the more HNO_3 dissolved in aqueous phase can also explain the observed nitrate mass concentration increase.

The simulation results shown above demonstrate that repartitioning of HNO_3 (total HNO_3 in aerosol and gas phase remain unchanged) from gas phase to aerosol phase can explain the observed nitrate mass concentration increase under both assumptions that aerosols are completely aqueous or solid. The actual atmospheric case is more likely that both solid (freshly emitted and crystallized) and aqueous particles (dehydration branch, not crystallized) exist, however, does not affect conclusion that the repartitioning of HNO_3 in gas and aerosol phase can explain the observed nitrate increase. In addition, NO concentration is quite low (almost zero) after 16:00 which might be favorable of N_2O_5 formation and the N_2O_5 hydrolysis as a possible nitrate

formation pathway cannot be excluded. Thus, texts in the manuscript which are related to the delineation of nitrate formation are revised accordingly. For example, the second paragraph of Sect 4.1 is revised as:

"The average diurnal variations of NO_2, NO, O_3, CO, aerosol chemical compositions, $\kappa_{f(RH)}$ and meteorological parameters are shown in Fig.2. O3 concentrations began to increase after sunrise, peaked near 15:00 and then began to decrease quickly but drops slower after midnight. Meanwhile, NO concentration began to decrease quickly after sunrise, reached and remained near zero after noontime, and began to slightly increase after 21:00. NO_2 concentration increased quickly after 15:00 and reached a plateau after 21:00. Variation characteristics of NO, O_3, and NO_2 suggest that the relatively low NO concentration resulted in weak titration effects on O_3, whereupon typical NO_3 chemistry and subsequent N_2O_5 chemistry might occur, which might contribute to the observed nitrate increase after sunset. However, nitrate concentrations increased quickly after about 16:00 and peaked after midnight (about 03:00 LT), indicating that there must be a mechanism is responsible for the observed nitrate increase at least before sunset. To dig more into this, the possible pathways of nitrate formation since 16:00 was simulated and discussed in Sect.3 of the supplement. The results demonstrate that the repartitioning of HNO_3 in gas and aerosol phase due to the temperature decrease and RH increase can mainly explain the observed nitrate increase. And the strong daytime photochemistry and decrease of NO_2 concentration might result in significant production of gas phase before about 16:00. However, the possible contribution of N_2O_5 hydrolysis to nitrate formation cannot be excluded."

Comment: Lines 307-310: This explanation is not sufficient stated. If this is the case (i.e., N2O5 production during the night, then NO2 should be reduced, but Figure 2f shows that NO2 increases during the night. Please make sure that you explain the phenomena correctly.

Response: Thanks for your comment, but we cannot agree with the reviewer on this. The N_2O_5 production does not require that NO_2 should be reduced. The variation of NO_2 mass concentration depends on its sources and sinks, if its source is relatively stronger then the N_2O_5 production only slows down the NO_2 concentration increase instead of reducing NO_2 mass concentration. The main factor that drives the speculation that NO_3 and N_2O_5 will be produced is that during night the O_3 concentration is still higher than 25 ppb before 24:00, however the NO concentration is almost zero since 16:00.

In addition, we have analyzed the possible mechanism behind the observed nitrate increase after 16:00 as suggested by the reviewer in the previous comment, and have revised this part as the following:

"The average diurnal variations of NO_2, NO, O_3, CO, aerosol chemical compositions, $\kappa_{f(\mathrm{RH})}$ and meteorological parameters are shown in Fig.2. O3 concentrations began to increase after sunrise, peaked near 15:00 and then began to decrease quickly but drops slower after midnight. Meanwhile, NO concentration began to decrease quickly after sunrise, reached and remained near zero after noontime, and began to slightly increase after 21:00. NO_2 concentration increased quickly after 15:00 and reached a plateau after 21:00. Variation characteristics of NO, O_3, and NO_2 suggest

that the relatively low NO concentration resulted in weak titration effects on O_3, where upon typical NO_3 chemistry and subsequent N_2O_5 chemistry might occur, which might contribute to the observed nitrate increase after sunset. However, nitrate concentrations increased quickly after about 16:00 and peaked after midnight (about 03:00 LT), indicating that there must be a mechanism is responsible for the observed nitrate increase at least before sunset. To dig more into this, the possible pathways of nitrate formation since 16:00 was simulated and discussed in Sect.3 of the supplement. The results demonstrate that the repartitioning of HNO_3 in gas and aerosol phase due to the temperature decrease and RH increase can mainly explain the observed nitrate increase. And the strong daytime photochemistry and decrease of NO_2 concentration might result in significant production of gas phase before about 16:00. However, the possible contribution of N_2O_5 hydrolysis to nitrate formation cannot be excluded."

Comment: Line 310: "Nitrate concentrations increased quickly since 16:00:". This is contradictory to Figure 2c, where nitrate decreases from 09:00 to 16:00 and then starts increasing, reaching its maximum at 03:00.

Response: Thanks, we have revised this sentence as "However, nitrate concentrations increased quickly after about 16:00 and peaked after midnight (about 03:00 LT), indicating that there must be a mechanism is responsible for the observed nitrate increase at least before sunset."

Comment: Line 312: "nighttime heterogenous formation of nitrate, " please check if

this is the case.

Response: we rephrased this sentence as "Under the strong daytime photochemistry and nighttime increase of nitrate"

Comment: Line 337: "On average, κ_{OA} increased slowly during the nighttime". From Figure 3c it seems that kOA is rather stable during the night and it increases after 03:00 (a.m.).

Response: It can be seen from Fig.3c, that κ_{OA} increased slowly since 18:00 and remain stable during 0 to 3:00 and then increase.

Comment: Lines 357-359: "However, κ_{OA} was also negatively correlated with LOOA 357 (Fig.4d), whose mass concentration increase rapidly after sunrise and are likely secondary due to local photochemistry with potential precursors such as isoprene and anthropogenic VOCs." How do you support this? Could you discuss any results from the gas-phase?

Response: The observation site is on a small mountain of Heshan county, is about 55 km away from megacity Guangzhou and is surrounded by villages and small residential towns, and the surrounding areas are covered with trees as shown in Fig.S1. Thus, the VOC precursors of observed quick LOOA formation are likely both biogenic and anthropogenic. Though we do not have detailed results about VOCs at this moment, a similar diurnal pattern of LOOA and ozone (Figure 3) may indicate that these two secondary products were formed in local photochemical process with VOC precursors

emitted from surrounding area. So, we rephrased this sentence as "whose mass concentration increase rapidly after sunrise and are likely secondary due to local photochemistry with potential precursors such as isoprene of both biogenic and anthropogenic VOCs as the observation site is surrounded by small towns and areas with high percentage cover of trees as shown in Fig.S1"
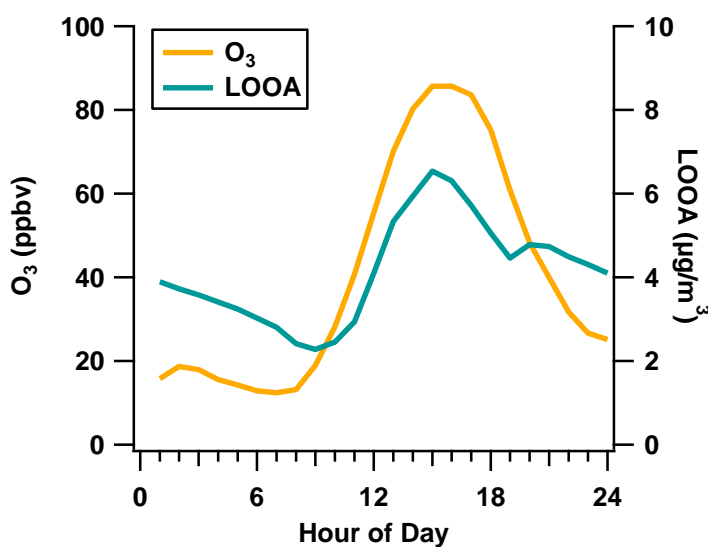

Figure 4. Diurnal patterns of O_3 and LOOA during the campaign.

Minor Comments:

Comment: Please use past tense throughout the whole manuscript. There are parts that the present tense alternates with past tense (e.g., section 2.2).

Response: Thanks, we went through the manuscript and used past tense throughout the whole manuscript.

Comment: Line 62: Please replace "evolvement" with "evolution".

Response: changed accordingly.

Comment: Lines 69-71: Please add here that the volatility could be another factor that affects the hygroscopicity of the SOA.

Response: We did not add volatility because volatility itself is also determined by carbon-chain length, functional groups, etc. Both volatility and hygroscopicity are physical properties of organic aerosol and determined by organic aerosol structure, they may be related, but the hygroscopicity is not essentially affected by volatility.

However, the reviewer raised a good point about organic aerosol volatility and hygroscopicity, thus we have added this point in Sect 4.3 as introduced in the response of the next comment.

Comment: Lines 71-72: Please cite here the following 4 papers:

Kuwata, M., Kondo, Y., Mochida, M., Takegawa, N., and Kawamura, K.: Dependence of CCN activity of less volatile particles on the amount of coating observed in Tokyo, J. Geophys. Res., 112, D11207, doi:10.1029/2006JD007758, 2007.

Asa-Awuku, A., Engelhart, G. J., Lee, B. H., Pandis, S. N., and Nenes, A.: Relating CCN activity, volatility, and droplet growth kinetics β-caryophyllene secondary organic aerosol, Atmos. Chem. Phys., 9, 795–812, 2009.

Frosch, M., Bilde, M., Nenes, A., Praplan, A. P., Jurányi, Z., Dommen, J., Gysel, M., Weingartner, E., and Baltensperger, U.: CCN activity and volatility of β-caryophyllene secondary organic aerosol, Atmos. Chem. Phys., 13, 2283–2297, 2013.

Kostenidou, E., Karnezi, E., Hite Jr., J. R., Bougiatioti, A., Cerully, K., Xu, L., Ng, N. L., Nenes, A., and Pandis, S. N.: Organic aerosol in the summertime southeastern United States: components and their link to volatility distribution, oxidation state and hygroscopicity, Atmos. Chem. Phys., 18, 5799–5819, 2018.

Response: Thanks, the reviewer mentioned several important papers about organic aerosol hygroscopicity and volatility. We did not cite these papers in Lines 71-72, but we have cited these papers in Sect 4.3 as the following "It seems more plausible to find parameters other than O/C ratio to parameterize κ_{OA}, which should be independent of sources and associated with the physical properties of OA, such as volatility (Kuwata et al., 2007;Asa-Awuku et al., 2009;Frosch et al., 2013;Kostenidou et al., 2018)"

Comment: Lines 106-110: Please rewrite this part. It should be not mentioned any sections, but rather describe and explain what it will follow in the next.

Response: Thanks, we have revised this part as " We described details on aerosol measurements and the κ_{OA} estimation method in measurements and method part. In the results and discussion section, we first sketched out the overview of campaign measurements and then discussed the κ_{OA} variation characteristics as well as its influencing factors, and in the last part, the complexity regarding κ_{OA} parameterization was further demonstrated and elucidated. The summaries are provided in the conclusion part."

Comment: Line 116: Please replace "locates" with "was located".

Response: changed accordingly.

Comment: Line 117: Please add "the" before "megacity".

Response: added accordingly.

Comment: Line 130: Please add "the" before "physical".

Response: added accordingly.

Comment: Line 132: Please add "the" before "aerosol".

Response: added accordingly.

Comment: Line 134: Please replace "of" with "the".

Response: replaced accordingly.

Comment: Line 135: Please add "the" before "aerosol".

Response: added accordingly.

Comment: Lines 137-139: Please rephrase this sentence.

Response: This sentence is rephrased as "To make sure the accuracy of the measured RH in the sensing volume of the wet Nephelometer, three Vaisala HMP110 sensors with accuracies of ($\pm$0.2 ℃ and $\pm$1.7 % for RH between 0 to 90%) were used to monitor the RH at different parts of the wet nephelometer."

Comment: Line 139: Please replace "Two" with "Two sensors".

Response: added accordingly.

Comment: Line 146: Please add "a" before "flow".

Response: added accordingly.

Comment: Line 146: Please add "the" before "sampling".

Response: added accordingly.

Comment: Line 149: Please add "the" before "particle".

Response: added accordingly.

Comment: Line 152: Please add "the" before "size-resolved".

Response: added accordingly.

Comment: Line 153: Please delete "basically".

Response: deleted accordingly.

Comment: Lines 161-162: Please rephrase this sentence.

Response: This sentence is rephrased as "The air flow in the AMS was first controlled by the orifice and then focused through the aerodynamic lens of SP-AMS, and then

particles with diameter in sub-micrometer range were detected."

Comment: Line 183: "As a wildly used source analysis method" I am not sure what do you mean here.

Response: we deleted those words.

Comment: Lines 186-188: Please rephrase this sentence.

Response: Thanks, the sentence is rephrased as "In this study, PMF using high resolution AMS data including two matrices (organic ion mass concentrations and their uncertainties) were conducted by an Igor Pro-based panel, i.e., PMF Evaluation Tool (PET, v2.06, Ulbrich et al., 2009), following the instruction in Ulbrich et al. (2009)."

Comment: Lines 190-199: This is not the right place for this paragraph. It should be moved to the Results section.

Response: Thanks, we think this part should be in the measurements part, because this is the description of how PMF factors are determined.

Comment: Lines 206-207: Please rephrase this sentence.

Response: Thanks, this sentence is rephrased as "and $\kappa_{f\text{(RH)}}$ represents a diameter independent hygroscopicity parameter κ that fits the observed $f(80\%, 525\text{ nm})$ best and solved through iteration algorithm."

Comment: Lines 223-226: Please rephrase this sentence.

Response: Thanks, this sentence is rephrased as "The aerosol hygroscopicity parameter κ can be calculated from aerosol chemical composition measurements (κ_{chem}) on the basis of volume mixing rule, thus the organic aerosol hygroscopicity parameter κ_{OA} were usually estimated through closure between measured κ and estimated κ using aerosol chemical measurements."

Comment: Line 250: Please define better the parameters κ and ε.

Response: Thanks, the sentence is rephrased as "Where κ_i is hygroscopicity parameter κ of compound i , and ε_i is volume fraction of compound i in the mixture (Vi/Vtot, Vi and Vtot are volume of compound i and total aerosol volume of PM_1)."

Comment: Lines 255-256: Please rephrase this sentence.

Response: Thanks, this sentence is rephrased as "These unidentified aerosol species, in continental regions, likely be dust but still possible composed of other components such as biogenic primary aerosol."

Comment: Line 352: Please provide the corresponding literature.

Response: Reference is added.

Comment: Line 356: "It was generally thought that secondary aerosol formation would result…", This phrase is not well connected to the previous sentences.

Response: This sentence is put after the sentence that originally after it, and thus revised as "However, κ_{OA} was also negatively correlated with LOOA (Fig.4d), whose mass concentration increase rapidly after sunrise and are likely secondary due to local photochemistry with potential precursors of both biogenic and anthropogenic VOCs as the observation site is surrounded by small towns and areas with high percentage cover of trees as shown in Fig.S1. The negative correlation between κ_{OA} and LOOA is contradictory with the generally thought that secondary aerosol formation would result in increases of aerosol hygroscopicity."

Comment: Line 359: What do you mean by average O/C"? Each factor derived from PMF analysis has a constant O/C ratio.

Response: The word "average" is deleted.

Comment: Lines 360-363: This sentence is quite big and complicate. Please rephrase and simplify.

Response: Thanks, this sentence is revised as "The negative correlation between κ_{OA} and LOOA mass fraction explained why O/C failed to describe diurnal variations of κ_{OA}: the O/C ratio for LOOA is 0.72, which is only lower than that of MOOA, suggesting that the daytime LOOA formation and decrease of BBOA and HOA mass concentrations drove the increase of daytime O/C but the κ_{OA} didn't follow."

Comment: Line 394: Please replace "?" with ".".

Response: replaced.

Asa-Awuku, A., Engelhart, G. J., Lee, B. H., Pandis, S. N., and Nenes, A.: Relating CCN activity, volatility, and droplet growth kinetics of β-caryophyllene secondary organic aerosol, Atmos. Chem. Phys., 9, 795-812, 10.5194/acp-9-795-2009, 2009.

Frosch, M., Bilde, M., Nenes, A., Praplan, A. P., Jurányi, Z., Dommen, J., Gysel, M., Weingartner, E., and Baltensperger, U.: CCN activity and volatility of β-caryophyllene secondary organic aerosol, Atmos. Chem. Phys., 13, 2283-2297, 10.5194/acp-13-2283-2013, 2013.

Guo, H., Weber, R. J., and Nenes, A.: High levels of ammonia do not raise fine particle pH sufficiently to yield nitrogen oxide-dominated sulfate production, Scientific reports, 7, 12109, 10.1038/s41598-017-11704-0, 2017.

Gysel, M., Crosier, J., Topping, D. O., Whitehead, J. D., Bower, K. N., Cubison, M. J., Williams, P. I., Flynn, M. J., McFiggans, G. B., and Coe, H.: Closure study between chemical composition and hygroscopic growth of aerosol particles during TORCH2, Atmos. Chem. Phys., 7, 6131-6144, 10.5194/acp-7-6131-2007, 2007.

Kostenidou, E., Karnezi, E., Hite Jr, J. R., Bougiatioti, A., Cerully, K., Xu, L., Ng, N. L., Nenes, A., and Pandis, S. N.: Organic aerosol in the summertime southeastern United States: components and their link to volatility distribution, oxidation state and hygroscopicity, Atmos. Chem. Phys., 18, 5799-5819, 10.5194/acp-18-5799-2018, 2018.

Kuang, Y., Zhao, C. S., Ma, N., Liu, H. J., Bian, Y. X., Tao, J. C., and Hu, M.: Deliquescent phenomena of ambient aerosols on the North China Plain, Geophysical Research Letters, n/a-n/a, 10.1002/2016GL070273, 2016.

Kuwata, M., Kondo, Y., Mochida, M., Takegawa, N., and Kawamura, K.: Dependence of CCN activity of less volatile particles on the amount of coating observed in Tokyo, Journal of Geophysical Research: Atmospheres, 112, https://doi.org/10.1029/2006JD007758, 2007.

Seinfeld, J., and Pandis, S.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Third Edition, 2016.

Zheng, G., Su, H., Wang, S., Andreae, M. O., Pöschl, U., and Cheng, Y.: Multiphase buffer theory explains contrasts in atmospheric aerosol acidity, Science, 369, 1374, 10.1126/science.aba3719, 2020.