

Responses to the Editor and Referee(s)' Comments

We are grateful to you and the reviewers for the thoughtful comments and suggestions. We have revised the manuscript accordingly. The listed below in blue are our point-by-point responses to the comments. The modified parts in the revised manuscript and revised Supplement are highlighted in red.

Main comments:

1. My comment regarding the title was not addressed.

(i) If I understand your study correctly, the changes in atmospheric composition you observed during the lockdown were largely due to different meteorological conditions (temperature, RH, air masses etc) that led to different O₃ concentrations whereas emission reductions (i.e. the lockdown measures) were only minor. At least that is how I interpret your text at multiple places throughout the manuscript, e.g. lines 76ff: Online observations, model simulations, and satellite measurements have pointed out that the appearance of haze events during the LCD were mainly caused by the unfavorable meteorological conditions....

l. 84ff: In order to understand the effect of the reduced anthropogenic emissions during the LCD and different meteorological parameters....

l. 182: ‘The more favorable atmospheric conditions such as the higher temperature and stronger solar radiation during the LCD...’

l. 185: A recent study using the WRF–Chem model found that about 80% of the increased O₃ level in eastern China was mainly due to meteorological changes,...

(ii) What do you mean by ‘contribution of photooxidation’? Are you referring to your (wrongly inferred –see my comment on Sections 3.3 and 3.4 below) difference in aqueous vs photochemical diacid formation? Also, you only use ‘photooxidation’ at one other place in the whole manuscript. Be consistent in terminology.

(iii) Please modify the title and abstract such that they properly reflect the content of the article.

Response: (i) In Section 3.1, we mentioned that the enhanced O_3 concentration during the LCD was not only influenced by the concentrations of NO_2 and $PM_{2.5}$, but also was associated with meteorological conditions such as solar radiation and temperature. The O_3 formation is also influenced by volatile organic compounds (VOCs) and other key factors, which were not available during our field campaign. Indeed, we think the higher concentration of O_3 was associated with the enhanced formation of gaseous photo-oxidation of diacids and related species during the LCD. In this study, we focused on the enhanced contribution of gaseous photo-oxidation to diacids and related species in urban aerosols during the LCD rather than the formation mechanism of O_3 . Thus, we don't use aerosol chemical models (e.g., WRF-Chem model) to quantitatively analyze the contribution of meteorological factors and emissions reductions to an increase in O_3 during the LCD.

To avoid any potential misunderstanding, we have deleted “A recent study using the WRF–Chem model found that about 80% of the increased O_3 level in eastern China was mainly due to meteorological changes, and only 20% resulted from the reduced pollutant emissions (Wang et al., 2022).”.

(ii) As discussed in Sections 3.2 and 3.3, C_2 and related compounds before the LCD were significantly derived from the aqueous oxidation during which RH and LWC were important influencing factors. However, the gaseous photochemical oxidation contributed significantly to the formation of these organic acids during the LCD, and the stronger solar radiation and higher oxidants (e.g., O_3) were key influencing factors. To express more accurately, the secondary oxidation was divided into aqueous oxidation and gaseous photochemical oxidation. Yu et al. (2021) reported that gas-phase photooxidation contributed significantly to the formation of diacids and related SOA during the entire sampling period, and the aqueous-phase process played an important role over the severe haze event in January in Beijing, China. Additionally, such a classification was supported by the results of PMF model. Therefore, we have distinguished between aqueous oxidation and gaseous photochemical oxidation.

“Contribution of photooxidation” means “contribution of gaseous photooxidation”. We have changed them to “gaseous photochemical oxidation” or “gaseous photooxidation” in the revised manuscript. Please see Line 28 (Page 1), Line 36 (Page 1), Line 38 (Page 1), Line 374 (Page 10), Line 392 (Page 10),

Line 393 (Page 10), Line 395 (Page 10), Line 405 (Page 10), Line 424 (Page 11), Line 485 (Page 12), Line 517 (Page 13), respectively.

(iii) According to your comment, we have modified the title to “Investigation on the sources and formation processes of dicarboxylic acids and related species in urban aerosols before and during the COVID-19 lockdown in Jinan, East China” in the revised manuscript (see Lines 1-3, Page 1).

In addition, we have modified the abstract as follows:

...while the O_3 concentration increased by 1.3 times during the LCD possibly because of the weakened titration effect of O_3 via the reduced NO, alleviated aerosol effect via the dropped $PM_{2.5}$, and the more favorable atmospheric conditions (e.g., higher temperature and stronger solar radiation). Moreover, the proportion of secondary inorganic aerosols in $PM_{2.5}$, concentration levels, and relative abundance of diacid homologues in water-soluble organic compounds (WSOC) during the LCD were still 2–4 times higher than those before the LCD, despite the sharp decrease of primary air pollutants, indicating the enhanced formations of secondary aerosols during the LCD.

However, C_2 and related species before the LCD was dominantly derived from the acidic aqueous oxidation with α -dicarbonyls determined by relative humidity (RH) and liquid water content (LWC).

Please see Lines 19-24 (Page 1) and Lines 29-30 (Page 1), respectively.

2. The subsections of Section 3 still have very little connection to each other. Instead of being repetitive, previously identified findings should be used and referred to in the further discussion: what did you conclude on the formation processes of C_2 in Section 3.2, how are these conclusions corroborated (or contradicted) in Section 3.3, what additional insight is gained in Sections 3.3, 3.4 and 3.5 etc.

Response: Thanks for your kind suggestion. Based on the differences in the concentration levels, molecular compositions, and aging extents of diacids and related compounds before and during the LCD, we preliminarily conclude that these organic compounds during these two observation periods experienced different formation pathways. However, the detailed information about formation mechanisms and influencing factors has not been well known in Section 3.2. Thus we continued to perform the investigations about such scientific questions before the LCD in Section 3.3 and during the LCD in Section 3.4, respectively. In Section 3.3, we first obtained that C_2 and related species before the

LCD were associated with the secondary oxidation of organic precursors emitted from biomass burning, then we concluded that C₂ before the LCD was dominantly derived from the acidic-catalyzed aqueous oxidation with Gly and mGly where RH and LWC were key controlling factors. In section 3.4, we first concluded that the higher C₂ and related compounds may be derived from the gas-phase photochemical oxidation driven by the higher O₃ concentration and stronger solar radiation, then we confirmed such a deduction and obtained that C₂ during the LCD was primarily derived from the photochemical degradation of longer-chain homologous diacids driven by stronger solar radiation and higher O₃ concentration and other oxidants rather than higher temperature. Compound-specific stable carbon isotope analyses of specific diacids and related compounds can provide crucial information on the sources, photochemical aging level, and atmospheric processing of organic aerosols because the isotopic fractionation of carbon occurs upon chemical reactions or phase transfer, thus we compared the stable carbon isotopic compositions of diacids and related species before and during the LCD in Section 3.5.

In Section 3.5, we gained that tPh before and during the LCD was mainly from the primary sources of plastic waste burning. In addition, we obtained that the ¹³C values of major diacids and the related compounds during the LCD were less negative than those before the LCD, demonstrating that the gaseous photochemical oxidation was promoted because of the higher temperature and O₃ concentration under the clearer sky conditions during the LCD. To quantitatively determine the contribution of each source to diacids and related compounds, PMF analysis was conducted in Section 3.6. In such a section, we gained that major diacids and related compounds before the LCD were from the aqueous oxidation (47.2%), coal combustion (16.2%), dust emission (12.9%), gaseous photochemical oxidation (12.3%), and biomass burning (11.4%), and these organic compounds were from the gaseous photochemical oxidation (50.5%), aqueous oxidation (16.1%), biomass burning (13.6%), dust emission (12.6%), and coal combustion (7.2%).

3. The structure of Section 3 and some of the headings of the subsections add to the confusion:

(i) 3.1 Overview of observations

The header suggests that this section includes only observations. However, there is already a lot of discussion and interpretation (e.g. on reasons of different O₃ levels). This adds to later repetitiveness.

(ii) 3.2 Molecular distributions of diacids and related species

The header should clarify that the discussion that follows relates to both periods i.e. before and during lockdown. In addition, the header does not fully describe the content of the section: You discuss potential formation pathways. In addition, you compare to previous studies – which is great; however, the header does not reflect the fact that you discuss data and place them in the context of previous studies.

(iii) 3.3 Aqueous formation of SOA before the LCD

(iv) 3.4 Enhanced photochemical formation of SOA during the LCD

Why suddenly switching from ‘dicarboxylic acids’ to SOA? The acids only constitute a small fraction of SOA? Also, distinguishing between ‘aqueous formation’ and ‘photochemical formation’ implies that the former is not photochemical. This is not correct.

(v) 3.5 Stable carbon isotopic compositions of diacids In the heading you refer to diacids; however, in Table 3, there are also oxoacids.

Response: Suggestion is taken.

(i) We have changed the header to “3.1 Meteorological conditions and air pollutants before and during the LCD” in the revised manuscript (see Line 169, Page 5).

(ii) We have changed the header to “3.2 Comparison on molecular distributions of diacids and related species before and during the LCD” in the revised manuscript (see Line 210, Page 6).

(iii) and (iv) We agree with you. Thus, we use “diacids and related species” instead of “SOA”. In addition, we changed them to “3.3 The aqueous-phase formation of diacids and related species before the LCD” and “3.4 Enhanced gaseous photochemical formation of diacids and related species during the LCD”, respectively.

Please see Line 302 (Page 8) and Line 374 (Page 10) in the revised manuscript, respectively.

(v) We have changed it to “3.5 Stable carbon isotopic compositions of diacids and related species before and during the LCD” in the revised manuscript (see Line 426, Page 11).

4. Summary and conclusions

This last section is merely a summary of your results. However, conclusions are not given, e.g. How do your results compare with previous studies? What do your results mean for our understanding of the state and/or behaviour of the atmosphere?

Response: Thanks for your suggestion. We have changed some parts in this section as follows:

Additionally, diacids, oxoacids, and TDOCs increased in both concentration levels and percentage contributions during the LCD, indicating the enhanced secondary formation of diacids and related species determined by stronger solar radiation and higher O₃ could offset the significant decline of organic precursors from pollutant emissions during the LCD. Before the LCD, C₂ was significantly derived from the acidic aqueous-phase oxidation where RH and LWC played a significant role, while during the LCD it was dominantly generated from the gaseous photochemical oxidation driven by stronger solar radiation and higher O₃ concentration rather than higher temperature. The higher ratios of C₂/diacids and C₂/TDOCs, and more positive $\delta^{13}\text{C}$ values of major detected diacids during the LCD than before the LCD suggested the more aged organic aerosols during the LCD.

These results are helpful for better understanding the sources, formation processes, and driving factors of SOA in the urban regions of East China during the LCD. However, our study suggested that if the primary emissions are blindly reduced, we cannot completely solve the current air pollution in the North China Plain where the sources and precursors of secondary aerosols are extremely complicated. The balanced strategy and trans-regional joint control of major air pollutants are necessary to be considered together with meteorological conditions.

Please see Lines 528-535 (Page 13), Lines 540-544 (Pages 13-14) in the revised manuscript, respectively.

Minor comments

5. 1. 23: isn't 'clearer sky conditions' and 'haze events' a contradiction?

Response: No, it isn't. The sentence "Unexpectedly, several haze events driven by enhanced secondary organic aerosols (SOA) still took place in eastern China during the LCD." is cited from (Huang et al.,

2020; Le et al., 2020; Zhong et al., 2021). In this study, the parameters of air quality including PM_{2.5}, PM₁₀, CO, SO₂, and NO₂ dropped sharply during the LCD. The haze event is defined as the daily average concentration of PM_{2.5} larger than the Grade II of the Chinese National Ambient Air Quality Standard (75 µg m⁻³) (Huang et al., 2014; Li et al., 2021). In this study, thirteen haze days before the LCD and four haze days during the LCD were observed in Jinan, respectively. Moreover, the solar radiation during the LCD was stronger than before the LCD. To sum up, the atmosphere during the LCD was clearer sky conditions in comparison with before the LCD, despite several haze days occurring during the LCD.

We have added related discussions as follows:

The haze event is defined as the daily average concentration of PM_{2.5} larger than the Grade II of the Chinese National Ambient Air Quality Standard (75 µg m⁻³) (Huang et al., 2014; Li et al., 2021b). Thirteen haze days before the LCD and four haze days during the LCD were observed in Jinan, respectively. Additionally, the solar radiation ($255 \pm 117 \text{ W m}^{-2}$) during the LCD was stronger than that ($164 \pm 70 \text{ W m}^{-2}$) before the LCD (Table 1). It could be concluded that the atmosphere was clearer sky conditions during the LCD in comparison with before the LCD, despite several haze days occurring during the LCD.

Please see Lines 179-184, Page 5 in the revised manuscript.

6. 1. 218: the new sentence: ‘Different secondary formation rates...’ is very vague. 1) The list of parameters includes basically all potential reasons why formation pathways may change in a qualitative sense. 2) In light of the previous discussion (see my comment 1 above), a more quantitative statement should be given.

Are the reasons for the differences the emission reductions? If yes, then the lockdown measures are the reason for the differences.

If all changes are due to differences in meteorological conditions (RH, photochemical activity), then the fact that emissions were reduced during the lockdown are not the main reason for the observed differences. – In this case, the title of the study is misleading.

Response: It is an excellent suggestion. Different secondary formation rates of TDOCs before and during the LCD were largely due to different meteorological factors (e.g., temperature, solar radiation, and RH), oxidants (e.g., O₃ and OH· radical), emission strengths and types of organic precursors, physicochemical properties of aerosols (e.g., pH_{is} and LWC), and other influencing factors. It is a pity that we do not obtain these important influencing factors such as OH· radical, emission strengths and types of organic precursors, and others during the field campaign. Thus, we can't give a more quantitative statement about the reasons for different secondary formation rates before and during the LCD. To express accurately, we changed it to "Different secondary formation rates of TDOCs between these two observation periods were possibly due to..." in the revised manuscript (see Line 227, Page 6).

7. 1. 179: The preceding lines say that high O₃ concentrations are caused by high NO_x concentrations. The sentence starting with 'Thus, ...' states the opposite. Be clear what you want to say here. I am not saying it is wrong but it is confusing.

Response: We have rephrased these sentences as follows:

Because of the lack of HO_x radicals during the wintertime, O₃ in Chinese megacities is primarily produced from the NO_x-saturated regime (Li et al., 2021), where the drop of NO_x can lead to the enhanced O₃ concentration (Liu and Wang, 2020). Firstly, the significant drop of NO₂ during the LCD resulted in the reduction of NO concentration (Xu et al., 2020), and further weakened the efficient titration effect of O₃ (Levy et al., 2014). Secondly, the reduction of NO₂ during the LCD could reduce the concentration of NO₃⁻, and in turn enhanced O₃ by alleviating the aerosol effects (Liu and Wang, 2020).

Please see Lines 185-190, Page 5 in the revised manuscript.

8. 1. 214: add 'more' (while these species are MORE stable in the absence of Fe) – all acids can be further oxidized by OH and by other oxidants.

Response: Suggestion is taken. We have added "more" in the revised manuscript (see Line 221, Page 6).

9. l. 250: ‘...because the hydroxylation of C₄ can be photodegraded into C₃ through the decarboxylation process’ – Hydroxylation is the addition of an OH group to a molecule. I do not think that this is the mechanism here. Do you mean simply ‘the photochemical degradation of C₄ leading to C₃...?’

Response: Yes, we do. We changed it to “the photochemical degradation of C₄ can lead to C₃...” in the revised manuscript (see Line 259, Page 7).

10. l. 276: what does ‘relative abundance’ refer to here? total organic aerosol mass?

Response: “Relative abundance” refers to “relative abundance in total diacids”. We have added it in the revised manuscript (see Line 284, Page 7).

11. l. 280: ‘heterogenous processes’ usually refer to processes on interfaces. However, the oxidation of oxoacids into diacids can also occur in the aqueous bulk phase (e.g. several studies by B. Turpin’s group, Carlton et al etc). – You correctly call it ‘aqueous oxidation’ in l. 284.

Response: Suggestion is taken. We have changed it to “aqueous oxidation (Carlton et al., 2007; Ervens et al., 2004)” in the revised manuscript (see Line 288, Page 7).

12. l. 334: There is a lot of literature on discussions that Henry’s law constants – that are usually defined for pure water – are not directly applicable for aerosol water (e.g., <https://doi.org/10.1021/es400083d>, <https://doi.org/10.1021/acs.est.5b02782> and references therein).

Response: We agree that Henry’s law constants are usually defined for pure water (Kampf et al., 2013; Waxman et al., 2015). In this study, we focused on the water solubility of Gly and mGly rather than the gas-to-aerosol aqueous phase partitioning of Gly and mGly by Henry’s law. We deleted the related text “In the presence of liquid water, both species can dissolve in the aqueous phase following Henry’s law (Myriokefalitakis et al., 2011)” because of the theme and limited length of our manuscript. Besides, we rephrased the sentence as follows:

... and thus can dissolve in the aqueous phase (Carlton et al., 2006; Myriokefalitakis et al., 2011).

Please see Lines 340-341, Page 9 in the revised manuscript.

13. 1. 337 – 339: This needs more discussion: On the one hand you say that Gly and LWC correlate, i.e. LWC is proportional to Gly. In the next sentence you say that C_2/Gly is proportional to LWC, which would imply that LWC is proportional to $1/\text{Gly}$ – unless the increase in C_2 is significantly higher than that of Gly. Can this be shown based on the observations?

Response: Yes, such strong correlations are based on the observations which are shown in Fig. 4a. When LWC increased from $4.3 \mu\text{g m}^{-3}$ to $172 \mu\text{g m}^{-3}$ before the LCD, C_2 increased by 1.9 times, which was higher than that of Gly (increased by 1.1 times) and mGly (increased by 1.2 times). We have added the above discussions as follows:

Indeed, the increase in C_2 (increased by 1.9 times) was significantly higher than that of Gly (increased by 1.1 times) and mGly (increased by 1.2 times) with the increase of LWC before the LCD.

Please see Lines 346-347, Page 9 in the revised manuscript.

14. 1. 339 – 341: I do not understand this sentence ('Such strong correlations...'. Please clarify.

Response: We have rephrased this sentence as follows:

These discussions suggest that the higher LWC concentration and RH could not only promote the formations of C_2 , Gly, and mGly, but also facilitate the transformation of C_2 from Gly and mGly.

Please see Lines 347-349, Page 9 in the revised manuscript.

15. 1. 356ff: (i) The sentence starting 'There, we conclude...' seems out of place because a lot of discussion actually comes after it.

(ii) Also the discussion should include the aspect that oxalic acid is not only formed more slowly at low pH but it is also oxidized more slowly at low pH.

Response: (i) Suggestion is taken. We have rephrased these sentences as follows:

However, Tan et al. (2009) reported that acidity had a minor effect on C_2 formation at cloud- and fog-relevant conditions via online experiments. Wang et al. (2015) suggested that the coarse particles during the dust period, which are alkaline, are favorable for the C_2 formation from ωC_2 , largely because the reaction rate constant ($3.6 \times 10^8 \text{ M}^{-1} \text{ s}^{-1}$) of ωC_2 with OH· radical to form C_2 is smaller than that ($2.9 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$) of its anion, glyoxylate. At very acidic pH, C_2 is not only formed more slowly but

also oxidized more slowly (Eugene et al., 2016; Herrmann, 2003). Those findings conflicted with each other, largely because the concentration levels of organic precursors, acidity, LWC, and other influencing factors were different from the aerosols in our field observations, thus further studies are necessary to elucidate the influencing mechanism of acidity on C₂ formation. To sum up, we could conclude that C₂ before the LCD was dominantly derived from the acidic-catalyzed aqueous oxidation with α -dicarbonyls rather than longer-chain diacids determined by RH and LWC.

Please see Lines 364-373, Page 9 in the revised manuscript.

(ii) Many thanks. We add this information in the revised manuscript as follows:

At very acidic pH, C₂ is not only formed more slowly but also oxidized more slowly (Eugene et al., 2016; Herrmann, 2003).

Please see Lines 367-368, Page 9.

16. 1. 365: (i) Why is this section entitled ‘Enhanced photochemical formation of SOA during the LCD’? You only talk about dicarboxylic acids – which are, of course, a fraction of SOA but if you were to replace ‘SOA’ by ‘dicarboxylic acid’ here, it is basically the title of your paper. (ii) What is the purpose of this section, and what additional information does it provide as compared to the preceding ones? It seems very repetitive, and in parts even contradicting.

Response: (i) We have changed the header to “3.4 Enhanced gaseous photochemical formation of diacids and related species during the LCD” in the revised manuscript. Please see Line 374, Page 10.

(ii) The purpose of Section 3.4 is to investigate the formation pathways via gas-phase photochemical oxidation and influencing factors of major diacids and related species during the LCD. Based on the differences in the diurnal variations, concentration levels, and molecular characteristics of diacids and related species, air pollutants, and meteorological factors before and during the LCD, we preliminarily concluded that the enhanced concentrations of diacids and related compounds, more aged organic aerosols during the LCD were due to the enhanced secondary formation of diacids and related compounds, despite the anthropogenic source strength dropping dramatically during the LCD. However, why does the concentration of diacids and related compounds increase when the anthropogenic source strength drops sharply during the LCD? How are these organic compounds formed? What are the

important influencing factors? These scientific questions have not been elucidated before, thus further discussions are conducted in Section 3.4. Therefore, it does not seem repetitive.

The goal of Section 3.3 is to analyze the formation pathways via aqueous oxidation and influencing factors of diacids and related compounds before the LCD. Different kinds and emission strengths of organic precursors, meteorological factors, physicochemical properties of aerosols (e.g., pH_{is} and LWC), and other influencing factors before and during the LCD could lead to different sources and formation mechanisms of diacids and related compounds in these two observation periods. Thus, these results before the LCD in Section 3.3 and during the LCD in Section 3.4 were not contradicting each other.

17. 1. 376: The next on OH levels is very confusing. Here you say that differences in OH levels are mainly due to differences in NO_2 Whereas higher above, you said that differences in O_3 levels are largely due to meteorological conditions. It is well known that O_3 and OH are closely related in the atmosphere. What is the main message you can and want to give?

Are the meteorological conditions more /equally/less important than the emission reductions as you point out at several places throughout the manuscript (see ‘Major comment’) or is the difference in chemical composition of the atmosphere (e.g. reduced NO_x etc) the main reason for enhanced dicarboxylic acid concentrations?

Response: Wang et al. (2020) reported that about 80% of the increased O_3 level in the eastern China was mainly due to meteorological changes, and only 20% resulted from the reduced pollutant emissions. In Section 3.1, we suggested that the enhanced O_3 concentration during the LCD was not only influenced by the concentrations of NO_2 and $\text{PM}_{2.5}$, but also was associated with meteorological conditions such as solar radiation and temperature. The O_3 formation is also influenced by volatile organic compounds (VOCs) and other key factors that we don’t obtain. Indeed, the higher concentration of O_3 is associated with the enhanced formation of photo-oxidation of diacids and related species during the LCD. In this study, we focused on the enhanced contribution of photo-oxidation to diacids and related species in urban aerosols during the LCD rather than the formation mechanism of O_3 . It’s a pity that we did not use aerosol chemical models (e.g., WRF-Chem model) to analyze quantitatively the contribution of meteorological factors and emissions reductions to an increase in O_3 during the LCD.

Similarly, we could not conclude that the meteorological conditions contribute more//equally/less than the emission reductions or not.

We have deleted “A recent study using the WRF–Chem model found that about 80% of the increased O₃ level in eastern China was mainly due to meteorological changes, and only 20% resulted from the reduced pollutant emissions (Wang et al., 2022).”.

18. l. 392: In your response to my previous round of reviews, you stated that O₃ is just a proxy for photochemical activity. The text here implies that O₃ was directly involved in the formation. Please be consistent.

Response: O₃ can be considered as a proxy for photochemical activity, which does not mean that O₃ can't be involved in the formation. For example, Mochizuki et al. (2017) suggested that C₂ is derived from the oxidation of isoprene and α -pinene with O₃ in the forest atmosphere of Mt. Fuji (Mochizuki et al., 2017). C₄, C₅, and C₆ are formed from the oxidation of cyclicolefins with O₃ (Kawamura and Ikushima, 1993; Kawamura and Usukura, 1993). The enhancement of diacids owing to O₃ has also been observed in many other studies (Kawamura and Yasui, 2005; Meng et al., 2018; Yu et al., 2021). Therefore, the expression “..., suggesting that O₃ played an important role in the formation of C₂ and other diacids.” is correct.

19. (i) l. 397: The sentence ‘the increase in the temperature can accelerate the volatilization of C₂, leading to the drop of C₂ concentration in the aerosol phase’ contradicts your earlier text (l. 216): ‘Therefore, the loss of diacids and related compounds may be negligible when the temperature increases’.

(ii) l. 445: But this statement ‘These results imply that the effect of photochemical decomposition of higher diacid homologues on C₂ before the LCD was minor’ is contradicting what you say in l. 404 “indicating that C₂ may be largely derived from the photochemical degradation of higher molecular weight homologues of diacids”

Response: (i) We are sorry for the mistakes. As discussed in Section 3.2, the loss of diacids and related compounds may be negligible when the temperature increases. The concentrations of C₂ and diacids did

not exhibit similar patterns of variations (Fig. 4b) and strong correlations with temperature ($R^2 < 0.2$, Fig. 5), again suggesting that the effect of temperature on the loss of C_2 and diacids was negligible. Therefore, we rephrased the sentences as follows:

However, such similarities and strong correlations of those were not observed with temperature ($R^2 < 0.2$, Fig. 4b and Fig. 5), again suggesting that the effect of temperature on the loss of C_2 and diacids was negligible.

Please see Lines 404-405, Page 10 in the revised manuscript.

(ii) As described in Line 445 of the old manuscript, we stated that the effect of photochemical decomposition of higher diacid homologues on C_2 before the LCD was minor. As described in Line 404 of the old manuscript, we stated that C_2 may be largely derived from the photochemical degradation of higher molecular weight homologues of diacids during the LCD. Different kinds and emission strengths of organic precursors, meteorological factors, physicochemical properties of aerosols (e.g., pH_{is} and LWC), and other influencing factors before and during the LCD could lead to different sources and formation mechanisms of C_2 in these two observation periods. The detailed information has described in Sections 3.2 - 3.4. Therefore, these results before the LCD and during the LCD were not contradicting each other. To express more accurately, we have added “during the LCD” behind “indicating that C_2 ” in the revised manuscript (see Line 410, Page 10).

20. l. 465: ‘each period suggests that C_2 in Jinan aerosols was mainly originated from aqueous oxidation of ωC_2 ’ – above you say that the main source were larger acids.

Response: Sorry for such a mistake. We have deleted this sentence “The more pronounced enhancement of $\delta^{13}C$ values in C_2 and ωC_2 in each period suggests that C_2 in Jinan aerosols was mainly originated from aqueous oxidation of ωC_2 (Meng et al., 2020).” in the revised manuscript.

21. l. 482: ‘more clear sky conditions’ implies higher visibility and less haze – however, in the beginning you state that during the lockdown enhanced haze formation was observed.

Response: Please see the reply to the fifth comment.

22. 1. 495: In several sections before, you discuss the correlations of O₃, Gly, wC₂ and argue that they point to efficient aqueous phase processing. Please clarify how the classification of factor 3 connects to the previous sections.

Response: The robust relationships of O₃, Gly, mGly, and ωC₂ were observed with the third factor, which indicated the contribution of gaseous photochemical oxidation before the LCD. In several sections before, we describe that C₂, C₃, ωC₂, Pyr, Gly, mGly, LWC, WSOC, and SO₄²⁻ exhibited relatively higher loadings in the third factor which was considered as the sources from aqueous oxidation. The following text describes that the aqueous oxidation made the greatest contribution (47.2%) to C₂ and related compounds before the LCD, while the gaseous photochemical oxidation contributed only 12.3%. Those expressions suggest that the aqueous oxidation contributes more than the gaseous photochemical oxidation to C₂, Gly, and ωC₂ before the LCD, again confirming the dominant formation of C₂ and its organic precursors (e.g., ωC₂, Pyr, Gly, and mGly), which is consistent with the results as discussed in Section 3.3.

We have added related discussions as follows:

The PMF-resolved relative contributions to the detected species before and during the LCD were presented in Fig. 9c and Fig. 9d, respectively. The aqueous oxidation made the greatest contribution (47.2%) to C₂ and related compounds, while the gaseous photochemical oxidation contributed only 12.3% to the total determined sources before the LCD, again suggesting that the aqueous oxidation was the dominant formation pathway for these organic compounds before the LCD. However, the gaseous photochemical oxidation contributed the largest percentage (50.5%) to the total identified sources, while the aqueous oxidation accounted for only 16.1% of the total identified sources during the LCD, confirming that the gaseous photochemical oxidation exerted a leading role in the formation of homologous diacids during the LCD, which was in agreement with the results as discussed above. The less contribution of C₂ and related compounds from aqueous oxidation during the LCD was likely because of the decrease of RH and LWC.

Please see Lines 512-520, Page 13 in the revised manuscript.

Technical comments

23. l. 22: remove 'the' before 'more aged organic aerosols'.

Response: Corrected.

24. l. 27 ff: 'Source apportionment using the molecular characteristics of organic compounds and positive matrix factorization (PMF) suggest...?'

Response: Corrected.

25. l. 41: 'marine area' sounds odd. Either 'marine regions' or 'marine air'.

Response: Suggestion is taken. We have changed it to "marine regions". Please see Line 46, Page 2 in the revised manuscript.

26. l. 48: Cloud observations are also field measurements. Better: 'field measurements in- and outside clouds...?'

Response: Thanks for your suggestion. We have changed it to "field measurements in- and outside clouds..." in the revised manuscript. Please see Line 53, Page 2.

27. l. 54: replace 'proved' by 'proven'.

Response: Corrected.

28. l. 56: 'aqueous' misspelled.

Response: Corrected.

29. l. 66: replace 'photodegraded under the O₃ chemical pathway' by 'oxidized by O₃'.

Response: Corrected.

30. l. 170: Define PSCF.

Response: Suggestion is taken. We have defined PSCF as “potential source contribution function” in the revised manuscript. Please see Line 176, Page 5.

31. 1. 173: either ‘were reduced by’ or ‘decreased by’.

Response: Thanks for your suggestion. We have changed it to “decreased by” in the revised manuscript. Please see Line 177, Page 5.

32. 1. 176: ‘O₃ is produced...’ : This sentence repeats nearly verbatim the information just a couple of lines above. If there is no additional information, remove one of the sentences.

Response: Suggestion is taken. We have deleted “O₃ is produced from nonlinear processes, and O₃ in Chinese cities is largely produced from NO_x (NO + NO₂)-saturated regime attributed to the scarce HO_x radicals during the wintertime (Le et al., 2020).”.

33. 1. 208: Arrhenius is misspelled.

Response: Corrected.

34. 1. 315: You refer here to Section 3.3. but it is in Section 3.3.

Response: Suggestion is taken. We have deleted it.

35. 1. 328: ‘slope’ misspelled.

Response: Corrected.

36. 1. 348: Remove ‘formation’

Response: Corrected.

37. 1. 382: ‘Because OH· radical was unavailable in this work...’ You mean ‘Because measurements of the OH radical were unavailable in this work...’

Response: Corrected.

38. l. 385: ‘significant escalation’ is not the right term here.

Response: Suggestion is taken. We have changed it to “significant enhancement” in the revised manuscript. Please see Line 394, Page 10.

39. l. 433: ‘heaviest’ does not seem the right word here. DO you mean ‘highest’?

Response: Yes, we do. Therefore, we changed it to “highest” in the revised manuscript. Please see Line 439, Page 11.

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