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 are our point-by-point responses in blue to the comments. The modified parts in the revised manuscript and revised Supplement are highlighted in red.

General comments:

1. Several previous referee comments were not sufficiently addressed. I agree with the main referees' comments that many arguments and conclusions are hand waiving and based on generic statement taken from the literature. These criticisms were only partially addressed. In addition, I have numerous other comments that need to be addressed before the manuscript may be accepted for publication. If you consider revision, please upload a track-change manuscript where all changes (adding/deleting/moving text) can be clearly seen.

Response: We thank the editor's encouragement. All comments and suggestions have been considered carefully and well addressed below.

I Previous referee comments:

2. Please improve upon the responses to the following comments by Referees 1 and 2. The numbering refers to the reviewer reports by reviewers 1 (R1) and 2 (R2) and their comments as numbered in their original reports. If you choose not to change the manuscript text according to the comments, please justify this decision.

R1, C2: reasoning for 10 min blank samples – where is the information as given in your response found in the manuscript?

Response: The field blank is sampled to see whether the aerosol samples have been polluted during the operation process, including the placing and collecting processes of the filter, which takes about a few minutes. This sampling procedure doesn't aim to see the environmental impact during sampling time.

Please see the definition of field blank at website (https://www.lcslaboratory.com/field-blank/). The operating procedure of collecting field blank samples for 10 min in this study is conventional and scientifically sound and was also confirmed in other studies (Qi et al., 2022; Yi et al., 2021). Therefore, we chose 10 min to collect field blank samples.

In the revised manuscript, we have added this discussion in Lines 103-108, Page 3: "The field blank is sampled to see whether the aerosol samples have been polluted during the operation process, including the placing and collecting processes of the filter, which takes about a few minutes. The operating procedure of collecting field blank samples for 10 min in this study is conventional and scientifically sound and was also confirmed in other studies (Qi et al., 2022; Yi et al., 2021). Therefore, field blank samples were also collected by mounting the blank filter onto the sampler for 10 min without turning on the sampler before, during, and after the sampling campaign, respectively.".

3. R1, C4 iii: Where was this information added in the text? How do you define 'good enough'?

Response: We have added related information in the revised manuscript as follows:

As described in previous studies, the recoveries of C_2 ranged from 70% to 83% and other target compounds were better than 80% (Ding et al., 2021; Kawamura et al., 2013; Kawamura and Yasui, 2005; Meng et al., 2020; Zhao et al., 2020). Recoveries of the target compounds in this study were 80% for C_2 and higher than 85% for other organic species. Therefore, the percent recoveries mentioned in this study were good enough for such analysis.

Please see Lines 129-133, Page 4 in the revised manuscript.

4. R1, C6: Please provide more reasoning why you limited your discussion to ozone as oxidant only. I understand that you do not have measurements of other oxidants; however, what is the extent to which this limitation influences your conclusions?

Response: O_3 can be used to evaluate the atmospheric oxidizing capacity. There has been a great deal of research indicating that the enhanced O_3 during the LCD in urban areas could increase the atmospheric oxidizing capacity and further facilitate secondary aerosol formation (Huang et al., 2020; Le et al., 2020; Liu et al., 2020; Meng et al., 2021). As a crucial indicator for atmospheric oxidizing

capacity, O_3 concentration has been used in previous studies to understand the influencing mechanism of diacids (Yu et al., 2021) and evaluate the strength of photochemical oxidation (Herndon et al., 2008; Xu et al., 2017). Therefore, we used O_3 as a tracer for atmospheric oxidizing capacity to investigate the effect of the stronger atmospheric oxidizing capacity during the LCD than before the LCD on the formation mechanism of C_2 and related diacids. Many field observations have demonstrated that O_3 is a key driving factor during the photochemical formation process of diacids (Kawamura and Ikushima, 1993; Kawamura and Yasui, 2005; Meng et al., 2018, 2021; Mochizuki et al., 2017; Yu et al., 2021). In addition, the OH·radical during the LCD was higher than that before the LCD as discussed in Lines 376-383, Page 10, being consistent with O_3 concentration. Therefore, our discussions of O_3 as an oxidant didn't influence our conclusions.

5. R1, C11: Can you be more specific and quantitative here? The potential influence of long range transport requires more discussion than just half a sentence. How would it affect your results if indeed the effect of long-range transport had affected the C_2, C_3, C_4-content of the aerosol samples?

Response: We have added related discussion in the revised manuscript (Lines 259-267, Page 7) as follows:

Previous studies have demonstrated that the C_3/C_4 ratio presented a strong correlation with temperature when the contribution of local sources predominates over long-distance transport (Kawamura and Usukura, 1993; Pavuluri et al., 2010; Wang et al., 2020). In this study, the C_3/C_4 ratio was correlated strongly with temperature before the LCD ($R^2 = 0.54$, Fig. 4a), indicating that diacids before the LCD were largely influenced by local sources. However, the C_3/C_4 ratio was correlated moderately with temperature ($R^2 = 0.33$, Fig. 4b) during the LCD, suggesting that the contribution of local sources was equal to that of long-range transport to diacids during the LCD. These results were consistent with the results of backward trajectory and PSCF analysis (Fig. S1). The higher ratios of C_2/C_4 and C_3/C_4 during the LCD may be due to the local photooxidation and aging effects of long-distance transport.

6. R1, C11: The textbook by Seinfeld and Pandis does not explicitly discuss the conversion of S(IV) in aerosol water, only in clouds; however, it shows that the S(IV) oxidation rate is a function of LWC, which is smaller by several orders of magnitude for aerosol water than clouds. *"The SO_4^{2-} formation is considered to be mainly via gas-phase reactions at relatively low atmospheric RH levels (RH < 30%) and aqueous-phase reactions at relatively high atmospheric RH levels (RH > 60%) (Li et al., 2017; Liu et al., 2020). RH in this study ranged from 29.8% to 86.9% with an average of 52.2±10.2 %. It can be concluded that SO_4^{2-} in this study was dominantly from the aqueous oxidation."*

Response: We agree with you. The formation mechanism of SO_4^{2-} at different RH levels still remains controversial and unclear. For example, Li et al. (2017) proposed that the SO_4^{2-} formation was simply considered to be mainly via gas-phase reactions at relatively low RH levels (RH < 30%) and heterogeneous reactions and aqueous-phase reactions at relatively high RH levels (RH > 60%). However, Wang et al. (2014) reported that the SO_4^{2-} formation was dominantly derived from aqueous-phase reactions when RH was above 50%.

The aqueous phase conversion of dissolved SO_2 to SO_4^{2-} is probably the most important chemical formation pathway in the North China Plain during the wintertime (Sun et al., 2013; Wang et al., 2014; Zhang et al., 2015). A few studies have demonstrated that LWC and RH are considered as key factors determining SO_4^{2-} formation via aqueous reactions (Bikkina et al., 2017; Cheng et al., 2016). We observed a robust relationship of SO_4^{2-} with RH and LWC (R^2 = 0.58) and LWC (R^2 = 0.71) before the LCD (Fig. S3), indicating that the aqueous oxidation determined by LWC and RH played a dominant role in the formation of SO_4^{2-} before the LCD. Field observations have also revealed the importance of aqueous-phase production of C_2 and related compounds based on the correlation analysis with SO_4^{2-} (Bikkina et al., 2017; Kunwar et al., 2019; Meng et al., 2020; Shen et al., 2023; Wang et al., 2012, 2017, 2020; Yu et al., 2021). Therefore, the correlation analysis between SO_4^{2-} and C_2 can be used to evaluate the formation process of C_2 mainly via aqueous phase pathways. In addition, RH and LWC are crucial factors determining the aqueous-phase formation of C_2 (Bikkina et al., 2017; Wang et al., 2020; Yu et al., 2021). A strong correlation of C_2 with RH and LWC as discussed in the revised manuscript (Lines 335-338, Page 9) supported the aqueous formation of C_2 before the LCD. Moreover, C_2 and related

compounds, LWC, WSOC, and SO_4^{2-} exhibited the relatively higher loadings in the first factor of PMF model, again indicating the aqueous-phase formation of C_2 and SO_4^{2-}.

In the revised Supplementary, we have added related information (Lines 101-107, Page 7) as follows:

Text S5. Aqueous phase formation of SO_4^{2-} before the LCD

The aqueous phase conversion of dissolved SO_2 to SO_4^{2-} is probably the most important chemical formation pathway in the North China Plain during the wintertime (Sun et al., 2013; Wang et al., 2014; Zhang et al., 2015). A few studies have demonstrated that LWC and RH are considered as key factors determining SO_4^{2-} formation via aqueous reactions (Bikkina et al., 2017; Cheng et al., 2016). We observed a robust relationship of SO_4^{2-} with RH ($R^2 = 0.58$) and LWC ($R^2 = 0.71$) before the LCD (Fig. S3), indicating that the aqueous oxidation determined by LWC and RH played a dominant role in the formation of SO_4^{2-} before the LCD.
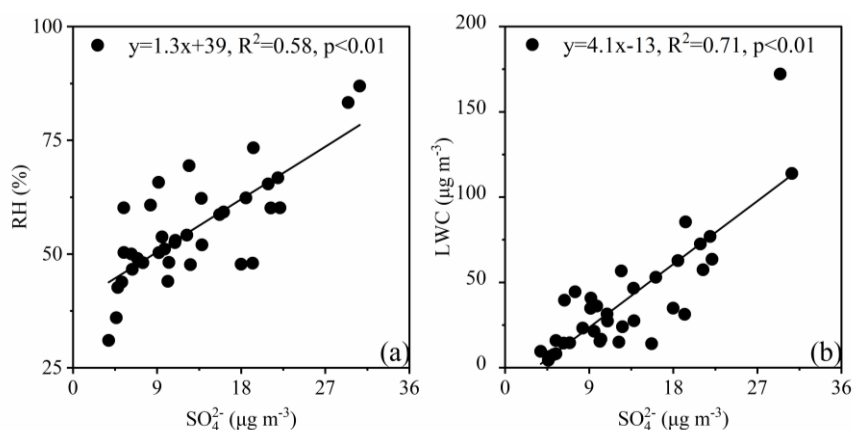

Figure S3. Relationships of SO_4^{2-} with (a) RH and (b) LWC before the LCD.

7. R2, C1 i: Where is this information stated in the manuscript? I could not find it. Apparently, it was not sufficiently clear for the referee. Please clarify.

Response: The mentioned question about the productive rate and fractionation constant for the individual reactant-product pairs is very critical for our study. We have added related discussions as follows:

In our manuscript, we stated that the recoveries of the target compounds were from 80% to 85%, which was not for yield during the derivatization reaction. The yield of derivatization reaction with 14%

BF$_3$/n-butanol was 100%, during which the carboxyl functional group was fully derivatized to butyl ester, and the aldehyde and keto groups were fully derivatized to dibutoxy acetal. The δ^{13}C values for diacids and related compounds could not be directly determined, so these organic species are derivatized with BF$_3$/n-butanol to dibutyl esters which are analyzed for the stable carbon isotopic composition using a GC-IR-MS. The δ^{13}C values for individual diacids are then calculated from δ^{13}C of 1-butanol and butyl ester derivative using a mass balance equation. The accuracy of the δ^{13}C measurement for these target compounds is within 0.8‰. The method developed for the determination of stable carbon isotopic composition of diacids and related compounds isolated from aerosols is reliable and scientific, and has been used widely in many studies (Aggarwal and Kawamura, 2008; Meng et al., 2020; Mkoma et al., 2014; Pavuluri et al., 2011; Qi et al., 2022; Shen et al., 2022, 2023; Wang et al., 2012; Wang and Kawamura, 2006; Xu et al., 2022a, 2022b; Zhang et al., 2016; Zhao et al., 2018).

Please see Lines 32-42, Page 2 in the revised Supplement.

8. R2, C3: The referee had suggested that parameters in columns, parameters in rows and sequence in both should be same in the panel that shows before lockdown and the panel that shows after lockdown. Please use this advice and rework the figures accordingly. Currently they are still too confusing as the reader does not have a clear comparison of the various dependencies and parameters.

Response: According to the editor's suggestion, we have modified the columns and rows in Fig. 5 and Fig. 6 and merged these two figures into one figure (Fig. 4, Page 28) as follows:
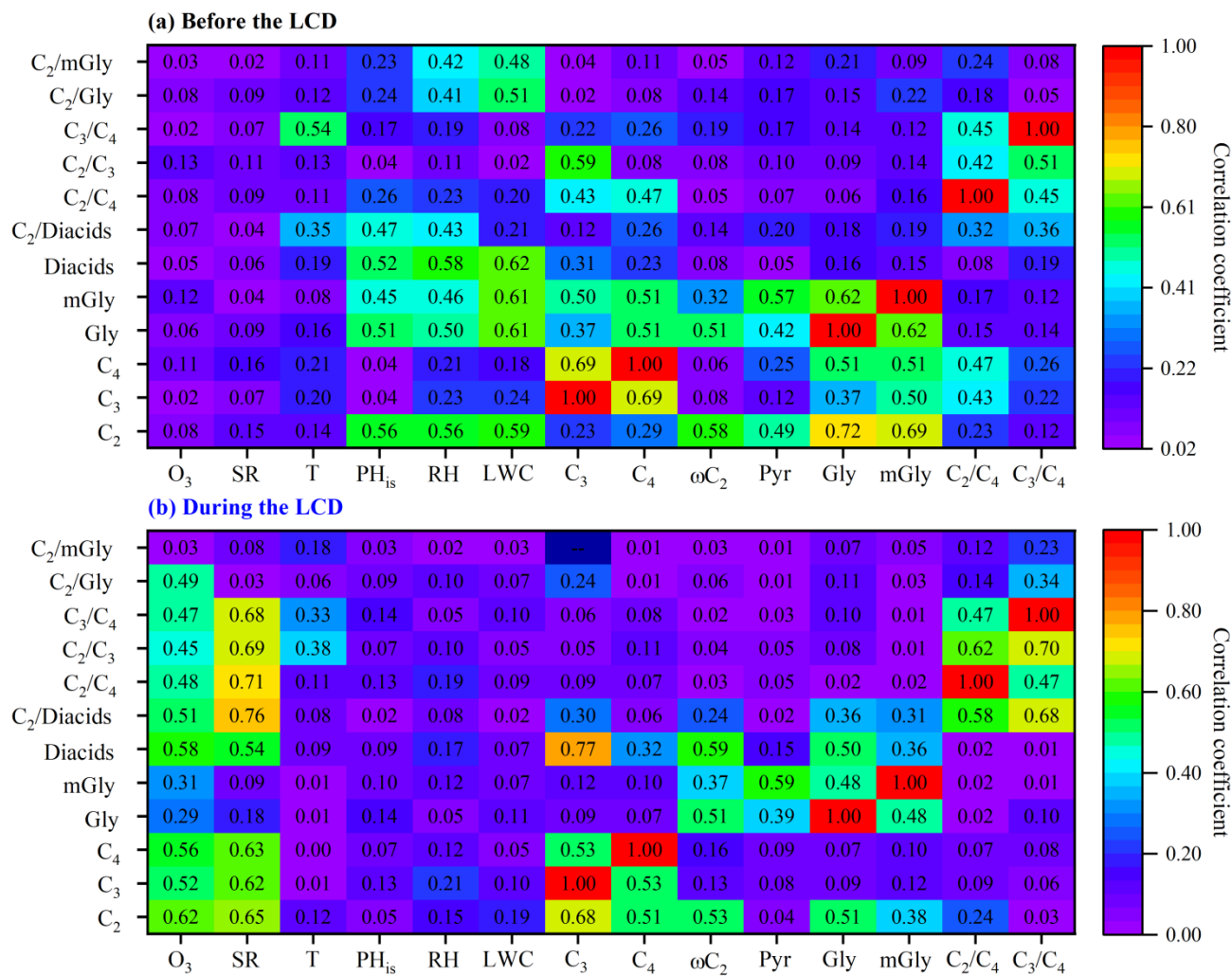
Figure 4. Correlation coefficients (R^2) of concentrations of C_2 and its organic precursors and selected ratios with influencing factors (a) before the LCD and (b) during the LCD.

9. R2, C4: Can you comment on the outlier? I agree with the referee's comment and your revision to remove the regression line. However, you should add a comment on the outlier either in the figure caption or main text.

Response: We have added a comment on the outlier in the revised manuscript as follows:

Moreover, the $\delta^{13}C$ value of tPh was negatively (one outlier that possessed a relatively high relative abundance of tPh in diacids was removed) in the daytime or poorly correlated with the ratio of

tPh/diacids at night before the LCD (Fig. 8f). The δ^{13}C value of tPh presented a negative correlation with the tPh/diacids during the LCD (Fig. 8l).

Please see Lines 458-461, Page 12 in the revised manuscript.

10. R2, C5: Given that you base a large part of your discussion on the ozone concentrations, you should discuss in more detail the reasons for the differences in ozone concentrations before and during LCD. I agree that the MLR/random forest analysis might get too far in the present manuscript; however, uncertainties associated with the various factors influencing the differences in ozone concentrations should be mentioned.

Response: Suggestion taken. We have added related discussions as follows:

O_3 is produced from nonlinear processes, and O_3 in Chinese cities is largely produced from NO_x (NO + NO_2)-saturated regime attributed to the scared HO_x radicals during the wintertime (Le et al., 2020). The significant drop of NO_2 during the LCD resulted in the reduction of NO concentration (Xu et al., 2020b), and further alleviated the efficient titration effect of O_3 (Levy et al., 2014). Thus, the lower concentration of NO_2 during the LCD could lead to an increase in O_3 concentration. Besides, O_3 exhibited a negative correlation with $PM_{2.5}$ mass concentration ($R^2 = 0.57$) during the LCD, due to the weakened aerosol radiative effect on the photochemical formation of O_3 (Wu et al., 2020) and the reduced precursors of O_3 (Li et al., 2019). The more favorable atmospheric conditions such as the higher temperature and stronger solar radiation during the LCD were beneficial for the generation and accumulation of O_3 (Li et al., 2019).

Please see Lines 176-184, Page 5 in the revised manuscript.

11. R2, C6 i: Where is this comment addressed in the revised manuscript? Even in the new text S3, I could not find any discussion on strong and weak species.

ii: Please add information on how you decided which acids to keep out of the models and why.

iii and iv: Is this information added to the manuscript? Where?

Response: (i) We have added related discussions on strong and weak species as follows:

Based on Norris et al. (2004) and Nayebare et al. (2018), the comparatively high signal-to-noise (S/N) ratios > 5 could be labeled as "strong" to enhance their effect on the model results for all species. The S/N ratios of species used in PMF model were higher than 8, thus these species were labeled as "strong".

Please see Lines 67-69, Page 4 in the revised Supplement.

(ii) We have added related information as follows:

The concentrations of selected major diacids (including C_2, C_3, C_4, C_9, and Ph), oxoacids (including Pyr and ωC_2), and α-dicarbonyls used in the PMF model accounted for 76.1 $\pm$ 6.0% and 82.8 $\pm$ 6.6% of total concentration of detected organic components (TDOCs) before and during the LCD, respectively. The other organic compounds (including C_5-C_8, C_{10}, C_{11}, iC_4-iC_6, M, F, mM, iPh, tPh, kC_3, kC_7, ωC_3, ωC_4 and ωC_7-ωC_9) presented relatively low concentrations and occasionally were below the detection limit, which could enhance the uncertainties. Moreover, these unconsidered organic compounds were less indicative for identified sources than the selected major acids. Thus, we kept those organic compounds with low concentrations out of the model.

Please see Lines 70-76, Page 4 in the revised Supplement.

(iii) We have added related information as follows:

As discussed in Section 3.3, SO_4^{2-} and LWC could be considered as significant influencing factors during the aqueous formation of C_2 before the LCD, and O_3 was a key influencing factor during the photochemical formation of C_2 during the LCD. Therefore, those species have been put in the PMF model to distinguish the aqueous pathway from the gaseous photochemical pathway of the diacids formation.

Please see Lines 77-80, Pages 4-5 in the revised Supplement.

(iv) We have added related information as follows:

The PMF model provides three uncertainty estimation methods to examine the robustness of the solution, namely, bootstrap (BS), displacement (DISP), and bootstrap combined displacement (BS-DISP), which can also provide a distribution range of the factor profiles through resampling for multi times and displacing with the Q value change within the preset value. Further details related to the error estimation methods can refer to Brown et al. (2015), Vossler et al. (2016), and Wang et al. (2018).

In this study, PMF solutions of 4-6 factors were analyzed and showed the convergence result. The relevant Q values and Q_{true}/Q_{robust} for these solutions were shown in Table S5. Finally, the five-factor solution was selected as the best solution with physical means before (Q_{true}/Q_{robust}=1.05, $Q_{true}/Q_{expected}$=1.27) and during the LCD (Q_{true}/Q_{robust}=1.05, $Q_{true}/Q_{expected}$=1.36), respectively. The error code of DISP before or during the LCD was 0 (Table S6), indicating no error. The values in the first row of dQ^{max} = 4 were zero before and during the LCD (Table S6), indicating that there was no significant rotational ambiguity and that the solution was sufficiently robust to be used. Mapping over 82% of the factors before or during the LCD indicated that the BS uncertainties could be interpreted and the number of factors may be appropriate. The Largest decrease in Q before and during the LCD was -0.048% and -0.061%, respectively (Table S6), suggesting the five-factor solution was more reliably attributed to sources than the four-factor solution and six-factor solution.

Please see Lines 81-93, Page 5 in the revised Supplement.

Table S5. Q values for PMF Analysis with different number of factors.

Number of factors	*R^2 for all input species	Q_{true}	Q_{robust}	$Q_{true}/Q_{expected}$	Q_{true}/Q_{robust}
(a) Before the LCD					
4	0.63-0.98	1556.1	1480.2	2.241	1.051
5	0.68-0.99	1176.9	1123.5	1.273	1.047
6	0.60-0.98	905.2	851.6	2.454	1.062
(a) During the LCD					
4	0.32-0.96	2722.0	2396.5	2.624	1.136
5	0.52-0.99	1859.3	1768.2	1.360	1.052
6	0.37-0.84	1334.1	1294.6	2.912	1.071

*R^2 between the measured and predicted species

Table S6. Summary of error estimation diagnostics from BS and DISP for PMF (a) Before the LCD and (b) During the LCD.

(a) Before the LCD						
DISP Diagnostics		Error code: 0		Largest decrease in Q:-0.048		
Factor Swaps	dQ^{max}=4	0	0	0	0	0
	dQ^{max}=8	0	0	0	1	1
	dQ^{max}=15	0	1	3	6	0
	dQ^{max}=25	0	1	5	8	10
(b) During the LCD						
DISP Diagnostics		Error code: 0		Largest decrease in Q:-0.061		
Factor Swaps	dQ^{max}=4	0	0	0	0	0
	dQ^{max}=8	0	0	0	0	0
	dQ^{max}=15	0	0	1	1	0
	dQ^{max}=25	0	1	8	10	6

12. R2, C7 ii: Please improve the captions of Tables S2 and S3. Did you use all, i.e. day and nighttime values in Table S2? How meaningful is such statistics given that there are significant differences in day vs night values as shown in Table S3? Add also sufficient information, either in the data base referred to in the data availability statement or in the supplement that the reader understands which statistical test was performed for which compounds and why.

Response: Thanks for your suggestion. We have revised captions of Table S3 to "Table S3. Statistic test for the day-night differences in concentrations and mass ratios of major species in $PM_{2.5}$ before and during the LCD, respectively." (see Lines 117-118, Page 10 in the revised Supplement).

Following the reviewer's suggestion, we added Table S3 to assess whether the day-night differences before or during the LCD are significant for concentrations and mass ratios of major species or not.

We have added information in Table S2 and Table S3 as follows:

[a]When the variables follow a normal distribution, ANOVA was employed to analyze the statistically significant differences between these two datasets before and during the LCD. [b]Otherwise, the Kruskal-Wallis test was used. Bold font represented the species (or ratios) that were statistically significantly different ($p < 0.05$).

[a]When the variables follow a normal distribution, ANOVA was employed to analyze the statistically significant differences during the daytime and nighttime. [b]Otherwise, the Kruskal-Wallis test was used. Bold font represented the species (or ratios) that were statistically significantly different ($p < 0.05$).

Please see Lines 111-122, Pages 9-10 in the revised Supplement.

13. Comments: R2, C8: I am not sure that the authors understood the referee's comment correctly. In the revised manuscript, the pH is now stated as 3.2 ± 3.0 which would correspond to an H^+ concentration of 6.3e-4 M $\pm$ 1e-3 M – but yet the reported range of measured values is $2.3 - 4.4$, i.e. $[H+] = $ 4e-5 – 5e-3 M. Please clarify.

Response: According to the reviewer's suggestion, values need to be converted back to the $[H^+]$ before averaging for the result to be mathematically correct because the pH scale is logarithmic and not a ratio variable. Sorry for the mistake. The pH_{is} value ranged from 2.3 to 7.7 (average value: 3.2 ± 3.0) which corresponded to an H^+ concentration (range: 2.0e-8 M to 5.6 e-8 M; average: 6.6e-4 M $\pm$ 1.1e-3 M).

We have changed it in Table 1 (see Page 22).

II Additional editor comments

Major comments

1. In the title and text throughout the manuscript, you imply that the lockdown led to differences in the concentrations of diacids before and during the lockdown. However, given that the meteorological conditions (T, RH) were also vastly different during the two periods, the diacid levels were likely also affected by these conditions. Your statement in lines 173/174 point to this but this is too brief. Please emphasize this in the discussion and possibly adjust the title accordingly.

Response: We agree that the meteorological conditions (T, RH) were also vastly different during the two periods, the diacid levels were likely also affected by these conditions. So, we discussed different meteorological conditions before and during the LCD on the formation of diacids and related compounds in Sections 3.3-3.4. For example:

Our results suggest that these species had different sources and underwent different formation processes because of different concentration levels of organic precursors and meteorological conditions before and during the LCD (see Lines 248-250, Page 7).

Considering the higher RH and LWC concentration at night, the increased concentrations of C_2 and related SOA during the nighttime may be closely linked to the accelerated aqueous production (Cheng et al., 2015; Meng et al., 2020) (see Lines 301-302, Page 8).

To explore the formation pathways and contributing factors of C_2 and related SOA before the LCD, the temporal variations of major diacids, LWC, pH_{is}, and meteorological parameters (e.g., solar radiation, temperature, and RH) were illustrated in Fig. 5 (see Lines 322-324, Page 8).

Noticeably, the concentrations of C_2 and diacids, as well as C_2/diacids ratio culminated on the nighttime of January 23, which was characterized by significantly higher LWC concentration (172 μg m^{-3}) and RH (86.9%) (Fig. 5) (see Lines 330-332, Page 9).

The higher RH and LWC concentration were favorable for the partitioning of Gly and mGly from gaseous phase to aqueous phase and forming C_2. As shown in Fig. 5, the enhanced concentrations of Gly and mGly in $PM_{2.5}$ before the LCD were observed when RH and LWC increased. Thus, C_2 and its

precursors (including Gly and mGly) were positively correlated with RH and LWC, respectively ($R^2 >$ 0.45, Fig. 4a) (see Lines 335-338, Page 9).

Therefore, LWC and RH can be regarded as vitally important factors controlling the aqueous production of C_2. The nighttime concentrations of LWC and RH were higher than those during the daytime, which led to the higher concentration and percentage contribution of C_2 in the nighttime (see Lines 341-343, Page 9).

Given the higher O_3 concentration and stronger solar radiation during the LCD (Table 1), it can be expected that the enhanced concentration and contribution of C_2 were driven by the promoted photochemical oxidation, which was supported by the significantly higher C_3/C_4 ratio (1.6 ± 0.4) during the LCD than that (0.3 ± 0.1) before the LCD (see Lines 368-370, Page 10).

In view of the significant escalation of O_3 concentration and solar radiation during the LCD, it could be concluded that the productions of C_2 and related compounds may be closely involved in the photochemical pathways driven by the higher O_3 concentration and stronger solar radiation (see Lines 354-387, Page 10).

Additionally, C_2 and diacids concentrations exhibited similar patterns of variations (Fig. 5) and strong correlations ($R^2 > 0.5$, Fig. 4b) with solar radiation during the daytime. However, such similarities and strong correlations of those were not observed with temperature ($R^2 < 0.2$, Fig. 4b and Fig. 5), suggesting that the effect of temperature on the photochemical formation of C_2 was negligible (see Lines 393-396, Page 10).

2. (i) Throughout the manuscript, you refer to ozone concentrations that may be responsible for different patterns of the diacid concentrations. Section 3.4 is even named 'O_3-dominated formation of SOA'. This is misleading as most likely many oxidation pathways of the organics are by the OH radical and/or other oxidants. Only in line 321, you state that OH measurements were unavailable. Please rephrase all instances where you refer to O_3-dominated pathways and O_3 as the main oxidant. (ii) Can you refer to any OH measurements before and during the lockdown? Could it be simply possible that OH during the lockdown was higher because fewer sinks were present?

Response: (i) Thanks for your suggestion. We have rephrased all instances which referred to O_3-dominated pathways and O_3 as the main oxidant as follows:

We have changed "…O_3-dominated formation pathways are involved in…" to "…formation pathways influenced by O_3 are involved in…" in the revised manuscript (see Line 67, Page 2).

We have changed "Enhanced O_3-dominated formation of SOA during the LCD" to "Enhanced photochemical formation of SOA during the LCD" in the revised manuscript (see Line 365, Page 9).

We have changed "…involved in the O_3-dominated photochemical pathways under the stronger solar radiation." to "…involved in the photochemical pathways driven by the higher O_3 concentration and stronger solar radiation." in the revised manuscript (see Lines 386-387, Page 10).

We have changed "…which represented photochemical oxidation dominated by O_3." to "…which represented gaseous photochemical oxidation" in the revised manuscript (see Lines 501-502, Page 13).

We have changed "The photochemical oxidation dominated by O_3…" to "The gaseous photochemical oxidation…" in the revised manuscript (see Lines 509-510, Page 13).

We have changed "…from the O_3-dominated photochemical pathways during the LCD" to "…from the photochemical pathways during the LCD" in the revised manuscript (see Line 530, Page 13).

(ii) We have referred to OH·radical before and during the lockdown as follows:

A few studies have observed that the OH·radical in the North China Plain (e.g., Shandong Province) was higher during the LCD (e.g., in February 2020) than that before the LCD (Gaubert et al., 2021; Kang et al., 2021; Li et al., 2021). The reduced NO_x during the LCD could lead to higher OH·radical, because less OH·radical could be consumed with NO_2 to produce nitric acid (Gaubert et al., 2021). Besides, the elevated O_3 concentration during the LCD could result in the enhanced OH·radical, as OH·radical is mainly from the O_3 photolysis with the water vapor in the atmosphere (Kang et al., 2021). It could be concluded that the OH·radical during the LCD was higher than before the LCD, being consistent with O_3 concentration. Because OH·radical was unavailable in this work, O_3 was used here as a marker for the oxidant concentration of photochemical oxidation.

Please see Lines 376-383, Page 10 in the revised manuscript.

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3. The formation and ratios of diacids have been discussed in numerous previous studies for a wide range of conditions and locations (work by the same and other authors). How do the results from such studies compare to those found here?

Response: Suggestion taken. We have made a comparison of C_2, diacids, and TDOCs concentrations, and C_3/C_4 ratios in Jinan with those in other Asian megacities during the wintertime and illustrated in Table S4 as follows:

Table S4. Comparison of C_2, diacids, and TDOCs concentrations (ng m^{-3}), and C_3/C_4 ratio in Jinan with those in other Asian megacities during the wintertime.

Location	Period	Size	C_2	Diacids	TDOCs	C_2/C_4	C_3/C_4	Data source
14 Chinese cities	13–14 Jan 2003	PM$_{2.5}$	558±351	904±480	974±499	7.0	0.5	(Ho et al., 2007)
Beijing, China	Dec 2013	PM$_{2.5}$	149±123	366±261	450±368	4.7	0.6±0.1	(Zhao et al., 2018)
Chengdu, China	7–23 Jan 2013	PM$_{2.1}$	1380	3390	4285	3.2	0.5±0.1	(Li et al., 2015)
Xi'an, China	Jan–Feb 2009	PM$_{10}$	1162±570	1843±810	2207±1044	11.9	1.0	(Cheng et al., 2013)
Tianjin, China	23 Nov–11 Dec 2016	PM$_{2.1}$	526±335	1223±623	1438±739	3.2	0.3	(Devineni et al., 2023)
Liaocheng, China	17 Jan–16 Feb 2016	PM$_{2.5}$	817±544	1415±899	1677±1026	3.6±0.9	0.4±0.2	(Meng et al., 2020)
Guangzhou, China	Dec 2006–Jan 2007	PM$_{2.5}$	182±106	384±171	414±184	9.9	0.7	(Ho et al., 2011)
Tianjin, China	1–28 Feb 2019	PM$_{2.5}$	668±470	1137±707	1561	7.6	1.0	(Zhao et al., 2023)
Padori, South Korea	1–28 Feb 2019	PM$_{2.5}$	389±204	582±266	750	8.3	0.7	(Zhao et al., 2023)
Daejeon, South Korea	1–28 Feb 2019	PM$_{2.5}$	516±327	784±446	1108	10.0	0.7	(Zhao et al., 2023)
Ulaanbaatar, Mongolia	Nov 2007–Jan 2008	PM$_{2.5}$	107±28	536±156	697±207	1.8±0.8	0.2±0.1	(Jung et al., 2010)
Chennai, India	23–28 Jan 2007	PM$_{10}$	472±137	695 ±176	765	11.8	1.5	(Pavuluri et al., 2010)
Chennai, India	29 Jan–6 Feb 2007	PM$_{10}$	380±89	641 ±157	719	7.9	1.2	(Pavuluri et al., 2010)
Tokyo, Japan	20–21 November 1989	TSP	186	438	595	3.9	0.9	(Kawamura and Yasui, 2005)
Jinan, China	6–23 Jan 2020	PM$_{2.5}$	181±48	351±92	437±117	3.9±1.5	0.3±0.1	This study
Jinan, China	31 Jan–17 Feb 2020	PM$_{2.5}$	239±108	386±127	486±144	8.4±3.4	1.6±0.4	This study

In addition, we have added related discussions in the revised manuscript as follows:

The concentrations of diacids and TDOCs in this study were significantly lower than those in Xi'an (Cheng et al., 2013), Chengdu (Li et al., 2015), Tianjin (Devineni et al., 2023; Zhao et al., 2023), Liaocheng (Meng et al., 2020), 14 Chinese cities, and other Asian megacities such as Padori, Daejeon (Zhao et al., 2023), Ulaanbaatar (Jung et al., 2010), Chennai (Pavuluri et al., 2010), and Tokyo (Kawamura and Yasui, 2005), but similar to those in Beijing (Zhao et al., 2018) and Guangzhou (Ho et al., 2011) during the wintertime (Table S4) (see Lines 228-233, Page 6).

The C_3/C_4 ratio before the LCD was lower than that in other Asian megacities such as 14 Chinese cities (Ho et al., 2007), Beijing (Zhao et al., 2018), Daejeon (Devineni et al., 2023), and Chennai (Pavuluri et al., 2010), but comparable to that in Tianjin where biomass burning, biogenic sources, and their aging contributed significantly to diacids and related compounds (Devineni et al., 2023) (Table S4).

However, the C_3/C_4 ratio during the LCD was much higher than that in other Asian megacities (Table S4), again implying the significantly enhanced photochemical oxidation during the LCD (see Lines 254-259, Page 7).

5. l. 43: High solubility and hygroscopicity are not necessary for good ice nuclei.

Response: Suggestion taken. We have deleted "and ice nuclei (IN) formation" in the revised manuscript.

6. l. 53: what do you mean by 'oxidative capacity of aerosols'? This expression is not used in any of the cited references and not clear in this context.

Response: Thanks for your suggestion. We meant by "oxidative capacity of atmosphere". We have changed it in the revised manuscript (see Line 53, Page 2).

7. l. 58 – 67: (i) There are many lab and model studies that have explored the formation of oxalic acid, e.g. (Carlton et al., 2007; Crahan et al., 2004; Perri et al., 2009; Warneck, 2003). In these studies, the main oxidant was OH. Please comment on this and include appropriate references. (ii) Given the small proportion of oxalic acid to total aerosol loading, its effect on air quality can be assumed negligible.

Response: Suggestion taken.

(i) We have added comments about these references as follows:

Modeling studies and laboratory experiments also demonstrate the in-cloud C_2 formation (Crahan et al., 2004; Lim et al., 2005; Warneck, 2003) from the oxidation of Gly (Carlton et al., 2007) and other gaseous "Gly-like" precursors such as glycolaldehyde (Perri et al., 2009) and mGly (Altieri et al., 2008) with OH ·radical.

Please see Lines 59-62, Page 2 in the revised manuscript.

(ii) We have changed it as follows:

Therefore, further investigations on C_2 and related compounds are necessary to provide a knowledge base for better understanding SOA and improving the accuracy of model.

Please see Lines 69-70, Page 2 in the revised manuscript.

8. l. 83/4: Also aqueous phase processes can be photochemical. Do you mean 'oxidation in the aqueous vs gas phase'?

Response: Yes, we meant oxidation in the aqueous versus gas phase. We changed it to "…aqueous oxidation versus gas-phase photochemical oxidation to…" in the revised manuscript (see Lines 85-86, Page 3).

9. l. 168: SOA formation can be also a strong function of NOx level. Thus, enhanced ozone concentrations do not necessarily lead to higher SOA.

Response: We agreed with the editor's suggestion that SOA formation can be also a strong function of NO_x level. As discussed in Section 3.4, we observed that the highest O_3 concentration was observed on the daytime of January 31 when the concentrations of C_2 and diacids reached their peaks. Additionally, both C_2 and diacids concentrations as well as the C_2/diacids ratio during the LCD exhibited robust correlations with O_3 ($R^2 > 0.5$, Fig. 4b) during the LCD, suggesting that the increase of O_3 resulted in the higher concentration of diacids and related compounds. The enhanced concentrations of diacids and related compounds were also due to other oxidants such as $OH \cdot$ radical, NO_x, and so on. To express precisely, we deleted it in the revised manuscript.

10. l. 175 – 185: What is the extent to which the differences in EC, OC, WSOC, $PM_{2.5}$, levoglucosan etc canbe ascribed to different air masses rather than different emission strengths? (How) Can the back trajectories help in answering this?

Response: The Results of backward trajectory analysis showed that air masses before and during the LCD in Jinan were different (Fig. S1). Thus, the differences in EC, OC, WSOC, levoglucosan, and other species might be not only because of different emission strengths and types of sources, but also due to different air masses between these two periods. We have added it in the revised manuscript in Lines 194-196, Page 5.

11. l. 191 ff and Figure 2: (i) The text states that TDOC/CO is discussed whereas the figure caption only says TDOC. Please correct. (ii) Why do you expect the increase of diacids to follow Arrhenius Law?

While the formation of diacids might follow such a temperature dependence, so does also their loss by further oxidation. (iii) If the loss may be negligible, why should the temperature dependence during and before LCD have a different exponent?

Response: (i) Sorry for the mistake. We have changed it to "Correlation analysis between the ratio of total concentration of detected organic components (TDOCs) normalized by CO (TDOCs/CO) and temperature". Please see Lines 852-853, Page 26 in the revised manuscript.

(ii) When exploring the secondary formation of diacids and related compounds, the total concentration of detected organic components (TDOCs) was usually normalized by CO (Yu et al., 2021) to eliminate the effect of atmospheric dilution and better assess the secondary formation rate. The ratio of TDOCs to CO (TDOCs/CO) increased exponentially with the increase of temperature before ($y = 257.46e^{0.019x}$, $R^2 = 0.56$, Fig. 5a) and during ($y = 301.49e^{0.067x}$, $R^2 = 0.58$, Fig. 5c) the LCD, which was consistent with the Arrhennius Law.

We have added related discussions in the revised manuscript as follows:

As shown in Fig. 2, the ratio of total concentration of detected organic components (TDOCs) normalized by CO (TDOCs/CO) increased exponentially with the increase of temperature before ($y = 257.46e^{0.019x}$, $R^2 = 0.56$, Fig. 5a) and during ($y = 301.49e^{0.067x}$, $R^2 = 0.58$, Fig. 5c) the LCD, which was consistent with the Arrhennius Law, confirming that TDCOCs in this study were primarily derived from secondary formation and the contribution of primary emissions was insignificant.

Please Lines 206-210, Page 6 in the revised manuscript.

(iii) TDOCs are considered as stable products of secondary oxidation for a number of hydrocarbons (Martinelango et al., 2007). The loss of diacids (e.g., C_2) through the photolysis of iron oxalate complexes is a dominant sink from field observations and model studies (Cheng et al., 2017; Pavuluri and Kawamura, 2012; Weller et al., 2014; Zhou et al., 2015), while these species are stable in the absence of Fe (Kunwar et al., 2019). Previous studies have pointed out that diacids and related compounds presented a strong correlation with temperature, emphasizing the significance of secondary formation of those compounds with the increase of temperature (Kawamura and Bikkina, 2016; Kawamura and Yasui, 2005; Meng et al., 2014, 2018). Therefore, the loss of diacids and related compounds may be negligible when the temperature increases.

We have added above discussions Lines 210-216, Page 6 in the revised manuscript.

Different meteorological factors (e.g., temperature, solar radiation, and RH), oxidants (e.g., O_3 and OH · radical), emission strengths and types of organic precursors, physicochemical properties of aerosols (e.g., pH_{is} and LWC), and other influencing factors before and during the LCD could lead to different secondary formation rates of TDOCs between these two observation periods. Thus, the temperature dependence before and during LCD had a different exponent.

In the revised manuscript, we have added this discussion in Lines 218-221, Page 6: "Different secondary formation rates of TDOCs between these two observation periods were largely due to different meteorological factors (e.g., temperature, solar radiation, and RH), oxidants (e.g., O_3 and OH · radical), emission strengths and types of organic precursors, physicochemical properties of aerosols (e.g., pH_{is} and LWC), and other influencing factors. ".

12. l. 271: Aqueous phase formation of C_2 is also photochemical as it is driven by OH (and less by NO_3 or other dark processes). What is the basis of your assumption of an accelerated formation of C_2 during night?

Response: Considering the higher RH and LWC concentration at night, the increased concentrations of C_2 at night may be closely linked to the accelerated aqueous formation. As discussed in Section 3.3, the concentrations of C_2 and diacids, as well as C_2/diacids ratio culminated on the nighttime of January 23, which was characterized by significantly higher LWC concentration (172 μg m^{-3}) and RH (86.9%) (Fig. 4). The conditions of higher LWC and RH are favorable for the partitioning of Gly and mGly from gaseous phase to aqueous phase, so C_2 and its precursors (e.g., Gly and mGly) were positively correlated with RH and LWC, respectively ($R^2 > 0.45$, Fig. 5). Moreover, the ratios of C_2/Gly and C_2/mGly also showed a significant correlation with RH and LWC ($R^2 > 0.4$, Fig. 5). Such strong correlations suggest that the higher LWC concentration and RH could not only promote the formations of C_2 and its precursors, but also facilitate the transformation of C_2 from the organic precursors. Therefore, LWC and RH can be regarded as vitally important factors controlling the aqueous production of C_2. The nighttime concentration of LWC and RH were higher than those during the daytime, which

led to the higher concentration and percentage contribution of C_2 in the nighttime. To express precisely, we have rephrased these sentences in the revised manuscript as follows:

…, which was mainly ascribed to the accelerated aqueous formation of C_2 at night (see following discussions in Section 3.3).

Please see Lines 314-315, Page 8 in the revised manuscript.

13. l. 291: Why should Gly and mGly show a positive correlation with RH and LWC? Wouldn't one expect the opposite, i.e. more efficient oxidation in the presence of water? They are formed by gas phase processes, i.e. independently from the presence of water.

Response: Gly and mGly are gaseous oxidation products of biogenic and anthropogenic VOCs, and both are highly water-soluble (Carlton et al., 2006). In the presence of liquid water, both species can dissolve in the aqueous phase following Henry's law (Myriokefalitakis et al., 2011). The higher RH and LWC concentration were favorable for the partitioning of Gly and mGly from gaseous phase to aqueous phase. As shown in Fig. 4, the enhanced concentrations of Gly and mGly in $PM_{2.5}$ before the LCD were observed when RH and LWC increased. Thus, Gly and mGly presented robust correlations with RH and LWC ($R^2 > 0.45$, Fig. 4a)). Moreover, the ratios of C_2/Gly and C_2/mGly, which could evaluate the oxidation degree of organic aerosols, also showed a significant correlation with RH and LWC ($R^2 > 0.4$, Fig. 4a), suggesting the more efficient oxidation in the presence of higher water. Therefore, the higher LWC concentration and RH could not only promote the formations of Gly and mGly, but also facilitate the transformation of these two α-dicarbonyls to C_2.

We have added the above discussions in the revised manuscript. Please see Lines 332-338, Page 9.

14. l. 306ff: Both the oxidation of glyoxylic acid and the oxidation of oxalic acid are pH dependent since the rate constants of the anions differ from those of the undissociated acids. Could these differences in formation/loss rates explain the different concentrations at different pH?

Response: The reaction rate constant ($3.6 \times 10^8 \ M^{-1} \ s^{-1}$) of glyoxylic acid ($\omega C_2$) with OH·radical to form C_2 is smaller than that ($2.9 \times 10^9 \ M^{-1} \ s^{-1}$) of its anion, glyoxylate, thus the coarse particles during dust period, which are alkaline, are favorable for the C_2 formation from ωC_2 (Wang et al., 2015). Tan et

al. (2009) reported that acidity had a minor effect on C_2 formation at cloud- and fog-relevant conditions via online experiments. However, field measurements and chamber studies have suggested that aerosol acidity is favorable for the formation of C_2 and its precursors (Cheng et al., 2017; Jang et al., 2002; Meng et al., 2014; Surratt et al., 2007; Wang et al., 2016; Yu et al., 2021). Those findings were necessarily conflict with each other, largely because the concentration levels of organic precursors, H^+, LWC, and other influencing factors were different from the aerosols in our filed observations. Therefore, further field measurements of wet aerosols, laboratory experiments, and model simulations are urgently necessary to elucidate the influencing mechanism of acidity on C_2 formation. We have added related discussions in the revised manuscript as follows:

As shown in Fig. 4a, pH_{is} exhibited pronounced negative relationships with C_2 and its precursors such as Gly and mGly ($R^2 \geq 0.45$), which was also found in other field studies (Cheng et al., 2017; Meng et al., 2014; Wang et al., 2016; Yu et al., 2021), possibly because of the fact that aerosol acidity could favor the productions of C_2 and its precursors (see Lines 353-356, Page 9).

The reaction rate constant (3.6×10^8 M^{-1} s^{-1}) of ωC_2 with OH $\cdot$radical to form C_2 is smaller than that (2.9×10^9 M^{-1} s^{-1}) of its anion, glyoxylate, thus the coarse particles during the dust period, which are alkaline, are favorable for the C_2 formation from ωC_2 (Wang et al., 2015). Tan et al. (2009) reported that acidity had a minor effect on C_2 formation at cloud- and fog-relevant conditions via online experiments. Those findings necessarily 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 filed observations. Therefore, further field measurements of wet aerosols, laboratory experiments, and model simulations are urgently necessary to elucidate the influencing mechanism of acidity on C_2 formation (see Lines 357-364, Page 9).

15. l. 308 vs 338: These lines are contradicting each other. In the former, you write that the degradation of longer acids is not a source of C_2 whereas in the latter you state that it is likely a major source.

Response: As discussed in Section 3.3, we stated that the degradation of longer-chain diacids was not a source of C_2 before the LCD. As discussed in Section 3.4, we confirmed that C_2 during the LCD was primarily originated from the photochemical degradation of longer-chain diacids. 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

16. l. 349 – 356: This text is very unclear. What is the relevance of the R^2 = 0.49 for O3 and C2/Gly in relation to the lab study by Carlton who used OH as an oxidant?

Response: We cited the reference by Carlton et al. (2007) to express that the oxidation rate of Gly with oxidants is faster than that of mGly. We are sorry to ignore that different oxidants could result in different oxidation rates. Thus, we deleted "Carlton et al. (2007) have reported that the oxidation rate of Gly (3×10^{10} M^{-1} s^{-1}) with OH·radical is faster than that of mGly (1.1×10^9 M^{-1} s^{-1})." in the revised manuscript.

17. Sections 3.5.1 and 3.5.2. Given that the latter subsections repeats quite a bit of information from the former one, I suggest merging both sections into Section 3.5.

Response: Suggestion taken. We have merged both sections into Section 3.5. Please see Pages 11-12.

18. Section 3.6: I do not fully follow the categorization into the different factors: (i) Given that sulfate may be also formed by gas phase oxidation and/or originates from long range transport, it seems questionable to based the 'aqueous phase factor' solely on the presence of sulfate. (ii) Again, I do not understand why you distinguish between 'aqueous oxidation' and 'photochemical oxidation', (iii) biomass mass burning emissions might be also processed by oxidation processes in the gas and/or aqueous phases. Why is biomass burning a separate factor?

Response: (i) We agree that it seems questionable to base the 'aqueous phase factor' solely on the presence of sulfate. As discussed in Section 3.3, LWC could be considered as a significant indicator for the aqueous formation. Thus, LWC has been put in the PMF model to identify the aqueous pathway. It

is worth mentioning that the results re-run by the PMF model (Fig. 9) in the revised manuscript are similar to that in the original manuscript.

(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 accurately, the secondary oxidation was divided into aqueous oxidation and gaseous photochemical oxidation. Yu et al. (2021) have 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.

We have modified related discussion and Fig. 9 in the revised manuscript. Please see the reply of the second comment of editor and Fig. 9.

(iii) Diacids and related compounds can be produced from the photochemical oxidation of volatile and semi-volatile organic precursors emitted from biomass burning as well as dark ozonolysis of VOCs and other organics, accretion reactions and oxidation of nonvolatile organics such as unsaturated fatty acids (Cao et al., 2017; Kawamura et al., 2013; Meng et al., 2013; Xu et al., 2020a). In addition, biomass burning can directly emit diacids and related compounds (Fu et al., 2008; Gao et al., 2003; Kundu et al., 2010; Narukawa et al., 1999; Shen et al., 2022). Therefore, biomass burning which is considered as an important primary source in Section 3.6 is an independent source to separate from secondary oxidation. To better distinguish primary sources and secondary sources, primary sources (including dust emission, coal combustion, and biomass burning) were highlighted by light green color, and secondary sources (including aqueous oxidation and gaseous photochemical oxidation) were highlighted by light blue color in Fig. 9.

We have changed related discussions in the revised manuscript as follows:

Although diacids and related compounds can be produced from the secondary oxidation of organic precursors from biomass burning (Cao et al., 2017; Kawamura et al., 2013), biomass burning can

directly emit those compounds (Fu et al., 2008; Gao et al., 2003; Kundu et al., 2010; Narukawa et al., 1999; Shen et al., 2022). Thus, the fifth factor could be regarded as a primary source of biomass burning (see Lines 497-500, Page 13).

Levoglucosan, C_9, EC, and OC presented the stronger loadings in the third factor, indicating a primary source of biomass burning (see Lines 503-504, Page 13).
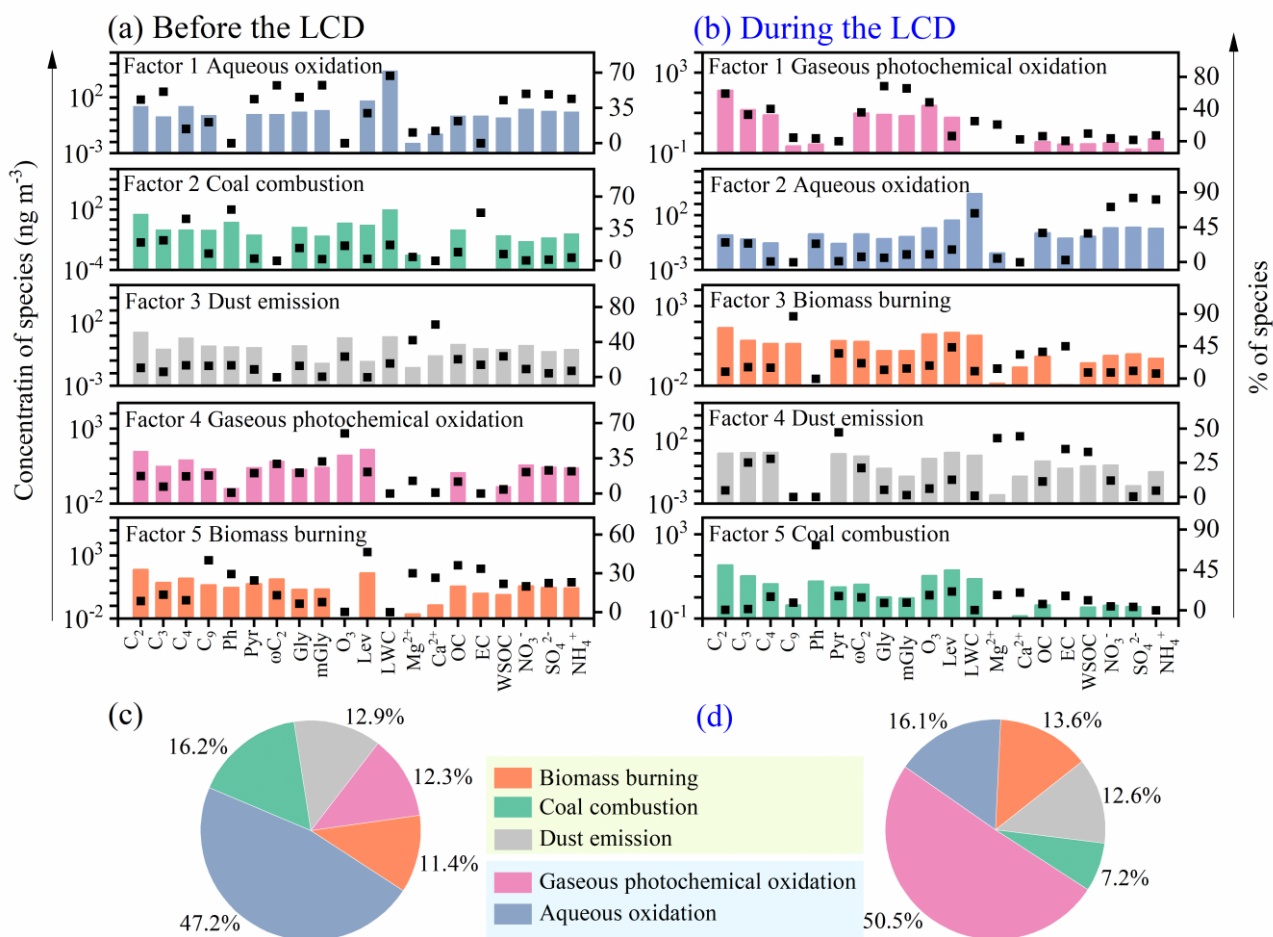

Figure 9. Source profiles of major chemical components in the PM$_{2.5}$ samples from Jinan (a, c) before the LCD and (b, d) during the LCD (BB: biomass burning).

Technical comments

Please carefully proofread your manuscript. There are many grammar and language mistakes as well as numerous typos. I list a few below but this list is by no means complete. (line numbers refer to the manuscript version without annotation).

Response: Thanks for your suggestion. We have carefully proofread our manuscript and changed the grammar and language mistakes as well as numerous typos.

19. l. 15: remove 'the' before eastern China.

Response: Corrected.

20. l. 16: 'evolutionary process' could be replaced by 'formation'.

Response: Corrected.

21. l. 17: Reword this sentence, e.g. 'Samples of ... were taken...' (samples are not measured).

Response: Suggestion is taken. We have changed it to "The samples were analyzed for…" in the revised manuscript. Please see Line 16, Page 1.

22. l. 19: replace 'element' by 'elemental'.

Response: Corrected.

23. l. 45: replace 'activity' by 'sources'.

Response: Corrected.

24. l. 49: replace 'aids' by 'acids'.

Response: Corrected.

25. l. 115: should 'dervied' be 'derivatized'?

Response: Corrected.

26. 116: replace 'quantificationally' by 'quantitatively'.

Response: Corrected.

27. l. 127: remove 'and' in 'and the derivatized'.

Response: Corrected.

28. l. 169: do you mean 'solar'?

Response: Yes. We have corrected it to "solar".

29. l. 170: concentration

Response: Corrected.

30. l. 262: replace 'makers' by 'markers'.

Response: Corrected.

31. Round numbers to significant digits. E.g., 22.7 ± 8.9 should be 23 ± 9 etc.

Response: Suggestion is taken. We have changed them in the revised manuscript.

32. Please carefully check all units in the text and tables for consistency and correctness. Individual acids are certainly not present in concentrations on the order of micrograms (e.g. l. 234)

Response: Suggestion is taken. We have carefully checked all units in the text and tables for consistency and correctness.

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