Dear ACP editor:

After reading the comments from you and the referees, we have carefully revised our manuscript. Our responses to the comments are itemized below.

Anything for our paper, please feel free to contact Prof. Pingqing Fu via fupingqing@tju.edu.cn.

All the best

Jingjing Meng
On behalf of Prof. Pingqing Fu
April 8, 2023

Referee(s)' Comments to Author:

Referee 1

We are thankful to the reviewer 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. This is an interesting article and provides some rare insights into secondary aerosols during the covid lockdown. However, authors need to do a better job to provide more robust statistical backing to their

dataset. Also, some of the conclusions made seem farfetched without strong evidence and rely solely on the previous literature. A major revision is suggested, and comments are provided in the attached document.

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

Scientific Comments:

2. 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. Why 10 min?

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.

3. After the collection, each filter was sealed in an aluminum foil bag and stored in a freezer (−20°C) prior to analysis. How long was the storage period?

Response: $PM_{2.5}$ samples in this study were collected from 6 January to 17 February 2020. The chemical analysis of these collected samples started from May to July in 2021. Therefore, the samples were stored for about 16 months before analysis (Line 104, Page 3). Each filter was sealed in an aluminum foil bag and stored in a freezer (−20°C) to prevent the volatilization of organic compounds.

4. Line 164: i) The same analytical method was also applied for field blank filters. What's the method? ii) provide brief details about GC-MS operating parameters for the analysis. How these parameters were selected? iii) Are the percent recoveries mentioned here good enough for such analysis? please reason.

Response: i) The field blank filters were treated as the ambient samples and analyzed by the same analytical procedure for the ambient samples. The method has been described in Lines 112-118, Pages 3-4.

ii) As for the details about GC-MS operating parameters, we have added it in the revised manuscript as follows:

GC-MS was performed on a Hewlett-Packard model Agilent 7890A GC coupled to a Hewlett-Packard model Agilent 5975C mass selective detector (MSD). GC separation was equipped with a split/splitless injection and a fused silica capillary column (DB-5MS, 30 m $\times$ 0.25 mm i.d., 0.25 μm film thickness). The GC oven temperature was programmed from 50 °C for 2 min to 120 °C at a rate of 15°C min^{-1}, and then to 300°C at a rate of 5°C min^{-1} with a final hold at 300°C for 16 min. The mass spectrometer was operated on the electron impact (IE) mode at 70 eV and scanned from 50 to 650 Da. These parameters were determined by a comparison of references, a library, and authentic standards (Fu et al., 2013; Kawamura and Ikushima, 1993; Kawamura et al., 2013).

Please see Lines 118-123, Pages 4.

iii) As described in previous studies, the recovery of oxalic acid (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 the manuscript are good enough for such analysis.

5. Line 177: Elemental carbon (EC), organic carbon (OC), and inorganic ion. Add WSOC also in the title.

Response: We have added it in the revised manuscript (Line 142, Page 4).

6. Line 154: atm oxi capacity is not only a function of O_3 concentration, authors need to substantiate the same more rigorously.

Response: We agree with that the atmospheric oxidation capacity is not only a function of O_3 concentration. It was regretful that we did not obtain the data of radicals, NO_x, and other atmospheric

oxidants. A few studies have reported that O_3 presented a clear relationship with atmospheric oxidative capacity (Li et al., 2021; Xu et al., 2006; Zhao et al., 2021). However, to express more precisely, we deleted the discussions about "O_3 is considered as a significant indicator of atmospheric oxidizing capacity" and changed these descriptions to "The decreased concentrations of EC, OC, and WSOC in $PM_{2.5}$ but the enhanced ratios of OC/EC and WSOC/OC during the LCD (Table 1, Fig. 1) indicated more SOA productions due to the stronger photochemical oxidation during the LCD", "the higher temperature and O_3 concentration during the LCD provided a favorable condition for α-dicarbonyls productions via secondary oxidation", and "because of the higher temperature and O_3 concentration under the more clear sky conditions", respectively.

7. Comments: Line 156: correct the spelling of "solor".

Response: We have corrected it in the revised manuscript (Line 169, Page 5).

8. Line 182: Full form of "TDOCs" should be explained at the very first instance itself.

Response: Suggestion taken. Please see Line 191 and Line 193 in Page 5.

9. Lines 185-186: "Additionally, the exponent number (0.067) of the regression trend line during the LCD was 3.5 times higher than that (0.019)". Are these regression values statistically significant?

Response: We did a statistic analysis ($p < 0.05$), which indicated that these regression values were statistically significant. We have added "$p < 0.05$" in the revised manuscript (Line 195, Page 6).

10. Line 188: "from 437 ± 117 μg m^{-3} before the LCD to 486 ± 144 μg m^{-3} during the LCD". ls the difference statistically significant? given their large spread? please provide statistical significance for such instances throughout the manuscript.

Response: Thanks for your suggestion. We did a statistic analysis ($p < 0.05$), which indicated that the difference was statistically significant. We have added the spread of TDOCs concentrations before and during the LCD in the revised manuscript (Lines 196-202, Page 6). Additionally, we have provided

statistical significance for such instances throughout the manuscript. Please see Table S2 and S3 in the revised Supplement.

11. Comments: Lines 207-209: Howe about the same effect due to long range transport of aged air mass? how that could be ruled out?

Response: C_4 photodegraded into C_3 and C_3 photochemically oxidized into C_2 can occur both via local oxidation and long-range transport. Therefore, we could not rule out about the effect of long range transport of aged air mass. As suggested by the reviewer, we changed it to "indicating the stronger photochemical transformation of organic aerosols from the local oxidation and long-range transport during the LCD" (Line 224, Page 6) in the revised manuscript.

12. Comments: Line 268: SO_2 to SO_4^{2-} conversion could happen due to photochemistry in dry environment as well, why did authors assume that the entire SO_4^{2-} is formed via aqueous chemistry only?

Response: Thanks for your suggestion. The SO_2 to SO_4^{2-} conversion could happen due to photochemistry in dry environment as well. The gas-phase oxidation of SO_2 is dominated by the reaction with OH radical. Compared with the aqueous formation, the reaction rate for gas-phase oxidation is slower and the lifetime of SO_2 from the reaction with OH is longer (one week) (Seinfeld and Pandis, 2006). 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). In the urban atmosphere, sharply increased SO_4^{2-} is often accompanied by a high relative humidity (RH) (Sun et al., 2013; Wang et al., 2014). 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. The correlation analysis between SO_4^{2-} and C_2 was used to evaluate the aqueous formation process of C_2 in previous studies (Ding et al., 2021; Jung et al., 2010; Meng et al., 2020; Shen et al., 2023; Yu et al.,

2005). In this study, C_2 before the LCD was mainly derived from the aqueous phase pathway which was supported by C_2 and its precursors (e.g., Gly and mGly) positively correlated with RH and LWC as discussed in Lines 287-296, Page 8.

Therefore, we have changed it to "The SO_4^{2-} formation was largely from aqueous phase oxidation (Sun et al., 2013; Zhang et al., 2015), thus the correlation analysis between SO_4^{2-} and C_2 can used to evaluate the formation process of C_2 mainly via aqueous phase pathways (Sorathia et al., 2018)." in the revised manuscript (Lines 280-282, Page 8)

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Referee 2

We are thankful to the reviewer 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. The paper entitled "Measurement Report: Enhanced contribution of photooxidation to dicarboxylic acids in urban aerosols during the COVID-19 lockdown in Jinan, East China" by Meng et al. is generally well written and falls within the scope of ACP. However, there are may small issues that overall sum up to the need of major revisions for this manuscript.

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

Major scientific concerns:

2. Analysis of delta^{13}C:

i) If I understand the procedure correctly then the yield during derivatization ranges from 80% to 85%. Have the authors considered the possibility that the derivatization reaction itself may introduce an isotope fractionation and that there could be a Rayleigh fractionation effect. At yield of 80% to 85%, the

isotopic composition of the pooled product is not yet identical to the isotopic composition of the reactant before the start of the reaction. Depending on the presence and magnitude of the fractionation there could be a significant effect. Have the authors investigated the possibility of a fractionation for this reaction (if yes can they report the fractionation constant for the individual reactant-product pairs reported in this study) and factored the Rayleigh effect into their mass balance calculations in section 2.2.2. These crucial details are completely omitted.

ii) Normally, when isotope ratios are measured with an accuracy of 0.2 permil then it is necessary that some type of standard is measured alongside the sample in the same mass spectrometer. Can the authors please elaborate on the standard they used (which is hopefully traceable to VPDB) and provide additional details. Did they spike the sample with a standard?

iii) The GC part of their GC-isotope ratio MS would have some type of injector, and a column temperature program as well as carrier gas flow rates. The authors need to provide these details.

Response: i) Thanks for the reviewer's suggestion. The mentioned question about productive rate and fractionation constant for the individual reactant-product pairs is very critical for our study. 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 derived to dibutoxy acetal. The $\delta^{13}C$ 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 $\delta^{13}C$ values for individual diacids are then calculated from $\delta^{13}C$ of 1-butanol and butyl ester derivative using a mass balance equation. The accuracy of the $\delta^{13}C$ 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; Shen et al., 2023; Wang et al., 2012; Wang and Kawamura, 2006; Xu et al., 2022a; Xu et al., 2022b; Zhang et al., 2016; Zhao et al., 2018).

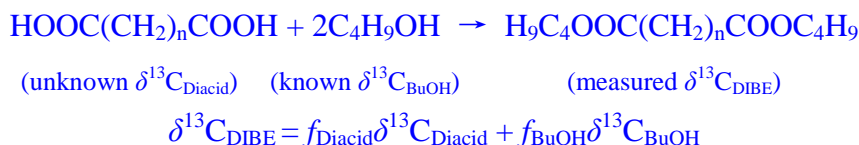
ii) The isotopic analytical precision was ≤ 0.5‰ for a peak height (*m/z* 44) of 0.2 – 8 V. The isotopic composition of diacids and their derivatives is reported in the δ notation relative to the Pee Dee Belemnite (PDB) standard as follows:

$$\delta^{13}C\ (‰) = [(^{13}C/^{12}C)_{sample}/(^{13}C/^{12}C)_{PDB} - 1] \times 10^3$$

After the measurement of isotopic compositions of butyl esters, $\delta^{13}C$ of individual diacids are calculated by the following mass balance equation:

$$HOOC(CH_2)_nCOOH + 2C_4H_9OH \rightarrow H_9C_4OOC(CH_2)_nCOOC_4H_9$$

$$\text{(unknown } \delta^{13}C_{Diacid}) \quad \text{(known } \delta^{13}C_{BuOH}) \quad \text{(measured } \delta^{13}C_{DIBE})$$

$$\delta^{13}C_{DIBE} = f_{Diacid}\delta^{13}C_{Diacid} + f_{BuOH}\delta^{13}C_{BuOH}$$

where $\delta^{13}C_{Diacid}$, $\delta^{13}C_{BuOH}$, and $\delta^{13}C_{DIBE}$ represent the carbon isotopic composition of diacid, 1-butanol, and diacid dibutyl ester, respectively; f_{Diacid} and f_{BuOH} are fractions of carbon in the ester derived from diacid and butanol, respectively. f_{Diacid} and f_{BuOH} of C_2 are 0.2 and 0.8, respectively. The f_{Diacid} and f_{BuOH} values of other organic acids have been described in Kawamura and Watanabe (2004). Before actual sample analysis, we confirmed that $\delta^{13}C$ values of the working standards (a mixture of normal $C_{16} - C_{30}$ alkanes with 0.55 – 2.83 ng μL^{-1}) were equivalent to the theoretical values within an analytical error of <0.2‰.

In addition, 2 µL internal standard (*n*-C_{13} alkane, -27.24 ‰) was spiked to the ester fraction, the esters were injected to a gas chromatograph interfaced to isotope ratio mass spectrometer to determine their stable carbon isotopic composition. We have added it in Text S1 of the revised Supplement.

iii) We have added these details in Text S1 of the revised Supplement as follows:

GC was installed with a HP manual on-column injector and a capillary column (CIP-Sil 8CB, 60 m × 0.32 mm × 0.25 µm) was used with a column oven temperature programmed from 50 to 120 ℃ at a rate of 30 ℃ min^{-1} and then to 300 ℃ at a rate of 6 ℃ min^{-1}. Flow rate of carrier gas (He) was maintained at 1.7 mL min^{-1}.

3. Figure 5: This figure is far too confusing. The authors need to settle on one set of organic compounds/compound ratios and influencing factors for which they want to compare before and after lockdown. They need to keep the arrangement of columns and rows identical both for the panels.

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. Having some unique parameters that are only there in the "before" or the "after" and having parameters that are common but swapped around between the columns and rows makes the visual comparison very difficult for the reader.

Response: Thanks for the review's suggestion. C_2 before the LCD was dominantly derived from the acidic-catalyzed aqueous oxidation with α-dicarbonyls determined by relative humidity (RH) and aerosol liquid water content (ALWC). However, C_2 during the LCD was largely produced from the photochemical oxidation driven by O_3 concentration and solar radiation. Figure 5 was used to investigate the formation mechanism and influencing factors before and during the LCD. Therefore, the arrangement of columns and rows for the panels was different between Fig. 5a and Fig. 5b. As the reviewer's suggestion, we have modified the columns and rows in Fig. 5a and Fig. 5b. To make the visual comparison easy for the reader, we have divided Fig. 5a and Fig. 5b into Fig. 5 and Fig. 6 as follows:

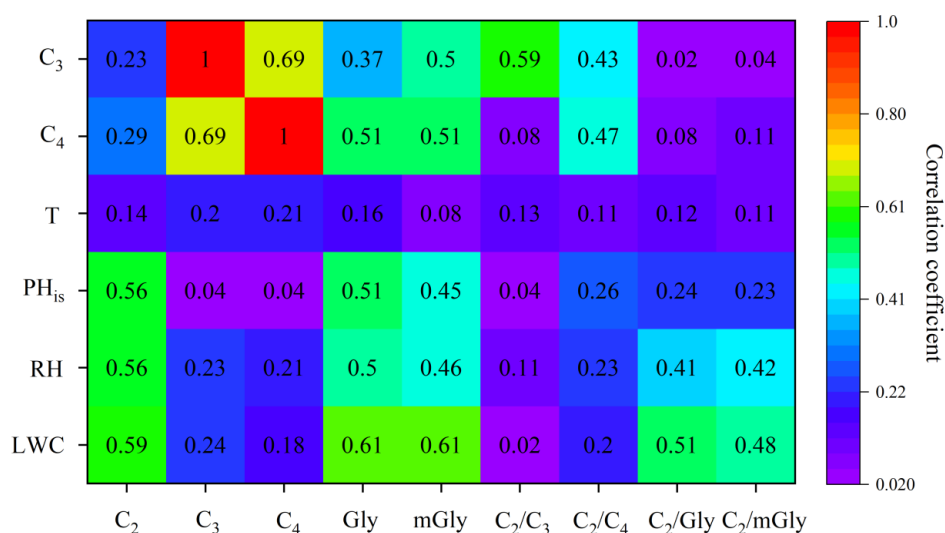



Figure 5. Correlation coefficients (R^2) of concentrations of C_2 and its organic precursors and selected ratios with influencing factors before the LCD.

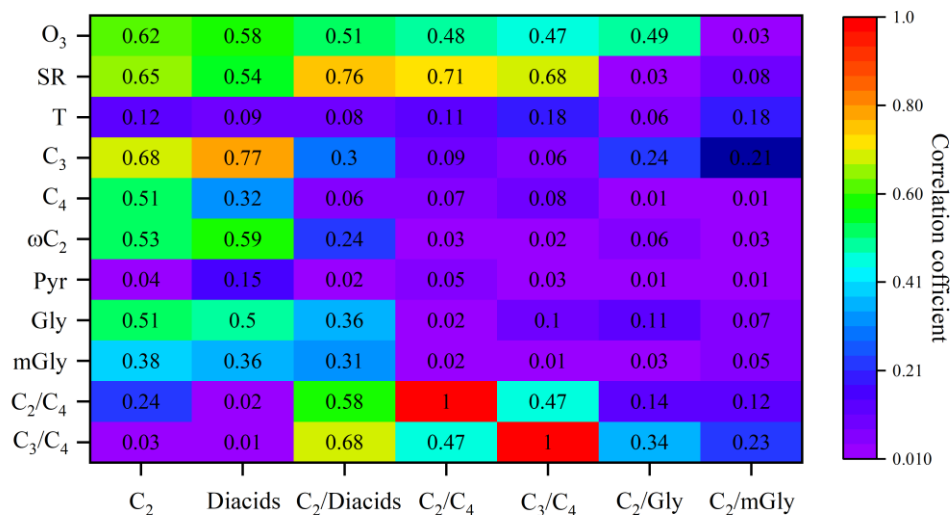



Figure 6. Correlation coefficients (R^2) of concentrations of C_2 and its organic precursors and selected ratios with influencing factors during the LCD.

4. Figure 8 panel f: I am not sure that fitting two different lines for day and night here is warranted. This could be the Simpson paradox in action. All day samples except one outlier fall onto the same line as the night samples just within a narrower range of that line.

Response: The data at night before the LCD presented a *p* value larger than 0.05, which statistically means there was no significant difference (Fig. 9f). Thus, we did not make regression analysis between the $\delta^{13}C$ value of tPh and tPh/diacids ratio at night before the LCD. When we made regression analysis during the day before the LCD, we eliminated one outlier. The modified slope (0.003) and correlation coefficient ($R^2 = 0.45$) of fitting line did not change greatly from those (0.0045, $R^2 = 0.46$) of the original fitting line, indicating this could not be due to the Simpson paradox in action. As the reviewer's suggestion, we have modified this figure in the revised manuscript. Please see Fig. 9f in Page 29 as follows:

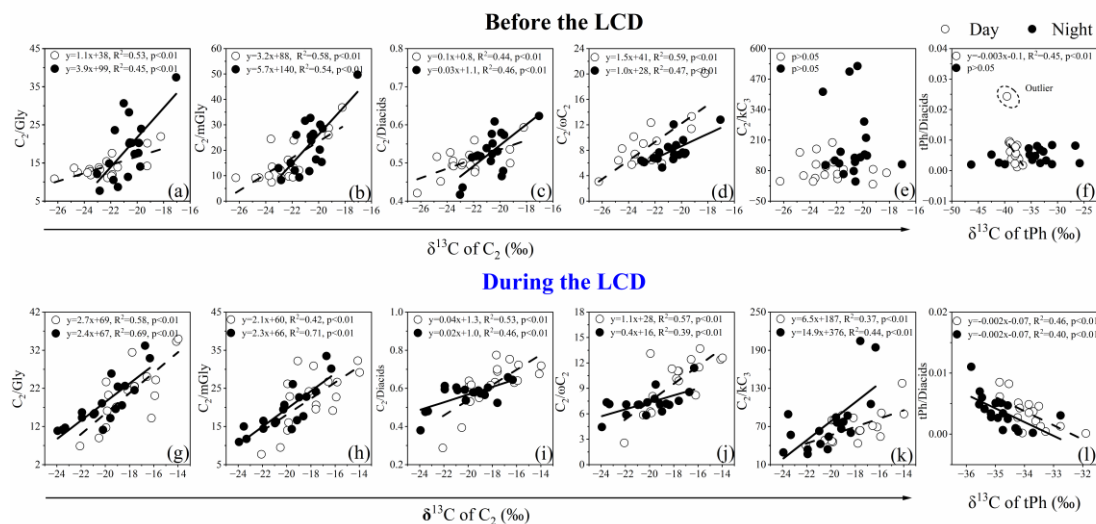



Figure 9. Correlations of the $\delta^{13}C$ of C_2 with the mass ratios of C_2/Gly, C_2/mGly, C_2/Diacids, C_2/ωC_2, and C_2/kC_3, and the $\delta^{13}C$ of tPh with the mass ratio of tPh/Diacids before and during the LCD in January to February 2020.

5. Statistical analysis: i) There are no details on the MLR analysis and random forest machine learning analysis in the methods section. 2) Line 163 states "The results showed that in Jinan city only 18.2% of the enhanced O_3 concentration was resulted from the meteorological variations, and the other 81.8% was ascribed to the reductions of anthropogenic emissions (Fig. S1)". Firstly, the supplement does not appear to contain what it is supposed to according to the main text. Fig. S1 contains only the Back trajectory cluster analysis and the PSCF of the trajectories, Figure S2 pertains to the MLR/Random forest results. Secondly Figure. S2, which does appear to show the results of the poorly described Random forest? or MLR? analysis shows the highest importance for temperature and solar radiation. So, I have several problems with the sentence above and the figure:

i) Figure numbering in the supplement and main manuscript inconsistent. Not only for this case but overall.

ii) Figure S2 directly contradicts the statement in the text. Temperature and solar radiation appear to jointly explain more than 60% of the ozone variability. When one throws in RH and WS > 80% appears to be explained by meteorology and <20% by emission changes. Hence it appears this sentence, the abstract and conclusions need to be revised.

iii) There is only one histogram which shows either the results of the MLR analysis or the results of the random forest analysis or some average in the supplement. I can't imagine both these methods would 100% agree on the relative importance of different factors. Both should be shown separately and contrasted if both techniques were used.

iv) The implementation of the MLR and random forest analysis is not described in the methods section.

v) The points raised above have implications for the abstract and also for the conclusions line 440 "However, the O3 concentration increased by 1.3 times synchronously during the LCD, which was largely launched by the reduction of anthropogenic emissions whose contribution reached up to 81.8%."

Response: Thanks for the reviewer's comments and suggestions. We are sorry to make mistakes in the manuscript. We compared the results between Random Forest and multiple linear regression analysis. However, we found that the results from both methods could not match well with the relative importance of different factors. The O_3 formation was not only influenced by temperature, wind speed, solar radiation, relative humidity, $PM_{2.5}$, CO, NO_2, and SO_2 discussed in our manuscript, but also influenced by volatile organic compounds (VOCs), nitrogen oxides (NO_x) and other key factors. It is a pity that we do not obtain these important influencing factors of O_3 formation during the field campaign. Besides, aerosol chemical models (e.g., WRF-Chem model) are necessary to use to better understand the formation mechanism and influencing factors of enhanced concentration of O_3 during the LCD. Indeed, we think the higher concentration of O_3 is associated with the enhanced contribution of photooxidation to dicarboxylic acids during the LCD. In this study, we focused on the enhanced contribution of photo-oxidation to dicarboxylic acids in urban aerosols during the LCD rather than the formation mechanism of O_3. Thus, we deleted the related text about "The results showed that in Jinan city only 19.8% of the enhanced O_3 concentration was resulted from the reductions of anthropogenic emissions, and the other 80.2% was ascribed to the meteorological variations" because of the theme and limited length of our manuscript.

6. PMF analysis: i) The details of the PMF analysis are completely missing in the method section. Which species were weak and which ones were strong. ii) Why exactly did the authors decide to keep NH_4^+ out of their PMF model? Why did they keep K^+ (a species that should be a decent biomass

burning tracer) out of their model? Same question for why did they keep about half their measured acids out of the model? iii) What was the rational for keeping O_3 and LWC in the model instead of using them as independent tracers to verify factor profiles. iv) How did the authors decide on 5 factors as the right number of factors. The PMF mathematically solves the equation "explain all my data under the assumption that there are X number of sources impacting my site" The choice of X as 4,5,6, …n is the largest assumption that modifies the PMF outcome. Second largest is inclusion and exclusion of species and third the choice of weak and strong species and the percent uncertainty of each individual species. Unequal uncertainties can heavily drive the PMF outcome. So, all these details of PMF implementation need to be provided. Particularity, why 5 why not 4 or 6 factors needs a justification. Also, what about the error analysis for the PMF? Was boot strapping done.

Response: i) We have added detailed methods for PMF model in Text S3 of the revised Supplement. 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".

ii) K^+ is not only from biomass burning, but also from mineral dust and sea salt. Compared with K^+, levoglucosan is regarded as a more reliable tracer for biomass burning because of its single source. Therefore, we chose levoglucosan rather than K^+ as a tracer for biomass burning in this study.

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 acids presented relatively low concentrations and occasionally were below the detection limit, which could enhance the uncertainties. Moreover, these unconsidered organic acids were less indicative for identified sources than the selected major acids. Thus, we kept those organic acids with low concentrations out of the model.

As suggested by the reviewer, we have kept NH_4^+ in the PMF model, and we have re-run the PMF model (Fig. 10).

iii) As discussed in Sections 3.3, SO_4^{2-} and LWC could be considered as significant influencing factor 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 photochemical pathway of the diacids formation. ALWC could not be directly measured and be calculated by ISORROPIA model as described in Section 2.3. To reduce the uncertainties, we have kept LWC out of the model. Thus, we have retained SO_4^{2-} in the model to identify the aqueous pathway. It is worth mentioning that the results re-run by the PMF model (Fig. 10) in the revised manuscript were similar to that in the old manuscript.

iv) 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), and Vossler et al. (2016).

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 S4. 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 S5), indicating no error. The values in the first row of $dQ^{max} = 4$ were zero before and during the LCD (Table S5), 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 S5), suggesting the five-factor solution was more reliably attributed to sources than four-factor solution and six-factor solution.

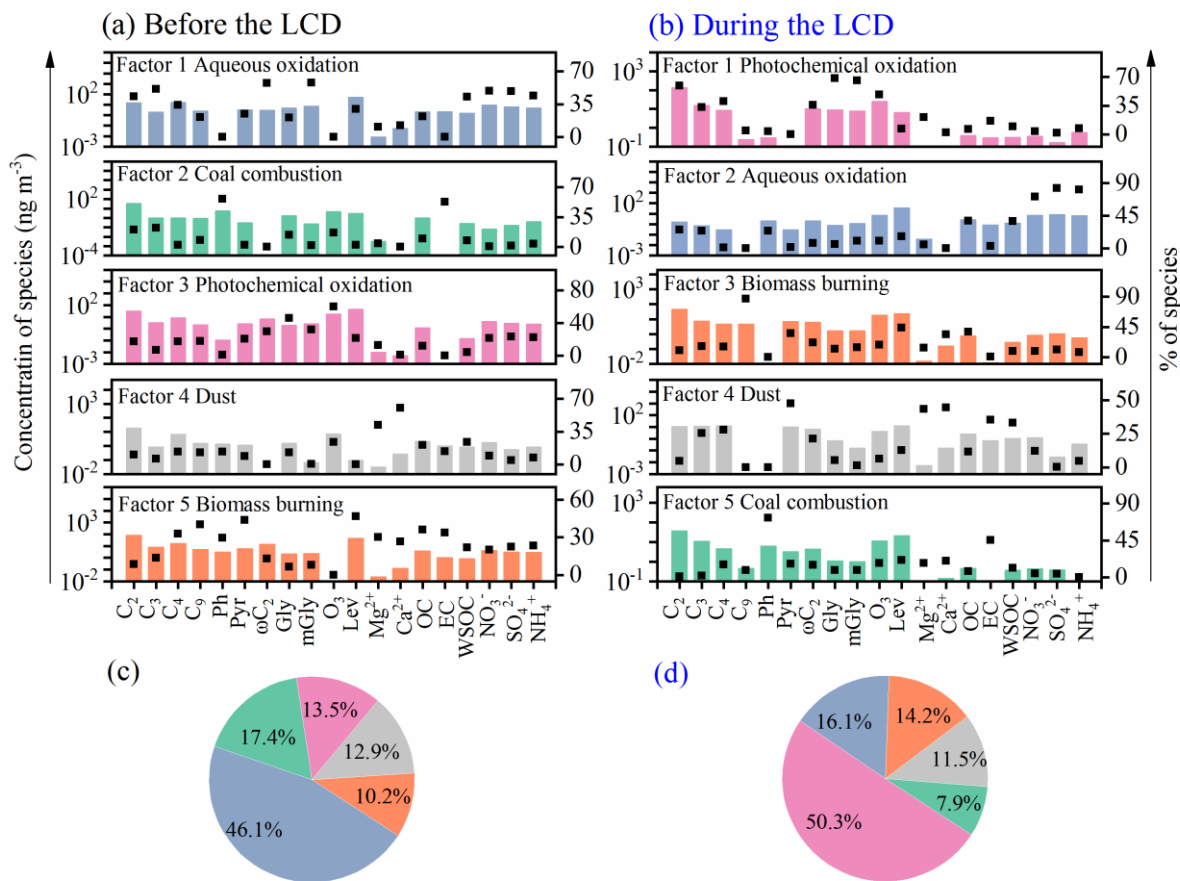



Figure 10. Source profiles of major chemical components in the PM$_{2.5}$ samples from Jinan (a, c) before the LCD and (b, d) during the LCD (BB: biomass burning).

Table S4. 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 S5. 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

7. i) There are a lot of statements that the concentration of some measured species increased/decreased from before the LCD to during the LCD or from day to night. Most of the parameters have substantial ambient variability as can be seen in Figure 3a & b. Maybe the differences will become more visibly clear if the authors chose to show a box and whisker plot instead of a bar graph with the standard deviation. ii) However, even if it looks less as if the concentration before and during the LCD overlaps within the variability, the authors would still need to use an appropriate statistical test to assess whether the differences between the two periods are significant for each of these cases or not. If the variables follow a normal distribution ANOVA may be appropriate. If they do not follow a normal distribution then a more robust test e.g. Kruskal-Wallis test may be required.

Examples of statements without significance level of the difference include:

(1) Line 187: "upward trend from 437 ± 117 µg m^{-3} before the LCD to 486 ± 144 µg m^{-3} during the LCD."

(2) Line 188 "concentrations of diacids and oxoacids during the LCD increased by 1.1 and 2.1 times"

(3) Line 191 "The daytime concentration of diacids before the LCD was 17% lower than that at night"

(4) Line 193 "C2 concentration increased from 181 ± 47.5 m^{-3} before the LCD to 239 ± 108 μg m^{-3} during the LCD"

(5) Line 197 "Both ratios of C_2/diacids and C_2/TDOCs during the LCD were higher than those before the LCD"

(6) Line 201 "daytime concentration of C_2 and the ratios of C_2/TDOCs and C_2/diacids were lower than those at night before the LCD but an opposite trends were found during the LCD"

(7) Line 209 "Both C_2/C_4 (8.4 ± 3.4) and C_3/C_4 (1.6 ± 0.4) ratios during the LCD were higher than those (3.9 ± 1.5, 0.3 ± 0.1) before the LCD (Fig. 3b),"

(8) Line 214 "It is noteworthy that C_9 concentration (12.0 ± 4.0) before the LCD was 2.0 times higher than that (5.9 ± 4.8) during the LCD (Table 2)," apart from significance test the plusminus is missing in the first bracket.

(9) Line 220 "its concentration (8.8 ± 11.0 μg m^{-3}) and relative abundance (0.03 ± 0.01) during the LCD were lower than those (11.0 ± 6.1 μg m^{-3}, 0.02 ± 0.01) before the LCD"

(10) Line 257 "Both ratios of C_2/levoglucosan (1.7 ± 0.6) and C_2/K^+ (0.2 ± 0.02) at night before the LCD exhibited larger values than those (1.3 ± 0.5, 0.16 ± 0.02) in the day, which was mainly ascribed to the accelerated aqueous formation of C2 at night. "

Response: i) Suggestion taken. We have changed Fig. 3 to a box and whisker plot as follows:

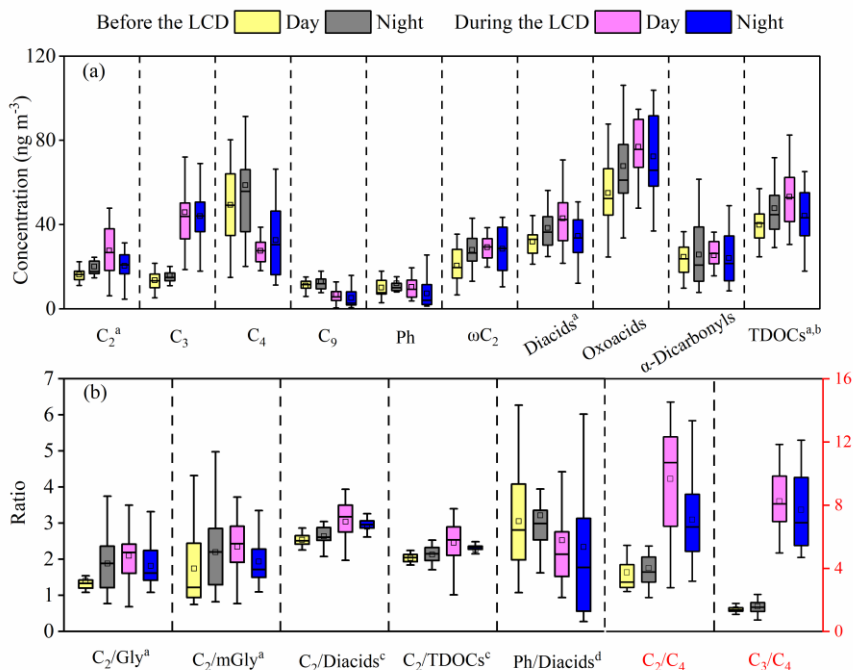



Figure 3. Diurnal changes of (a) major organic compounds and (b) selected mass ratios before and during the LCD ([a] the concentrations reduced by 10 times; [b] TDOCs: total detected organic components; [c] the mass ratios enlarged by 5 times; [d] the mass ratios enlarged by 100 times).

ii) We thank the reviewer for the helpful comments above. In the revised manuscript, we have performed a statistic test (e.g., ANOVA and Kruskal-Wallis test) to verify if the concentrations of target compounds and major ratios were of significant difference. As shown in Table S2, the concentrations of organic species (except for α-dicarbonyls) and major ratios in $PM_{2.5}$ before and during the LCD presented a p value less than 0.05, indicating that the abundances and compositions of the major species before and during the LCD were statistically different. Besides, statistic test was also carried out for the day and night samples before and during the LCD, and the results were illustrated in Table S3.

Minor scientific comments:

8. Line 102: it appears that the online air quality monitoring data for $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2, is from an air quality monitoring station associated with some network. Since the referenced web link is in Chinese I could not follow it. Could you please briefly elaborate in this paper on.

i) the location of the monitoring station used and how it relates to your PM off line sampling location (collocated with your PM sampling equipment or somewhere nearby?).

ii) Inlet height of the air quality monitoring station.

iii) Measurement equipment used to measure those parameters and Q/A Q/C protocol followed by the primary data generator.

Response: i) The monitoring station is located in the Jinan Environment Monitoring Center, which is one of the State Controlling Air Sampling Sites in Jinan. The $PM_{2.5}$ sampling was conducted on the rooftop of a six-story building (36.67 °N, 117.06 °E) that is about 40 meter away from the Jinan Environment Monitoring Center. We have added this information in the revised manuscript (Lines 92-94, Page 3).

ii) The inlet height of the air quality monitoring station was approximately 20 m above the ground level (Lines 107-108, Page 3).

iii) Those measured parameters were obtained from one of the State Controlling Air Sampling Sites in Jinan. The quality assurance/quality control (QA/QC) was operated strictly by professionals followed by "Technical Specification for Automatic Monitoring of Ambient Air Quality", "Technical Standard for Ambient Air Quality Monitoring", and "Technical Instruction" formulated by the Ministry of Ecology and Environment of the People's Republic of China. Thus, the liability and validity of those data could be ensured. We have added it in Text S4 of the revised Supplement (Lines 106-107, Page 3).

9. Table 1: Hope the authors have not taken the simple average and standard deviation of the pH. The high standard deviation suggests they may have. The pH scale is logarithmic and not a ratio variable. Values need to be converted back to the $[H^+]$ before averaging for the result to be mathematically correct.

Response: Suggestion taken. $[H^+]$ have been converted back before averaging the values of pH. We have changed pH_{is} values to "3.2 ± 3.0 before the LCD and 3.5 ± 3.1 during the LCD" in Table 1 and "However, pH_{is} remained similar before (3.2 ± 3.0) and during the LCD (3.5 ± 3.1) with no significant statistical difference ($p > 0.05$, Table S2)" in the revised manuscript (Lines 184-185, Page 5).

10. Line 230 "> 0.5, Fig. S2)." Wrong figure number.

Response: Corrected.

11. Line 243 Please look into the following sentence "However, the increase in the ratios of C_2/diacids and C_2/TDOCs at night indicates that the lowered nighttime PBL height was not the case, which could be supported by the insignificant diurnal differences of primary pollutant markers such as Na^+, Ca^{2+}, and Mg^{2+} (t test, $p > 0.01$) between the daytime and nighttime." You can't state that the PBL height was not more shallow than daytime PBL based on this evidence. You can, however, make a case that the change in PBL height was not the only cause of the change in the observed concentration. Also, before you get into these details a statistical test is required to establish that the night time concentration is significantly higher than the daytime concentration. Since the text may get messy with too many numbers, you may have to put the appropriate tables in the supplement.

Response: We are sorry for this misunderstanding. We have rephrased it to "However, the increase in the ratios of C_2/diacids and C_2/TDOCs at night indicates that the effect of lowered nighttime PBL height was minor, which could be supported by the insignificant diurnal differences of primary pollutant markers such as Na^+, Ca^{2+}, and Mg^{2+} ($p > 0.05$, Table S3) between the daytime and nighttime." (Lines 255-258, Page 7).

In addition, as suggested by the reviewer, we have made a statistical test (Table S2 and Table S3).

12. Line 269 (Fig. S2) wrong figure number.

Response: Corrected.

13. Line 293 "Therefore, pH_{is} exhibited pronounced negative relationships with C_2 and its precursors such as Gly and mGly ($R^2 \geqslant 0.45$, Fig. 3a)" which figure is this referring to. Figure 2 does have a correlation analysis but not with pH_{is} included. Fig. 3a has a histogram of concentrations.

Response: Sorry for the mistake. The correlations of pH_{is} with C_2 and its precursors such as Gly and mGly were shown in Fig. 5. We have changed it (Line 307, Page 8).

Minor language related comments:

14. Line 30: "photochemical oxidation lunched by the higher O_3 concentration" please revise lunched is not the right word. It's an ancient past tense of the verb to eat. "triggered by higher O_3 mixing ratios" or "promoted by higher O_3 mixing ratios" may be a better wording.

Response: Suggestion taken. We have changed "lunched" to "promoted" (Line 30, Page 1).

15. Line 109: "The water extracts were concentrated to near dryness and then reacted with 14% BF3/n-butanol at 100°C for 1 hour to convert butyl esters or dibutoxy acetals" This sentence is not clear and grammatically incomplete. Please clearly state which compound(s) you are converted to which compound(s).

Response: Suggestion taken. The water extracts were concentrated to near dryness and then reacted with 14% BF3/n-butanol at 100 ℃ for 1 hour. During this process, the carboxyl functional group was derivatized to butyl ester, and the aldehyde and keto groups were derived to dibutoxyacetals.

We have added it in the revised manuscript. Please see Lines 114-115 in Page 3.

16. Line 128: "Levoglucosan in the field blank samples is 4% less than the ambient samples." Please check the language here. Do you really mean to say that your field blank is so high that the levoglucosan in the blank is only 4% less than the levoglucosan in your ambient samples. If that is what you meant to say then consider this comment under major scientific concerns.

Response: Thanks for the reviewer's suggestion. We meant that the concentration of levoglucosan in the field blank samples was so low that the levoglucosan concentration in the field blank samples was lower than 4% compared with that in the ambient samples. Therefore, we have changed it to "Compared with the ambient samples, the concentration of levoglucosan in the field blank samples was lower than 4%."

Please see Lines 128-129 in Page 4.

17. Line 159: "A rent study" the authors probably mean to say "A recent study".

Response: Corrected.

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