Title: Simultaneous observations of peroxyacetyl nitrate and ozone in central China during static management of COVID-19: Regional transport and thermal decomposition.

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Manuscript number: egusphere-2024-575

Dear Editor

Thank you for encouraging us to resubmit the above-mentioned manuscript. We also thank the reviewers for reading our manuscripts and providing valuable comments and suggestions in enhancing the quality of our paper. We believe that all reviewer's comments have been addressed and the itemized replies to each comment are as follows.

To incorporate reviewers' comments into the revised manuscript, we will certainly overhaul our manuscript and provide more in-depth analysis of the data. In addition, we will seek a professional expert to edit the text for clarity and for better comprehension. Changes made in response to these responses are marked in yellow in the highlighted copy of the revised version. Our own minor changes are marked in red.

Below are the point-by-point responses to the comments for each reviewer.

Reviewer #1:

We do appreciate your constructive and useful comments. To better reply to your general comments in your long paragraph, we have divided your comments into serval parts with superscript $^{a, b, c}$, etc., and correspondingly addressed your comments in a separate paragraph $^{a, b}$, etc. More detailed replies for the same topic are shown in your specific comments.

Detailed comments:

The article explored the relationship between VOCs and $PM_{2.5}$ with abundant VOCs species observed in Zhengzhou during the COVID-19 and made recommendations for the control of VOCs source emissions. ^{a}The current discussion may not be sufficiently supportive, please add more details to each section to make the entire article more logical.

^{b}Basic details regarding instrumentation and data collection are missing. The authors need to supplement materials related to the reliability of the PMF results.

^{c}Further more, more work is needed to elucidate the relationship between VOCs and haze pollution, as well as the influencing factors. And it is suggested that model simulation on SOA formation potential be added to the manuscript.

While the theme and results of the study are interesting, I have provided a few suggestions for improvement.

Response: We are very grateful for the positive comments and suggestions. We have separately replied your suggestions into three parts as following:

^{a}The current discussion may not be sufficiently supportive, please add more details to each section to make the entire article more logical.

We will overhaul every section of the revised version. In each chapter we will add more discussion to make the entire article more logical and comprehensive. Details can be found in the following point-to-point response.

[b]Basic details regarding instrumentation and data collection are missing. The authors need to supplement materials related to the reliability of the PMF results.

Additional details about the instruments and data collection are provided below.

For instrumentation comments, please see our replies in the following specific comments.

Reliability of PMF results will be added to the text with relevant figures and tables in the supplementary materials.

We used displacement of factor elements (DISP) to assess PMF modelling uncertainty (for a description, see Paatero et al. (2014)). Q was less than 1% and no swaps occurred for the small est dQmax in DISP. Fpeak values from -2 to 2 were tested to explore the rotational stability of the solutions. Q_{true}/Q_{exp} is lowest when Fpeak = 0, so we chose the PMF results for that case.

After examining 3-8 factors, 20 base runs with 5 factors eventually selected to represent the final result. We provide an explanation of factor selection in the supplementary materials. Figure 3(a) includes Q_{true}/Q_{exp}, Q_{robust}/Q_{exp} for factors 3-8. The slopes of these two ratios in changed at five factors, and we found that five factors were more realistic after repeated comparisons of the results at four, five and six factors. These five factors eventually selected as potential sources for the observed VOCs are: (1) Fuel evaporation; (2) Solvent usage; (3) Vehicular emission; (4) Industrial source; and (5) Combustion.

References:

Paatero, P., Eberly, S., Brown, S. G., Norris, G. A.: Methods for estimating uncertainty in factor analytic solutions, Atmospheric Measurement Techniques, Volume 7, 781-797, https:// 10.5194/amt-7-781-2014, 2014.

[c]Further more, more work is needed to elucidate the relationship between VOCs and haze pollution, as well as the influencing factors. And it is

suggested that model simulation on SOA formation potential be added to the manuscript.

It is well known that VOCs are precursors for ozone formation and generation of secondary organic aerosols (SOAs). It is incorrect to state that O_3 pollution is a true haze event. However, O_3 can assist the formation of fine particulates; there are numerous studies about the so-called double pollution of O_3 and $PM_{2.5}$. O_3 as an oxidant can improve the oxidation capacity and promote the oxidation of SO_2 and NO_2 (Li et al., 2023). On the other hand, the suppression of O_3 formation due to the presence of $PM_{2.5}$ has recently been highlighted for further O_3 pollution controls in regions that suffer high ozone concentrations (Zhang et al., 2024). Furthermore, $PM_{2.5}$ decreased the surface photolysis rates J_{NO2} and J_{O1D}, resulting in a decrease in O_3 concentration in the VOC-sensitive area and a slight increase in the NO_x-sensitive area (Qu et al., 2023.). SOAs themselves are of course part of organic aerosols in $PM_{2.5}$ haze conditions.

The factors affecting VOC-haze interactions are typically atmospheric photochemistry and the mixing ratio of NOx and type of VOCs in generating SOAs. However, most VOC species posed no non-carcinogenic risk during haze events (Zhang et al., 2021).

Additionally, we have included quantitative analysis for SOA as well. In particular, Figure 1 shows the SOAP concentrations and contribution rates of the top ten species throughout the entire process, during two pollution processes, and clean days. The top ten species all reached close to 100% of the total SOAP contribution, with Case 1 reaching 98%. The composition of the top ten species is basically the same for each process. Toluene, m/p-xylene, and benzene were consistently the top three species. Toluene, the highest contributing species, reached a SOAP value of 49.4 μg/m^3 in the most polluted Case 2, which was 3.2 times higher than the SOAP sum of all species on the clean day (15.5 μg/m^3). The SOAP value for Case 1, which is also a contaminated process, was 67 μg/m^3, and the main species including toluene (34.6 μg/m^3) were lower than those for Case 2 (m/xylene: 9.8 μg/m^3, benzene: 8.5 μg/m^3) (m/xylene: 19.4 μg/m^3, benzene: 13.4 μg/m^3).

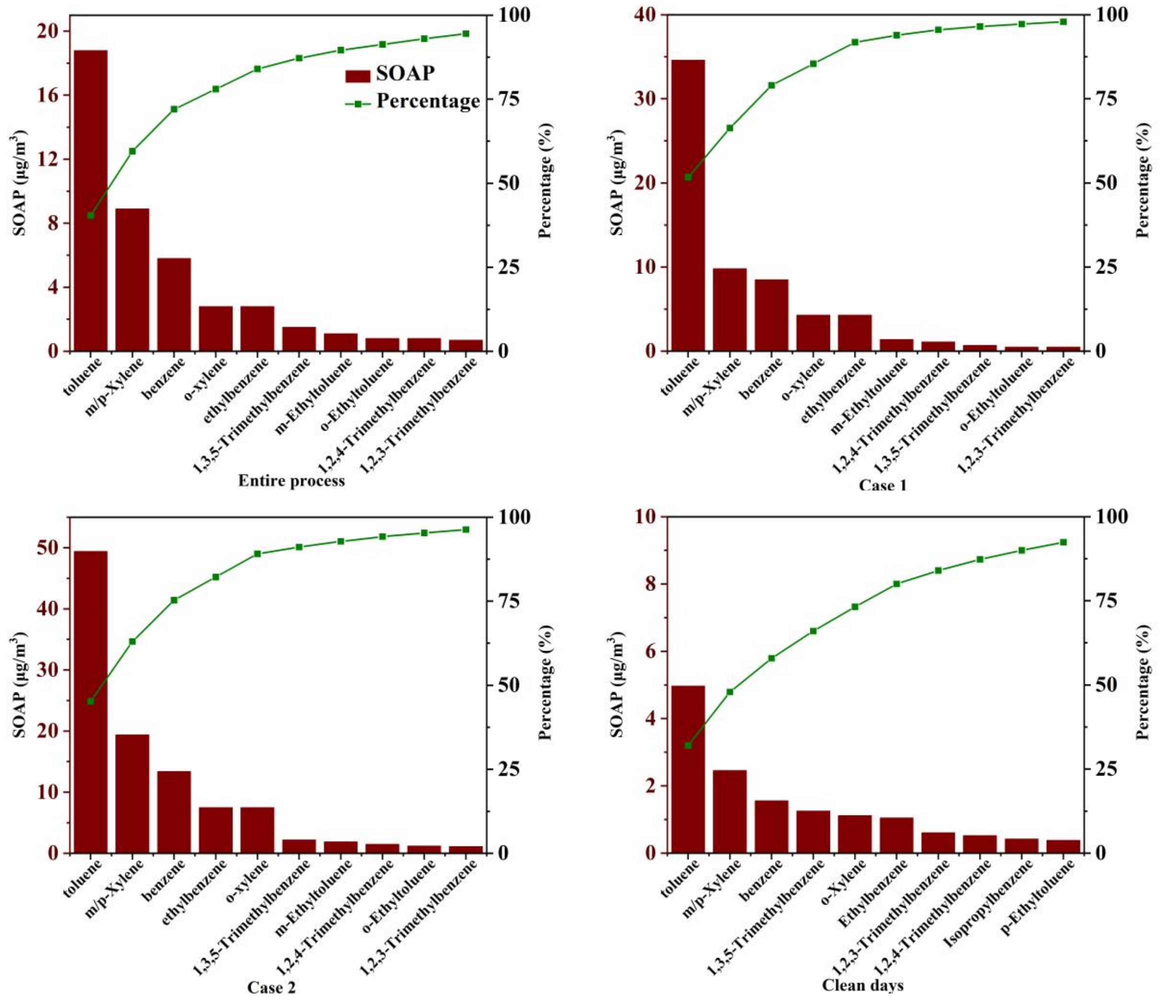


Figure 1. SOAP dominant species in different processes

The following is point-by-point responses to all your comments and valuable suggestions.

References:

Qu, Y.: The underlying mechanisms of $PM_{2.5}$ and O_3 synergistic pollution in East China: Photochemical and heterogeneous interactions, Science of The Total Environment, Volume 873, https://doi.org/10.1016/j.scitotenv.2023.162434, 2023.

Li, Y.: Spatiotemporal Variations of $PM_{2.5}$ and O_3 Relationship during 2014–2021 in Eastern China, Aerosol and Air Quality Research, https://doi.org/10.4209/aaqr.230060, 2023.

Zhang, D.: Characteristics, sources and health risks assessment of VOCs in Zhengzhou, China during haze pollution season, Journal of Environmental Sciences, Volume 108, https://doi.org/10.1016/j.jes.2021.01.035, 2021.

Zhang, J.: Enhanced summertime $PM_{2.5}$-suppression of O_3 formation over the Eastern U.S. following the O_3-sensitivity variations, Environmental Science: Atmospheres, 2024.

1. Line 124-135: The authors lack more detailed descriptions of the instrumentation. What are the working procedures of the instruments? What is the time resolution of the samples? How long were the samples collected for? Where were they captured? It is recommended to include information about instrument quality control methods.

Response: As per your comments, we have added a description of instrumental details including time resolution to the Materials and Methods section:

The VOCs were measured hourly using a GC-FID/MS (*TH-PKU 300 b*, Wuhan Tianhong Instruments Co., China). The instrument TH-PKU300b includes electronic refrigeration ultra-low temperature pre-concentration sampling system, analysis system and system control software. The ambient VOCs in the first 5 minutes of each hour were collected by the sampling system and then entered the concentration system. Under low temperature conditions, the VOCs samples collected were frozen in the capillary capture column, and then quickly heated and resolved, so that the compounds entered the analysis system. After separation by chromatographic column, the compounds were monitored by FID and MS detectors. During the detection process, the atmospheric samples collected undergo analysis through two distinct pathways. C2-C5 hydrocarbons are analyzed using FID, while C5-C12 hydrocarbons, halocarbons, and OVOCs are analyzed with a MS detector. After excluding species with missing data exceeding 10%, the detected volatile organic compounds include 29 alkanes, 11 alkenes, 17 aromatics, 35halocarbons, 12 OVOCs, 1 alkyne (acetylene), and 1 sulfide (CS_2) with a total of 106 compounds.

As for information on instrument quality control methods, the revised text shall be:

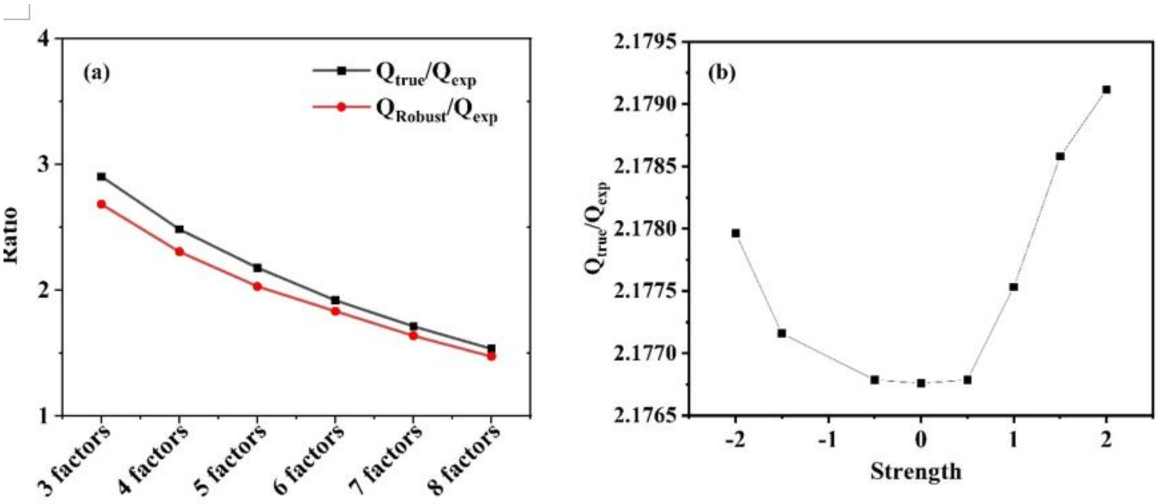
The instrument was calibrated per week to ensure the accuracy of VOCs by injecting standard gases with a five-point calibration curve. The detection limit of C2-C5 hydrocarbons ranges from 0.007 to 0.099 ppbv, other hydrocarbons are 0.004–0.045 ppbv, halogenated hydrocarbons 0.009-0.099 ppbv, OVOCs and other compounds of 0.006–0.095 ppbv. Thirty-two of the monitored VOCs had more than 90% of their data greater than the detection limit, and 34 had more than 50% of their data greater than the detection limit.

2. Section 2.2 Positive Matrix Factorization (PMF) model

How did the authors conduct factor selection, and why did not choose the 5-factor solution, 6-factor solution, and 7-factor solution? The authors need to provide more explanations and justifications in the manuscript.

Response: After examining 3-6 factors, 20 base runs with 5 factors eventually selected to represent the final result. We provide an explanation of factor selection in the Supplementary Materials. Figure 2(a) includes Q_{true}/Q_{exp}, Q_{robust}/Q_{exp} for factors 3-8. The slopes of these two ratios in changed at five factors, and we found that five factors were more realistic after repeated comparisons of the results at four, five and six factors. These five factors eventually selected as potential sources for the observed VOCs are: (1) Fuel evaporation; (2) Solvent usage; (3) Vehicular emission; (4) Industrial source; and (5) Combustion. Five factors have been commonly reported before, e.g., in Shijiazhuang, northern China (Guan et al, 2023) and in Beijing (Cui et al., 2022). Figure 2(b) shows the result of Fpeak model run; Q_{true}/Q_{exp} is lowest when Fpeak = 0, so we chose the PMF results for that case.

The above statement will be incorporated into the revised text.



References:

Cui, L., Wu, D., Wang, S., Xu, Q., Hu, R., and Hao, J.: Measurement report: Ambient volatile organic compound (VOC) pollution in urban Beijing: characteristics, sources, and implications for pollution control, Atmospheric Chemistry and Physics, 22, 11931-11944, https://doi.org/10.5194/acp-22-11931-2022, 2022.

Guan, Y., Liu, X., Zheng, Z., Dai, Y., Du, G., Han, J., Hou, L. a., and Duan, E.: Summer O3 pollution cycle characteristics and VOCs sources in a central city of Beijing-Tianjin-Hebei area, China, Environmental Pollution, 323, 121293, https://doi.org/10.1016/j.envpol.2023.121293, 2023.

Figure 2. (a) The $Q_{true}/Q_{expected}$ ratios in different solutions; (b) the $Q_{true}/Q_{expected}$ ratio for different Fpeak value solutions.

3. Section 3.1 Pollution characteristics

Line 194: Ensure that the font in the figures is consistently in Times New Roman. The y-axis labels do not match the legend (NO and NO_x).

Response: We have revised the manuscript according to your comments.

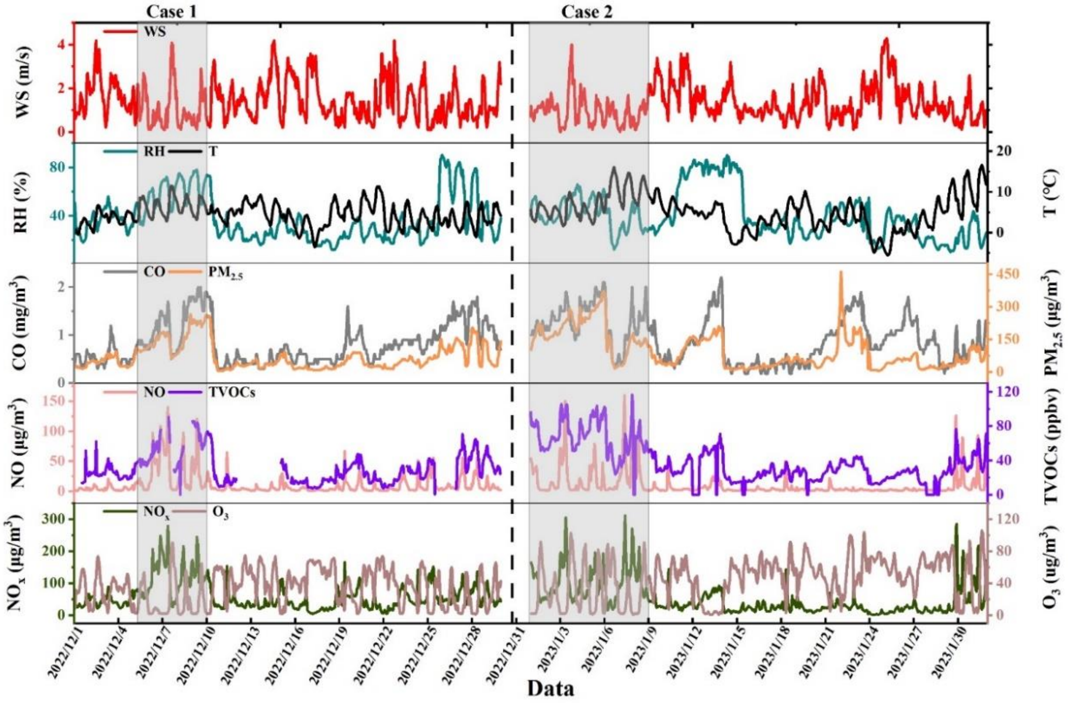


Figure 3. Time series of WS, WD, T, RH, CO, $PM_{2.5}$, NO, TVOCs, NO_x and O_3 during the observation period.

4. What does the shading in Figure 1 represent? What are Case 1, Case 2, Case 3, Case 4, and Case 5? Clear explanations need to be provided. If these cases represent haze pollution processes, how do you define your pollution processes? Please include the references you consulted.

Response: The shadow section in Figure 3 represents two haze pollution events during the monitoring period. A pollution event is determined when the daily average concentration of $PM_{2.5}$ exceeds 75 μg/m^3 (China's II-level standard) for at least three consecutive days. We apologize for the unclear statement and recognize that the original annotations might confuse readers, so we simplify the labeling in Figure 1. To avoid misinterpretation, we deleted processes with no more than 3 days of continuous contamination in Figure 3. In the revised version, we focus on the distinct characteristics of Case 1, Case 2, and Clean days as depicted in the figure. Case 1 (December 5 to December 10 with daily average $PM_{2.5}$ = 142.5 μg/m^3) and Case 2 (January 1 to January 8 with daily average $PM_{2.5}$ = 181.5 μg/m^3) were selected as they represent the pollution events in infection and recovery periods, respectively, due to their long duration and high pollution levels. We divided this period into an infection period (1-30 December 2022) and a recovery period (1 January 2023-31 January 2023) based on Chinese Center for Disease Control and Prevention's December 2022-January 2023 infection data statistics (Figure 4). Any days with a $PM_{2.5}$ concentration lower than 35 μg/m^3 (China's I-level standard) is considered as Clean days.

The above definition of pollution process will be incorporated into the revised manuscript.

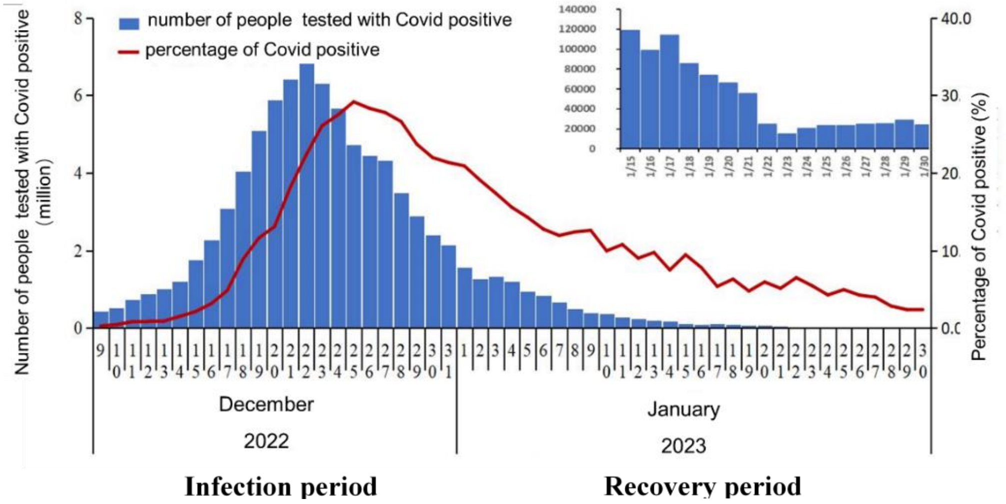


Figure 4. Trend of Omicron infection in China from 9 Dec. 2022 to 1 Jan. 2023 (CCDCP, 2023)

5. Line 217-225: Why did you only discuss Case 1 and Case 3? Are these two periods particularly significant? Provide your reasoning.

<u>Response:</u> In this study, a continuous online observation of VOCs was carried out, which covered the abolishment of lockdown measures in Zhengzhou. A two-month-long lockdown measure was applied after first Omicron case of student in Zhengzhou University was confirmed on October 8, 2022. Lockdown measure was abolished from the beginning of December in 2022, which resulted in a sharp increase of Omicron-infected people and a decrease in daily social production activities. In fact, the “Nucleic Acid Screening

Measures for all staff’ policy was also canceled on December 8 in 2022. People are basically homebound after the lifting of the lockdown policy due to either infection or fear of infection of Omicron variant. Due to herd immunization, people resumed normal life and industry normal activity. Therefore, the characteristics and variations of VOCs during different periods were investigated to assess their impact on pollution in general and on the formation of SOA in particular and to provide data support for future pollution control policies in Zhengzhou.

During the pollution events that occurred in the observation phase, Case 1 (December 5 to December 10) and Case 2 (January 1 to January 8) were considered to be in the early stages of infection and recovery periods, respectively. These two cases have long durations and high pollution levels, making them representative pollution processes for the infection and recovery periods. To avoid misinterpretation, we deleted processes with no more than 3 days of continuous contamination in Figure 1. Essentially, Case 3 in the original paper now is Case 2.

6. In Figure 2, the font should be changed to Times New Roman.

Response: We have modified in the revised manuscript according to your suggestions for the consistent font.

7. In this section, you only analyzed the variations in pollutant concentrations and meteorological conditions. What is the relationship between them? Which factors are crucial causes of pollution? You have not provided analysis and explanations.

Response: The pollutant emission from different sources is the main cause of pollution. Indeed, meteorological conditions play an important role in the extent of pollution. But we know that the changes in emissions from pollution sources over a period of time are usually small, and meteorological conditions play a very important role in the formation of pollution. And previous studies have also shown that low wind speed, high relative humidity, and low precipitation are meteorological factors that contribute to the worsening of particulate matter pollution in Zhengzhou during winter (Duan et al., 2019).

The meteorological conditions in the two periods are generally similar, and the Case 2 in the recovery periods are slightly more prone to atmospheric stability, high relative humidity and other meteorological conditions that are not conducive to the dispersion of pollutants than Case 1 in the infection periods. However, this slight meteorological difference cannot directly lead to a significant change in the degree of pollution we have observed. Clearly, the extent of pollution in different periods is mainly due to anthropogenic activities and to a lesser extent, regional transport (see the following reply), and not meteorological conditions. The reason for providing meteorological data is to add supplementary information for these events.

Based on your comments, we have studied the relationship between meteorological conditions and the concentration of different pollutants. We found a significant correlation between relative humidity and the following three pollutants (Figure 5). It shows that changes in relative humidity have an important effect on pollution formation.

We will supplement this part according to your comments as: We analyzed the relationship between meteorological parameters and pollutant concentrations and found correlations between $PM_{2.5}$, TVOCs and NO_x and RH, suggesting that meteorological conditions have an important influence on pollution formation.

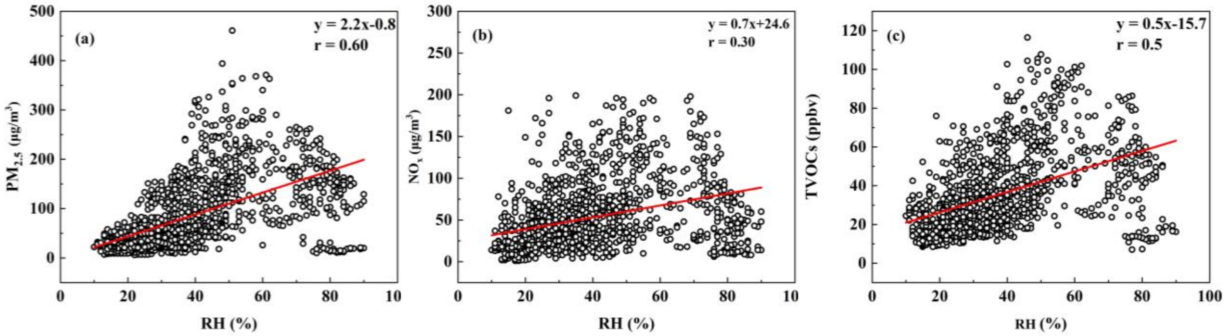


Figure 5. Relative humidity and (a) $PM_{2.5}$, (b) NO_x, (c) TVOCs correlation

References:

Duan, S., Jiang, N., Yang, L., Zhang, R.: Transport Pathways and Potential Sources of $PM_{2.5}$ During the Winter in Zhengzhou, Environmental Science, Jan 8;40(1):86-93, https://doi.org/10.13227/j.hjkx.201805187, 2019.

8. Line 222-223: "[…] Among them, Case 1 (from December 5 to December 10 and […]" A closing bracket is missed.

Response**:** We have revised it in the manuscript.

9. Section 3.2 Source appointment

Line 272: 'indicating that the measured air VOC content was influenced by both remote sources and urban area emissions.', Are you referring to all VOCs? Or specifically to m/p-xylene and ethylbenzene?

Response**:** We apologize for the impact on your understanding due to our negligence. We are referring to m/p-xylene and ethylbenzene here.

10. Line 271-273: Your conclusion indicates that VOCs are influenced by transport and emissions from distant regions. Can this be further substantiated through transport or other means?

Response**:** We infer the photochemical age of the air mass by the ratio of X/E. When the ratio is significantly lower than 3, it indicates that VOC mainly migrates from long-distance sources (aged air masses) (Kumar et al., 2018; Cerón Bretón et al., 2020). The average X/E value in this study was 2.0, indicating that the measured air VOCs content was affected by transport of nearby or long-distance source emissions.

To further confirm that VOCs are affected by long-range transport, we conducted a potential source analysis of VOCs.

The area covered by the airflow trajectory was gridded, and the 80th percentile values of TVOCs for each process were set as standard values to obtain a map of the potential source distribution of TVOCs. Areas with high PSCF values indicate potential source areas of VOCs pollution (Figure 6).

Figure 6 (a) shows the potential source analysis of VOCs during the infection period. The areas with the highest PSCF values (> 0.36, red) are found in Jincheng and Xi'an, northwest of Zhengzhou, and the areas with high PSCF values (> 0.28, orange) include Luoyang, Jiyuan, and north of Xuchang, which are all industrial-intensive cities. Figure 5 (b) shows the results of the recovery period, with a wider distribution of potential sources than the former, and a greater variation in the areas of high PSCF values. Compared with the previous month, Handan and Liaocheng areas become new high PSCF areas, the influence of Xi'an is weakened, and the yellow area (PSCF > 0.2) is shifted from the northwest to the northeast of Zhengzhou.

The above analysis can also show that the VOCs at the observation sites are mainly influenced by the transmission from the distant areas.

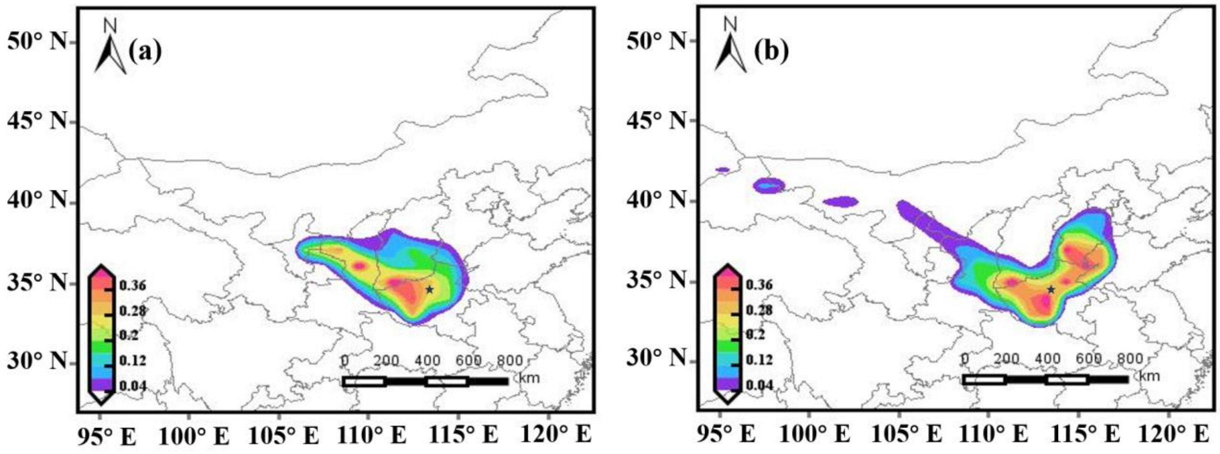


Figure 6. Potential source areas for VOCs (a) Infection period (b) Recovery period (Black pentagrams represent sampling locations)

Reference:

Cerón Bretón, J. G.: Health Risk Assessment of the Levels of BTEX in Ambient Air of One Urban Site Located in Leon, Guanajuato, Mexico

during Two Climatic Seasons, Atmosphere, 11, 165, https://doi.org/10.3390/atmos11020165, 2020.

Kumar, A., Singh, D., Kumar, K., Singh, B. B., and Jain, V. K.: Distribution of VOCs in urban and rural atmospheres of subtropical India: Temporal variation, source attribution, ratios, OFP and risk assessment, Science of the Total Environment, 613-614, 492-501, https://doi.org/10.1016/j.scitotenv.2017.09.096, 2018.

11. Line 306: 'olefins' should be corrected to 'alkenes'.

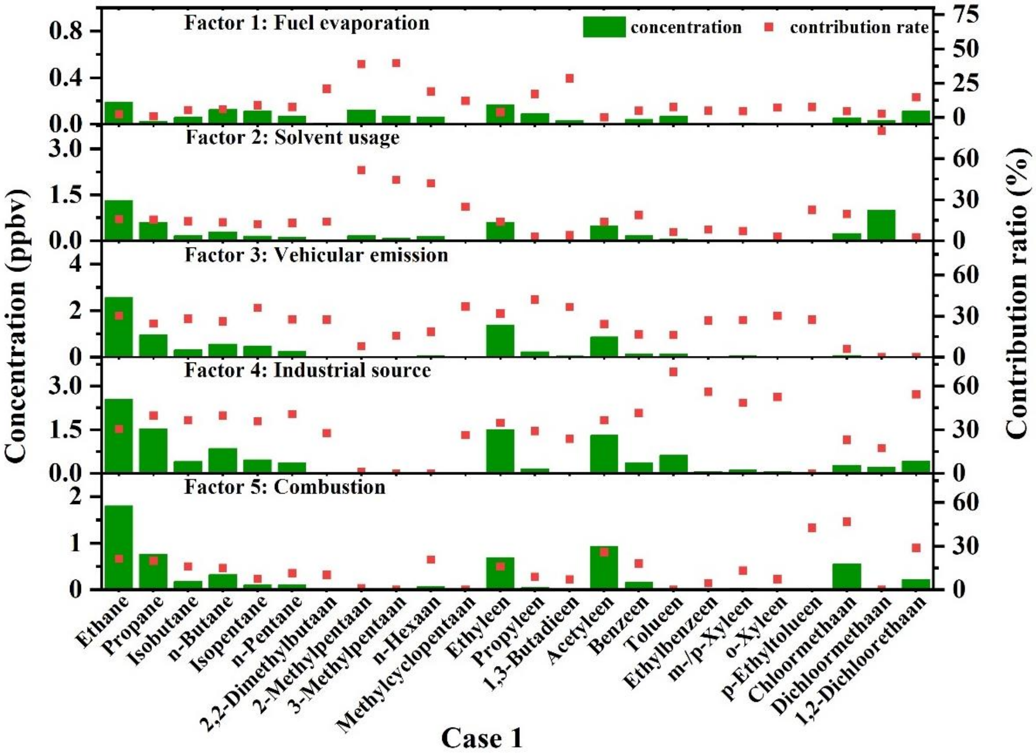
Response**:** We have modified it in the revised version.

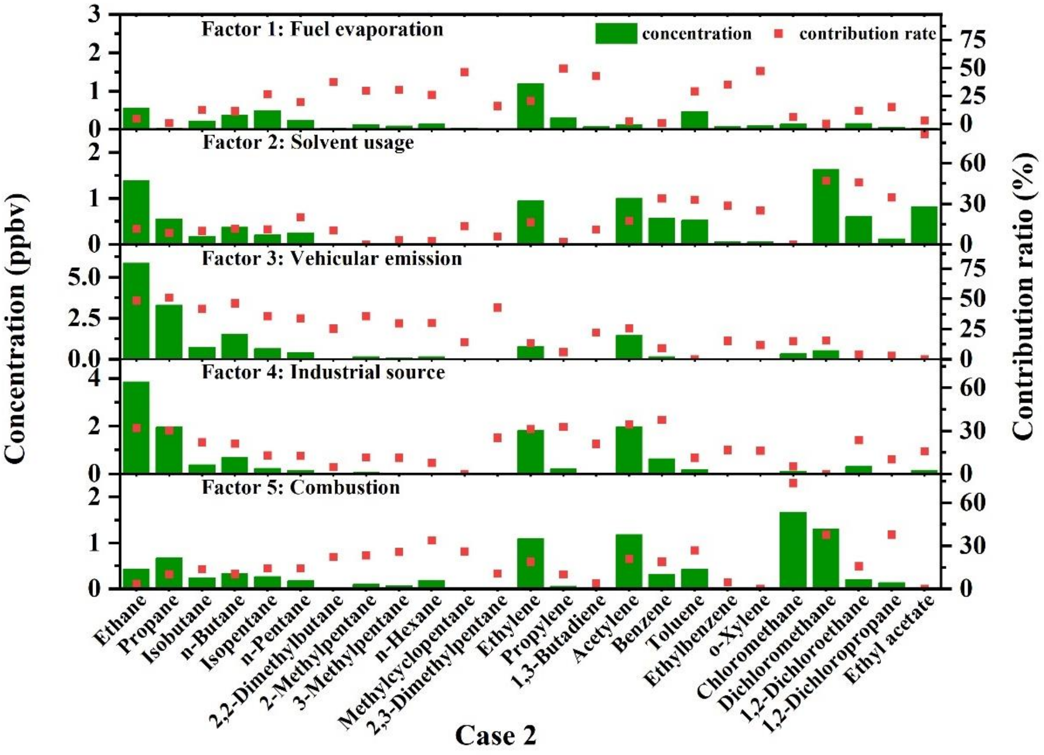
12. Line 316-325: Have you performed PMF in Case 1, Case 3, and clean days? It is recommended to check whether the results of factor analysis are consistent in different conditions (Case 1, Case 2, and clean days) and compare the results.

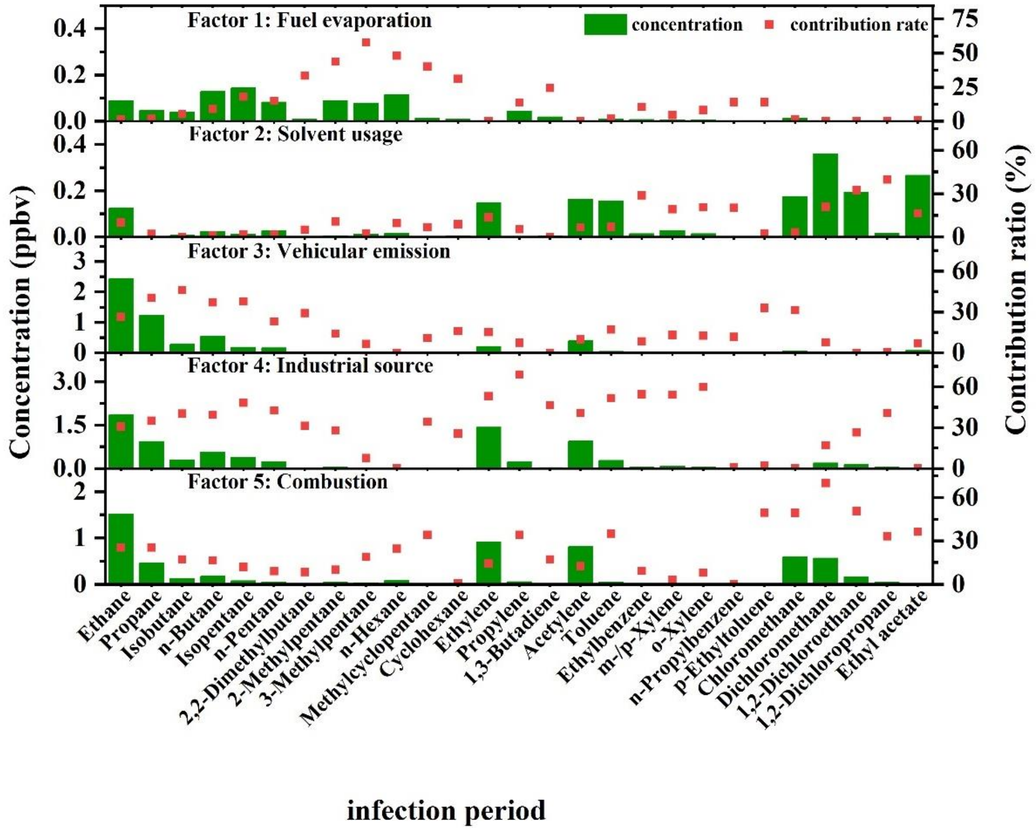
Response**:** We have indeed performed PMF on infection period, recovery period, Case 1, Case 2 and clean days.

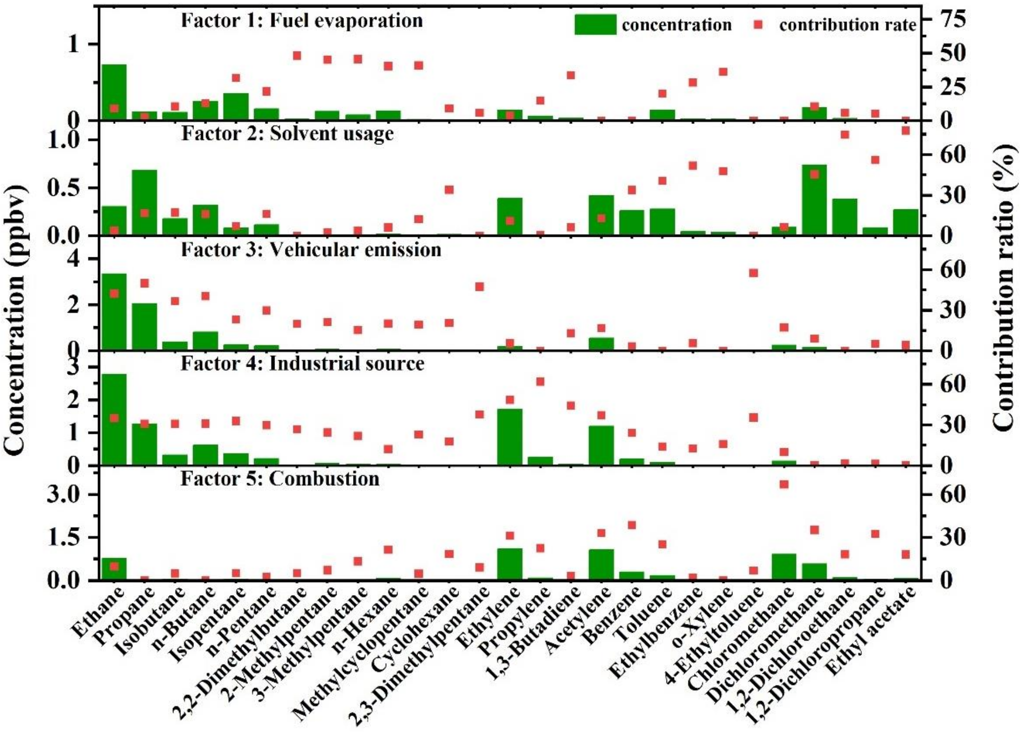
The PMF results for infection period (Dec 1 to 30, 2022), and recovery period (Jan 1 to 31), as well as the two pollution events and clean days, are shown in the figures below (Figure 7). They all exhibit the same 5 factors. It is worth noting that there are two y-axes in Figure 6: the left side represents the concentration of VOCs in units of ppbv, and the right side represents the percentage of specific VOCs within that factor. Additionally, the concentration scales of some figures also differ.

Concentrations of most species were significantly higher during the recovery period than during the infection period. The representative pollution processes in both periods showed the same results as well, with a 79% higher concentration of TVOCs in Case 2 (65.1 ppbv) compared to Case 1 (36.3 ppbv) (Figure 8). While in Case 1 industry was the dominant source of VOCs, by Case 2 motorized sources reached a concentration value of 21.2 ppbv, accounting for 33% of the observed VOCs, and became the dominant source of emissions. This is consistent with the fact that people's mobility activities have increased after the epidemic has entered the recovery period. As a group of VOCs species with the highest concentration share, ethane and propane contributed more to the clean day motor vehicle sources than other processes, which also resulted in a 34% clean day motor vehicle source share.

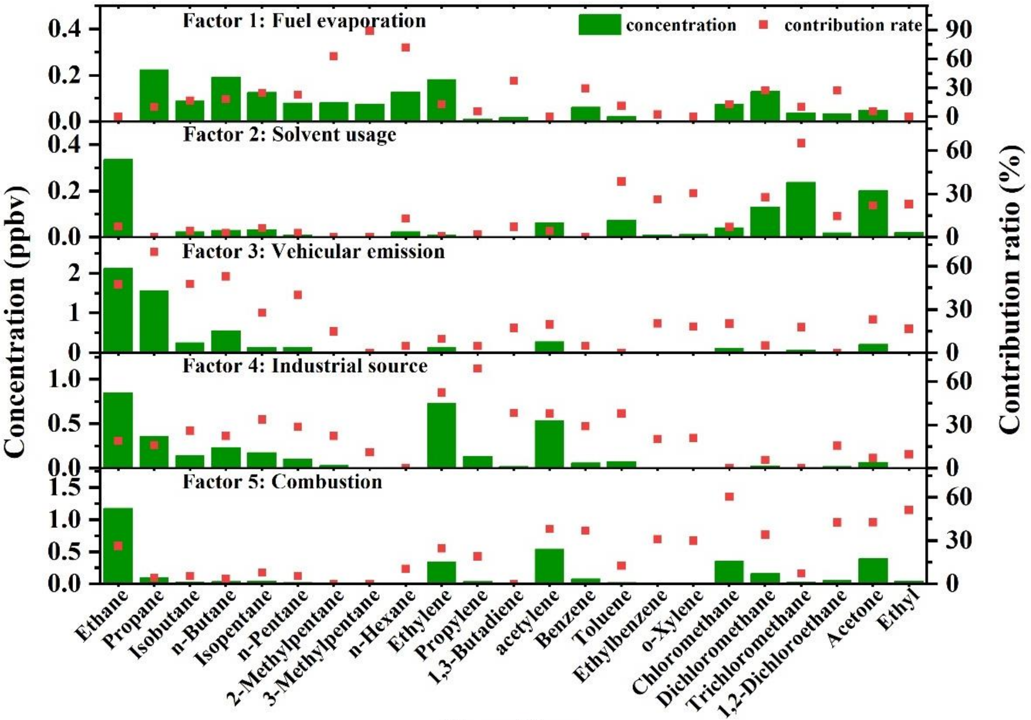



Factor 1: Fuel evaporation
Factor 2: Solvent usage
Factor 3: Vehicular emission
Factor 4: Industrial source
Factor 5: Combustion
concentration
contribution rate
Concentration (ppbv)
Contribution ratio (%)
Ethane
Propane
Isobutane
n-Butane
Isopentane
n-Pentane
2,2-Dimethylbutane
2-Methylpentane
3-Methylpentane
n-Hexane
Methylcyclopentane
2,3-Dimethylpentane
Ethylene
Propylene
1,3-Butadiene
Acetylene
Benzene
Toluene
Ethylbenzene
o-Xylene
Chloromethane
Dichloromethane
1,2-Dichloroethane
1,2-Dichloropropane
Ethyl acetate
Case 2


Factor 1: Fuel evaporation
Factor 2: Solvent usage
Factor 3: Vehicular emission
Factor 4: Industrial source
Factor 5: Combustion
concentration
contribution rate
Concentration (ppbv)
Contribution ratio (%)
Ethane
Propane
Isobutane
n-Butane
Isopentane
n-Pentane
2,2-Dimethylbutane
2-Methylpentane
3-Methylpentane
n-Hexane
Methylcyclopentane
Cyclohexane
Ethylene
Propylene
1,3-Butadiene
Acetylene
Toluene
Ethylbenzene
m-/p-Xylene
o-Xylene
n-Propylbenzene
p-Ethyltoluene
Chloromethane
Dichloromethane
1,2-Dichloroethane
1,2-Dichloropropane
Ethyl acetate
infection period



recovery period



Clean Days

Figure 7. Infection period, recovery period, high pollution events, and

clean days PMF source analysis

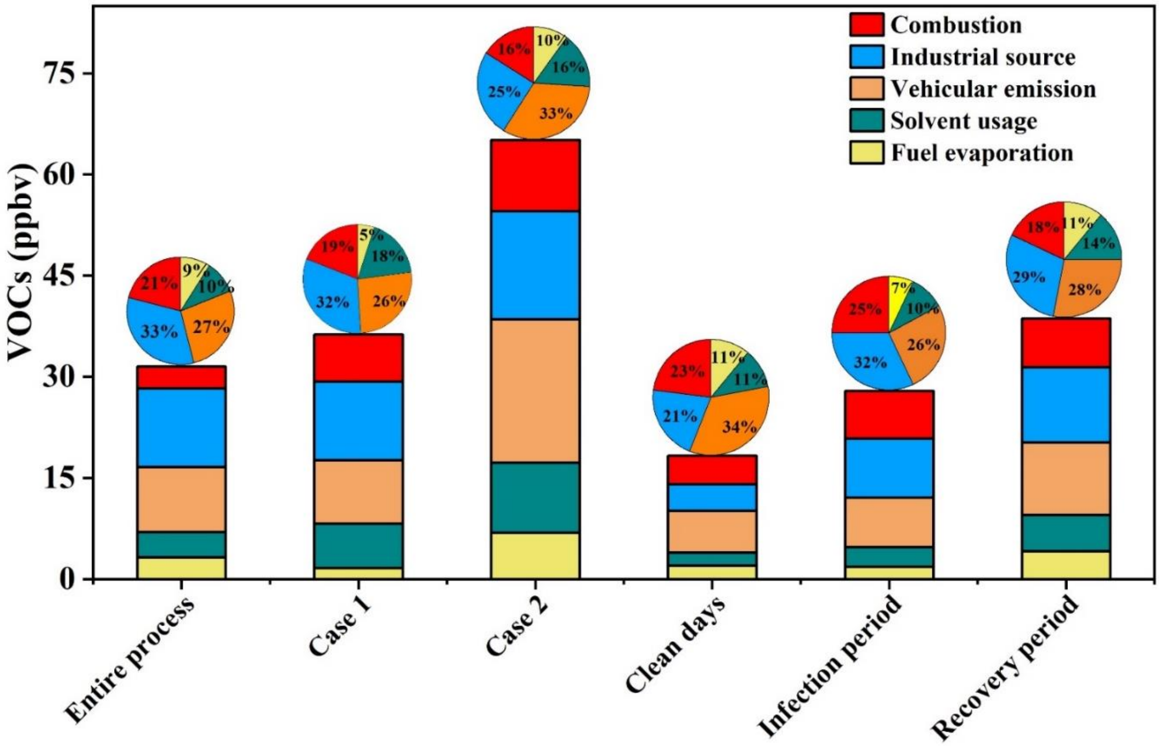


Figure 8. Contribution of each source to VOCs for different processes

13. Section 3.3 SOAFP

In this part, you only discuss the Case 1 and Case 2 processes, and you think that the control of $PM_{2.5}$ pollution in winter should focus on controlling vehicle emissions, solvent use, and combustion. I don't think it's convincing enough. It is recommended to add analysis of clean days. Contrast the pollution process with the clean day.

Response**:** VOCs are estimated to contribute about 16–30% or more of $PM_{2.5}$ by mass through SOA production (Huang et al., 2014). Therefore, by calculating the SOAP value, the influence of different sources on $PM_{2.5}$ production can be reflected to a certain extent.

We calculated the SOAP for the different processes from the PMF results in the previous question and added the results for the clean days as you suggested.

The modified results are shown in Figure 9.

The SOAP of Case 2 was 65.6 μg/m^3, which was much higher than that of Case 1 (37.6 μg/m^3), and the main sources of SOAP differed significantly between the two pollution processes on the clean days. Industrial sources were absolutely dominant in Case 1 (63%). While in Case 2 the contribution of each pollution source is relatively more even, the contribution of solvent use sources and fuel volatilization sources increases to 32% and 26% as the major SOAP sources. The result of clean day with SOAP of 8.8 μg/m^3 also shows that industrial and solvent use sources are the most dominant SOAP sources. Therefore, there is a need to reduce PM$_{2.5}$ pollution by controlling emissions from industrial and solvent use sources.

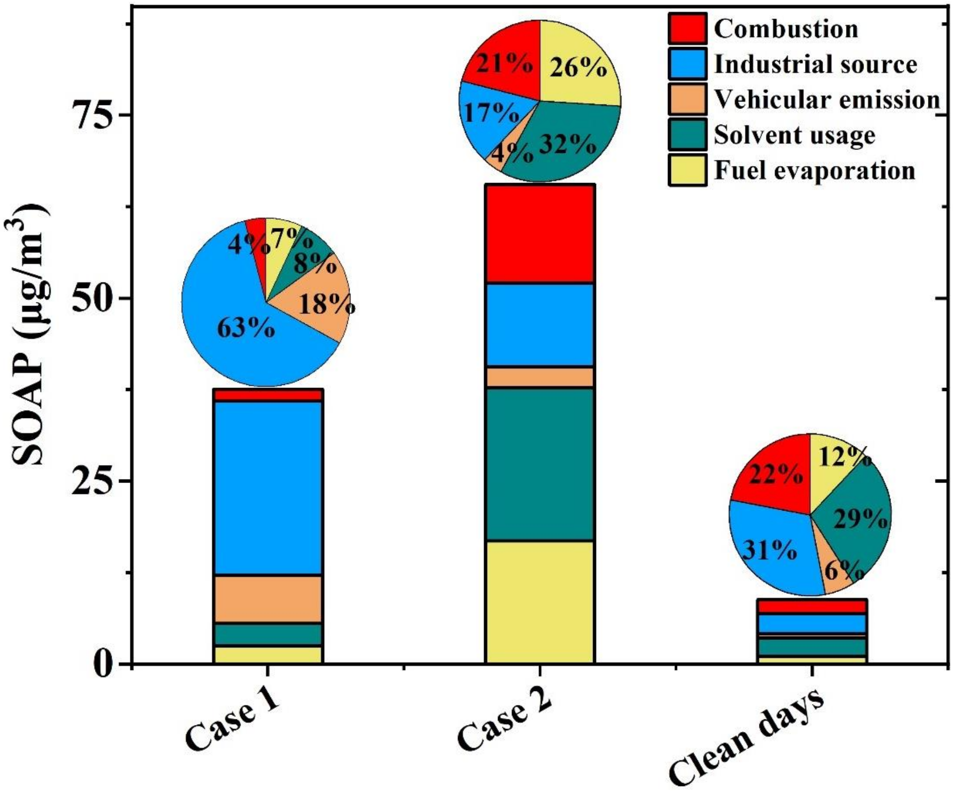


Figure 9. SOAP value and contribution ratio of each component

References:

Huang, R. J.: High secondary aerosol contribution to particulate pollution during haze events in China, Nature 2014, 514 (7521), 218−22.

Reviewer #2:

We do appreciate your constructive and useful comments. To better reply to your overall comments in your long paragraph, we have divided your comments into serval parts with superscript [a, b, c], etc., and correspondingly addressed your comments in a separate paragraph [a, b], etc. More detailed replies are shown in your specific comments.

Overall comment:

The COVID-19 lockdown measures provide a natural experiment for probing air quality changes under substantial emission reductions. Zhang et al. investigate the variations of VOCs in response to the policy-driven emission changes in Zhengzhou city of China by using online ambient measurements, and the PMF model. [a]While this paper is within the scope of ACP, the present manuscript is limited to a cursory data analysis (simply reporting measurement results), without convincing evidence and in-depth discussion, which makes this paper unpublishable in the present form. [b]Further, the innovation of this work is far below the standard required to be published on ACP, which is even not qualified as a measurement report. [c]Though addressing the specific comments below may improve the paper, I don't think these improvements could justify publication in a high-standard journal such as ACP. Concerning the major flaws and the lack of innovation, I think this paper should be rejected.

[a]While this paper is within the scope of ACP, the present manuscript is limited to a cursory data analysis (simply reporting measurement results), without convincing evidence and in-depth discussion, which makes this paper unpublishable in the present form.

We extend our heartfelt appreciation for your insightful comments. In response, we are committed to augmenting the manuscript with a more rigorous quantitative analysis and a profound exploration of the subject matter.

Additionally, we undertook a comprehensive overhaul of the article to elevate its scholarly merit and overall quality.

We are sorry for the unclear and confusing statements in our original draft. Initially we had many cases (5 cases) in different studied periods exhibiting $PM_{2.5}$ pollution, and did not clearly explain why only discussing Case 1 and 3. We also did not clearly state the infection and recovery periods. These shortcomings including the annotations in Fig. 1 certainly confuse readers/reviewers.

Due to the lack of sufficient sampling days in other cases, we only discuss VOCs, and to a lesser extent, $PM_{2.5}$ changes in two major cases (Case 1 and 2 which is previous Case 3) along with clean days as well as infection and recovery periods; all due to the impact of ending China's zero- COVID policy.

In the analysis section of the results discussion, we added quantitative analyses of the main VOC and SOAP species for the clean days and for the two pollution processes; in the PMF source analysis section we added CPF plots and in the supplementary Materials added plots of daily trends in the source analysis results, as well as the rationale for the selection of the PMF factors. The results of the infection period and the recovery period are also compared according to the updated VOCs source analysis results. In addition, the correlation analysis between meteorological conditions and pollutant concentrations, the analysis of potential pollution sources, the PMF factor profiles of different pollution processes, and the concentrations of the main tracers of different processes are added in the supplementary materials, which provide a more scientific basis for the conclusions in our manuscript.

[b]Further, the innovation of this work is far below the standard required to be published on ACP, which is even not qualified as a measurement report.

It is our fault not to clearly show the rationale for our study. This research investigation is centered on the examination of the fluctuations in VOCs and $PM_{2.5}$ pollution levels within Zhengzhou, following the relaxation of COVID-19 control measures with the emergence of COVID-19 variant.

While some atmospheric VOC studies involving the impact of Covid-19 lockdown have been performed in India (Singh et al., 2023a), in China (e.g., Pei et al., 2022; Jensen et al., 2023; Zuo et al., 2024), or with respect to BETX only (e.g., Sahu et al., 2022; Singh et al., 2023b), a gap persisted in the investigation of VOCs due to the impact of abolishment of China's zero-policy. Furthermore, the present study is focused on the period dominated by the COVID-19 Omicron variant, which exhibited distinct characteristics in terms of geographical spread, infected population size, and symptomatology compared to earlier strains (Petersen et al., 2022; Merino et al., 2023). This period also witnessed substantial alterations in China's pandemic zero-Covid control policy, resulting in significant changes in societal activities (Figure 1). Consequently, this study aims to a detailed examination of how the alteration influenced atmospheric pollution, particularly regarding VOCs.

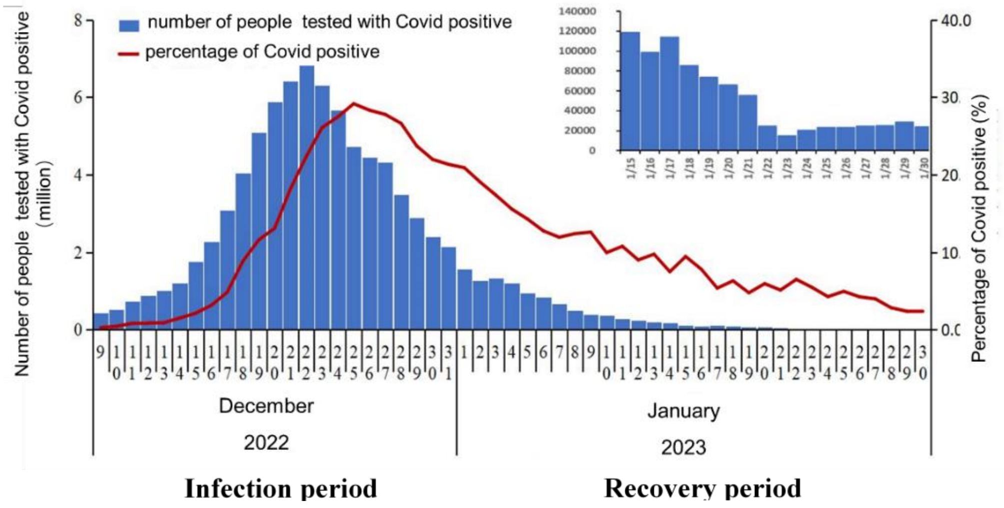


Figure 1. Trend of Omicron infection in China from 9 Dec. 2022 to 1 Jan. 2023 (CCDCP, 2023)

The shadow section in Figure 2 represents two haze pollution events during the monitoring period. A pollution event is determined when the daily average concentration of $PM_{2.5}$ exceeds 75 μg/m^3 (China's II-level standard) for at least three consecutive days. We apologize for the unclear statement and recognize that the original annotations might confuse readers, so we simplify the labeling in Figure 2. To avoid misinterpretation, we deleted processes with no more than 3 days of continuous contamination in Figure 2. In the revised version, we focus on the distinct characteristics of Case 1, Case 2, and Clean days as depicted in the figure. Case 1 (December 5 to December 10 with daily average $PM_{2.5}$ = 142.5 μg/m^3) and Case 2 (January 1 to January 8 with daily average $PM_{2.5}$ = 181.5 μg/m^3) were selected as they represent the pollution events in infection and recovery periods, respectively, due to their long duration and high pollution levels. We divided this period into an infection period (1-30 December 2022) and a recovery period (1 January 2023-31 January 2023) based on Chinese Center for Disease Control and Prevention's December 2022-January 2023 infection data statistics (Figure 1). Any days with a $PM_{2.5}$ concentration lower than 35 μg/m^3 (China's I-level standard) is considered as Clean days.

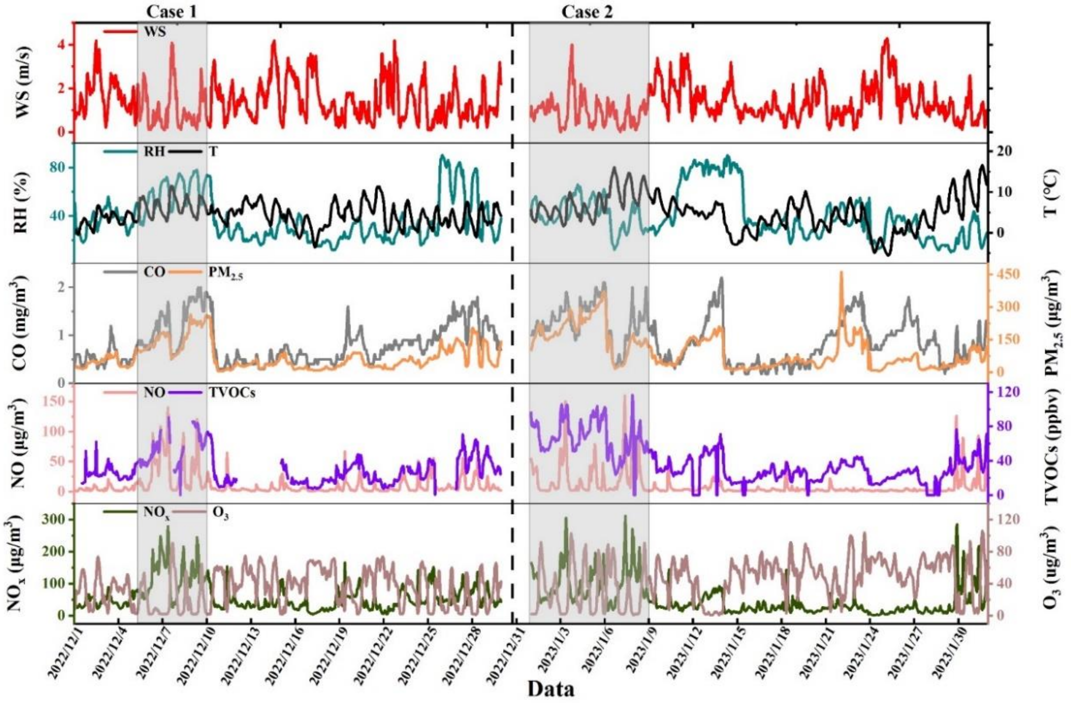


Figure 2. Time series of WS, WD, T, RH, CO, $PM_{2.5}$, NO, TVOCs, NO_x and O_3 during the observation period.

The above definition of pollution process will be incorporated into the revised manuscript.

References:

Jensen et al., 2023. Measurements of Volatile Organic Compounds During the COVID-19 Lockdown in Changzhou, China.

Pei et al., 2022. Decrease in ambient volatile organic compounds during the COVID-19 lockdown period in the Pearl River Delta region, south China.

Sahu et al., 2022. Impact of COVID-19 Pandemic Lockdown in Ambient Concentrations of Aromatic Volatile Organic Compounds in a Metropolitan City of Western India.

Singh et al., 2023a. Substantial Changes in Selected Volatile Organic Compounds (VOCs) and Associations with Health Risk Assessments in Industrial Areas during the COVID-19 Pandemic.

Singh et al., 2023b. Distribution and temporal variation of total volatile organic compounds concentrations associated with health risk in Punjab, India

Zuo et al., 2024. Pollution characteristics and source differences of VOCs before and after COVID-19 in Beijing

Merino et al., 2023. Evaluating the spread of Omicron COVID-19 variant in Spain

Petersen et al., 2022. Clinical characteristics of the Omicron variant - results from a Nationwide Symptoms Survey in the Faroe Islands, International Journal of Infectious Diseases,

[c]The major flaws and the lack of innovation.

The rationale for our study involved three major tasks: (1) Omicron variant; (2) abolishment of China's zero policy; and (3) detailed VOC/$PM_{2.5}$ analysis. To our best knowledge this is the first attempt to evaluate the Omicron variant impact of ending China's zero-Covid policy on ambient VOCs and $PM_{2.5}$. We do hope that through refining and overhauling the article's content, we can deepen our analysis/discussion and potentially alter your negative view.

Please refer to the above brief rationale (innovation) for our study. We will try to explain the innovation of our work in more details below.

China lifted the zero-COVID strategies, notably by announcing the '10 measures' about the optimization of COVID-19 rules on 7 December 2022 (Xinhua, 2022). After that, China experiences a nationwide outbreak of COVID-19. Leung et al. (2023) estimated that the cumulative infection attack rate in Beijing was 75.7% (95% credible interval (CrI): 60.7–84.4) on 22 December 2022 and 92.3% (95% CrI: 91.4–93.1) on 31 January 2023. A recent study by Liang et al. (2023) showed that the cumulative SARS-CoV-2 infection rate rose rapidly to 70% within three weeks after the ending of the zero-COVID policy in Macao. A study conducted in Guangzhou also revealed that the infection attack ratio reached to 80.7% (95% CrI: 72.2–86.8) at 30 days after easing the zero-COVID policy (Huang et al., 2023)

Indeed, there have been some studies discussing the impact of human factors on air pollution during and after the outbreak of the Coronavirus disease (e.g., Ma et al., 2022; Jiang et al., 2023; Song et al., 2023), but as mentioned earlier, only a few studies with in-depth exploration of the changes in VOCs and none dealing with ending the zero-Covid policy during Omicron variant infection period.

Our research primarily concentrates on the period dominated by COVID-19 Omicron variant, where they demonstrate notable differences from the early virus strains (i.e., original SARS-CoV-2 virus and Delta) in terms of geographical transmission, the scale of the infected population, and symptom manifestation.

The 7th announcement of 2022 issued by the National Health Commission of China states that, starting from January 8, 2023, the Class A infectious disease prevention and control measures specified in the Infectious Disease Prevention and Control Law of the People's Republic of China for COVID-19 will be lifted; COVID-19 will no longer be included in the quarantine infectious disease management stipulated by the Frontier Health and Quarantine Law of the People's Republic of China. This signifies a significant shift in China's pandemic control policy in comparison to the period preceding the issuance of the announcement. We believe that this change is worth exploring in terms of its impact on transportation and industrial production emissions. Essentially, this research serves to address the existing gap in the literature concerning the effects of the Omicron variant on VOCs and $PM_{2.5}$ pollution levels in Zhengzhou amidst policy fluctuations, specifically the end of zero-Covid policy.

Our research findings also confirm that traffic emissions remain the primary source of pollution in Zhengzhou, thus providing valuable insights for formulating control measures.

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Jiang, N., Hao, X., Hao, Q., Wei, Y., Zhang, Y., Lyu, Z., Zhang, R.: Changes in Secondary Inorganic Ions in $PM_{2.5}$ at Different Pollution Stages Before and After COVID-19 Control, Environmental Science, 0250-3301(2023)05-2430-11, https://doi.org/10.13227/j.hjkx.202206170, 2023.

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Ma, Q., Wang, W., Wu, Y., Wang, F., Jin, L., Song, Y., Han, Y., Zhang, R., Zhang, D.: Haze caused by NO_x oxidation under restricted residential and industrial activities in a mega city in the south of North China Plain, Chemosphere, Volume 305, 135489, https://doi.org/10.1016/j.chemosphere.2022.135489, 2022.

Song, X., Zhang, D., Li, X., Lu, X., Wang, M., Zhang, B., Zhang, R.: Simultaneous observations of peroxyacetyl nitrate and ozone in Central China during static management of COVID-19: Regional transport and thermal decomposition, Atmospheric Research, Volume 294, 106958, https://doi.org/10.1016/j.atmosres.2023.106958, 2023.

Xinhua News Agency, The "new ten" to optimize the implementation of epidemic prevention and control is here, http://www.news.cn/politics/2022-12/07/c_1129189285.htm, 2022.

Major comments:

1) The major weakness of this work is the lack of innovation. The impacts of the Omicron outbreak on Chinese cities are already well-documented and extensive studies have been conducted to elucidate the role of the anthropogenic sector on air pollution during- and post-outbreak periods. The authors claimed that industrial and vehicular emissions are dominant sectors contributing to ambient VOC, which is quite clear in prior studies. Further, the changes in $PM_{2.5}$ and VOCs in response to the lockdown are broadly consistent with previous findings in Zhengzhou (even in Chinese literature). What is the innovation of this work and what are the new findings from this work that contribute to the air quality community?

Response: Again, we apologize for the lack of description of the rationale for our study and lack of in-depth analysis of our VOC results in our original draft. We have added the distribution of major flaws and the lack of innovation in the above comment, please see our point-by-point responses of ^{c}The major flaws and the lack of innovation.

The usage of SOAP should be revisited. The authors should be aware that SOAP is a very simple metric that provides limited information on SOA formation potential because SOA yield for individual VOCs in China may vary significantly in other countries due to the different levels of NOx and other oxidants. SOAP is generally adopted to reflect the SOA production potential based on bottom-up emission inventory (see Wu & Xie, ES&T), rather than using short-time ambient measurements. Therefore, I doubt the conclusion driven by the simple SOAP calculation. The authors should consider using F0AM and PBM-MCM for examining SOA production changes rather than SOAP.

Response: Thank you very much for your valuable advice. After carefully reading the literature you recommended, we found that the analysis conclusions about SOAP in Wu & Xie 's research have some similarities with ours (Wu et al., 2018). For example, Wu & Xie 's research found that aromatics contribute the most to SOAP, followed by alkanes and alkenes. Similarly, the results calculated using the toluene weighted mass contributions method (Derwent et al., 2010) also indicate that aromatics contribute the most to SOAP, followed by alkanes and alkenes. The toluene weighted mass contributions method has been widely used in calculating SOAP based on observed VOCs (Zhang et al., 2017; Hui et al., 2019; Li et al., 2020). Therefore, this method also has a certain representativeness. Of course, as you said, this is not the most appropriate method, and using F0AM and PBM-MCM for examining SOA production changes is a very good suggestion. However, due to the limitations of our related technologies, we are unable to use F0AM or PBM-MCM for examining SOA production changes. This is a very regrettable thing. Your suggestion has pointed out a very good direction for our future research.

On the other hand, PBM-MCM can indeed effectively simulate atmospheric chemical processes in the troposphere under certain circumstances. Taking MCMv3.2 as an example, it includes 5900 species of reactants and 16500 chemical reactions. During the modeling process, a large number of model parameters need to be set, and it is influenced by various environmental variables such as temperature, atmospheric pressure, relative humidity, boundary layer height (Lam et al., 2013), among others. The-results may have significant errors compared to the true values. Furthermore, this model is often used to analyze the sources of atmospheric O_3 (Xie et al., 2021).

The issues faced when applying the F0AM model are similar as well. For example, the observed photodissociation frequency (J value) needs to be input. The photodissociation frequency controls the generation of free radicals and the lifetimes of many compounds. Due to the various influencing factors, the accurate simulation of J values is challenging. A major shortcoming of the modeling approach is the lack of explicit representation of transport processes (entrainment, dilution, etc.), which has several practical consequences. First, primary emissions like NO_x and hydrocarbons must be constrained or otherwise re-supplied to compensate for chemical loss. Emissions can also be parameterized explicitly but require knowledge of the boundary layer depth and assumed instantaneous mixing. Second, a generic "physical loss" lifetime of 6–48 h is often assigned to all species to mitigate build-up of long-lived oxidation products over multiple days of integration. Model users must be aware of the limitations imposed by these choices (Wolfe et al., 2016).

Even though the SOAP calculation process based on a coefficient of individual VOC species developed by Derwent et al. (2010) certainly has errors, it is our belief that SOA production obtained from F0AM and PBM-MCM models exhibited as many uncertainties as a simple Derwent's SOAP approach.

The importance of SOA to atmospheric problems is well known. Previous studies have used SOAP calculations to investigate the contribution of atmospheric VOCs to $PM_{2.5}$ production, demonstrating that contribution of different sources to the formation of SOA. (Shi et al., 2015; Liu et al., 2022; Liang et al., 2023). In our paper, SOAP values were determined to reflect the impact of the end of China's zero-Covid policy. But we will continue to work hard, hoping to include the analysis of F0AM and PBM-MCM in our future research.

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Liang, S., Gao, S., Wang, S., Chai, W., Chen, W., Tang, G.: Characteristics, sources of volatile organic compounds, and their contributions to secondary air pollution during different periods in Beijing, China, Science of the Total Environment, Volume 858, Part 2, https://doi.org/10.1016/j.scitotenv.2022.159831, 2023.

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Wolfe, G.M., Marvin, M.R., Roberts, S.J., Travis, K.R., Liao, J.: The Framework for 0-D Atmospheric Modeling (F0AM) v3.1, Geoscientific Model Development, 9, 3309–3319, https://doi:10.5194/gmd-9-3309-2016, 2016.

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Xie, Y., Cheng, C., Wang, Z., Wang, K., Wang, Y., Zhang, X., Li, X., Ren, L., Liu, M., Li, M.: Exploration of O_3-precursor relationship and observation-oriented O_3 control strategies in a non-provincial capital city, southwestern China, Science of The Total Environment,800, 149422, https://doi.org/10.1016/j.scitotenv.2021.149422, 2021.

Zhang, Z., Wang, H., Chen, D.: Emission characteristics of volatile organic compounds and their secondary organic aerosol formation potentials from a petroleum refinery in Pearl River Delta, China, Science of The Total Environment, 584–585, 1162-1174, https://doi.org/10.1016/j.scitotenv.2017.01.179, 2017.

3) The captions of the results section are meaningless. "Pollution characteristics" and "Source appointment" is not clear to the readers and should be rewritten for clarification.

Response: We have modified “Pollution characteristics” to “Overview of variation in pollutants and meteorological parameters”, and “Source appointment” to “Source Analysis of VOCs”.

4) The writing of this paper is in need of much attention. Specifically, the writing suffers from a series of fundamental issues, including a lack of clear organization, pervasive grammatical, and stylistic errors. I suggest the authors carefully read through the manuscript and rephrase the results section. There is substantial awkward phrasing throughout the paper which is confusing and misleading to the readers.

Response: We apologize for the grammatical and stylistic errors in the manuscript, and unclear statement which certainly confuses readers. We have addressed your comments in serval parts with superscript $^{a, b}$ above.

We will undertake extensive revisions and proofreading to enhance the clarity and coherence of the manuscript. This will ensure that the article is free from grammatical errors and stylistic issues, making it easier for readers to understand.

Minor comments:

1) The SOA formation potential is called "SOAP" rather than "SOAFP". The author should correct this abbreviation.

Response: After thorough examination of the literature, we found both usages in circulation. However, in the revised text, we shall adopt your suggested term of SOAP, as demonstrated in our replies to reviewers' comments.

Reviewer #3:

We do appreciate your constructive and useful comments. To better reply to your detailed comments in your long paragraph, we have divided your comments into serval parts with superscript a, b, c, and correspondingly addressed your comments in a separate paragraph a, b, etc. More detailed replies are shown in your specific comments.

Detailed comments:

To provide a fair assessment, I refrained from reading previous reviews of the manuscript. Zhang et al. investigated VOC emissions during winter in Zhengzhou, China, likely aiming to understand the impact of the Omicron lockdown on city pollutants. [a]However, given the extensive documentation of air quality studies during COVID pandemic and its variants, including Omicron, the manuscript lacks novelty in this context. [b]The discussion on source apportionment also falls short, lacking depth and quantitative analysis. [c]Overall, the manuscript does not meet the standards for publication in ACP. I recommend the authors revise the manuscript, enhance data analysis and interpretation, present their findings in a more scientifically rigorous manner, and plan to resubmit as a new submission.

Response: We first express gratitude for your encouragement of our paper revision for resubmittal. The following replies are for your general comments.

[a]However, given the extensive documentation of air quality studies during COVID pandemic and its variants, including Omicron, the manuscript lacks novelty in this context.

Response: It is undeniable that there have been numerous studies on air quality during the COVID-19 pandemic and its variants (including the Omicron variant). However, there is still a gap in the investigation of VOCs during the epidemic period in Zhengzhou; almost no VOC study before/after the impact of ending China's zero- COVID policy.

During the studied period, China experienced significant shifts in its control policies regarding the Omicron variant, which in turn caused substantial changes in social activities. Consequently, this study aims to delve into the impact~~s~~ of the zero-COVID policy change on atmospheric pollution, with a particular focus on VOCs.

[b]The discussion on source apportionment also falls short, lacking depth and quantitative analysis.

Response: In the analysis section of the results discussion, we added quantitative analyses of the main VOC and SOAP species for the clean days and for the two pollution processes; in the PMF source analysis section we added CPF plots and in the supplementary Materials added plots of daily trends in the source analysis results, as well as the rationale for the selection of the PMF factors. The results of the infection period and the recovery period are also compared according to the updated VOCs source analysis results. In addition, the correlation analysis between meteorological conditions and pollutant concentrations, the analysis of potential pollution sources, the PMF factor profiles of different pollution processes, and the concentrations of the main tracers of different processes are added in the supplementary materials, which provide a more scientific basis for the conclusions in our manuscript.

cOverall, the ion in ACP. I recommend the authors revise the manuscript, enhance data analysis and interpretation, present their findings in a more scientifically rigorous manner, and plan to resubmit as a new submission.

Response: Your encouragement is greatly appreciated. We will overhaul the manuscript and plan to resubmit it.

The followings are our responses to all of your comments and valuable suggestions.

1、The manuscript lacks analysis of measurements during or post-Omicron period to provide relevant insights for policy and management. Authors have broadly compared two pollution cases with a clean day during the sampling period. Even in this regard, the poor labeling technique of Figure 1, makes it very confusing what are Cases 1 through 5. Discussions for Cases 2 and 4 are missing. I feel that the title is misleading as the data has not been leveraged to present relevant results related to the Omicron period and policy relevance.

Response**:** We are sorry for the unclear and confusing statements in our original draft. Initially we had many cases (5 cases) in different studied periods exhibiting $PM_{2.5}$ pollution, and did not clearly explain why only discussing Case 1 and 3. We also did not clearly state the infection and recovery periods. These shortcomings including the annotations in Fig. 1 certainly confuse readers/reviewers.

In our revised text, due to the lack of sufficient sampling days in other cases, we only discuss VOCs, and to a lesser extent, $PM_{2.5}$ changes in two major cases (Case 1 and 2 which is previous Case 3) along with clean days as well as infection and recovery periods; all due to the impact of ending China’s zero-COVID policy.

We have also added a quantitative analysis of the dominant species of VOCs and SOAP during the Case 1 and Case 2.

China lifted the zero-COVID strategies, notably by announcing the ‘10 measures' about the optimization of COVID-19 rules on 7 December 2022 (Xinhua, 2022). After that, China experiences a nationwide outbreak of COVID-19. We divided this period into an infection period (1-30 December 2022) and a recovery period (1 January 2023-31 January 2023) based on Chinese Center for Disease Control and Prevention's December 2022-January 2023 infection data statistics (Figure 1). The data in Figure 1 shows that during the initial phase when the containment had just been lifted and Omicron was not widely spread, there were long periods of pollution (Case 1, December 5 to December 10, daily mean $PM_{2.5}$ = 142.5 μg/m^3). While during the peak of Omicron infections, there were several consecutive clean days. When the peak of Omicron infection ended and the recovery phase began, there was another prolonged period of pollution lasting 8 days (Case 2, January 1 to January 8 with daily average $PM_{2.5}$ = 181.5 μg/m^3), which aligns with the actual situation of increased emission intensity due to intensified human activities. The aim of this data analysis is to confirm the correlation between the series of phenomena and the policies and Omicron infections.

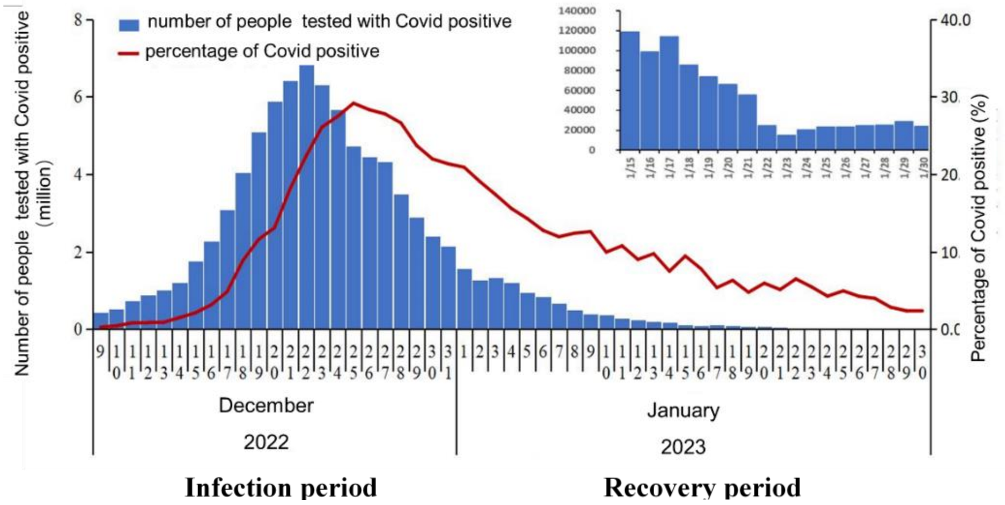

Figure 1. Trend of Omicron infection in China from 9 Dec. 2022 to 1 Jan. 2023 (CCDCP, 2023)

References:

Xinhua News Agency: the "new ten" to optimize the implementation of epidemic prevention and control is here, http://www.news.cn/politics/2022-

12/07/c_1129189285.htm, 2022.

2、In the source apportionment section, the authors seem to have limited knowledge of using the VOC ratios. The results presented are very vague and do not seem to add any quantitative information.

Response: We appreciate your feedback and acknowledge that there may have been limitations in our use of VOC ratios.

The ratios of specific species are commonly employed to assess the sources of atmospheric VOCs and the degree to which air masses have aged (Xiong et al., 2020). However, this method only provides a preliminary assessment of VOC sources. For example, Yang et al. (2023) found that the T/B ratio in the Ningbo area was 0.97, indicating a strong influence of vehicular emissions on VOC emissions in that region. Zhang et al. (2023) identified X/E ratios in the range of 3.33–5.68 in the Rizhao area, suggesting a significant influence of local emissions on VOCs in that area. Wu et al. (2023) discovered a ratio of isopentane and n-pentane of 1.8 in the Huairou area, indicating that n-pentane was more likely to originate from a mix of gasoline and fuel evaporation sources. They also found an isobutane/n-butane ratio of 0.52 in Huairou, suggesting that LPG might be the main source of the two species.

To address your concerns about the vague presentation of results, we have revisited our analysis and improved the presentation of our results to provide more quantitative information (Table 1). We have made the necessary revisions to the manuscript in line with your suggestions. (Line 296-315)

Line 296-315: Specific VOC ratios can be used for initial source identification of VOCs and determination of photochemical ages of air masses (Monod et al., 2001; An et al., 2014; Li et al., 2019). Table 1 lists the species concentrations and four ratios used to identify potential sources of VOCs.

Toluene-to-benzene ratio (T/B ratio) was widely used to assess the relative importance of different sources. Specifically, T/B ratio with the value of 1.3–3.0 was observed in vehicle emissions for vehicles with different fuel types

(Schauer et al., 2002; Wang et al., 2015). The reported T/B ratio for combustion processes was between 0.13 and 0.7 (Li et al., 2011; Wang et al., 2014). The average T/B value for the entire period was 1.0, indicating that both traffic emissions and combustion are significant sources of VOCs.

The isopentane/n-pentane concentration ratios of 0.6-0.8 represent mainly coal combustion emissions, ratios of 0.8-0.9 represent liquefied petroleum gas (LPG) emissions, 2.2-3.8 represent vehicle exhaust emissions, and 1.8-4.6 represent fuel evaporation (Conner et al., 1995; Liu et al., 2008; Li et al., 2019). The overall ratio of i-pentane/n-pentane is 1.4, indicating that pentane is mainly derived from the combined effects of liquid petrol and fuel evaporation.

Isobutane/n-butane concentration ratios of 0.2-0.3 represent vehicle emissions, 0.4-0.6 LPG use , and 0.6-1.0 represent natural gas emissions (Russo et al., 2010; Zheng et al., 2018). The ratio of isobutane/n-butane in this study was 0.50, which suggests that the VOC concentrations at the observation sites are influenced by natural gas emissions (Shao et al., 2016; Zeng et al., 2023).

Table 1. Specific VOCs concentrations and ratios

species	Concentration (ppbv)	Ratio
toluene	0.7	toluene/benzene = **1.0**
benzene	0.7	
isopentane	1.0	isopentane/n-pentane = **1.4**
n-pentane	0.7	
isobutane	0.9	Isobutane/n-butane = **0.5**
n-butane	1.8	
m/p-xylene	0.2	m/p-xylene/ethylbenzene = **2.0**
ethylbenzene	0.1	

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3、The PMF source apportionment is weak, lacking statistical analysis and error estimation. There is no statistical analysis that supports why 5 factor was the best solution. The use of median value to replace missing values is not a justifiable way to treat the data, if the authors think so then needs to be discussed. Authors should examine at least 100 base runs with different seed numbers to find the best solution. Authors should discuss uncertainty and error estimations, and rotation ambiguity analysis.

Response: Thank you very much for your pertinent advice and we will answer your questions point by point:

[a]The PMF source apportionment is weak, lacking statistical analysis and error estimation. There is no statistical analysis that supports why 5 factor was the best solution. Authors should discuss uncertainty and error estimations, and rotation ambiguity analysis.

We used displacement of factor elements (DISP) to assess PMF modelling uncertainty (for a description, see Paatero et al. (2014)). Q was less than 1% and no swaps occurred for the small est dQ^{max} in DISP. Fpeak values from -2 to 2 were tested to explore the rotational stability of the solutions (Figure 2b). Q_{true}/Q_{exp} is lowest when Fpeak = 0, so we chose the PMF results for that case.

After examining 3-8 factors, 20 base runs with 5 factors eventually selected to represent the final result. We provide an explanation of factor selection in the Supplementary Materials. Figure 2(a) includes Q_{true}/Q_{exp}, Q_{robust}/Q_{exp} for factors 3-8. The slopes of these two ratios in changed at five factors, and we found that five factors were more realistic after repeated comparisons of the results at four, five and six factors. These five factors eventually selected as potential sources for the observed VOCs are: (1) Fuel evaporation; (2) Solvent usage; (3) Vehicular emission; (4) Industrial source; and (5) Combustion. Five factors have been commonly reported before, e.g., in Shijiazhuang, northern China (Guan et al, 2023) and in Beijing (Cui et al., 2022).

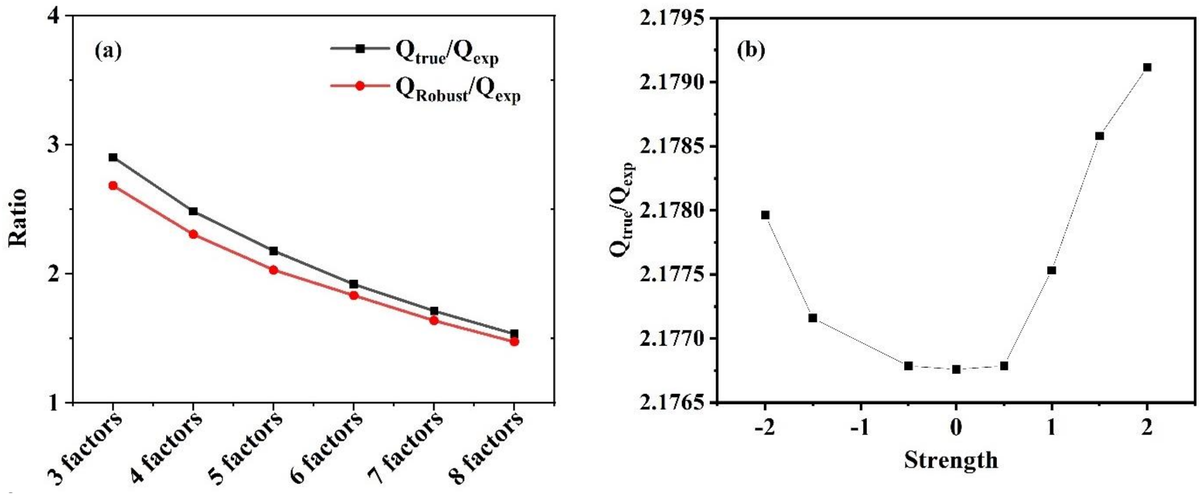

Figure 2. (a) The $Q_{true}/Q_{expected}$ ratios in different solutions; (b) the $Q_{true}/Q_{expected}$ ratio for different Fpeak value solutions.

[b]The use of median value to replace missing values is not a justifiable way to treat the data, if the authors think so then needs to be discussed.

We reviewed the literature of relevant studies based on your suggestion and found that there have been previous studies that chose to use the median as a replacement for missing (Baudic et al., 2016). In addition, the EPA PMF 5.0 User Guide also recommends using the median as a proxy for missing values (Norris et al., 2014). Therefore, we believe this is a reasonable approach to the data.

[c]Authors should examine at least 100 base runs with different seed numbers to find the best solution.

The EPA PMF 5.0 User Guide recommends 20 base runs. We reviewed studies using the PMF model and found that many of the results were obtained from 20 base runs (Qu et al., 2018; Li et al., 2015). Therefore, the results obtained from 20 base runs are credible.

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4、While analyzing PMF factors, authors should use the time series trend, diurnal variations, use of wind speed and direction for identifying possible source sectors, and comparison with other inorganic tracers like trace gases to parameterize the PMF factors. Without some of these analyses, naming the factors just using the VOC profile may be inaccurate as there can be several sources for an individual VOC.

Response: Based on your suggestions, we have updated the PMF spectra and plotted the daily trends for the different sources and the CPF plots for each source.

Figure 3 shows the chemical profiles of individual VOCs resolved by the PMF model during the entire observation period. These five factors eventually selected as potential sources for the observed VOCs are: (1) Fuel evaporation; (2) Solvent usage; (3) Industrial source; (4) Vehicular emission; (5) Combustion.

Alkanes of C4-C6 substances were predominant in factor 1, including 2-methylpentane, 3-methylpentane, isobutane, n-butane, isopentane and n-pentane from oil and gas (Xiong et al., 2020). Figure 4 shows that emissions from this source peak at midday, when fuel volatilization is high, The CPF plot shows that south-east is the dominant direction at wind speeds of less than 2 m/s (Figure 5a). Therefore, factor 1 was identified as the source of oil and gas volatilization.

The contribution of benzene, toluene, methylene chloride, 1,2-dichloroethane and ethyl acetate was high in factor 2. It has been shown that Benzene, Toluene, Ethylbenzene, and Xylene is an important component in the use of solvents (Li et al., 2015); methylene chloride is often used as a chemical solvent, while esters are mostly used as industrial solvents or adhesives (Li et al., 2015). Factor 2 is determined to be a solvent usage source. The CPF plot shows that local sources with wind speeds less than 1 m/s are the main sources (Figure 5b).

Factor 3 contains predominantly C3-C8 alkanes, olefins and alkynes, and relatively high concentrations of benzene. These substances are usually emitted by industrial processes (Shao et al., 2016), so Factor 4 is defined as an industrial source. The CPF plots indicate that a local source at low wind speeds is the dominant sources (Figure 5c).

Factor 4 is characterized by relatively high levels of C2-C6 low-carbon alkanes (ethane, propane, isopentane, n-pentane, isobutane and n-butane), olefins (ethylene and propylene), and benzene and toluene, which are important automotive exhaust tracers (Song et al., 2021; Zhang et al., 2021b). Ethylene and propylene are important components derived from vehicle-related activities. Previous studies of VOCs in Zhengzhou have shown a high percentage of VOCs emitted from gasoline vehicles, with the main source of alkanes being on-road mobile sources (Bai et al., 2020). The daily variation of this source in Figure 3 shows a bimodal trend, with peaks occurring in the morning and evening peaks of traffic, consistent with motor vehicle emissions. Figure 5d shows that this source is mainly from the west where wind speeds are below 2 m/s, and in this direction, there are a number of urban arterial roads with high traffic volumes. Therefore, factor 4 was defined as vehicular emission source.

The highest contribution to Factor 5 is chloromethane (62%). Benzene (46%) and acetylene (41%) also contribute highly to factor 5. Chloromethane is the key tracer for biomass combustion and acetylene is the key tracer for coal combustion (Xiong et al., 2020). Therefore, Factor 5 is defined as a combustion source. The CPF plot shows that at wind speeds below 2 m/s, the north-east direction is the dominant source direction (Figure 5e).

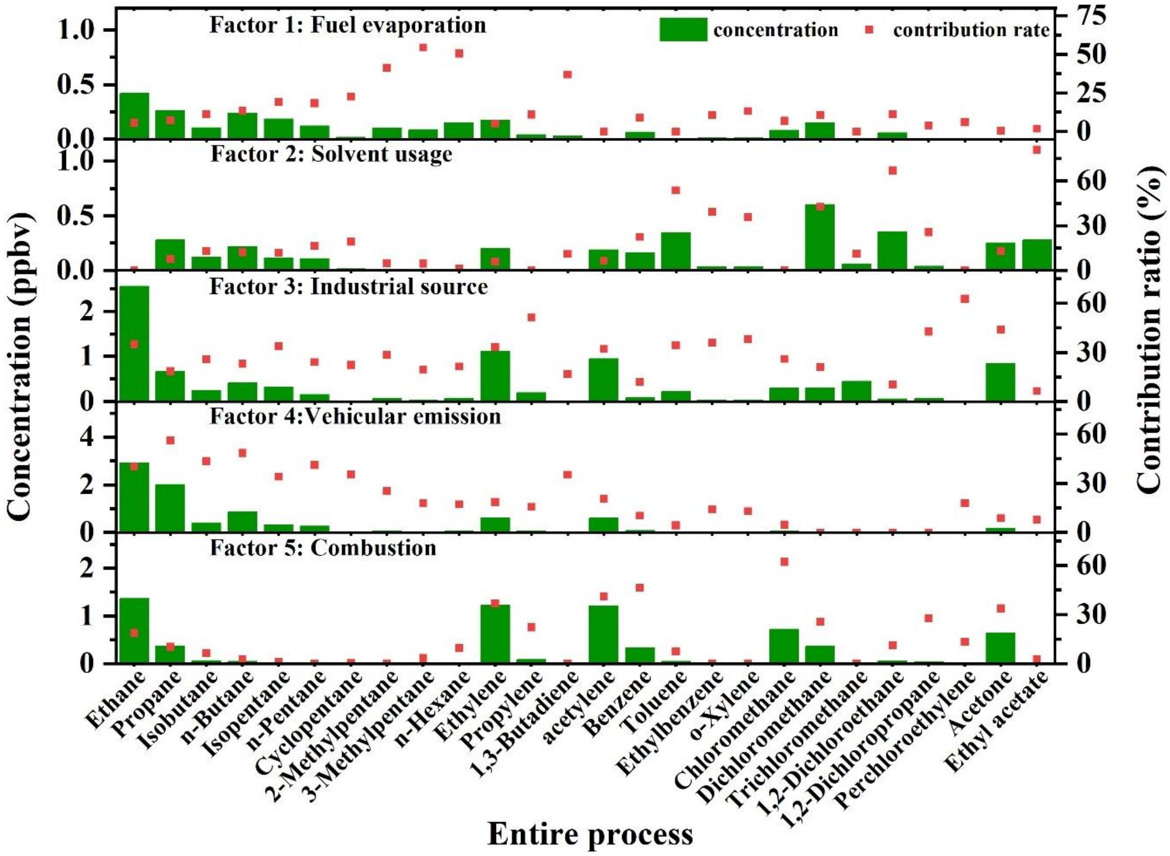


Figure 3. Concentration of VOC species in each factor and contribution to each source

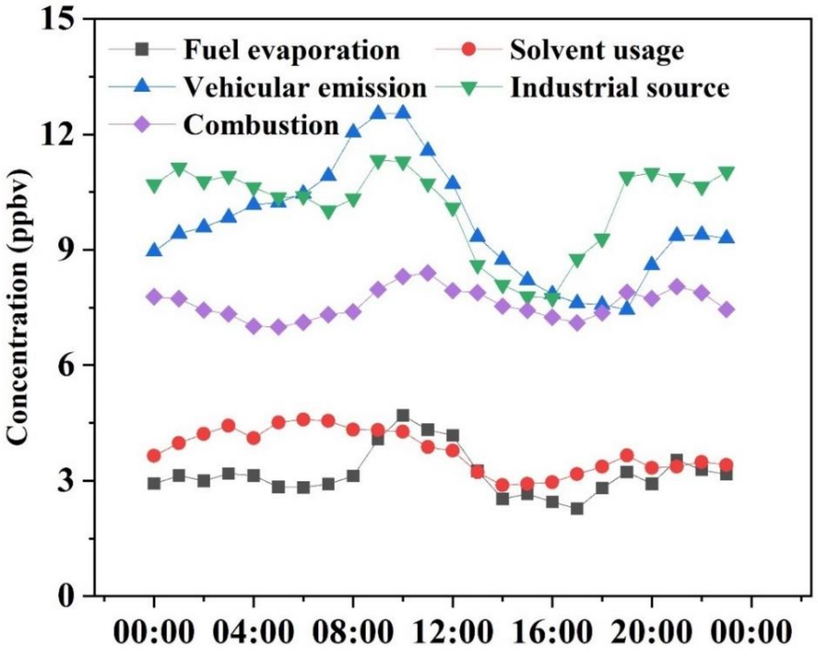
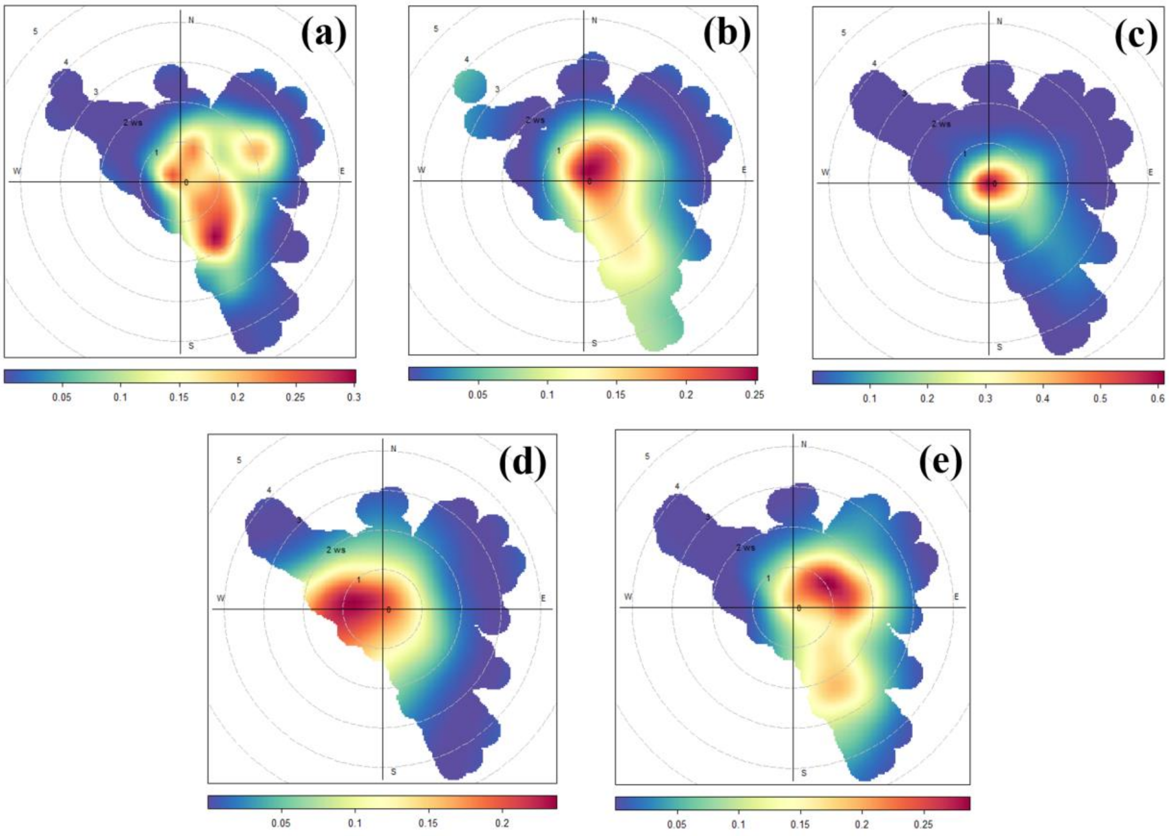


Figure 4. Characteristics of daily changes in different sources obtained using the PMF model



Note: a: Fuel evaporation; b: Solvent usage; c: Industrial source; d: Vehicular emission; e: Combustion.

Figure 5. CPF plots of five VOCs sources obtained using the PMF model

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Song, M., Li, X., Yang, S., Yu, X., Zhou, S., Yang, Y., Chen, S., Dong, H., Liao, K., Chen, Q., Lu, K., Zhang, N., Cao, J., Zeng, L., and Zhang, Y.: Spatiotemporal variation, sources, and secondary transformation potential of volatile organic compounds in Xi'an, China, Atmospheric Chemistry and Physics, 21, 4939-4958, https://doi.org/10.5194/acp-21-4939-2021, 2021.

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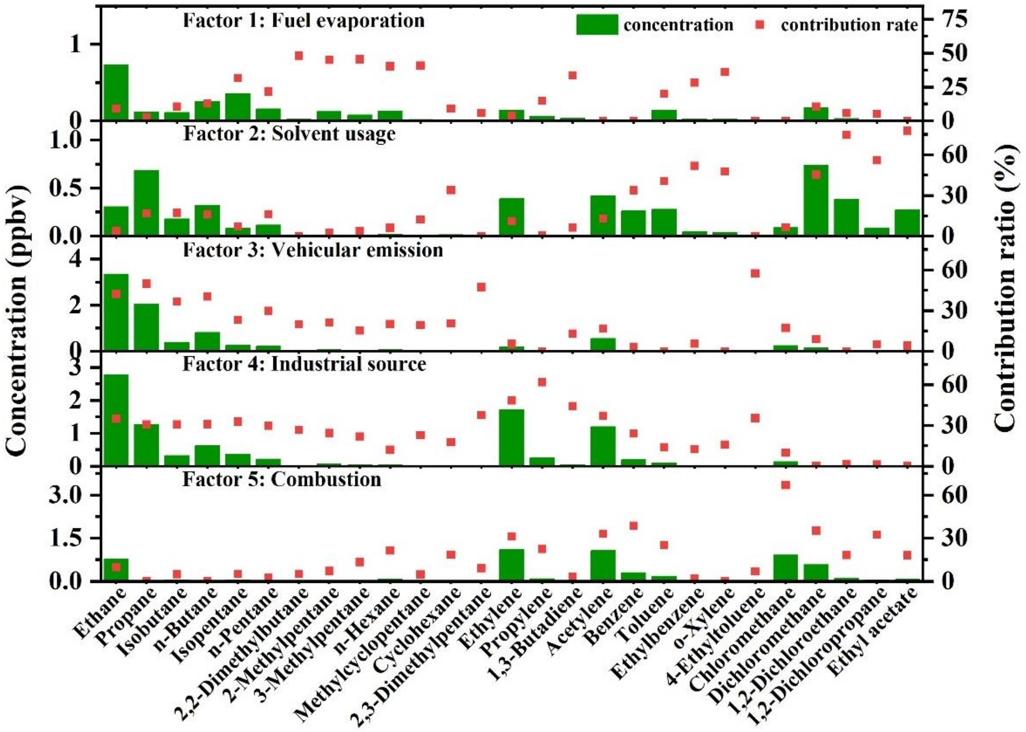
5、The authors should analyze differences in PMF factors/source profiles during and post-Omicron lockdown days and between high pollution and clean days.

Response**:** The following additions are based on your comments. Figure 6 compares the differences in PMF factor/source profiles during the peak of Omicron infection with those during the recovery phase after the peak, as well as between contaminated and clean days.

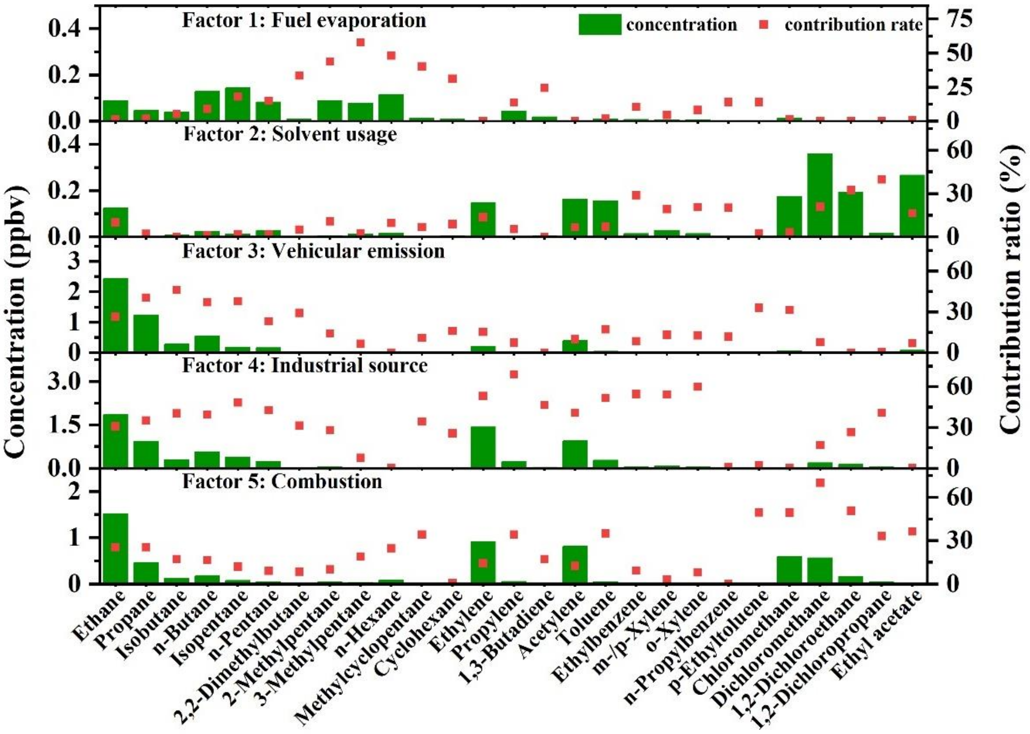
The screening of observed VOC species and their inclusion into PMF model, followed by the application of the random seed approach for the examination of 20 baseline runs per process using 3-6 factors, resulted in the selection of 5 factors from the 20 baseline runs to represent the final results of 5 factors. These five factors included: (1) Fuel evaporation; (2) Solvent usage; (3) Vehicular emission; (4) Industrial source; and (5) Combustion (Figure 6). These 5 factors have been commonly reported before, e.g., in Shijiazhuang, northern China (Guan et al, 2023) and in Beijing (Cui et al., 2022). It is worth noting that there are two y-axes in Figure 6: the left side represents the concentration of VOCs in units of ppbv, and the right side represents the percentage of specific VOCs within that factor. Additionally, the concentration scales of some figures also differ. We present the concentrations of the five main VOCs in all five factors in Table 2. Ethane (vehicular emission), 2-methylpentane (fuel evaporation), benzene (industry source), chloromethane (combustion), and ethyl acetate (solvent usage) were selected as tracers for five sources.

Concentrations of most species were significantly higher during the recovery period than during the infection period. The representative pollution processes in both periods showed the same results as well, with a 79% higher concentration of TVOCs in Case 2 (65.1 ppbv) compared to Case 1 (36.3 ppbv) (Figure 7). While in Case 1 industry was the dominant source of VOCs, by Case 2 motorized sources reached a concentration value of 21.2 ppbv, accounting for 33% of the observed VOCs, and became the dominant source of emissions. This is consistent with the fact that people's mobility activities have increased after the epidemic has entered the recovery period. As a group of VOCs species with the highest concentration share, ethane and propane contributed more to the clean day motor vehicle sources than other processes, which also resulted in a 34% clean day motor vehicle source share.

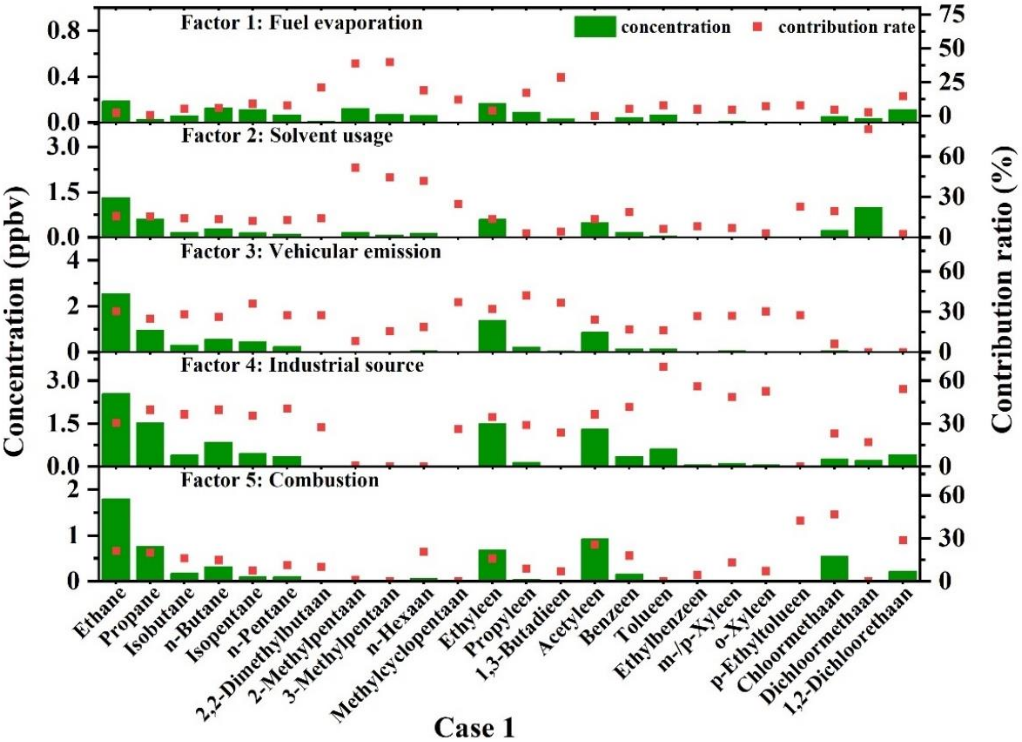
It can be anticipated that certain sources may overlap, meaning that some VOCs emissions undoubtedly come from multiple sources. Taking ethane in Case 1 as an example, the largest source is vehicle exhaust emissions (2.55 ppbv, 30%), followed by industrial emissions (2.54 ppbv, 30%), combustion sources (1.80 ppbv, 21%), solvent usage (1.32 ppbv, 16%), and fuel evaporation (0.19 ppbv, 2%). The total was 8.4 ppbv, which is somewhat different from the observed values (Table 2). At the same time, there are cases where the observed values are perfectly matched, e.g., for 2-methylpentane in the whole process. Similarly, this discrepancy is due to the simple fact that the PMF model cannot fully explain the observed values at 100%.



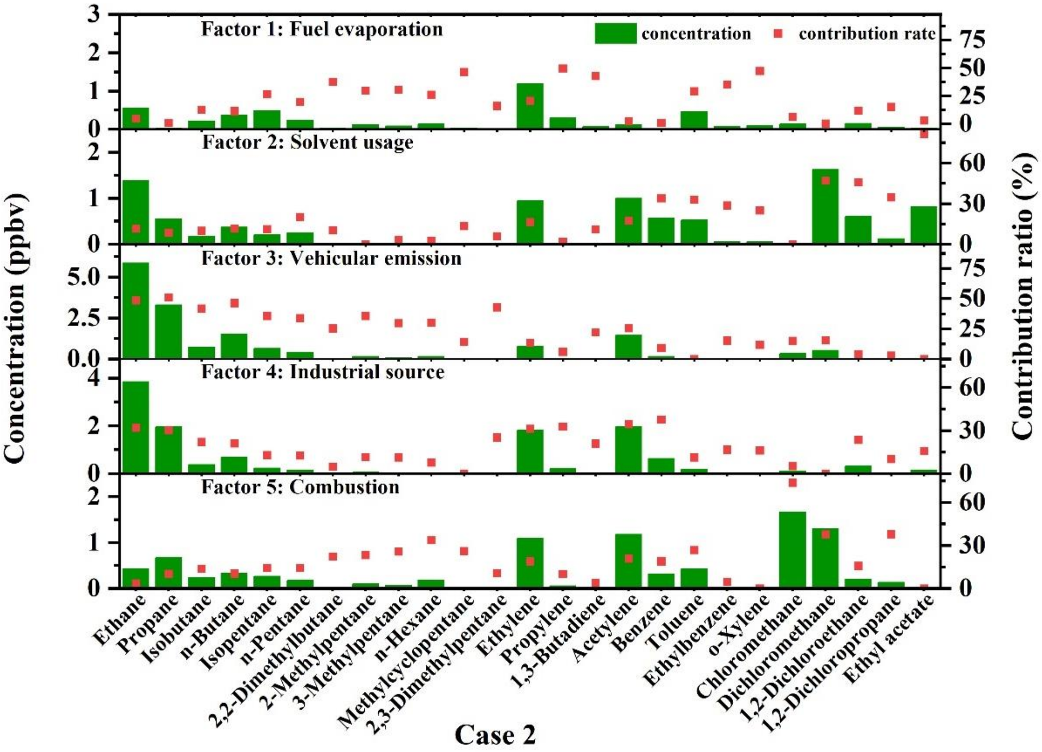
recovery period



infection period



Case 1


Factor 1: Fuel evaporation
Factor 2: Solvent usage
Factor 3: Vehicular emission
Factor 4: Industrial source
Factor 5: Combustion
concentration
contribution rate
Concentration (ppbv)
Contribution ratio (%)
Ethane
Propane
Isobutane
n-Butane
Isopentane
n-Pentane
2,2-Dimethylbutane
2-Methylpentane
3-Methylpentane
n-Hexane
Methylcyclopentane
2,3-Dimethylpentane
Ethylene
Propylene
1,3-Butadiene
Acetylene
Benzene
Toluene
Ethylbenzene
o-Xylene
Chloromethane
Dichloromethane
1,2-Dichloroethane
1,2-Dichloropropane
Ethyl acetate
Case 2

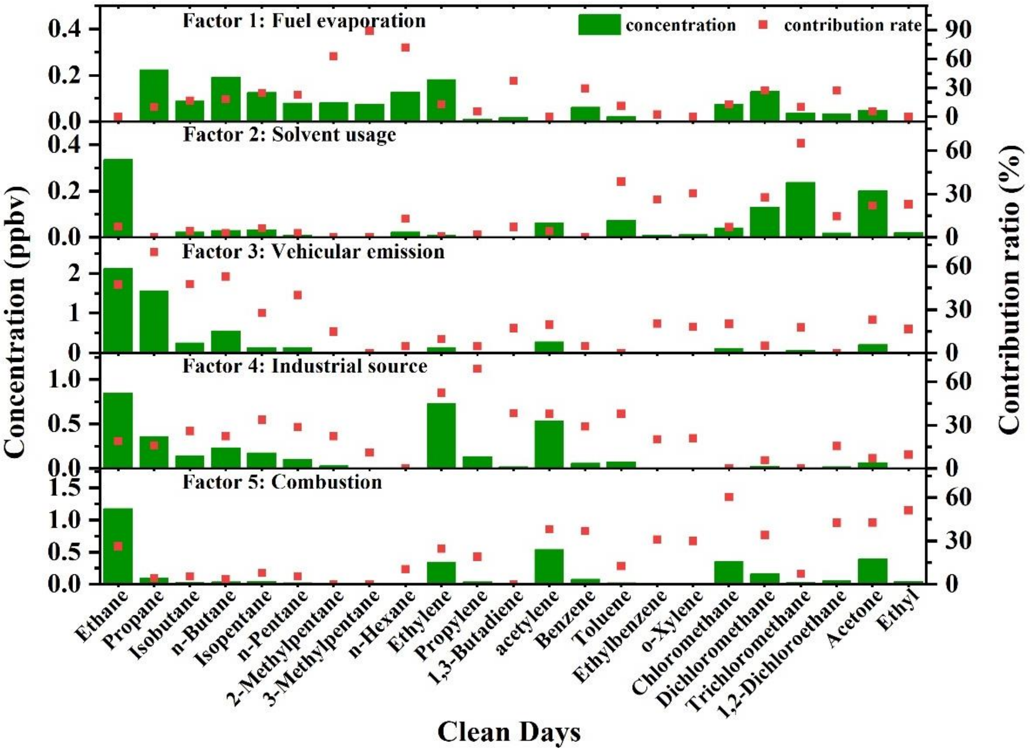

Factor 1: Fuel evaporation
Factor 2: Solvent usage
Factor 3: Vehicular emission
Factor 4: Industrial source
Factor 5: Combustion
concentration
contribution rate
Concentration (ppbv)
Contribution ratio (%)
Ethane
Propane
Isobutane
n-Butane
Isopentane
n-Pentane
2-Methylpentane
3-Methylpentane
n-Hexane
Ethylene
Propylene
1,3-Butadiene
acetylene
Benzene
Toluene
Ethylbenzene
o-Xylene
Chloromethane
Dichloromethane
Trichloromethane
1,2-Dichloroethane
Acetone
Ethyl
Clean Days

Figure 6. Infection period, recovery period, high pollution events, and clean days PMF source analysis

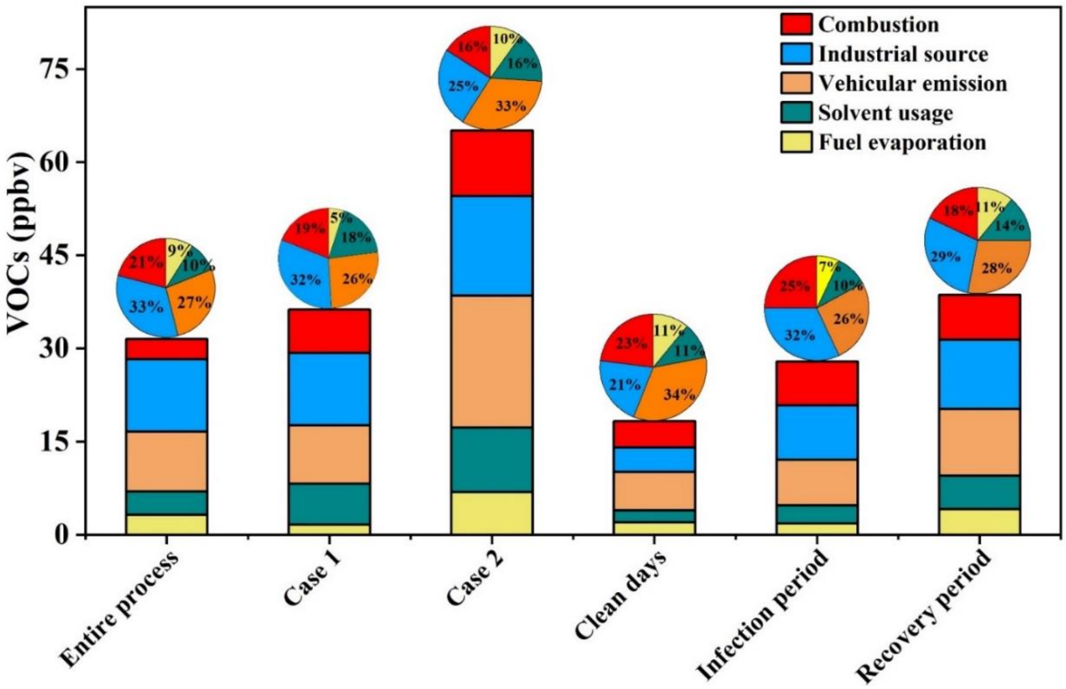


Figure 7. Contribution of each source to VOCs for different processes

Table 2. Concentrations of important tracer substances in different processes (ppbv) (observations in parentheses, red text indicates the corresponding source concentration of the substance)

	ethane						**2-Methylpentane**					
Source	**Infection**	**Recovery**	**Entire**	**Case 1**	**Case 2**	**Clean**	**Infection**	**Recovery**	**Entire**	**Case 1**	**Case 2**	**Clean**
Factor 1 Fuel evaporation	0.09	0.73	0.41	0.19	0.55	0	**0.09**	**0.12**	**0.10**	**0.12**	**0.13**	**0.08**
Factor 2 Solvent usage	0.14	0.30	0	1.32	1.38	0.34	0.01	0.01	0.01	0.16	0	0
Factor 3 Vehicle emission	**2.39**	**3.35**	**2.91**	**2.55**	**5.85**	**2.12**	0.02	0.06	0.06	0.03	0.16	0.02
Factor 4 Industrial source	1.83	2.77	2.5	2.54	3.84	0.85	0.06	0.07	0.07	0.01	0.05	0.03
Factor 5 Combustion	1.55	0.76	1.36	1.80	0.43	1.17	0.04	0.02	0	0	0.10	0
sum	**6.00 (6.80)**	**7.91 (7.81)**	**7.18 (6.80)**	**8.40 (10.06)**	**12.05 (12.17)**	**4.48 (4.30)**	**0.22 (0.25)**	**0.28 (0.26)**	**0.24 (0.24)**	**0.32 (0.37)**	**0.44 (0.45)**	**0.13 (0.14)**
	benzene						**methyl chloride**					
Factor 1 Fuel evaporation	0.02	0	0.06	0.04	0.01	0.06	0.02	0	0.08	0.05	0.14	0.07
Factor 2 Solvent usage	0.13	0.26	0.16	0.17	0.57	0	0.18	0.09	0	0.23	0	0.04
Factor 3 Vehicle emission	0.01	0.03	0.07	0.15	0.15	0.01	0.06	0.23	0.06	0.07	0.34	0.12
Factor 4 Industrial source	**0.16**	**0.19**	**0.09**	**0.36**	**0.63**	**0.06**	0	0.13	0.30	0.27	0.11	0
Factor 5 Combustion	0.24	0.3	0.33	0.16	0.31	0.08	**0.58**	**0.91**	**0.72**	**0.55**	**1.67**	**0.35**
sum	**0.56 (0.65)**	**0.78 (0.83)**	**0.71 (0.69)**	**0.88 (1.10)**	**1.67 (1.74)**	**0.21 (0.20)**	**0.84 (0.99)**	**1.36 (1.43)**	**1.16 (1.14)**	**1.17 (1.37)**	**2.26 (2.35)**	**0.58 (0.54)**
	ethyl acetate											
	Infection	**Recovery**	**Entire**	**Case 1**	**Case 2**	**Clean**						

Factor 1 Fuel evaporation	0	0	0.01	0.02	0.03	0						
Factor 2 Solvent usage	**0.27**	**0.27**	**0.72**	**0.63**	**0.80**	**0.02**						
Factor 3 Vehicle emission	0.08	0.01	0.03	0.01	0	0.01						
Factor 4 Industrial source	0	0	0.02	0.08	0.16	0.01						
Factor 5 Combustion	0	0.06	0.01	0.01	0	0.04						
sum	**0.35 (0.45)**	**0.34 (0.40)**	**0.79 (0.68)**	**0.75 (0.81)**	**0.99 (1.09)**	**0.08 (0.06)**						

References:

Cui, L., Wu, D., Wang, S., Xu, Q., Hu, R., and Hao, J.: Measurement report: Ambient volatile organic compound (VOC) pollution in urban Beijing: characteristics, sources, and implications for pollution control, Atmospheric Chemistry and Physics, 22, 11931-11944, https://doi.org/10.5194/acp-22-11931-2022, 2022.

Guan, Y., Liu, X., Zheng, Z., Dai, Y., Du, G., Han, J., Hou, L. a., and Duan, E.: Summer O3 pollution cycle characteristics and VOCs sources in a central city of Beijing-Tianjin-Hebei area, China, Environmental Pollution, 323, 121293, https://doi.org/10.1016/j.envpol.2023.121293, 2023.