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. To facilitate your review, the comments are in black, and the responses are in blue.

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 impacts 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.

[c]Overall, 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.

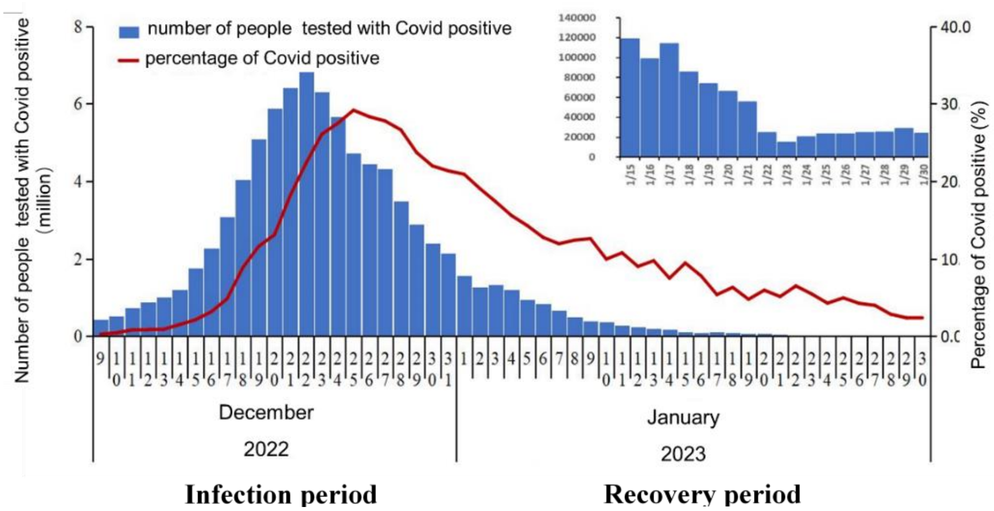

Figure 1. Trend of Omicron infection in China from 9 Dec. 2022 to 1 Jan. 2023 (CCDCP, 2023)

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 251-277)

Line 251-277: 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	

References:

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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).

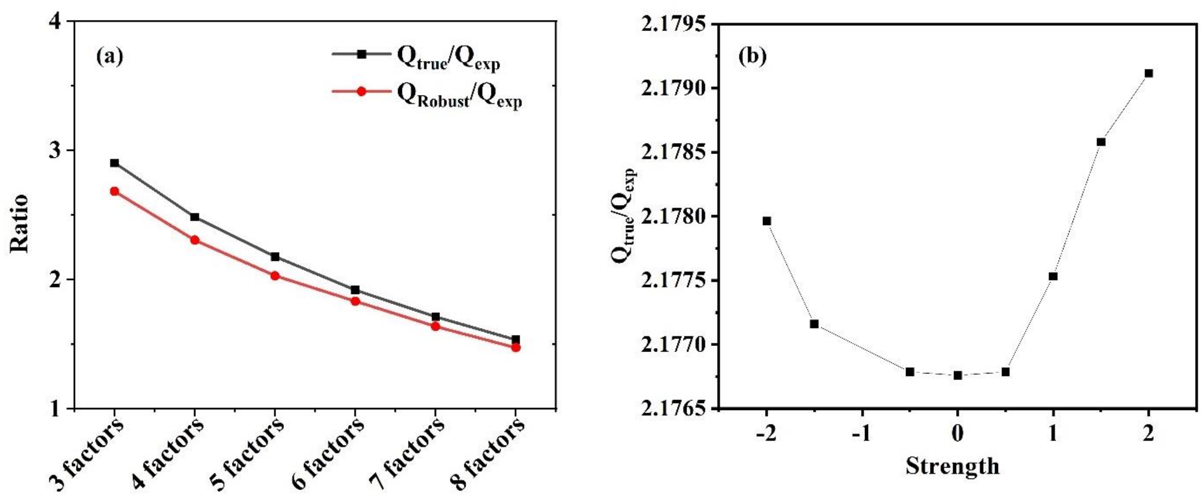

Figure 2. (a) The $Q_{true}/Q_{expected}$ ratios in different solutions; (b) the $Q_{true}/Q_{expected}$ ratio for different Fpeak value solutions.

[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).

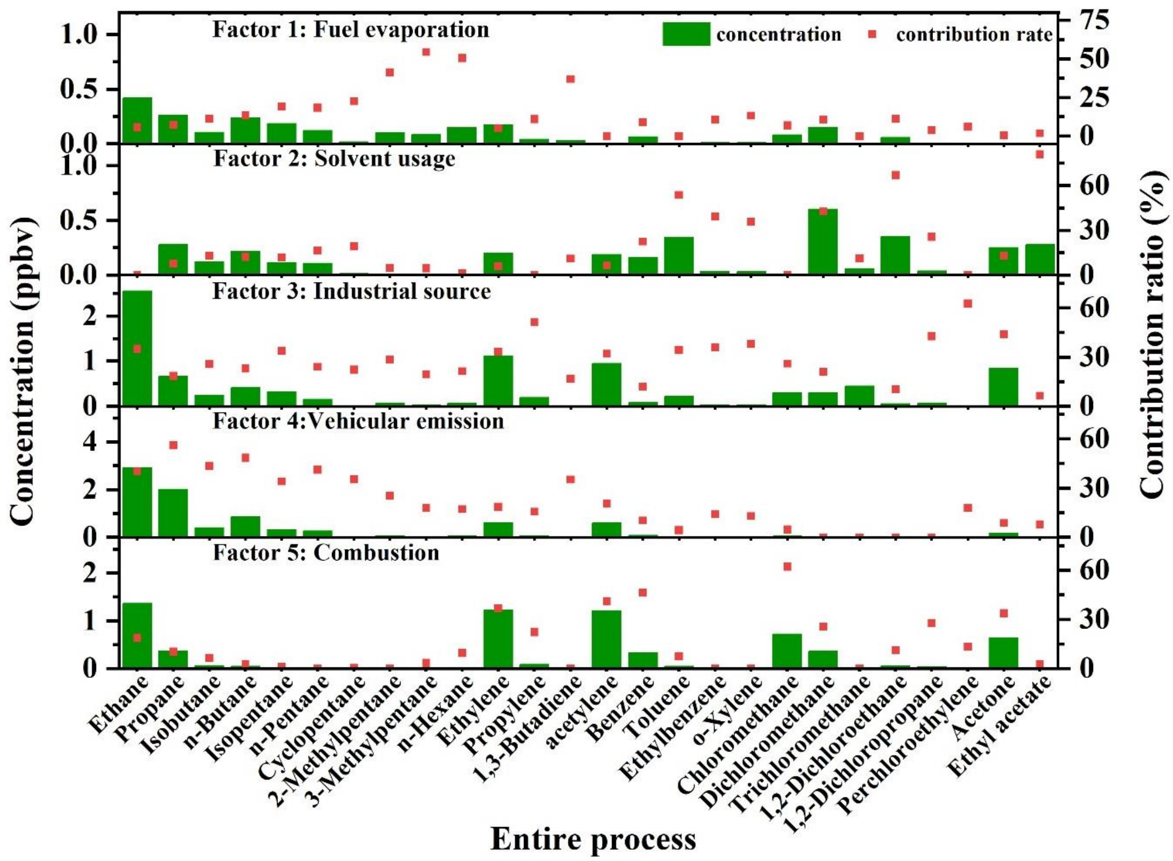


Figure 3. Concentration of VOC species in each factor and contribution

to each source

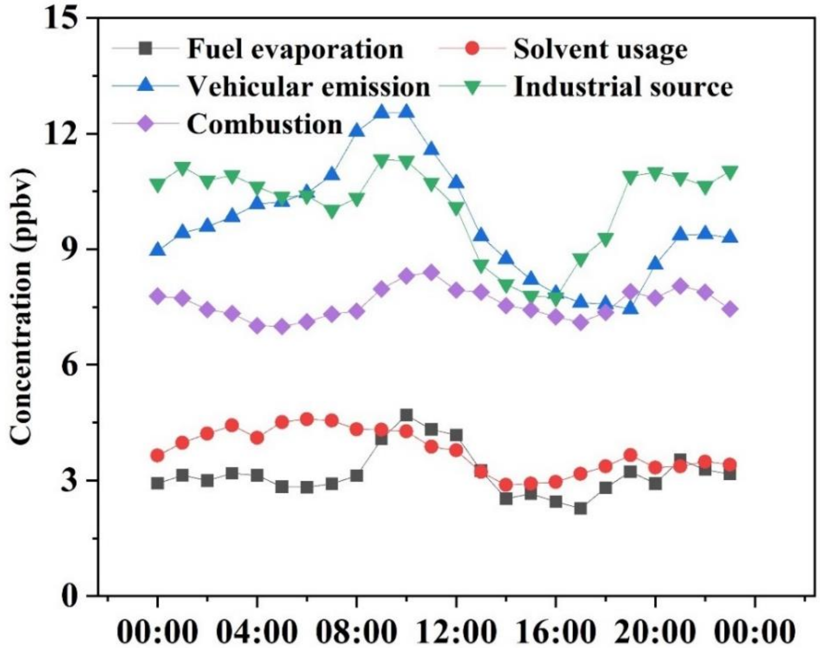
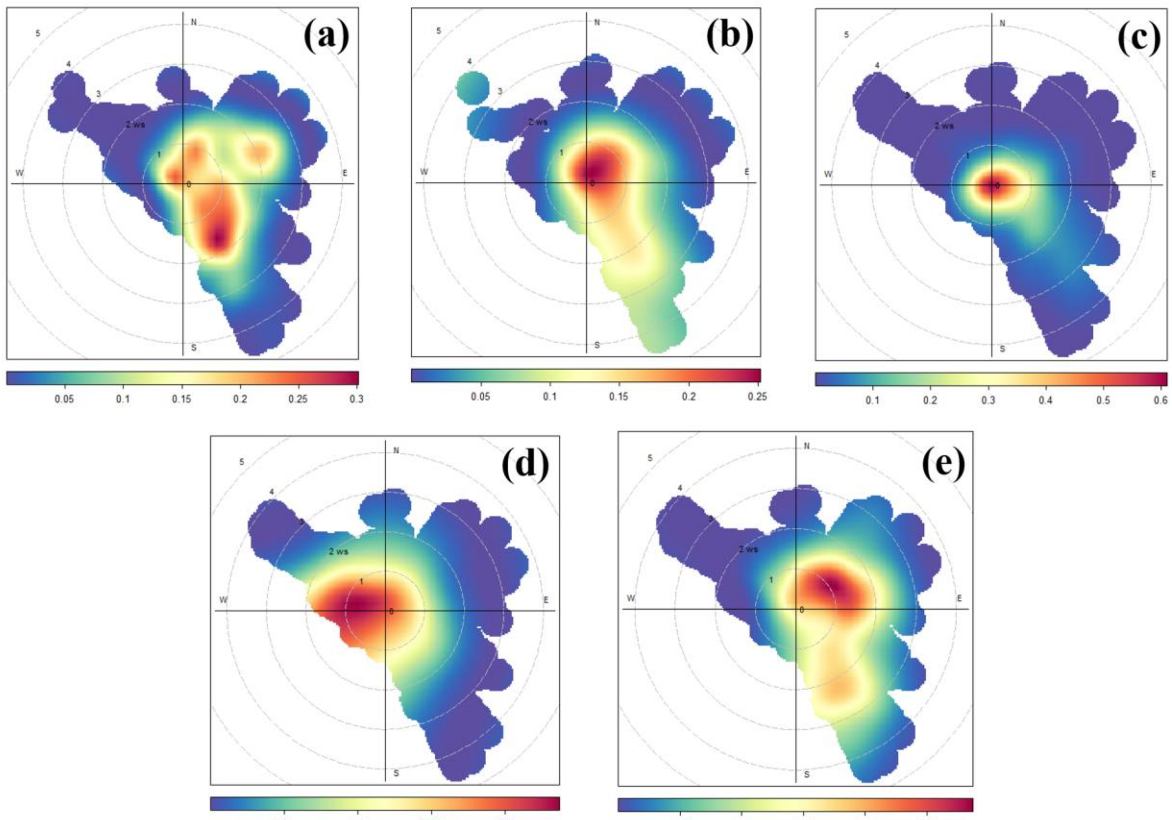


Figure 4. Characteristics of daily changes in different sources obtained using the PMF model



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

References:

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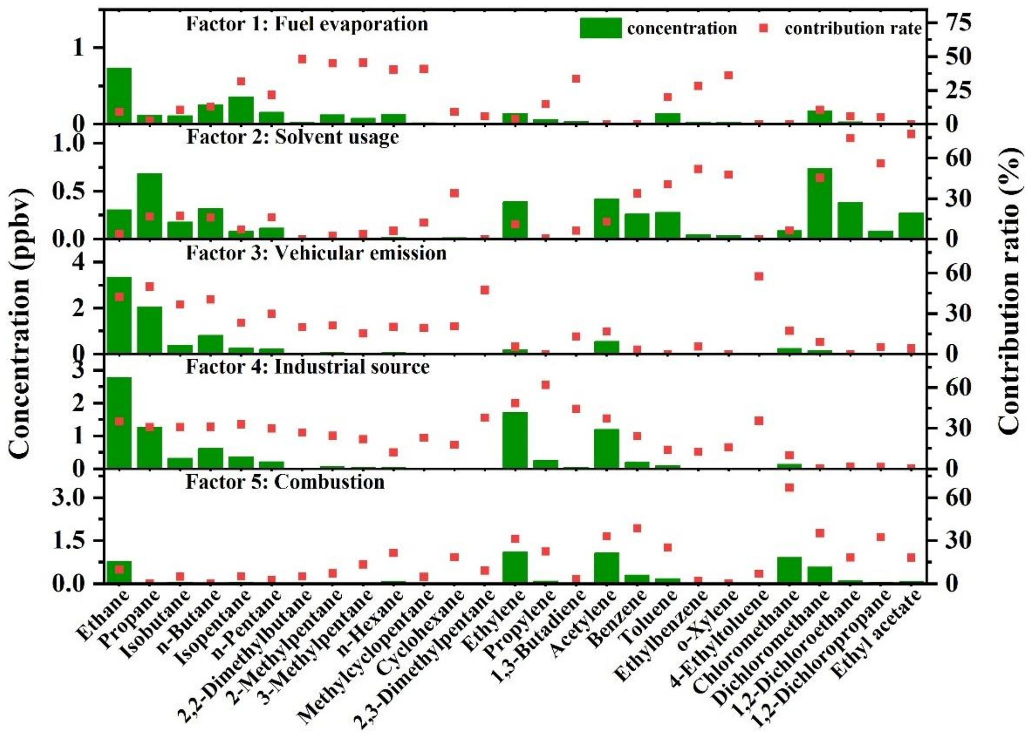
5、The authors should analyze differences in PMF factors/source profiles during and post-Omicron lockdown days and between high pollution and clean days.

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.

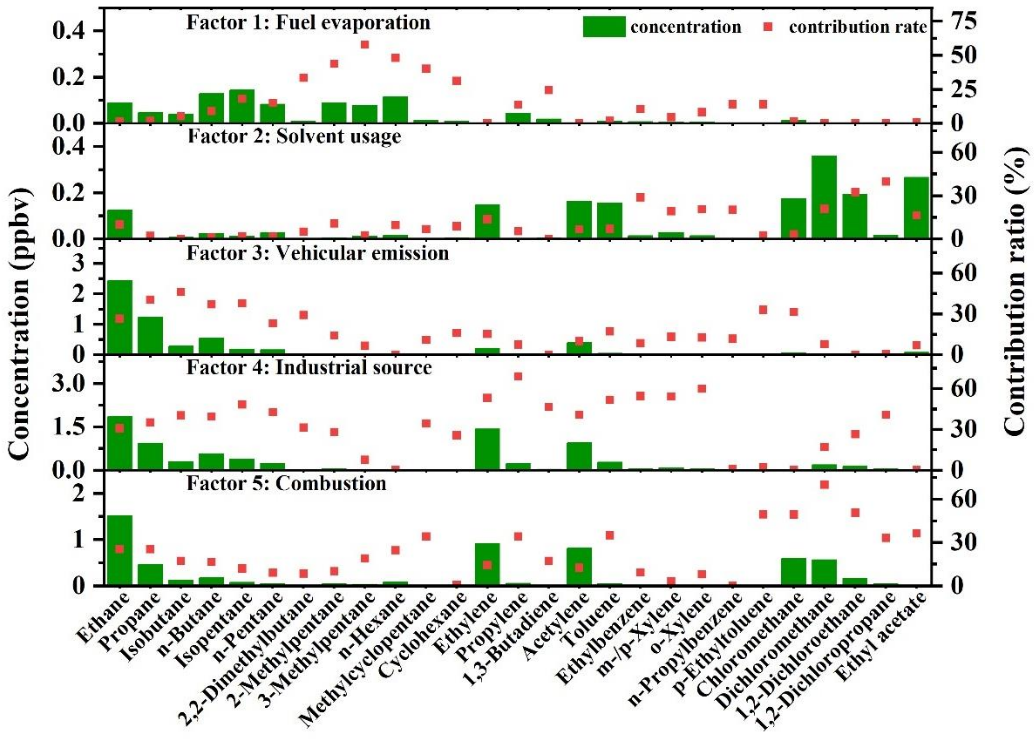
The screening of observed VOC species and their inclusion into PMF model, followed by the application of the random seed approach for the examination of 20 baseline runs per process using 3-6 factors, resulted in the selection of 5 factors from the 20 baseline runs to represent the final results of 5 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.

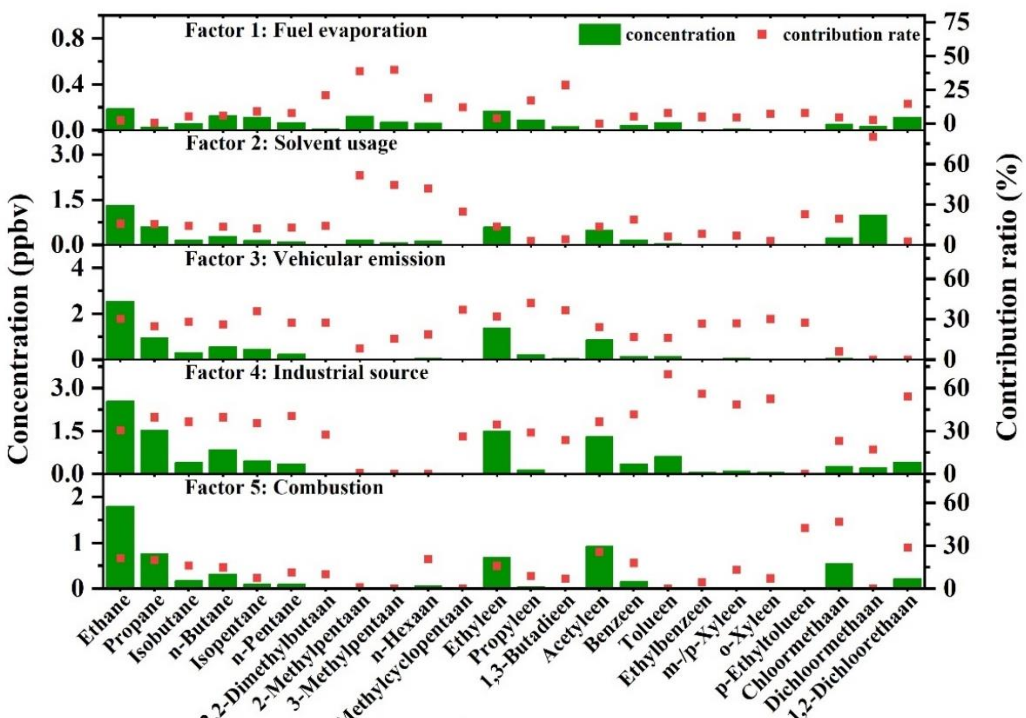
It can be anticipated that certain sources may overlap, meaning that some VOCs emissions undoubtedly come from multiple sources. Taking ethane in Case 1 as an example, the largest source is vehicle exhaust emissions (2.55 ppbv, 30%), followed by industrial emissions (2.54 ppbv, 30%), combustion sources (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%.



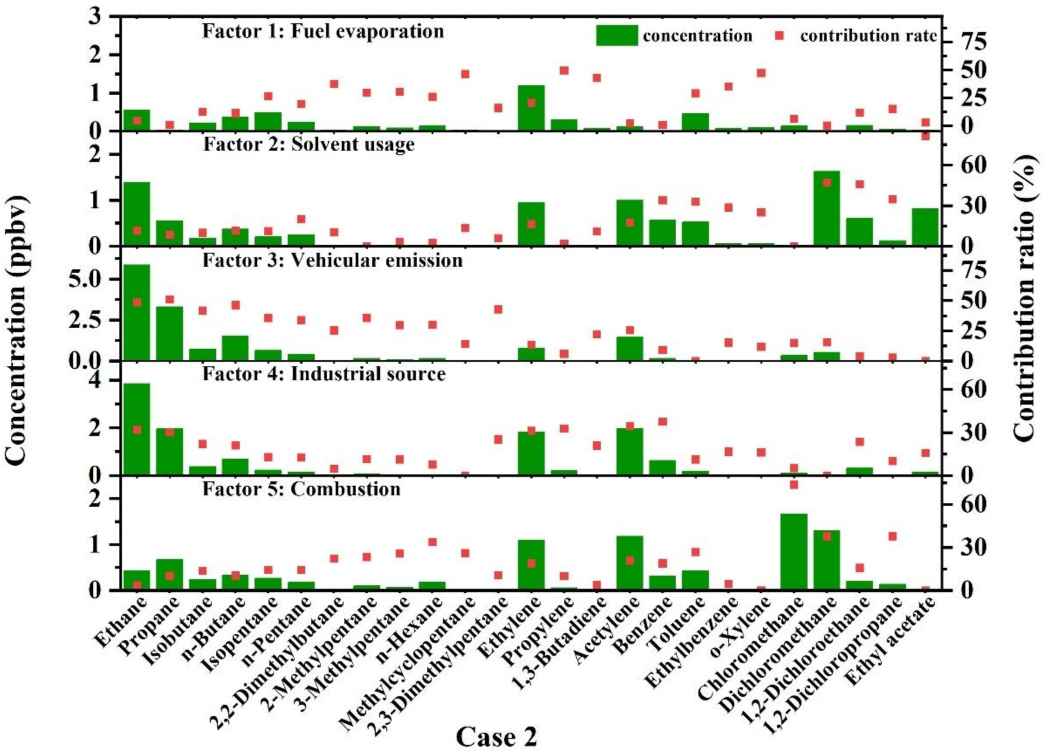
recovery period



infection period



Case 1


Factor 1: Fuel evaporation
concentration
contribution rate
Factor 2: Solvent usage
Factor 3: Vehicular emission
Factor 4: Industrial source
Factor 5: Combustion
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

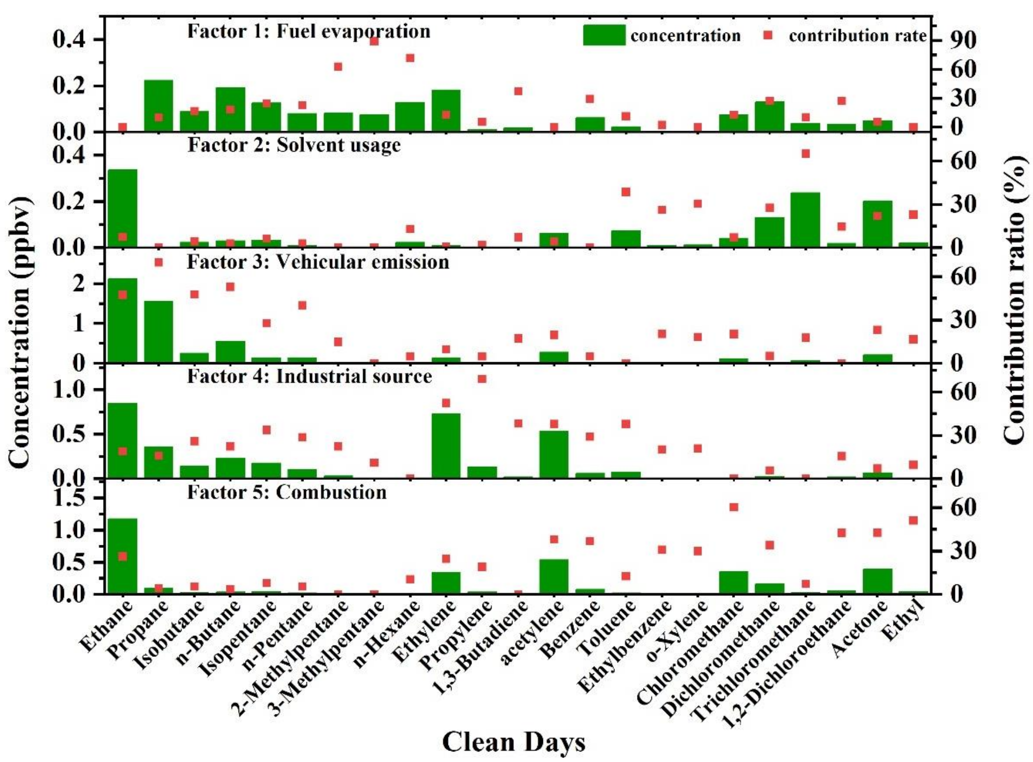

Factor 1: Fuel evaporation
concentration
contribution rate
Factor 2: Solvent usage
Factor 3: Vehicular emission
Factor 4: Industrial source
Factor 5: Combustion
Concentration (ppbv)
Contribution ratio (%)
Ethane
Propane
Isobutane
n-Butane
Isopentane
n-Pentane
2-Methylpentane
3-Methylpentane
n-Hexane
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

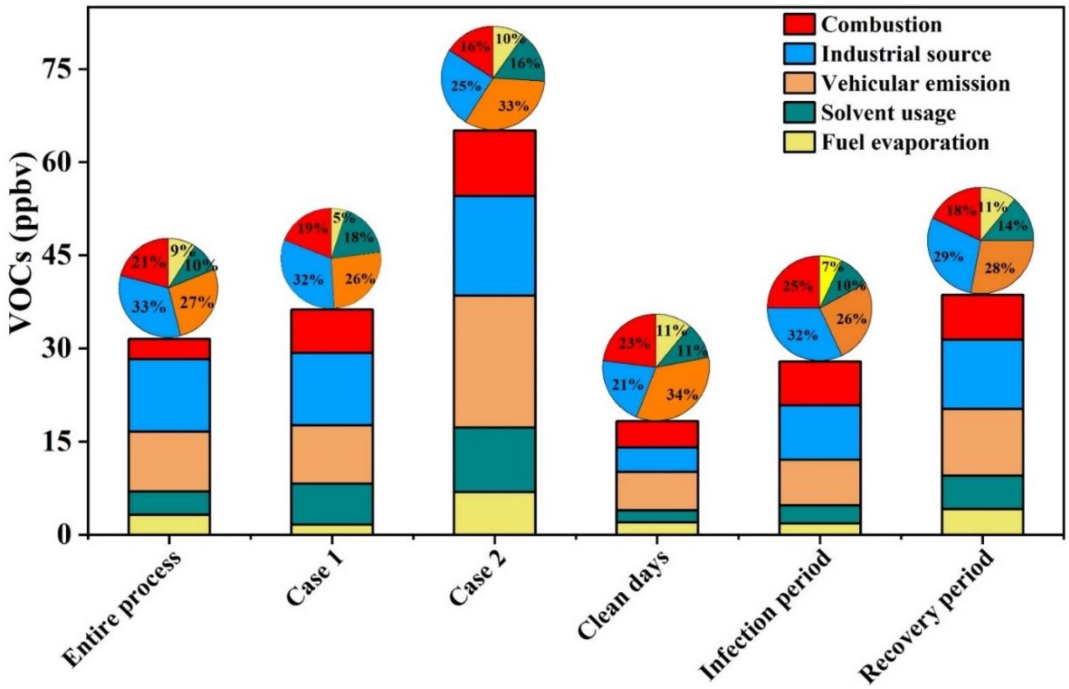


Figure 7. Contribution of each source to VOCs for different processes

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	0.02	0	0.06	0.04	0.01	0.06	0.02	0	0.08	0.05	0.14	0.07

Fuel evaporation												
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)**						

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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.