The variations of VOCs based on the policy change of Omicron in traffic-hub city Zhengzhou

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Abstract: Online volatile organic compounds (VOCs) were monitored before and after the Omicron policy change at an urban site in polluted Zhengzhou from December 1, 2022, to January 31, 2023. The characteristics and sources of VOCs were explored. The daily average concentration of $PM_{2.5}$ and total VOCs (TVOCs) ranged from 53.5 to 239.4 μg/m^3 and from 15.6 to 57.1 ppbv with an average value of 111.5 ± 45.1 μg/m^3 and 36.1 ± 21.0 ppbv, respectively during the entire period. The values of $PM_{2.5}$ and TVOCs in Case 2 (pollution episode after the abolishment of "Nucleic Acid Screening Measures for all staff" policy) were 1.3 and 1.8 times of the values in the Case 1 (pollution episode during "Nucleic Acid Screening Measures for all staff" policy). The concentration of TVOCs in Case 1 and Case 2 were 48.4 ± 20.4 and 67.6 ± 19.6 ppbv, respectively, increased by 63% and 188% compared with values during clean days. Alkanes were found to be the most abundant compounds during the entire period. Equivalent volume contribution of halogenated hydrocarbon and oxygenated VOCs (15%) were found the most in Case 2, followed by alkenes (10%). Though the volume contributions of aromatics were the lowest (6% in Case 1 and 7% in Case 2), the highest increasing ratio was found from clean days to polluted episodes. Positive Matrix Factor model results showed that the main source of VOCs during the observation period was industrial emissions, which accounted for 32% of the TVOCs, followed by vehicular emission (27%) and combustion (21%). In Case 1, industrial emissions constituted the largest contributor, accounting for 32% of the total VOCs. In Case 2, however, the share of vehicular emission source increased to 33%, becoming the primary source of VOCs. Secondary organic aerosol formation potential (SOAP) values were 37.6 and 65.6 μg/m^3 in Case 1 and Case 2, respectively. In Case 1, industrial source accounted for the overwhelming majority (63%, 23.8 μg/m^3), while vehicular source, as the second largest source, accounted for only 18%. In Case 2, the distribution of contributions is more uniform, with solvent usage source and fuel evaporation source accounting for the majority of SOAP, at 32% (20.9 μg/m³) and 26% (16.8 μg/m³), respectively. Industrial source and solvent usage continue to be the main contributors to SOAP on clean days. It is crucial to prioritize the regulation of emissions from industrial and solvent-using sectors as a means of curbing $PM_{2.5}$ pollution in Zhengzhou. Additionally, it is imperative to consider the impact of rising vehicular emissions on air quality.

Keywords: Volatile organic compounds; Pollution episode; Source apportionment;

1. Introduction

Volatile organic compounds (VOCs) in the atmosphere have high reactivity and can react with nitrogen oxides (NO_x) to form a series of secondary pollutants such as ozone (O_3) and secondary organic aerosol (SOA), resulting in regional air pollution (Li et al., 2019; Hui et al., 2020). The problem of O_3 pollution has been plaguing major urban agglomerations in China (Zheng et al., 2010; Li et al., 2014; Wang et al., 2017). SOA is an important component of fine particulate matter ($PM_{2.5}$) and contributes significantly to haze pollution (Liu et al., 2019). $PM_{2.5}$ remains the most significant air pollutant in many Chinese cities for years (Shao et al., 2016; Wu et al., 2016). In addition, VOCs, represented by the benzene homologues, can cause damage to kidneys, liver, and nervous system of humans when they enter the body (Zhang et al., 2018).

Studies have shown that the most common VOC components in China are alkanes, olefins, aromatic hydrocarbons, oxygenated VOCs (OVOCs), and halogenated hydrocarbons, among which alkanes are the most abundant species (Liu et al., 2020; Zhang et al., 2021a). VOCs in the atmosphere have a wide range of sources, and VOCs in different regions are affected by multiple factors such as local geography, climate, and human activities (Mu et al., 2023; Zou et al., 2023). The above reasons lead to significant regional and seasonal differences in the characteristics of VOCs (Song et al., 2021). For example, the annual average concentration of VOCs in the coastal background area of the Pearl River Delta is 9.3 ppbv. The seasonal variation trend of VOCs is high in autumn and winter and low in summer (Yun et al., 2021). In contrast, the average VOC concentration in autumn and winter in Beijing was 22.6 ± 12.6 ppbv, and the VOC concentration in the winter heating period was twice that in the autumn non-heating period (Niu et al., 2022).

Moreover, the sources of VOC components in different regions are also related to the local industrial structure and living habits. In rural areas of North China Plain in winter, it is found that the SOA formation potential (SOAP) of VOCs under low NO_x conditions is significantly higher than that under high NO_x conditions, and the increase of aromatic hydrocarbon emissions caused by coal combustion is the main reason for the higher SOAP in winter (Zhang et al., 2020). Li et al. (2022) found that the average increased concentration of acetylene was 4.8 times from autumn to winter in the Guanzhong Plain, indicating that fuel combustion during the heating period in winter has a significant impact on the composition of VOCs. In contrast, continuous observations conducted by Zhou et al. (2022) in the suburbs of Dongguan in summer found that industrial solvent usage, liquefied petroleum gas (LPG) and oil and gas volatilization were the main sources of VOCs. The results highlighted a wide variation of characteristics, sources and chemical reactions of VOCs in the atmosphere, thus it is necessary to investigate VOCs in different cities when formulating control measures.

Zhengzhou, as the capital of Henan Province, is an important transportation hub and economic center in the Central Plains region. Zhengzhou is currently facing significant air pollution problems, with the Air Quality Index at the bottom of the national ranking of 168 cities for many years. In January 2023, for example, the number of polluted days with $PM_{2.5}$ as the primary pollutant was 17, and the daily average value of $PM_{2.5}$ reached a maximum of 298 μg/m^3 (https://www.aqistudy.cn/historydata/daydata.php?city=%E9%83%91%E5%B7%9E&month=202301, Accessed Jan 2024), which is almost 300% higher than the Chinese daily average standard (grade II, 75 μg/m^3). The studies of VOCs were carried out in Zhengzhou in recent years, which focused on the characteristics and sources of VOCs during pollution episodes (Lai et al., 2024) or before the coronavirus epidemic outbreak (Li et al., 2020; Zhang et al., 2021b). While some atmospheric VOCs 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 toluene, benzene, m/p-xylene and ethylbenzene 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. In addition, 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.

In this study, a continuous online observation of VOCs in polluted winter at an urban site was carried out, which covered the abolishment of lockdown measures in Zhengzhou. China lifted the zero-COVID strategies, notably by announcing the '10 measures' about the optimization of COVID-19 rules on 7 December 2022 (http://www.news.cn/politics/2022-12/07/c_1129189285.htm, Accessed Jan 2024), which led to significant changes in social activities. After that, China experiences a nationwide outbreak of COVID-19. 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 (Petersen et al., 2022; Merino et al., 2023). A two-month-long lockdown measure was applied to 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 at 8 October in 2022. People are basically homebound after the lifting of the lockdown policy due to infection or fear of infection of Omicron. The resumption of normal production and livelihoods was based on the assumption of herd immunization. This change is worth exploring in terms of its impact on transportation and industrial production emissions. Therefore, the characteristics and variations of VOCs during different periods were investigated to assess their impact on the formation of SOA and to provide data support for future pollution control policies in Zhengzhou.

2. Materials and methods

2.1 Sample collection and Chemical analysis

The online VOCs observation station is located on the roof of the Zhengzhou Environmental Protection Monitoring Center, which is in the urban area. The sampling site is close to main roads on three sides (150 m away from Funiu Road on the east side, 200 m away from Qinling Road on the west side, and connected to Zhongyuan Road on the south side), and surrounded by residential areas and commercial areas without other large nearby stationary sources. The sampling period for this study was from December 1, 2022, to January 31, 2023, which is always the most polluted period in the entire year. Apart from a brief occurrence of rain and snow on December 25, the sampling days were either sunny or cloudy. The wind speed (WS), temperature (Temp) and relative humidity (RH) during this period were 1.3 ± 0.9 m/s, 5.3 ± 3.2 ℃ and 38.9 ± 19.0%), respectively, similar to the values observed in previous years in Zhengzhou. It is interesting to point out that the sampling period in the present study covered the entire infection period of Omicron in Zhengzhou, including the phase of surge in infected population (Infection period, from 2022.12.01 to 2022.12.31) and restoration of production and livelihood phase (Recovery period, from 2023.1.1 to 2023.1.31 in 2023) (Fig. S1, Chinese Center for Disease Control and Prevention, 2023).

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, 35 halocarbons, 12 OVOCs, 1 alkyne (acetylene), and 1 sulfide (CS_2) with a total of 106 compounds.

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 over 90% observed data greater than the detection limit, and 34 had more than 50% observed data greater than the detection limit.

Simultaneous observations at the same site were also carried out for particulate matter ($PM_{2.5}$, PM_{10}), other trace gases (carbon monoxide (CO), O_3, nitric oxide (NO), nitrogen dioxide (NO_2)), and meteorological data (Temperature, RH, WS, and wind direction (WD)) based on 1 h resolution.

2.2 Positive Matrix Factorization (PMF) model

EPA PMF5.0 model was used for the quantitative source analysis of VOCs (Norris et al., 2014). The principles and methods have been described in detail in previous studies (Mozaffar et al., 2020; Zhang et al., 2021b). The decomposition of the PMF mass balance equations is simplified as follows (Norris et al., 2014):

$$x_{ij} = \sum_{k=1}^{p} g_{ik}\, f_{kj} + e_{ij} \qquad (1)$$

where x_{ij} is the mass concentration of species j measured in sample i; g_{ik} is the contribution of factor k to the sample i ; f_{kj} represents the content of the jth species in factor k; e_{ij} is the residual of species j in sample i; p represents the number of factors. The fitting objective of the PMF model is to minimize the function Q to obtain the factor contributions and contours. The formula for Q is given in Eq. (2):

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}}\right]^2 \qquad (2)$$

where n and m denote the number of samples and VOC species, respectively.

Concentrations and uncertainty data are required for the PMF model. In this study, the median concentration of a given species is used to replace missing values with an uncertainty of four times of the median values; data less than the Method Detection Limit (MDL) were replaced with half the MDL, with an uncertainty of 5/6 of the MDL; and the uncertainty for values greater than the MDL was calculated using Eq. (3). In Eq. (3), EF is error fraction, expressed as the precision of VOCs species, and the setting range can be adjusted from 5 to 20% according to the concentration difference (Buzcu et al., 2006; Song et al., 2007); and c_{ij} is the concentration of species j in sample i:

$$U_{ij} = \sqrt{\left(EF \times c_{ij}\right)^2 + (0.5 \times MDL)^2} \tag{3}$$

when the concentration of VOCs in the species is less than the value of the detection limit U_{ij} is calculated using Eq. (4):

$$U_{ij} = \left(\frac{5}{6}\right) MDL \tag{4}$$

VOC species and concentration input into PMF were carefully selected to ensure the accuracy of the PMF results. Species were excluded when over 25% of the samples were missing or concentrations values were below the MDL (Gao et al., 2018); VOCs with a short lifetime in the atmosphere were also excluded unless they are source-relative species (Zhang et al., 2014; Shao et al., 2016). After that, retained VOC species were categorized according to the signal-to-noise ratio (S/N) with S/N < 0.2 species categorized as bad, 0.2 < S/N < 2 species categorized as weak; and S/N > 2 species categorized as strong (Shao et al., 2016).

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. Q_{true}/Q_{exp} is lowest when Fpeak = 0, so we chose the PMF results for that case (Fig. S2a). After examining 3-8 factors, 20 base runs with 5 factors eventually selected to represent final result. We provide an explanation of factor selection in the supplementary materials. Fig. S2(b) 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.

2.3 SOA generation potential

The contributions of VOC species to SOAP were calculated based on the toluene weighted mass contributions (TMC) method (Derwent et al., 2010). The methodology for calculating SOAP is as follows:

$$SOAPF_i = \frac{VOCs\ component\ i\ to\ SOA\ mass\ concentration\ increments}{Toluene\ to\ SOA\ mass\ concentration\ increment} \times 100 \quad (5)$$

$SOAPF_i$ for each VOC is taken from the literature (Derwent et al., 2010). The SOAP was estimated by multiplying the $SOAPF_i$ value by the concentration of individual VOC species. The SOAP calculations through each VOC are as follows:

$$SOAP = \sum E_i \times SOAPF_i \quad (6)$$

E_i is the concentration of species *i*.

3. Results and discussion

3.1 Overview of variation in pollutants and meteorological parameters

Figure 1 shows the time series of meteorological parameters, TVOCs, O_3, NO_x, SO_2, CO and $PM_{2.5}$ during the observed periods. Low WS and Temperature were found with an average value of 1.3 ± 0.6 m/s and 5.0 ± 2.5 °C, respectively, during the entire period, comparable with observations at the same site in 2021 (Lai et al., 2024). A total of 62 days of valid data was acquired with the daily average concentration of $PM_{2.5}$ ranging from 53 to 239 μg/m^3, with the average value of 111 ± 45 μg/m^3. The concentration of TVOCs ranged from 15.6 to 57.1 ppbv with an average of 36.1 ± 21.0 ppbv, higher than the same period in last year (27.9 ± 12.7 ppbv, Lai et al., 2024). During the observation period, the average values of T, WS and RH were 5.0 ± 2.5 °C, 1.3 ± 0.6 m/s and 38.9 ± 16.7%, respectively.

The relationship between meteorological parameters and pollutant concentrations were analyzed and correlations between $PM_{2.5}$, TVOCs and NO_x and RH were found (Fig. S3), suggesting that meteorological conditions have an important influence on pollution formation. The comparisons of average concentrations of different periods between different periods are presented in Tables 1 and 2. WS, Temp and RH conditions during infection and recovery periods were generally similar. However, the average concentration of $PM_{2.5}$ during the recovery period was 1.6 times the value during the infection period. Furthermore, the concentrations of other pollutants including SO_2, NO_2, CO, and O_3 all showed a similar trend between infection and recovery periods. The TVOCs concentration during the recovery period was 1.2 times the value during the infection period, showing an obvious increase trend after resuming production. Decreased trends of air pollutants were found in other studies before and after the outbreak of the novel coronavirus (COVID-19) in early 2020 (Qi et al., 2021; Wang et al., 2021).

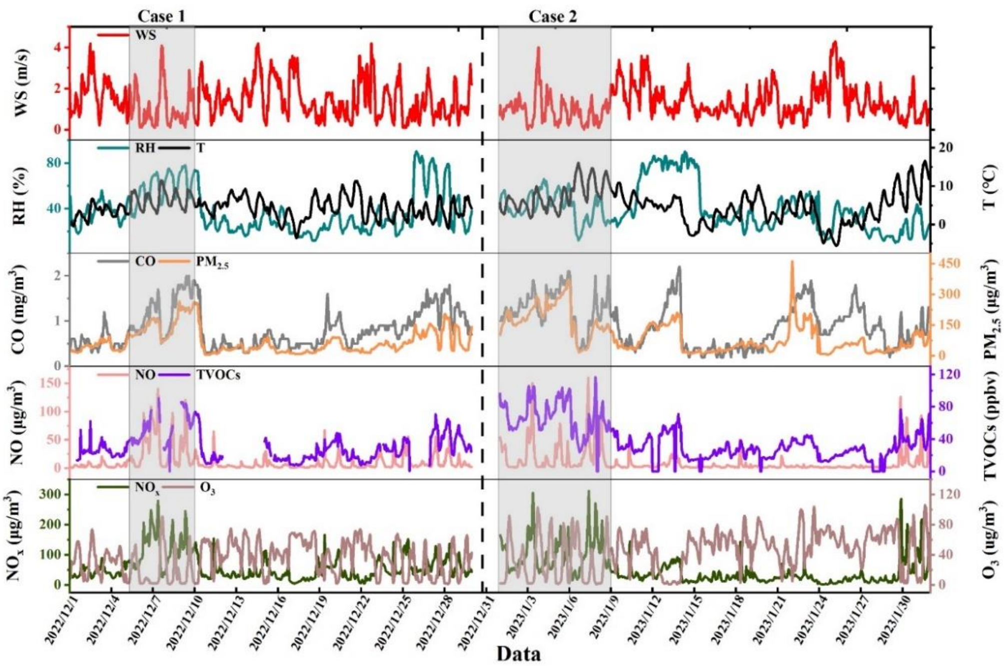


Fig. 1. Time series of WS, T, RH, CO, $PM_{2.5}$, NO, TVOCs, NO_x and O_3 during the observation period.

The shadow section in Fig. 1 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. 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. Any days with a $PM_{2.5}$ concentration lower than 35 μg/m^3 (China's I-level standard) is considered as Clean days.

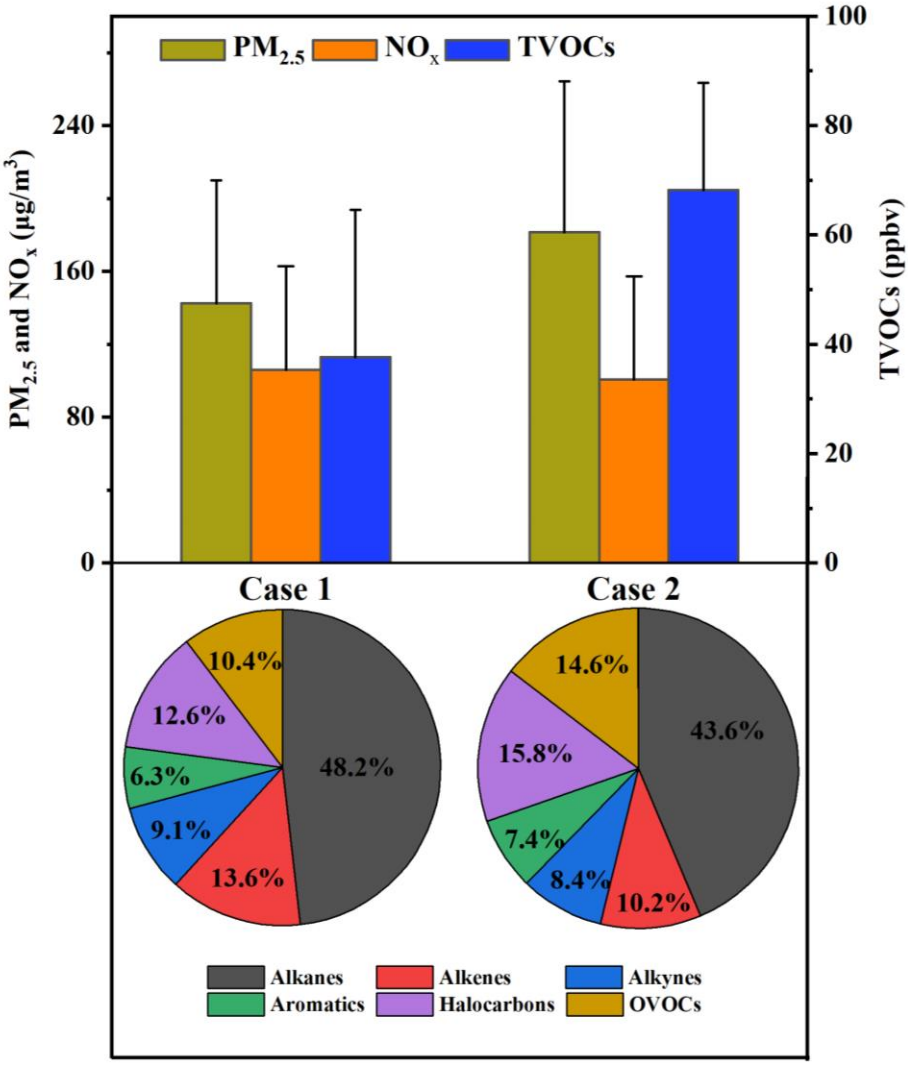


Fig. 2. The concentration of $PM_{2.5}$, NO_x, TVOCs and the composition ratio of VOCs in Case 1 and Case 2.

As for the two representative pollution processes (Case 1 during the infection period and Case 2 during the recovery period), the concentration of TVOCs in Case 1 and Case 2 were 48.4 ± 20.4 and 67.6 ± 19.6 ppbv (Fig. 2), respectively, increased by 63% and 188% compared with values during clean days. The average concentrations of $PM_{2.5}$ and TVOCs during Case 2 were 1.3 and 1.8 times the values in Case 1. The highest volume contributions of alkanes were found both in Case 1 (48%) and Case 2 (44%), consistent with the results in the Yangtze River Delta region (36-43%, Liu et al., 2023). While alkenes exhibited higher volume percentages of 13% in Case 1, followed by halogenated hydrocarbon (12%) and OVOCs (10%). Higher volume percentages of alkanes and alkenes in Case 1 were similar to the results in the gasoline evaporation site in winter (Niu et al., 2022). Equivalent volume contribution of halogenated hydrocarbon and OVOCs (15%) were found in Case 2, followed by alkenes (10%).

Though the volume contributions of aromatics were the lowest (6% in Case 1 and 7% in Case 2), the highest increase ratio was found from clean days to polluted episodes.

Table 1 The average concentrations of meteorological parameters and pollutants during different processes.

Category	Entire process	Infection period	Recovery period	Case 1	Case 2
	N = 62 days	N = 31 days	N = 31 days	N = 6 days	N = 8 days
WS (m/s)	1.3 ± 0.6	1.4 ± 0.6	1.3 ± 0.6	1.2 ± 0.9	0.9 ± 0.7
T (℃)	5.0 ± 2.5	4.7 ± 1.7	5.4 ± 3.1	6.1 ± 2.2	7.4 ± 3.5
RH (%)	38.9 ± 16.7	37.6 ± 15.5	40.2 ± 18.2	55.7 ± 14.7	42.0 ± 12.1
TVOCs (ppbv)	36.1 ± 21.0	31.9 ± 18.1	39.8 ± 22.4	37.6 ± 27.0	68.2 ± 19.6
SO_2 (μg/m^3)	11.4 ± 2.7	10.2 ± 2.8	12.7 ± 2.3	11.0 ± 3.7	16.2 ± 6.1
NO_2 (μg/m^3)	47.2 ± 10.0	46.8 ± 8.6	47.8 ± 11.7	62.7 ± 20.5	65.0 ± 21.3
CO (mg/m^3)	0.9 ± 0.2	0.8 ± 0.2	1.1 ± 0.2	1.2 ± 0.5	1.3 ± 0.4
O_3 (μg/m^3)	34.9 ± 6.0	31.1 ± 4.5	39.0 ± 4.6	21.8 ± 23.7	32.5 ± 29.6
$PM_{2.5}$ (μg/m^3)	111.5 ± 45.1	86.6 ± 34.6	138.3 ± 39.6	142.5 ± 67.4	181.5 ± 82.7

Table 2 Concentration of VOC species during different processes (ppbv).

Category	Entire process	Infection period	Recovery period	Case 1	Case 2	Clean days
TVOCs	36.1 ± 21.0	31.9 ± 18.1	39.8 ± 22.4	48.4 ± 20.4	67.6 ± 19.6	17.5 ± 9.5
alkanes	16.8 ± 9.2	15.0 ± 8.4	18.4 ± 9.5	23.1 ± 10.0	29.5 ± 8.4	9.2 ± 5.6
alkenes	4.1 ± 2.7	3.8 ± 2.6	4.4 ± 2.7	6.5 ± 2.9	7.0 ± 2.6	1.7 ± 1.3
alkynes	3.1 ± 2.0	2.7 ± 1.7	3.4 ± 2.1	4.3 ± 2.0	5.8 ± 1.9	1.3 ± 0.8
aromatics	2.1 ± 2.0	1.8 ± 1.5	2.3 ± 2.2	3.0 ± 1.8	4.9 ± 2.8	0.7 ± 0.5
halogenated hydrocarbon	5.4 ± 3.3	4.4 ± 2.3	6.2 ± 3.8	6.0 ± 1.9	10.7 ± 3.6	2.7 ± 1.4
OVOCs	4.6 ± 3.2	3.5 ± 2.7	5.1 ± 3.5	5.0 ± 2.4	9.7 ± 2.8	1.9 ± 1.1

3.2 Source Analysis of VOCs

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). In this study, the ratios of toluene/benzene (T/B), isopentane/n-pentane, isobutane/n-butane, and m/p-xylene/ethylbenzene (X/E) were selected to initially identify the potential sources of VOCs (Fig. 3). Concentrations of selected pollutants and ratios used are shown in Table S1.

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 isopentane/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 represent LPG usage, 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).

The ratio of X/E can be used to infer the photochemical age of the air mass. X/E ratios around 2.5-2.9 are typical of urban areas, indicating that VOCs are mainly from the urban area (fresh air mass) (Kumar et al., 2018). When this ratio is significantly lower than 3, it indicates that VOCs are mainly transported from distant sources (aging air masses) (Kumar et al., 2018). The average X/E value in this study was 2.0 (Fig. 3(d)), indicating low photochemical activity and aging of the air mass at the observation site. Potential source analyses also indicate that air masses are affected by long-range transport (Fig. S4).

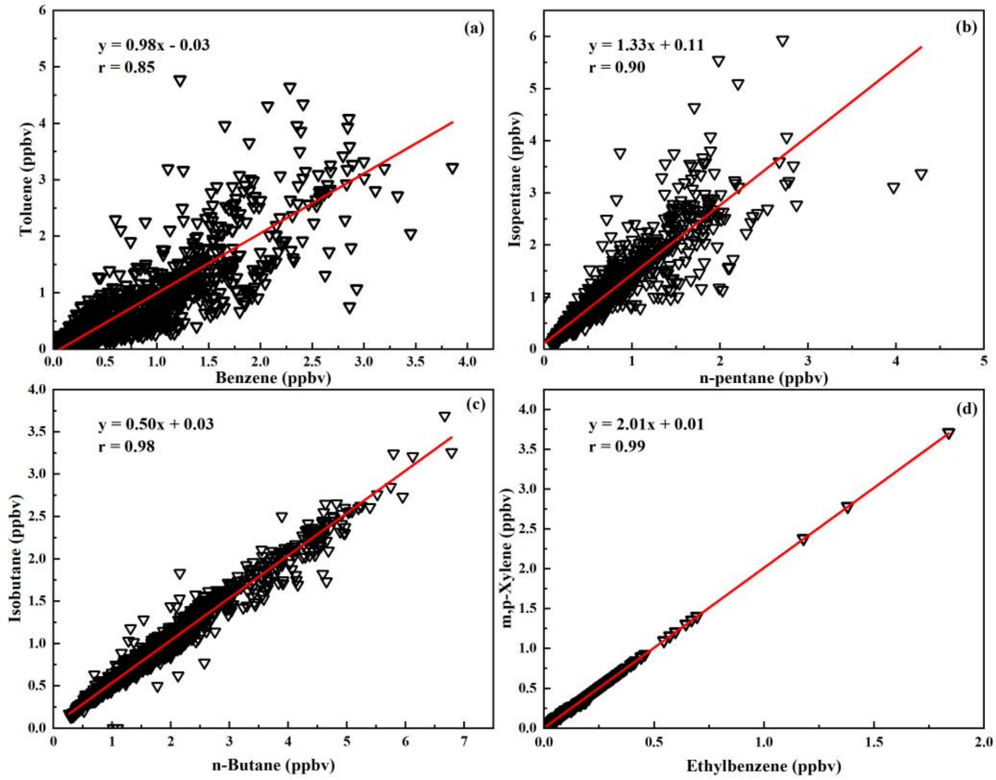


Fig. 3. Correlation analysis between specific VOC species.

Figure 4 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) Vehicular emission; (4) Industrial source; and (5) Combustion. 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).

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). Fig. S5 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 (Fig. 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 solvent usage source.

The CPF plot shows that local sources with wind speeds less than 1 m/s are the main sources (Fig. 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 (Fig. 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 Fig. S5 shows a bimodal trend, with peaks occurring in the morning and evening peaks of traffic, consistent with motor vehicle emissions. Fig. 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 (Fig. 5e).

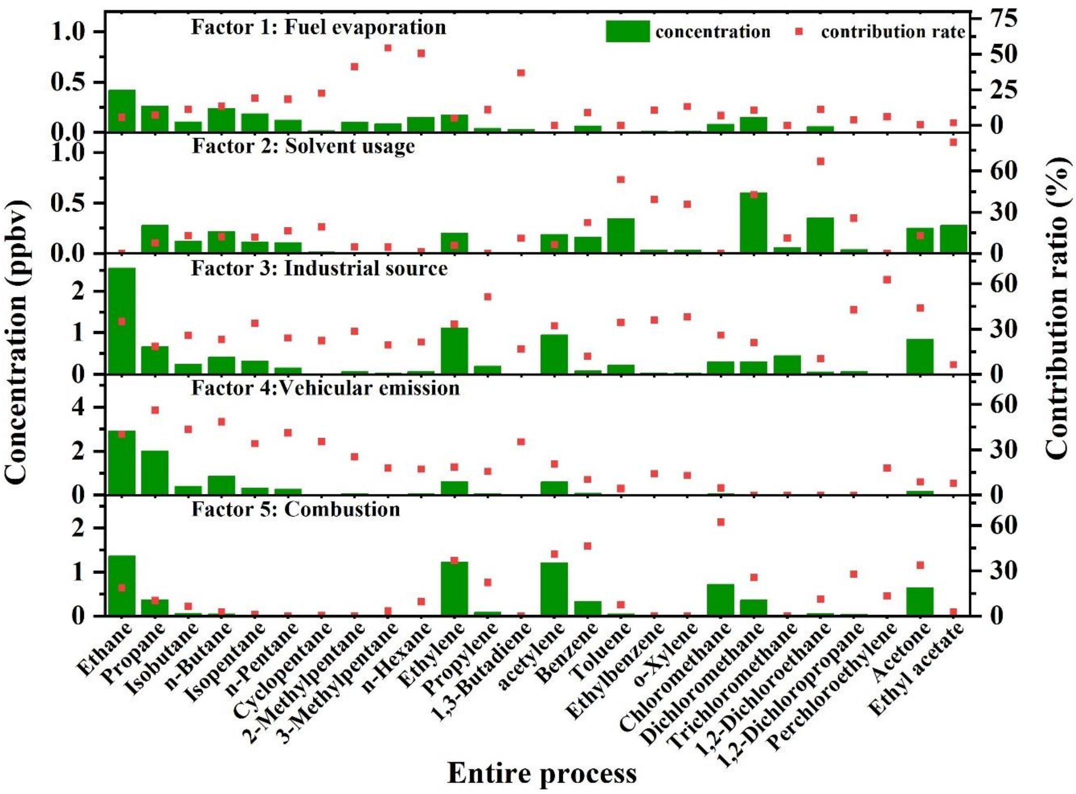
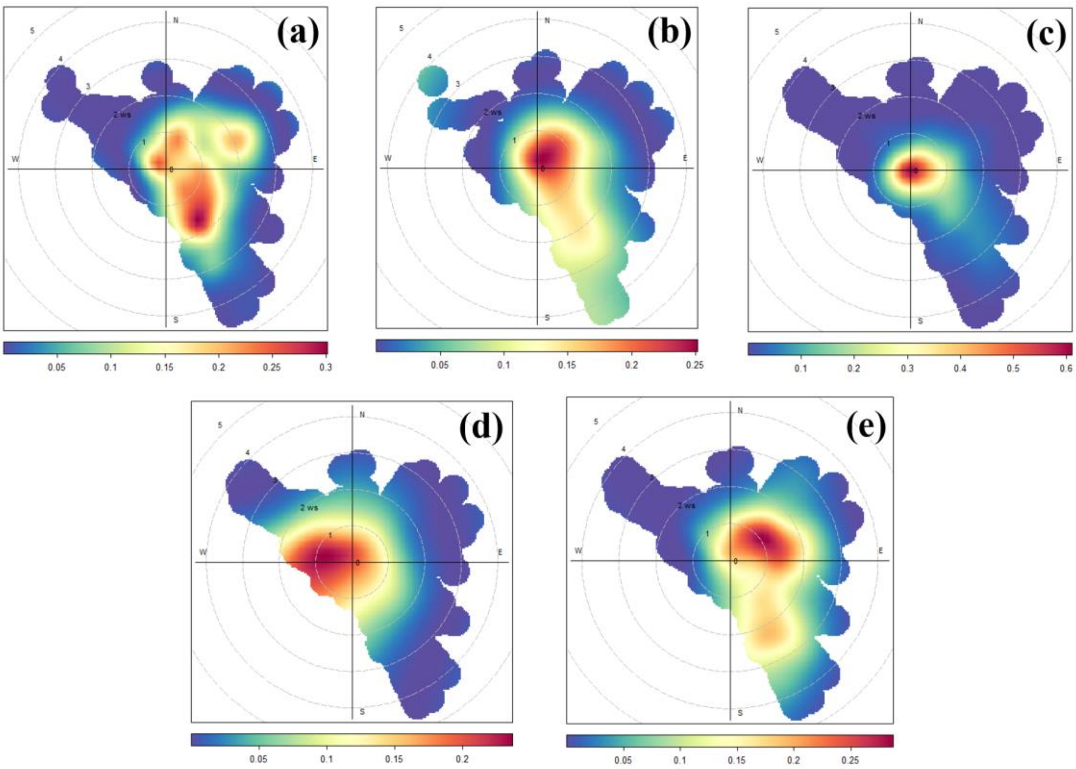


Fig. 4. Concentration of VOC species in each factor and contribution to each source.



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

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

Figure S6 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. We present the concentrations of the five main VOCs in all five factors in Table S2. Ethane (vehicular emission), 2-methylpentane (fuel evaporation), benzene (industry source), chloromethane (combustion), and ethyl acetate (solvent usage) were selected as tracers for five sources. Ethane concentration in Case 2 (5.9 ppbv) is much higher than in other processes, and ethane concentration during the recovery period (3.4 ppbv) is also higher than during the infection period (2.4 ppbv), which may to some extent reflect increased vehicular emissions during the recovery period.

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) (Fig. 6). 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 days motor vehicle source than other processes, which also resulted in a 34% clean days motor vehicle source share.

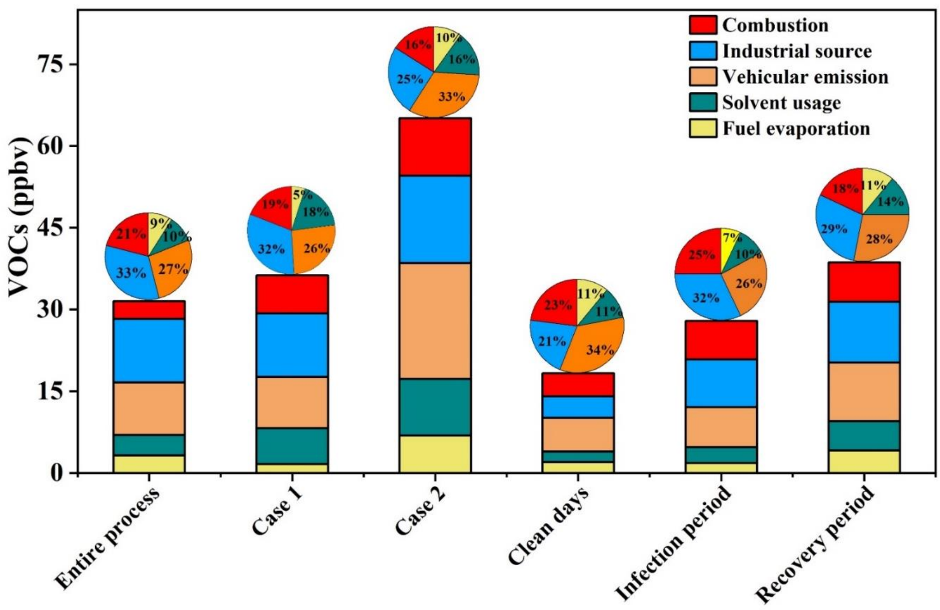


Fig. 6. Contribution of each to VOCs for different processes.

3.3 SOAP

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 have included quantitative analysis for SOAP as well. Fig. 7 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%. In each process, the composition of the top ten substances is essentially the same. Aromatic hydrocarbons contributed the most, with BTEX always occupying the top five positions and toluene the most. The SOAP values of the top ten contributing species for the two polluting processes are shown in Tables S3 and S4. Toluene, the highest contributing species, reached a SOAP value of 49.4 $\mu g/m^3$ 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 $\mu g/m^3$). The SOAP value for Case 1, which is also a contaminated process, was 67 $\mu g/m^3$, and the main species (m/xylene: 9.8 $\mu g/m^3$, benzene: 8.5 $\mu g/m^3$) including toluene (34.6 $\mu g/m^3$) were lower than those for Case 2 (m/xylene: 19.4 $\mu g/m^3$, benzene: 13.4 $\mu g/m^3$).

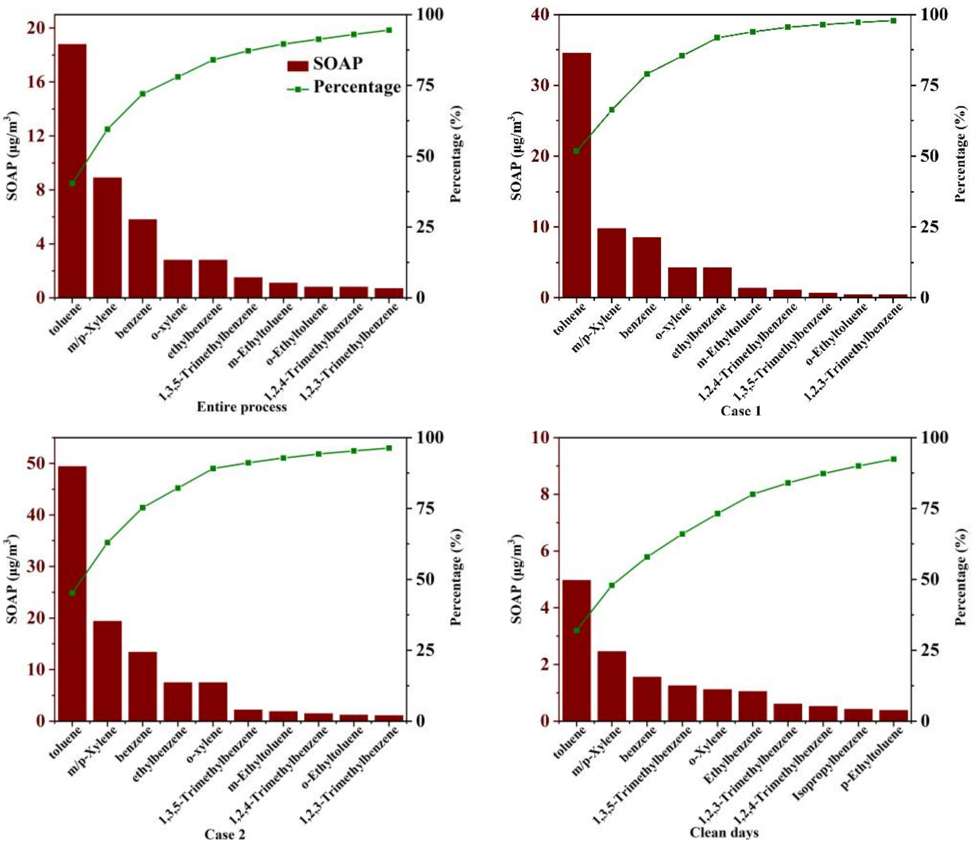


Fig. 7. SOAP dominant species in different processes

Figure 8 shows the SOAP calculated after source resolution of the two pollution processes by PMF for clean days, respectively. In Case 1, industrial source is the dominant source with a contribution ratio of 63%. In Case 2, the pollution sources exhibit a more evenly distributed contribution, where the solvent usage and fuel evaporation sources emerge as the primary contributors to SOAP, with their respective contribution levels rising to 32% and 26%. The clean day result with a SOAP of 8.8 μg/m^3 also indicates that industrial and solvent usage sources are the most dominant SOAP sources. The primary sources of aromatic compounds, which are the most significant contributors to SOAP, are solvent usage and industrial process emissions. This finding aligns with the results of other studies (Wu et al., 2017). Consequently, it is imperative to implement measures to reduce $PM_{2.5}$ pollution by regulating emissions from industrial and solvent usage sources.

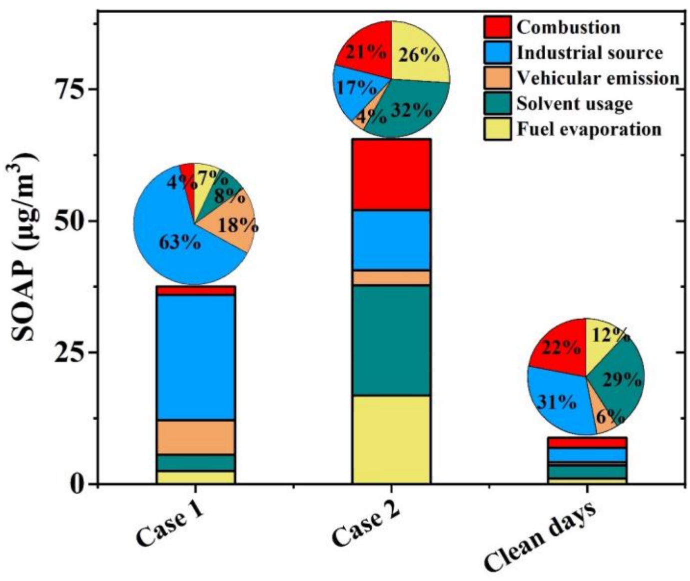


Fig. 8. SOAP value and contribution ratio of each process

4. Conclusions

Continuous observation of VOCs during the infection of the Omicron epidemic was carried out at an urban site in polluted Zhengzhou from December 1, 2022, to January 31, 2023. The daily average concentration of $PM_{2.5}$ ranged from 53.5 to 239.4 μg/m^3 with an average value of 111.5 ± 45.1 μg/m^3 during the whole period. The concentration of TVOCs ranged from 15.6 to 57.1 ppbv with an average of 36.1 ± 21.0 ppbv, higher than the same period in last year (27.9 ± 12.7 ppbv, Lai et al., 2024).

Two representative contamination processes were identified (Case 1 during the infection period and Case 2 during the recovery period). The concentration of TVOCs in Case 1 and Case 2 were 48.4 ± 20.4 and 67.6 ± 19.6 ppbv, respectively, increased by 63% and 188% compared with values during clean days. The average concentrations of $PM_{2.5}$ and TVOCs during Case 2 were 1.3 and 1.8 times of the values in Case 1. The highest volume contributions of alkanes were found both in Case 1 (48%) and Case 2 (44%). Though the volume contribution of aromatics were the lowest (6% in Case 1 and 7% in Case 2), the highest increase ratio was found from clean days to polluted episodes.

Local sources were initially identified through T/B, isopentane/n-pentane, isobutane/n-butane, and X/E ratios. The average X/E value was 2.0, indicating that measured levels of airborne VOCs were influenced by emissions from remote sources and urban areas. The PMF receptor modeling yielded five major sources of pollution, which included industrial emissions (32%), vehicular emissions (27%), combustion

(21%), solvent usage (11%), and fuel evaporation (9%). Significant differences were observed in the sources of VOCs across different pollution periods. In Case 1, industrial emissions constituted the largest contributor, accounting for 32% of the total VOCs. In contrast, in Case 2, the proportion of vehicular emissions increased to 33%, becoming the primary source of VOCs.

Aromatic compounds are the main contributors to SOAP, with BTEX being the main contributor during the entire period. SOAP values reached 37.6 and 65.6 $\mu g/m^3$, respectively in Case 1 and Case 2. In Case 1, industrial source accounted for a substantial majority (63%, 23.8 $\mu g/m^3$), while vehicular source, as the second most significant contributor, made up only 18%. In Case 2, the distribution of contribution rates was more uniform, with solvent usage source and fuel evaporation source becoming the primary contributors to SOAP, at 32% (20.9 $\mu g/m^3$) and 26% (16.8 $\mu g/m^3$), respectively. The SOAP result for the Clean Day was 8.8 $\mu g/m^3$, with industrial source and solvent usage remaining the primary contributors. Consequently, it is of paramount importance to regulate emissions from the industrial and solvent usage sectors with the objective of reducing PM2.5 pollution.

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