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Characterization of Ambient Volatile Organic Compounds (VOCs) in the Area Adjacent to a Petroleum Refinery in Jinan, China

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ABSTRACT

56 VOCs were monitored for a whole year in a refinery vicinity area in suburban of Jinan, as well as $PM_{2.5}$, PM_{10}, and ozone. The results of VOCs and particulate matters showed that January was the most polluted month during the sampling period, possibly resulting from coal burning in the district heating period of Jinan. According to the concentration profiles of monthly variety of the ambient pollutants, the change of VOCs showed a certain extent of positive correlation with $PM_{2.5}$ and PM_{10}, while negative correlation with ozone. It was found that the average concentration of total VOCs was 50.58 μg m^{-3} while ethane and ethene were the most abundant VOC species with the concentrations of 24.58 and 3.94 μg m^{-3}, respectively. The high relative contribution of ethane was unusual, compared with the previous related VOC research in urban area of same city or other cities in China. Based on the analysis of the monitoring data, this area was not majorly affected by major VOCs pollutants from refinery emission. m & p-Xylene of 1.34 μg m^{-3} was the most abundant pollutant of BTEX. The BTEX ratio analysis presented that traffic exhaust was not the major VOCs source.

Keywords: VOCs; PM; Ozone; Refinery; Ethane.

INTRODUCTION

Air pollution is attracting huge attention in current China. Volatile organic compounds (VOCs) are defined as chemicals with boiling points less than 260 degree Celsius and room temperature vapor pressures greater than 0.52 mmHg (Cai *et al.*, 2006), are a group of ambient air pollutants threatening human health (Liu *et al.*, 2005). Additionally, VOCs negatively affect environment and human welfare by leading to the formation of ozone (Wei *et al.*, 2014b), which was also a main pollutant in Chinese cities.

Human activities such as industrial emission, vehicle exhaust and gasoline storage have been thought to affect the quality of the urban atmosphere. Industrial and traffic emissions were responsible for the increasing ambient VOCs in cities or industrial area (Zheng *et al.*, 2009). Refineries and petrochemical plants are important sources of VOCs, sulfur compounds, nitrogen oxides, and particulate matters, which were emitted from production units, storage tanks, transport pipelines, etc. (Gariazzo *et al.*, 2005). Many works have been carried out to monitor air quality in the adjacent area of refineries (Wei *et al.*, 2014b). The VOCs pollution became severe and has been concerned by the citizen and city managers in China, especially in some metropolises for recent years (Liu *et al.*, 2008). Understanding the profile and distribution of VOCs would be important to identify the pollution source, and furtherly give guidance to formulate the air pollutant abatement strategies in a city (Liu *et al.*, 2005). Besides, VOCs contribute to formation of the secondary organic aerosols, which was usually measured as part of particulate matter. However, there were few researches, focused on the studies of relation between VOCs and particulate matter (such as $PM_{2.5}$ and PM_{10}). It was unclear whether both pollutants originated from similar sources, making it necessary to investigate their relationship during the same sampling period.

In this research, based on one-year observation of VOCs, $PM_{2.5}$ and PM_{10}, and ozone, the distribution of VOCs was investigated and the relationship between VOCs and other atmospheric pollutants were discussed to evaluate the influence of a refinery on the suburban vicinity area.

METHODS AND MATERIALS

VOC Sampling Site

Jinan is the capital of Shandong provinces, 400 km south of Beijing, and 700 km north of Shanghai. Due to heavy industries and traffic, this city was listed as one of top 10 most air-polluted cities in the world in 2013, reported

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in the "Country Environmental Analysis" of the Asian Development Bank together with Tsinghua University (Zhang and Crooks, 2012). According to the online data of the Ministry of Environmental Protection of the PR China, there were 39 days when the air quality index (AQI) was larger than 300, meaning serious air pollution, during 2015 in the city. To solve the problem, the local government is endeavoring on the emission inventory and source control of air pollutants, including PM, SO_2, NO_x, VOCs, etc. In addition, Jinan implemented a city-wide coal-burning district heating system each winter between Nov and Jan in next year as other northern cities in China. The monitoring/sample site (E 117.03, N 36.66) was located in the east outskirts in Jinan, China, which was on the top of a 3 floor building (10 m) and approximately 2 km south to a refinery. The refinery, belonging to a large Chinese petroleum and chemical company, covers 2,400 m^2 with the annual production capacity of 5 MTons. The refinery has more than 30 sets of production units, supplying petroleum, kerosene, diesel, liquefied petroleum gas, propene, light oil, lubricant, asphalt and other productions. Within a range of 5 km centering the refinery, there are a hospital, three colleges, several residential districts, and other sensitive area. Some residents doubt the refinery could influence their health negatively. In our previous research (Wang *et al.*, 2016), the ambient VOCs in urban of the same city was investigated, which was used as a reference. Although the sampling site was close to an eight-lane road, the intensity of the daily traffic was low and there were no high buildings besides, since the site was around 10 km away from the urban center. Significantly, the sample site was not close to any coal-burning emission in a range of approximately 10 km.

On-line Sampling and Analysis

Fifty-six ambient VOCs were sampled and determined at hourly interval with the on-line ozone precursors analyzer: airmOzone C_2–C_{12} (CHROMATOTEC Group, France), consisting of a light VOC analyzer airmoVOC C_2–C_6 and a heavy VOC analyzer airmoVOC C_6–C_{12}. The system could trap ambient VOCs with Carbotrap C, Carbopack B, and Carboxen with air sample volume of 80 mL and 120 mL for both groups of VOCs respectively; after thermal desorption, separate VOCs with capillary columns and detect them with FID detector. The system has been widely used for VOC monitoring and more details could be found in previous studies (Wang *et al.*, 2013; Liu *et al.*, 2016; Wang *et al.*, 2016). The daily concentration of VOCs was obtained by averaging 24 hourly interval data, therefore the monthly concentration was an average value of the daily ones in the target month. Considering abnormal data and the incidental errors of instrument, 315 available daily data sets covering 2014 were used in this study.

Two on-line particulate matter monitors BAM-1020 (Met One Instrument, Inc., USA) were used to collect $PM_{2.5}$ and PM_{10} data, with BX-2.5 and BX-10 sampling kit, separately. The model BAM-1020 can continuously provide airborne particulate mass/volume concentration using beta ray attenuation. Air was automatically pumped through glass fiber filter tape at 16.7 L min^{-1} and the penetrated beta rays were counted with a sensitive scintillation detector and reported daily. The filter tape was changed every two months. The 24-hour detection limit was 2.0 μg m^{-3} and the measurement cycle time was 1 hour. The span calibration check was conducted with automatic 0.8 mg span membrane verification (BAM-1020 Operation Manual for more details).

Ozone (O_3) was detected with a photometric ozone analyzer Model 400E (Teledyne Advanced Pollution Instruments (TAPI), Teledyne Technologies Inc., USA). The concentration of ambient ozone was measured by detecting the intensity attenuation of passing ultra-violate light following Beer-Lambert equation. The sampling flow rate was 0.75 L m^{-3} and the detection limit was 0.6 ppb. The O_3 data was also reported daily.

RESULTS AND DISCUSSIONS

As shown in Table 1, ethane dominated the ambient VOCs pollution in the area, 24.58 μgm^{-3} out of 50.68 μg m^{-3} of total VOCs (sum of concentrations of all 56 VOCs), while it was only in 1.7 μg m^{-3} out of 71.94 μg m^{-3} in the referencing site in urban area of the same city (Liu *et al.*, 2015). Although ethane was listed as abundant VOC specie in China, the relative concentration of ethane was significantly higher in this work, compared with 1.92 ppbv (2.58 μgm^{-3}) out of 27.82 ppbv of 56 VOCs in urban area of Shanghai (Wang *et al.*, 2013), 4.47 out of 101.77 μg m^{-3} of 31 VOCs in urban area of Beijing (Song *et al.*, 2008). According to previous report (Cetin *et al.*, 2003; Song *et al.*, 2008), ethane majorly originated from gasoline-related emission, natural gas, or petroleum product. Then, the possible major pollution sources of ethane in the sampling site could be the traffic on the road, a vehicle-natural gas station located 100 meters away, or the refinery. Because of the low the traffic intensity, it was hard to conclude that traffic emission was the major source of ethane, which was also supported by the fact that the concentration of total VOCs was lower than urban area, where traffic played an important role. Additionally, the ethane was not reported as a major VOC from petroleum refinery (Liu *et al.*, 2008) and a research conducted in area of petrochemical facilities in the Yangtze River Delta (Mo *et al.*, 2015) presented the relative concentration of ethane was only 9.5 %wt which was much smaller than 48.6% in this research. Sequentially, the vehicle-natural gas station likely contributed to the phenomena, since ethane was the second largest compound in natural gas. In Table 1, ethene was the second most abundant VOC in the observation, which majorly resulted from oil refinery, coal burning, and vehicle emission (Liu *et al.*, 2008). The concentration of 3.94 μg m^{-3} was comparable with the urban area in other Chinese cities; 5.18 μg m^{-3} in Beijing (Song *et al.*, 2008) and 1.73 ppbv (2.17 μg m^{-3}) in Shanghai (Wang *et al.*, 2013), but almost twice as that in the same city (2.10 μg m^{-3} in urban area (Wang *et al.*, 2016)), indicating the ethene could come from the refinery because the suburban site was less affected by traffic and coal burning. The other relative prevailing VOCs included n-dodecane, 2,3,4-trimethylpentane, m & p-xylene, trans-2-butene, cis-2-butene, 1-butene, etc., of which, only m & p-xylene, cis-2-butene,

Table 1. Average concentration of 56 VOC species and total VOCs in 2014, Jinan, China.

Species	Concentration ($\mu g\ m^{-3}$)	Species	Concentration ($\mu g\ m^{-3}$)	Species	Concentration ($\mu g\ m^{-3}$)
Ethane	24.58	i-Butane	0.52	3-Methlhexane	0.08
Ethene	3.94	Propane	0.52	3-Methylheptane	0.08
n-Dodecane	1.72	1-Pentene	0.51	o-Ethyltoluene	0.08
2,3,4-Trimethylpentane	1.60	2-methyl-1-Pentene	0.48	m-Ethyltoluene	0.08
m & p-Xylene	1.34	1-Pentene	0.46	Toluene	0.08
trans-2-Butene	1.28	3-methylpentane	0.37	Cyclohexane	0.08
cis-2-Butene	0.90	2,2-dimethylbutane	0.31	2,3-dimethylpentane & 2-methylhexane	0.07
1-Butene	0.89	Propene	0.29	n-Nonane	0.07
n-Pentane	0.86	Methyl-cyclopentane	0.28	p-Diethylbenzene	0.06
i-Pentane	0.83	Styrene	0.26	p-Ethyltoluene	0.05
Ethylbenzene	0.82	Bezene	0.24	n-Heptane	0.05
Cyclopentane	0.82	1,2,3-Trimethylbenzene	0.19	m-Diethylbenzene	0.05
1,3-Butadiene	0.82	1,2,4-Trimethylbenzene	0.19	i-Propylbenzene	0.04
Ethyne	0.80	2,3-dimethylbutane	0.14	n-Octane	0.04
Isoprene	0.78	1,3,5-Trimethylbenzene	0.13	2-Methylheptane	0.03
o-Xylene	0.60	n-Undecane	0.13	2,2,4-trimethylpentane	0.03
trans-2-Pentene	0.60	2-methylpentane	0.13	n-Decane	0.02
n-Butane	0.59	n-Propylbenzene	0.12	2,4-dimethlypentane	0.02
n-Hexane	0.54	Methylcyclohexane	0.10	Total VOCs	50.68

and 1-butene xylene was also abundant in Jinan's urban atmosphere (Liu *et al.*, 2016); m & p-xylene was abundant in Shanghai and Beijing (Song *et al.*, 2008; Wang *et al.*, 2013). The different profile implied that varied sources of ambient VOCs between urban or suburban area. Usually, the main VOCs in refinery exhaust were reported as propene, i-butane, benzene, n-butane, n-hexane, toluene, i-pentane, 1-butene,n-pentane, etc. (Cetin *et al.*, 2003; Lin *et al.*, 2004; Wei *et al.*, 2014a). Only 1-butene was found abundant in this research, meaning possibly that the contribution of the refinery to VOCs in the sampling was limited.

The monthly trend of total VOCs was shown in Fig. 1, presenting that the highest concentration appeared in January, which was in the period of citywide district heating, when more frequent north-west winds shown in the rose plots likely also brought some pollutants from the urban area to the north of the sampling site (shown in Fig. 2). The concentration in November and December was not as high as in January, which possibly attributed to stricter air control policies implemented by local government, such as compulsory improvement of the emission clean system of coal burning, etc. The low concentration in February could attribute to one week off for Chinese spring festival, when the industries, including the refinery, ceased the production and the traffic was significant low in the street. From March to May, the VOCs decreased gradually, which could attribute to the frequent south and north-east winds, bringing cleaner air from south recreation area and east rural area, in spring and the increasing temperature in Jinan (Liu *et al.*, 2016). The concentrations were relatively stable from June to October between 40–50 $\mu g\ m^{-3}$, when the north-west winds started taking over. In Fig. 1, a comparison between weekends and weekdays were also conducted. Only in April and November, the VOCs on weekends were lower than weekday, while the concentration was similar between on weekday and weekend in other months. This could imply that although the sampling site was close to a road, daily commuting (traffic) was possibly not the major source of VOCs in this area.

As shown in Fig. 3, the monthly change profile of particulate matter was similar to that of VOCs. Highest concentrations of both $PM_{2.5}$ and PM_{10} appeared in January (0.12 and 0.21 mg m^{-3}, respectively). Compared to PM_{10}, the change of $PM_{2.5}$ indicated more similar to that of VOCs, presenting lower concentrations in May and December and stable between June and September. It has been widely reported VOCs could lead to the production of secondary organic aerosols, possibly increasing the $PM_{2.5}$ (Zhu *et al.*, 2016). Particulate and VOCs emissions were also reported in high concentrations around refinery area or roadside (Gariazzo *et al.*, 2005; Wei *et al.*, 2014b). The unusual observation was in February, when the $PM_{2.5}$ kept in a high level while VOCs presented the lowest in the sampling period. As mentioned above, the decrease of VOCs could result from less industrial activities and traffic in the festival vacation, when, additionally, the area was less affected by fire works due to the smaller population density. It could be explained by that more $PM_{2.5}$ might travelled a long distance from the west urban area or else besides VOCs reaction, because smaller particulates transported longer distance than larger ones, which could be supported by PM_{10} presenting a concentration valley in February, though the positive relation between PM_{10} and VOCs were weaker as shown in Fig. 3. Construction work in surrounding area could result in the high PM_{10} in April and May, because cleaner south or east-north winds frequently occurred in the season while $PM_{2.5}/PM_{10}$ ratio was relatively lower for the mud and dirt from construction industries (Muleski *et al.*, 2005).

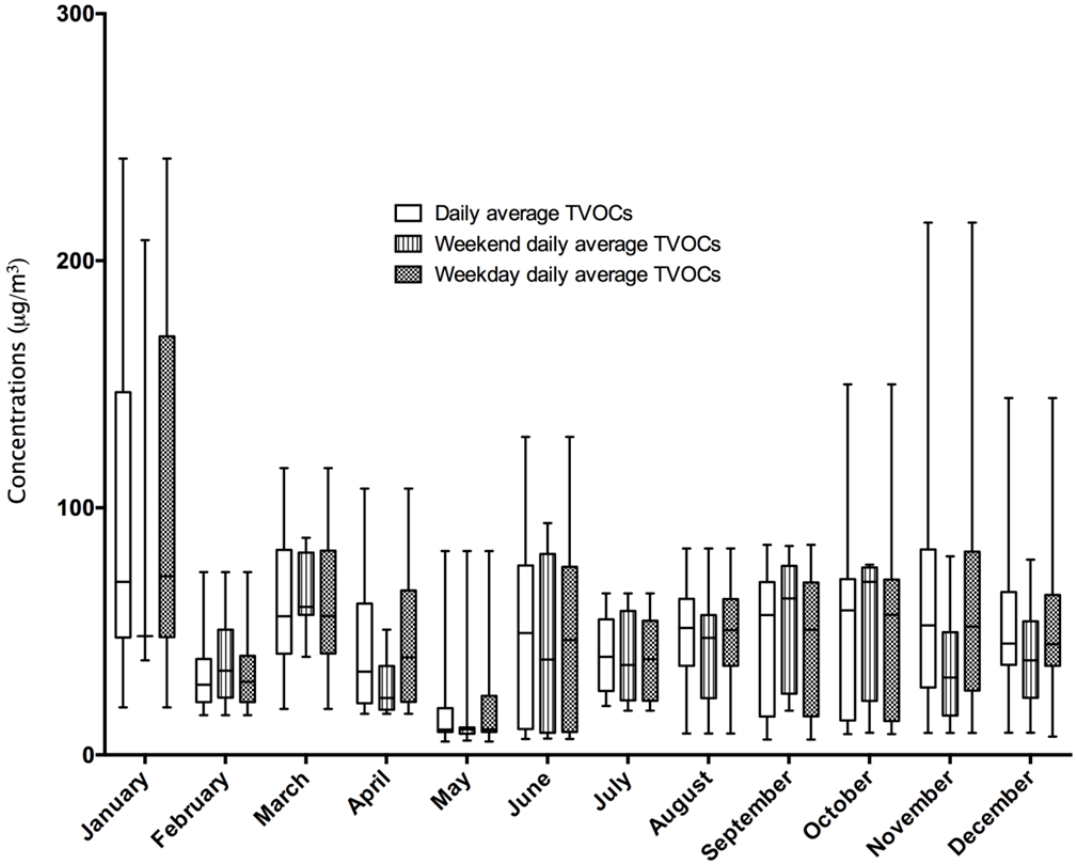

Fig. 1. Monthly change of VOCs and the comparison between average concentration on weekdays and weekends.

Fig. 3 also shows the negative relation between total VOCs and ozone. VOCs are important potential precursors of ozone (Zheng *et al.*, 2009). The profile of ozone in the sampling period favored higher temperature and more intense sunshine, which both benefit the reaction of VOCs to form ozone (Han *et al.*, 2013). As shown in Fig. 3, highest concentrations of ozone occurred in the summer months while VOCs were low. The monthly change of ozone did not fluctuate as sharp as VOCs, meaning VOCs was possibly not the only source, for instance, NO_x could also influence the depletion of ozone (Wei *et al.*, 2014a). Another explanation was that the chemical reactivity of VOCs affected the formation of ozone more than the mass of VOCs (Ou *et al.*, 2015).

BTEX (benzene, toluene, ethylbenzene, xylene) were found in the vicinity of refineries (Baltrėnas *et al.*, 2011), the ratio between them was used as pollution source identifier. As shown in Table 2, the ratio of average volumetric concentration of the four aromatics was approximately 0.4:0.1:1:2.4, which was different from previous reported obtained in urban area where traffic emission dominated, for instance 3:4:1:4 (Na, 2006), 3:4:1:3 (Baltrėnas *et al.*, 2011), or 2.3:5.5:1:2.3 (Wang and Zhao, 2008). It was argued that relative high concentrations of ethylbenzene and xylene could attribute to industrial solvent or gasoline evaporation (Louie *et al.*, 2013) rather than. Additionally, as the characteristic ratios of BTEX are shown in Fig. 4, toluene/benzene (T/B) ratios along the year were less than 2.0, meaning that traffic emission may not be the major VOCs source in this research. The refinery was doubt contributing to both pollutants. Lower ratio of xylene/ethylbenzene (X/E) in the earlier half year could demonstrate that the pollutants travel a long distance away from the monitoring site, since xylene was more photoactive than ethylbenzene and this ratio was used as indicators of chemical aging due to the different atmospheric lifetimes (Zhang *et al.*, 2015; Zhu *et al.*, 2016). In Fig. 4, the relative higher X/E value in later-half year was found, when the VOCs concentrations were relative higher than earlier-half year (except January) as shown in Table 1.Besides, more frequent north-west winds as shown in Fig. 1 in the same period. Consequently, it was doubt that the rising VOCs could come from adjacent source (higher X/E ratio) brought by north-west wind.

CONCLUSIONS

Based on the analysis of VOCs monitoring data in a year, some conclusions are listed as follows,

1. The suburban VOCs pollution was not serious as that in the urban in Jinan.
2. Refinery might affect the adjacent area, such as increasing ethene concentration, but it was not the major source of VOCs.
3. Traffic emission was not the major source, either, supported by lower T/B value and no significant difference between weekend and weekday VOC concentrations.

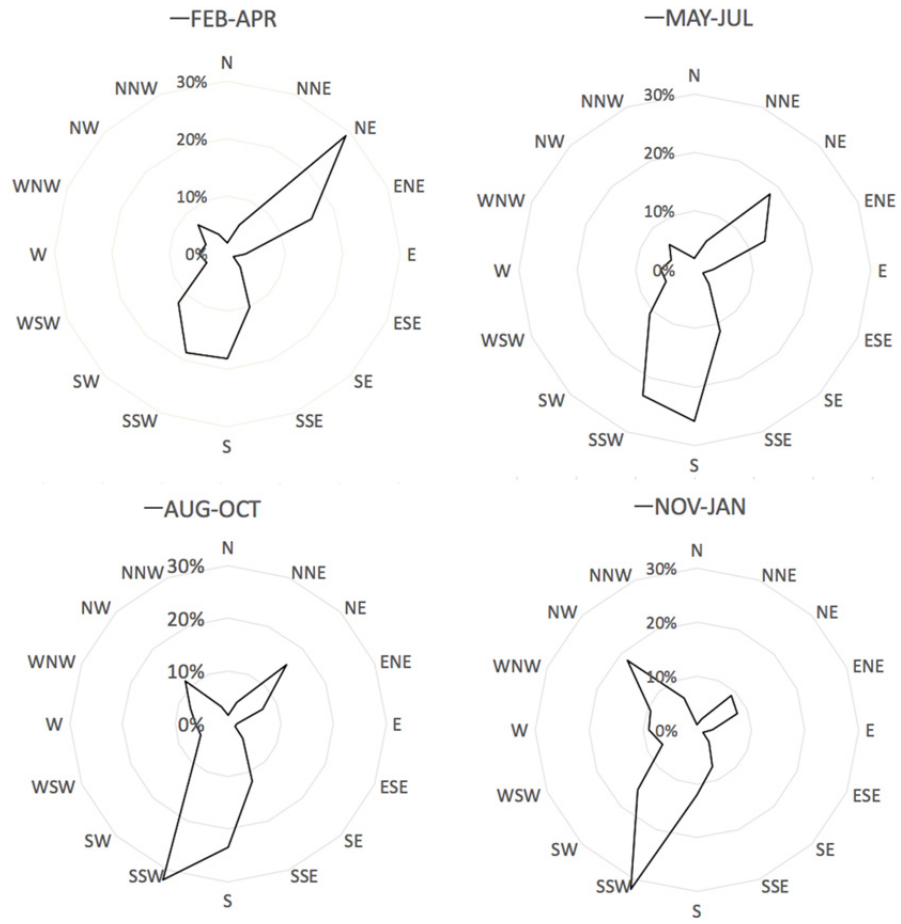

Fig. 2. Seasonal wind rose plots (wind frequency) during sampling period.

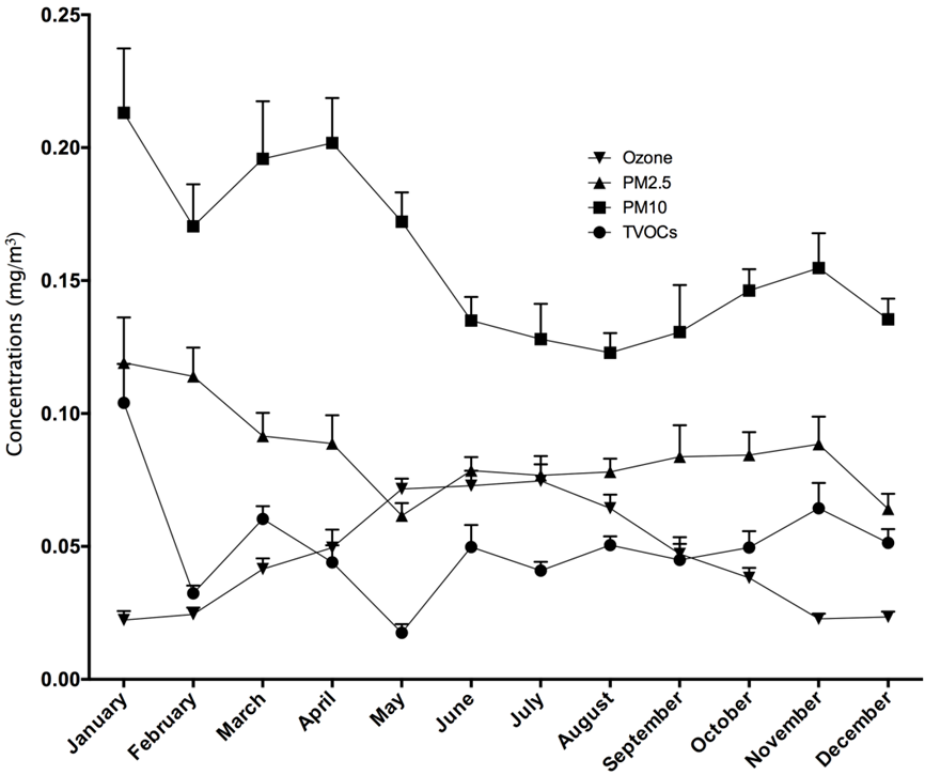

Fig. 3. Monthly concentration changes of PM_{10}, $PM_{2.5}$ and ozone compared with VOCs during the sampling period.

Table 2. Monthly BETX concentrations during the sampling period, Jinan, China.

	Benzene (μg m^{-3})	Toluene (μg m^{-3})	Ethylbenzene (μg m^{-3})	Xylene (μg m^{-3})
Jan	0.024	0.012	0.051	0.028
Feb	0.012	0.004	0.074	0.014
Mar	0.016	0.002	0.088	0.015
Apr	0.033	0.002	0.106	0.012
May	0.015	0.004	0.027	0.015
Jun	0.046	0.004	0.044	0.102
Jul	0.010	0.016	0.073	0.141
Aug	0.011	0.003	0.075	0.135
Sep	0.006	0.002	0.080	0.151
Oct	0.005	0.003	0.064	0.132
Nov	0.492	0.121	1.132	3.512
Dec	0.104	0.047	0.153	0.251
Yearly	0.069	0.019	0.173	0.409

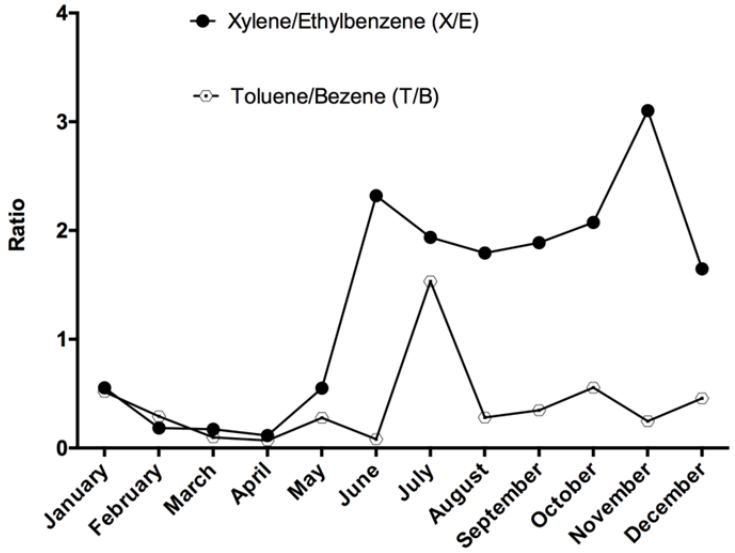

Fig. 4. Monthly change of xylene/ethylbenzene (T/B) and toluene/benzene (X/E) during the sampling period.

4. The abnormal high ethane concentration could result from the leaking of a vehicle-natural gas station.
5. The particulate matter and VOCs shared similar profiles but ozone related negatively to the VOCs due to the photochemistry reaction, especially in summer.

Generally, although no significant local VOC sources were found (except for ethane), meaning the source identification were complicate, adjacent sources, e.g., urban sources or the refinery, could affect the local suburban air quality with frequent north-west winds occurring.

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