Characteristics of Volatile Organic Compounds during Different Pollution Periods in Winter in Yuncheng, a Typical City in North China

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ABSTRACT

The composition characteristics and health risks of volatile organic compounds (VOCs) and their effects on secondary organic aerosol (SOA) during polluted and clean periods in winter 2018 for Yuncheng, a typical city in North China, were studied. The average concentration of VOCs was 2.3 times higher during the polluted period ($105.29 \mu\text{g m}^{-3}$) than the clean period ($45.78 \mu\text{g m}^{-3}$), whereas the average concentrations of the alkanes, alkenes, and aromatics were 3.4, 2.1, and 3.9 times higher, respectively, during the polluted period than the clean period. In addition, the VOC concentration was significantly influenced by meteorological conditions. Using positive matrix factorization (PMF), seven sources of atmospheric VOCs were identified, with the largest factors being vehicle emission (27.89%), coal combustion (23.37%), liquefied petroleum gas/natural gas evaporation (18.30%), and industrial emission (15.51%). Vehicle emission (30.04%) was the primary contributor during the polluted period, whereas coal combustion (33.53%) was the primary contributor during the clean period. The SOA formation potential (which almost exceeded 80%) was influenced by industrial emission (28.80%), solvent usage (26.30%), and vehicle emission (24.85%). Additionally, an assessment of the health risks of six aromatics based on the health risk exposure model of the United States Environmental Protection Agency revealed that the non-cancer risk was higher during the polluted period (6.61×10^{-2}) than the clean period (2.20×10^{-2}) but still below the Environmental Protection Agency (EPA) limit (1.00) and therefore negligible for the exposed population. However, the carcinogenic index of benzene (2.85×10^{-5} to 5×10^{-5}) exceeded 10^{-6} , suggesting a higher carcinogenic risk. Large-scale energy restructuring during recent years has sharply reduced coal combustion, but the VOC concentration has dramatically increased due to vehicle emission. Hence, regulating vehicle emissions is an effective strategy for controlling VOCs in Yuncheng.

Keywords: Volatile organic compounds; Source apportionment; Secondary organic aerosol formation potential; Health risk assessment; Pollution period.

INTRODUCTION

Volatile organic compounds, one of the important organic contaminants in the atmosphere, cause environmental pollution and are harmful to human health (Chang *et al.*, 2019). Numerous VOC species, including benzene and ethylbenzene, have been confirmed as carcinogens and potentially toxic substances (Yan *et al.*, 2015). Additionally, VOCs act as precursors of photochemically formed secondary pollutants such as secondary organic aerosol (SOA), which may contribute to haze days (Sun *et al.*, 2016). Understanding

VOC sources and the associated risk to human health is fundamental for establishing measures to reduce VOC emission into the environment.

In recent years, frequent haze events in winter in northern China have caused widespread concern regarding its effects on human health and atmospheric visibility (Liu *et al.*, 2016a; Li *et al.*, 2015; Zheng *et al.*, 2017; Hu *et al.*, 2018). Studies have indicated that haze days are largely affected by SOA formation and poor meteorological conditions (Guo *et al.*, 2014; Zheng *et al.*, 2015). As an important precursor of SOA, high VOC concentrations may largely contribute to PM_{2.5} generation in haze pollution. Liu *et al.* (2016) found that the VOC concentration in Beijing was obviously higher during haze-fog days ($461.61 \mu\text{g m}^{-3}$) than during clean days ($208.63 \mu\text{g m}^{-3}$). Studying VOCs in haze episodes is helpful for understanding VOC characteristics and providing guidance for VOC control strategies. In a recent study, Cai

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et al. (2010) used the PMF to reveal that VOCs in pollution days in Shanghai were derived from industrial sources (29%), vehicle emission (25%), solvent usage (17%), liquefied petroleum gas (LPG) and gasoline evaporation (15%), biomass/biofuel burning (9%), and coal combustion (7%). High levels of VOCs, especially toxic species such as benzene, toluene, ethylbenzene and xylene (BTEX), cause health risks in exposed people (Yan *et al.*, 2015). Dutta *et al.* (2009) investigated the health risks of BTEX in India; they found that high levels of VOCs were mainly derived from transportation sources, and the risk of carcinogenesis can reach 6.1×10^{-5} , which was 1.9 times that of the background point. In Guangzhou, the health risk of BTEX was 5.34×10^{-5} , which was much higher than the recommended level of 1×10^{-6} (Li *et al.*, 2015).

Based on the potential harm of VOCs in haze pollution, few studies have investigated the ability of VOCs to form SOA and the effects on public health under different pollution conditions in North China. In this study, characteristics, source contribution, SOA formation potential (SOA_{fp}), and health risk assessment of VOCs were analyzed during pollution and clean periods in Yuncheng. The source of VOCs was identified using the PMF, and combining the fractional aerosol coefficient (FAC) method, we also calculated the SOA_{fp} of each VOC source. In addition, we use the health risk exposure model of the United States Environmental Protection Agency to estimate the potential risks of BTEX in exposed people. Three main objectives were to determine the major sources of VOCs by comparing the proportion of sources in pollution and clean periods, to calculate SOA_{fp} and determine the qualitative relationship between VOCs and PM_{2.5}, and to explore the sources and compounds with higher contribution to public health risks.

MATERIALS AND METHODS

Sampling Sites

Yuncheng is located in the south of Shanxi Province, covering 13,968 km² with a population of approximately 5,600,000. With rapid development in economy and sharp increase in energy consumption, PM_{2.5} in Yuncheng is at a higher level than that in other cities in Shanxi in North China (Fig. 1). In this study, sampling was conducted from January 20 to 24, 2018. The sampling points were located on the top of Yuncheng Middle School (35.05°N, 111.03°E), 25 m above the ground level, and the sampling site was close to the central urban area, which includes residential, commercial, and office mixed areas and reflect the regional pollution level of Yuncheng. According to the air quality index (AQI) value, the VOC sampling time was divided into the polluted period with AQI > 200 (January 20–22) and clean period with AQI < 200 (January 23–24). VOCs were sampled from 06:00 to 20:00 (at a 2-h interval) using SUMMA canisters (Silonite® Canisters; Entech Instruments Inc., CA, USA).

Chemical Analysis

In this study, gas samples were preprocessed using a Model 7100 VOC preconcentrator (Entech Instruments Inc., USA) and analyzed through a gas chromatographer equipped with a mass-selective detector (7890A GC/5975C MSD/FID; Agilent Technologies, USA). Firstly, gas samples (300 mL) were drawn into a cold trap Module 1. The samples were cooled at −160°C and desorbed at 10°C. N₂, O₂, and some argon (Ar) were separated on the cold trap Module 1. Secondly, the samples desorbed from Module 1 were concentrated at Module 2 at −40°C. CH₄, CO₂, some water, and the rest of Ar were separated from the cold trap

Data Source: YunCheng's Environmental Protection Bureau, January 2018

Fig. 1. (a) The spatial distribution of PM_{2.5} in Yuncheng and surrounding cities during the whole observation. (b) The sampling site was in Yuncheng Middle School.

Module 2. Third, the samples were frozen in an empty capillary column (trap Module 3) at -150°C . After being frozen, the samples from empty capillary column were heated and vaporized quickly. All target species were transferred under high-purity (99.999%) helium to the DB-1 capillary column ($60\text{ m} \times 0.32\text{ mm} \times 1.0\text{ mm}$; Agilent Technologies, USA) for separation. Then, $\text{C}_2\text{--C}_3$ hydrocarbons were separated on a PLOT-Q column ($30\text{ m} \times 0.32\text{ mm} \times 2.0\text{ mm}$; Agilent Technologies, USA) and detected using an FID, whereas other species of $\text{C}_4\text{--C}_{12}$ hydrocarbons were trapped in the passivated empty capillary column and detected using an MSD. To ensure data validity, quality assurance and control (QA/QC) procedures were performed for the certification of all samples, and the QA/QC procedures performed in this study followed those by Yan *et al.* (2015).

Data Analysis

SOAfp

SOAfp represents the maximum propensity for VOCs to form SOA. In this study, FAC_i was used to estimate SOAfp directly. The equation is as follows:

$$\text{SOAfp}_i = \text{VOCs}_{0i} \times \text{FAC}_i \quad (1)$$

where SOAfp_i is the SOAfp of VOC species i , $\mu\text{g m}^{-3}$; VOCs_{0i} is the initial concentration of VOC species i calculated by Eq. (2), $\mu\text{g m}^{-3}$; and FAC_i is the SOA-generating coefficient of VOC species i .

$$\text{VOCs}_{ti} = \text{VOCs}_{0i} (1 - \text{F}_{\text{VOC}i}) \quad (2)$$

where VOCs_{ti} is the concentration of VOC species i measured in this study, $\mu\text{g m}^{-3}$. $\text{F}_{\text{VOC}i}$ is the reacted percentage of VOC species i . Among them, FAC_i and $\text{F}_{\text{VOC}i}$ are provided by Grosjean *et al.* (1989) from multiple smoke box experiments, which could estimate the SOA concentration from measured VOCs and reflect the contribution of SOA precursors. The equation is based on the following assumptions: SOA is only generated during daytime (08:00–17:00), and the removal of VOCs is governed by the reaction with OH radicals.

PMF

The PMF model is widely used for analyzing the source apportionment of VOCs in the atmosphere (Yan *et al.*, 2017; Hui *et al.*, 2018), especially suitable for the consecutive collecting of VOC samples like in this study (Norris *et al.*, 2014). PMF is an advanced receptor model that decomposes a matrix of sample data (X) into two matrices, the source contribution matrix (G) and the source matrix (F), the mass amount of a species can be calculated from the contribution of sources to samples and the species profile of each source, as shown in Eq. (3) (Norris *et al.*, 2014):

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (3)$$

where x_{ij} is the concentration of j^{th} species measured in the

i^{th} sample, g_{ik} is the contribution of the k^{th} source to the i^{th} sample, f_{kj} is the fraction of j^{th} species from the k^{th} source, e_{ij} is the residual for each sample/species, and p is the total number of the sources.

Eq. (4) usually leads to uncertainty due to the error fraction of species concentrations and the method detection limit (MDL; ppbv) (Norris *et al.*, 2014). The PMF analysis minimizes the objective function Q based on the uncertainties:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left[\frac{x_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right]^2 \quad (4)$$

where n and m is the number of samples and the number of VOC species, respectively, and u_{ij} is uncertainty of the j^{th} species measured in the i^{th} sample.

In the PMF analysis, the uncertainties of the input datasets were calculated using Eqs. (5) and (6) (Norris *et al.*, 2014). If the VOC concentration (Conc.) is less than MDL, Eq. (5) is adopted. Otherwise, Eq. (6) is adopted:

$$Unc = \frac{5}{6} \times MDL \quad (5)$$

$$Unc = \sqrt{(\text{Error Fraction} \times \text{concentration})^2 + (0.5 \times MDL)^2} \quad (6)$$

We used the USEPA PMF 5.0 model to perform source apportionment for VOCs in the atmosphere. The principles of PMF were as follows: Species with high proportions of missing samples were excluded, and data below the MDL were substituted with $0.5 \times MDL$; species with high reactivity were excluded because they were rapidly consumed in the atmosphere (exclude some trace compounds, such as isoprene); and species with a lower mixing ratio should be avoided because they were not important tracers of VOC sources. Eventually, considering the fundamental assumption of non-reactivity or mass conservation of the PMF model, 38 VOC species were identified and quantified for source apportionment in this study.

Health Risk Assessment

To identify the effects of pollution sources on human health, we used the Integrated Risk Information System (IRIS) by USEPA to estimate the health risk of specific VOCs in this study. The risk of inhalation of non-carcinogenic and carcinogenic VOCs was assessed using the following equation:

$$EC = (\text{CA} \times \text{ET} \times \text{EF} \times \text{ED}) / \text{AT} \quad (7)$$

where EC ($\mu\text{g m}^{-3}$) represents inhalation exposure concentrations, where CA represents the concentration of pollutants, $\mu\text{g m}^{-3}$. ET represents the exposure time, 24 h d^{-1} . EF represents the exposure frequency, 365 d y^{-1} . ED represents the exposure duration (70 y). AT represents the

average exposure time, $70 \times 365 \times 24$ h. Based on Eq. (1), the sustained exposure concentration is assessed.

In this study, carcinogenic (R) and non-carcinogenic risks (HQ) of all risk-posing VOC species were estimated using Eqs. (8), (9), (10), respectively:

$$R = EC \times IUR \quad (8)$$

$$HQ = EC / (RfC \times 1000) \quad (9)$$

$$HI = \sum HQ \quad (10)$$

where R is the estimated inhalation cancer risk. IUR ($\text{m}^3 \mu\text{g}^{-1}$) represents the inhalation unit risk, and data are from U.S. EPA's IRIS. Normally, cancer risks no higher than 1×10^{-6} was regarded as acceptable. Where HQ is the non-cancer risk hazard quotient (HQ) of specific VOCs, RfC (mg m^{-3}) represents chronic reference concentration, mg m^{-3} . Where HI represents the sum of HQs of several pollutants. Namely, if the value of HQ or HI is below 1, there is no chronic toxicity. On the contrary, if the value of HQ exceeds 1, chronic toxicity will occur after long-term exposure.

RESULTS AND DISCUSSION

Overall Level of VOCs in Yuncheng

The concentrations of VOCs were determined in pollution, clean, and whole observation periods in Yuncheng (Table 1). The total concentration of VOCs in the atmosphere was $87.70 \mu\text{g m}^{-3}$, and 47 major VOC species were measured, including 26 alkanes, 10 alkenes, 11 aromatics, and 1 alkyne. Alkanes and aromatics were the most abundant species, accounting for 61.04% and 22.84% of VOCs, respectively. Alkyne contained acetylene only but represented 6.30% of VOCs. In addition, among VOCs, 5 compounds, namely propane ($13.55 \mu\text{g m}^{-3}$), *n*-butane ($12.48 \mu\text{g m}^{-3}$), benzene ($8.89 \mu\text{g m}^{-3}$), ethane ($5.92 \mu\text{g m}^{-3}$), and acetylene ($5.52 \mu\text{g m}^{-3}$), were the dominant species, which accounted for 52.89% of VOCs. The level of VOCs in the polluted period ($105.29 \mu\text{g m}^{-3}$) was 2.3 times that in the clean period ($46.03 \mu\text{g m}^{-3}$). Alkanes showed the highest concentration, and VOC composition characteristics were basically same during the whole observation period, indicating that emission sources may be consistent in pollution and clean periods.

Comparisons of VOC level in winter in Yuncheng and other northern and southern cities in China are shown in Fig. 2. The VOC concentration during the observation period was higher in Yuncheng than in winter in Beijing ($66.90 \mu\text{g m}^{-3}$), Tianjin ($39.40 \mu\text{g m}^{-3}$), and Wuhan ($44.87 \mu\text{g m}^{-3}$) and was

lower to that in winter in Jincheng ($93.34 \mu\text{g m}^{-3}$) (Liu et al., 2016b; Lyu et al., 2016; Wei et al., 2018; Yang et al., 2019), which indicated that VOC concentration in Yuncheng was at a relatively middle level among northern cities. Concentrations of alkanes and VOCs in Yuncheng were prominently higher than those in the southern Chinese cities (An et al., 2011; Xu et al., 2015; Liu et al., 2011; Song et al., 2019b). Alkanes and aromatics in Yuncheng were higher than other cities, such as Beijing, Tianjin and Wuhan (Liu et al., 2016b; Lyu et al., 2016; Wei et al., 2018). Barletta et al. (2005) showed that coal combustion in winter leads to emit VOCs with high-content alkanes and aromatics. In addition, a high demand for coal combustion in winter may be a key factor for VOC pollution in Yuncheng.

Variation of VOCs in Different Pollution Periods

Daily variations of wind speed, relative humidity, temperature (T), air quality index (AQI), VOC species, and VOCs are shown in Fig. 3. We observed that the proportion of alkanes (61.04%) and aromatics (22.84%) was higher in the whole observation period. Nelson et al. (1984) reported that the high concentration of alkanes and aromatics highlighted the strong effects of coal combustion. The diurnal variation in the VOC concentration was consistent with AQI. Poor meteorological conditions may lead to the accumulation of pollutants, higher AQI, and excessive VOCs. During the polluted period, the average wind speed was 1.71 m s^{-1} . The concentrations of VOCs, AQI, and $\text{PM}_{2.5}$ were 105.29, 286, and $237 \mu\text{g m}^{-3}$, respectively. On January 23, the wind shifted from north to south, with a temporarily increasing wind speed (11.19 m s^{-1}), which rapidly reduced the concentration of VOCs in the atmosphere to $43.87 \mu\text{g m}^{-3}$. The concentrations of AQI and $\text{PM}_{2.5}$ were 124 and $94 \mu\text{g m}^{-3}$, respectively.

Snowfall occurring on January 24 was also responsible for the removal of pollutants, including VOCs. Fries et al. (2008) revealed that gas and particle scavenging by snow/ice have the potential to significantly contribute to the deposition of volatile organic compounds by washing out the aerosol particles. The concentration of VOCs decreased significantly after snowfall (Fig. 3). Because Yuncheng belongs to a basin type, the accumulation of pollutants is easy due to poor meteorological conditions such as low wind speed and high humidity, whereas the wind speed increases, pollutants rapidly cleared in a relatively short time. VOC concentrations during sampling time have a reverse tendency with wind speed during, especially while pollution happened and ended (Fig. 3).

Diurnal Variation during Pollution and Clean Periods

The diurnal variation characteristics of VOCs observed in

Table 1. VOC concentrations measured in pollution, clean period and whole observation in Yuncheng.

Species	Polluted period	Clean period	Whole observation
	Mean $\pm$ S.D. ($\mu\text{g m}^{-3}$)	Mean $\pm$ S.D. ($\mu\text{g m}^{-3}$)	Mean $\pm$ S.D. ($\mu\text{g m}^{-3}$)
Alkanes	64.40 ± 20.30	27.61 ± 8.22	53.5 ± 25.36
Alkenes	9.82 ± 4.87	5.78 ± 1.88	8.80 ± 4.79
Alkynes	6.37 ± 2.56	3.53 ± 1.23	2.60 ± 5.52
Aromatics	24.69 ± 9.63	9.10 ± 2.89	20.02 ± 11.12
VOCs	105.29 ± 37.37	46.03 ± 14.22	87.70 ± 45.87

Fig. 2. Comparison of the ambient levels of VOCs among the study area and other cities.

Fig. 3. Diurnal variation of the concentration of VOCs, AQI and meteorological conditions during whole observation.

this study were the same as those also reported in winter in Jincheng, a city just located near Yuncheng (Yang *et al.*, 2018), which revealed that the level of VOCs increased significantly at 08:00–10:00 and 18:00–20:00. Compared with the clean period, VOCs had obvious diurnal variation during the polluted period, as illustrated in Fig. 4. Na *et al.* (2001) demonstrated that the diurnal variation of VOCs was

influenced by relevant VOC source strength, photochemical reaction intensity, and diffusion conditions. The peak traffic of Yuncheng was at 08:00, which was consistent with the timing of increases in the VOC concentration. In addition, VOCs were less consumed in the atmosphere because of weak light intensity in the morning, resulting in a high concentration. In the afternoon, VOCs were obviously reduced

Fig. 4. Diurnal variation of VOCs during the observation: polluted period vs. clean period in Yuncheng.

by the increase in temperature and intense photochemical reactions, accelerating the conversion of VOCs into other substances.

Comparison of VOC Compounds in Different Periods

By studying VOC dominant species, we will be able to clearly understand which sources require controlling during heavy pollution to some extent. Thus, the top 15 VOC species were compared in pollution and clean periods. The composition of the top 15 species was basically consistent in pollution and clean periods (Fig. S1). Na *et al.* (2004) mentioned that aromatics in the atmosphere were mainly derived from coal combustion and solvents. Wang *et al.* (2017) found that vehicle sources emit more alkenes, such as ethene and propene. The high proportion of aromatics and alkynes in Yuncheng may be associated with the increased demand for coal combustion in winter and the rapid growth in vehicle ownership in the past few years. Propane, *n*-butane, and benzene reached a high level in the polluted period, which was much higher than that in the clean period. Song *et al.* (2019a) showed that propane, *n*-butane, and benzene were typical tracers of vehicle emission, which indicated vehicle source emission was significantly increased in the polluted period.

Source Analysis

Diagnostic Ratios

Diagnostic ratios were used to make simple identification and judgments on VOC sources. The B/T ratio is commonly used to identify the traffic source (Ho *et al.*, 2004; Bruno *et al.*, 2006; Lokesh *et al.*, 2015). In this study, the B/T ratio was observed to be 0.35–0.68, which was consistent with that reported in Beijing (Wang *et al.*, 2012), demonstrating that vehicle emission might be the main VOC source. In addition, the ratios of propane and *n*-butane to acetylene were used to determine the contributions of LPG leakage (Lai *et al.*, 2009). In this study, the ratios of propane and *n*-butane to acetylene were 3.08 and 2.89, respectively, during the polluted period, which was above the observation levels in Guangzhou (1.2 and 0.6, respectively; Zhang *et al.*, 2013). This phenomenon suggested that LPG/natural gas (NG)

evaporation may have a higher contribution during the polluted period in the atmosphere in Yuncheng.

PMF Analysis

In this study, source apportionment was conducted based on PMF 5.0. There was a good correlation ($R^2 = 0.96$) between the observed concentration and the simulated concentration by PMF, which showed the source analysis results could explain VOC pollution very well in Yuncheng. Seven possible VOC sources were identified, namely vehicle emission, coal combustion, LPG/NG, industrial sources, gasoline evaporation, solvent usage, and biogenic emission. The percentage contribution of the source characteristics is shown in Fig. S2.

Factor 1 was identified as the solvent usage source because of the high proportion of aromatics species, such as ethylbenzene (40.58%), *m*-xylene (63.39%), and *p*-xylene (73.97%), which are typical tracers of solvent usage (Yuan *et al.*, 2010). Factor 2 was characterized by high percentages of C₃–C₆ alkanes (propane (31.85%), isopentane (27.11%), and 2-methylpentane (65.45%)), and a few alkenes were closely related to vehicle emission; these findings are the same as those in previous studies (Grosjean *et al.*, 1989; Cui *et al.*, 2018; Deng *et al.*, 2018). Factor 3 was distinguished by a significant percentage of isoprene (80.91%). Because isoprene is an indicator of biogenic emission (Kesselmeier *et al.*, 1989), Factor 3 was mainly attributed to biogenic sources. Factor 4 was characterized by high percentages of C₅–C₇ alkanes, especially isopentane (50.90%) and 2,3-dimethyl-butane (50.31%). Isopentane is a typical tracer of volatile gasoline (Liu *et al.*, 2008; Eleonora *et al.*, 2008). Therefore, this factor was identified as gasoline evaporation. Factor 5 was identified by a high percentage of ethane (74.29%), ethene (63.09%), and acetylene (41.64%). Those species were all closely related to coal combustion (Zheng *et al.*, 2018). Thus, Factor 4 was likely derived from coal combustion. Factor 6 presented the main components from propane (34.10%), *n*-butane (53.67%), and *n*-hexane (36.28%). McCarthy *et al.* (2013) revealed that propane and *n*-butane are the most abundant species in LPG/NG. LPG/NG was the third largest source because LPG has been widely used as a

new energy source in North China Plain in the past few years. Thus, Factor 6 was suggested as the source of LPG/NG. Factor 7 was characterized by high values of C₆–C₉ alkanes and BTEX compounds (Hui *et al.*, 2019), such as *n*-nonane (54.10%), ethylbenzene (54.16%), 1,3,5-trimethyltoluene (54.91%), 1,2,4-trimethyltoluene (66.92%), and 1,2,3-trimethyltoluene (59.44%). Therefore, Factor 7 was identified as industrial emission.

The source contributions to VOCs during the whole observation period, polluted period, and clean period are given in Fig. 5, which revealed that vehicle emission were the largest contributors in the polluted period (responsible for 29.04% of VOCs), followed by coal combustion source (21.56%), LPG/NG (19.46%), industrial sources (16.25%), gasoline evaporation (6.51%), solvent usage (5.10%), and biogenic emission (2.08%). However, the proportions of vehicle emission decreased sharply in the clean period (16.75%) and industrial sources slightly reduced (11.88%). During January 20–22 in 2018 of sampling period, Yuncheng issued heavy atmospheric pollution warning announcement to reduce pollutant emission, including VOCs (<http://shanxi.sina.com.cn/news/report/2018-01-19/detail-ifyqtzww6025167.shtml>). The contribution of industrial emissions to VOCs in the atmosphere was gradually reduced, mainly due to the industrial production limit and implement the intermittent production of industrial, which was mandatory demanded by the announcement mentioned above. The contribution of LPG/NG to VOCs stabilized at a high degree in the whole observation period, accounting for 16.22–19.46% of VOCs at Yuncheng, which may be because LPG has been encouraged as a major clean fuel for cooking and heating in recent years. Therefore, we speculated that vehicle emission, coal combustion, and LPG/NG were the main contributors in Yuncheng during the whole observation period, which was consistent with the diagnostic ratios. The result suggested that restricting the emission of these three sources can effectively reduce VOC levels. With the full implementation of the “coal-to-gas” and “coal-to-electric” project in Yuncheng, coal used for combustion had decreased sharply from 2015, especially for residential combustion. In the winter of 2018, the coverage of centralized heat supply had increased to over 90% in Yuncheng (<https://www.yuncheng.gov.cn/article/18/06/36365.shtml>), which would significantly reduce burned

coal for decentralized and extensive heating. This would also cause less emission of pollutants, especially VOCs, a product of incomplete coal combustion. In addition, centralized heat supply was established in many northern cities, with an annual reduction of 74 million tons of coal in recent years (<http://www.mee.gov.cn>). In contrast to coal combustion, the number of vehicles increased gradually. In this study, the number of vehicles in Yuncheng increased to 1.15 million in 2018, with a continuous growth rate of 6.48% (<https://www.yuncheng.gov.cn/article/19/04/56225.shtml>). The increase in the vehicle population significantly increased VOC emission from vehicle direct exhaust and gasoline volatilization. From the result of VOC source apportionment in Yuncheng, the contribution of vehicle emission exceeded that of coal combustion in polluted period, which reflected the situation of variation of atmospheric pollution in this northern city in China. Thus, controlling vehicle emission is a key to reduce atmospheric VOCs in this northern Chinese city, which should be a priority in environmental management.

Apportionment of VOC Sources to SOA_{fp}

Yang *et al.* (2019) reported that through a photochemical reaction of VOCs, SOA_{fp} can be a remarkable contributing factor to haze events in winter. The contribution of aromatics in the top 10 species to total SOA_{fp} was 94.35% (Table 2). Toluene was the largest contributor to ambient SOA, contributing 31.28–31.35%. In addition, aromatics were the major component of SOA_{fp}, accounting for 95.96–97.21%. PMF model results indicated solvent usage (40.17%) and industrial emission (30.37%) were the main sources contributing to aromatic in the whole period. Therefore, the main step to reduce SOA in the atmosphere was to control the solvent usage and industrial emission.

Daily variations of SOA_{fp}, VOC, and PM_{2.5} concentrations are displayed in Fig. S3(a). SOA_{fp} peaked at 06:00–08:00 in both the pollution and clean periods. The determined SOA_{fp} values were 30–240 $\mu\text{g m}^{-3}$ during the whole observation period, with an average of 120 $\mu\text{g m}^{-3}$, which was higher than that reported in the Beijing haze episode, where the average level of SOA_{fp} was 50 $\mu\text{g m}^{-3}$ (Han *et al.*, 2015). However, SOA_{fp} obtained through the FAC method showed trends quite similar to VOC variation, illustrating the key role of VOC levels in SOA_{fp}. However, the time change trend of

Fig. 5. Source contributions to the VOC concentration in the PMF model during different pollution periods.

Table 2. The top 10 SOAfp species during pollution and clean periods.

VOCs species	Polluted period		Clean period	
	SOAfp ($\mu\text{g m}^{-3}$)	Contribution (%)	SOAfp ($\mu\text{g m}^{-3}$)	Contribution (%)
Toluene	33.07	31.35	12.63	31.28
Benzene	24.66	23.37	8.33	20.62
<i>m</i> -Xylene	13.43	12.74	4.22	10.45
1,3,5-Trimethylbenzene	2.57	2.44	1.14	2.83
<i>p</i> -Xylene	6.40	6.07	3.11	7.70
Ethylbenzene	7.41	7.03	3.40	8.40
<i>o</i> -Xylene	6.80	6.45	2.74	6.78
Methylcyclohexane	2.71	2.57	1.00	2.48
1,2,4-Trimethylbenzene	3.34	3.16	1.05	2.60
1,2,3-Trimethylbenzene	2.15	2.03	1.14	2.82
Sum of top 10	102.55	97.21	38.77	95.96

SOAfp and VOCs was 2–3 h earlier than the change trend of $\text{PM}_{2.5}$, indicating that SOA and VOC undergo complex multiphase physicochemical processing for the formation of $\text{PM}_{2.5}$ (Zhang *et al.*, 2016). In the whole observation period, alkanes and alkenes did not contribute significantly to SOAfp, with the relative contributions of 0.94% and 1.90%, respectively, whereas aromatics contributed 97.16% to SOAfp (Fig. S3(b)). During the polluted period, aromatics increased by 52.93% at 06:00–08:00 on January 22, and correspondingly, SOAfp rapidly increased from $107.10 \mu\text{g m}^{-3}$ to $227.04 \mu\text{g m}^{-3}$. Moreover, SOAfp significantly reduced from $100.97 \mu\text{g m}^{-3}$ to $32.68 \mu\text{g m}^{-3}$ as the concentration of VOCs decreased in the clean period. These results indicated that aromatics were the major contributors to SOA in the atmosphere, and identifying the aromatics emission source is important.

A strong correlation existed between SOAfp and VOCs ($R^2 = 0.86$), indicating that the increase in SOAfp was induced by higher levels of VOCs. The correlation between $\text{PM}_{2.5}$ and SOA was strong during the whole observation period ($R^2 = 0.54$). As an important component of $\text{PM}_{2.5}$, SOA had a positive correlation with $\text{PM}_{2.5}$, which indicated that SOA had a key role in $\text{PM}_{2.5}$ generation. Zhang *et al.* (2013) reported that the contribution of SOA to $\text{PM}_{2.5}$ mass was greater during the polluted period (31%) than during the clean period (6.82%) in the urban sites of Beijing.

The source apportionment results revealed that industrial emission apportioned 26.26% of SOAfp contributors during the whole observation period (Fig. 6). Industrial emission (28.62%) and solvent usage (37.07%) were the main sources contributing to SOAfp in the pollution and clean periods, respectively. In contrast to the polluted period, the contribution of vehicle emission and industrial emission to SOAfp decreased from 27.88% to 14.32% and from 28.62% to 20.75% in the clean period, which may result from the limited traffic and factory closedown. Solvent usage increased from 18.38% to 37.07%. Solvent usage was efficient to form SOA than other sources, which may include compounds with high reactivity that result in high SOAfp levels. In addition, the contribution of solvent usage to SOA was higher than that to VOCs. Thus, controlling the emission sources of solvent usage, vehicles, and industries can effectively reduce the SOAfp level during the polluted period.

Health Risk Assessment of Aromatics

As a standard to estimate the overall risk, 6 aromatics (benzene, toluene, ethylbenzene, *p*-xylene, *o*-xylene, and styrene) were characterized in health risk assessment. The HQ of non-cancer risk and carcinogenic risk (R) in the polluted period were 6.61×10^{-2} and 5×10^{-5} , which were higher than those in the clean period (2.20×10^{-2} and 2.85×10^{-5}), respectively. Benzene exhibited the maximum HQ and R, attributing to its greater toxicity. The HQ of individual compounds did not exceed unity, but the HI value (2.3) indicated that cumulative exposure to these benzene series may cause potential non-carcinogenic risk to the public. R (2.85×10^{-5} to 5×10^{-5}) of benzene was above the limit (1×10^{-6}), indicating that benzene had a potential carcinogenic risk to health if the population had long-term exposure of benzene in the study area. Hence, benzene emission sources should be controlled for the exposed population health. Reducing benzene from some particular emission sources, such as the solvent usage and industrial emission, was quite important for public health.

CONCLUSIONS

To determine the variation of VOCs during different pollution periods in winter, VOC samples were passively analyzed in Yuncheng.

The VOC concentration averaged $87.66 \mu\text{g m}^{-3}$ across the entire observation period. The levels of alkanes, alkenes, alkynes, and aromatics were substantially higher during the polluted period, when particularly high proportions of alkanes (61.04%) and aromatics (22.84%) were found. The VOC concentration was 2.3 times higher during the polluted period than the clean period; furthermore, it was affected by meteorological conditions, such as wind speed or snowfall. Vehicle emission (30.04%), coal combustion (21.56%), and LPG/NG evaporation (19.46%) were the dominant contributors of VOCs during the polluted period, which suggests that controlling these three factors would efficiently reduce VOCs.

Aromatics were the major precursors of the SOA. The source apportionment results revealed that solvent usage (40.17%) and industrial emission (30.37%) were the primary contributors to the SOAfp during the entire observation

Fig. 6. The potential contribution of sources of VOCs in Yuncheng to SOA during different pollution periods.

period. Thus, controlling vehicle emission and solvent usage could effectively decrease the SOA_{fp} during polluted periods.

Although the non-carcinogenic HQ was found to be acceptable, the HI value was 2.3, indicating that cumulative exposure to the aromatics in this city potentially results in non-carcinogenic risks to the public. Additionally, benzene exceeded the permissible limit (1×10^{-6}) in terms of carcinogenic risk (2.85×10^{-5} to 5×10^{-5}). Because aromatics are toxic substances as well as the major precursors of SOA, controlling solvent, vehicle, and industrial emissions will benefit public health and air quality.

In North China, large quantities of pollutants, including VOCs, are emitted due to the prevalence of coal combustion for heating during winter. However, the results of this study show that the contribution to VOCs from vehicle emission has surpassed that of coal combustion in Yuncheng, especially on heavily polluted days. A key factor has been the implementation of the “coal-to-gas” and “coal-to-electric” projects, which has reduced coal combustion during winter, and, based on the current policies, industrial restructuring will also decrease the contribution from this source (Yu *et al.*, 2018). Therefore, to control VOC pollution in Yuncheng, a typical city in North China, it is necessary to strengthen the regulation of vehicle emissions in addition to maintaining the limits on coal combustion.

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DISCLAIMER

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be

found in the online version at <http://www.aaqr.org>.

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