Spatiotemporal Distributions of Ambient Volatile Organic Compounds in China: Characteristics and Sources

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ABSTRACT

Ambient volatile organic compounds (VOCs) play a critical role in air quality as important precursors for the formation of ozone and secondary organic aerosols. The characteristics of ambient VOCs vary among different regions. In this study, characteristics and sources of VOCs from 33 sites in 32 major cities across China were compiled from field measurements and literature. The annual average VOC concentrations ranged from 6.7 to 75.0 ppbv and featured spatial variations, with a high value in northwestern China (62.7 ± 17.5 ppbv) but no significant difference among northern, central, southern, and southwestern China (32.0–36.0 ppbv). The VOC concentrations in Qingdao and Fuzhou were consistent with the background values for the region (7.6 ± 1.2 ppbv). VOC concentrations exhibited different seasonal variations across China. High concentrations occurred in winter and autumn, especially in cities in northern China, attributed to the combined contributions from weather and heating activities. The annual average chemical compositions of ambient VOCs across China were dominated by alkanes (36.5%–69.3%), followed by aromatics (7.7%–47.8%), alkenes (6.9%–53.4%), and alkynes (0.7%–15.0%). Chemical compositions also exhibited different seasonal variations across China, depending on the emission and consumption of VOCs. Vehicle exhaust, natural gas/liquefied petroleum gas, fuel combustion, and solvent usage were the dominant VOC contributors, followed by industrial, fuel evaporation, and biogenic sources. Listing VOCs as a mandatory monitoring pollutant for environmental monitoring stations in China and developing accurate source apportionment methods are critical to guide policy making and for understanding of the spatiotemporal distribution of ambient VOCs.

Keywords: VOCs, Chemical composition, Spatiotemporal distribution, Source apportionment

1 INTRODUCTION

Ambient volatile organic compounds (VOCs) have recently elicited public concern due to their important roles in the formation of ozone and secondary organic aerosols, as well as their potential adverse effects on public health.
Understanding the concentration and spatiotemporal distribution of ambient VOCs is conducive to formulating emission reduction schemes. Several studies have focused on the characteristics of ambient VOC concentrations, such as spatial variation (Wang et al., 2020b; Wang et al., 2017), seasonal variation (Chen et al., 2021; Liu et al., 2020; Sun et al., 2021), and diurnal variation (Qin et al., 2021; Yao et al., 2021a) during episode or non-episode days (Hui et al., 2021; Fan et al., 2021) or major events (e.g., Asia-Pacific Economic Cooperation and National Traditional Games; Wang et al., 2016; Zhao et al., 2020a). For example, significant seasonal differences were found for VOC concentrations and compositions in Beijing (Gu et al., 2019) and Xi’an (Sun et al., 2021), with winter values that were double those in summer. Additionally, average VOC concentrations have exhibited obvious diurnal variations, with high values in the morning and evening and low values in the afternoon due to photochemical reactions (Qin et al., 2021; Xiong et al., 2021).

Quantifying the chemical composition of VOCs is critical for the effective control of ozone and particle pollution because the contribution of each component to the formation of ozone and secondary organic aerosols varies greatly (Cheng et al., 2016; Han et al., 2017; Yao et al., 2021b). Alkanes were the most abundant components in Beijing (Liu et al., 2020), Chengdu (Simayi et al., 2020), Wuhan (Hui et al., 2020), Xi’an in winter and summer (Sun et al., 2021), and at a background site in central China (Li et al., 2021). However, in Xinxiang in autumn, the most abundant components were aromatics (Zhang et al., 2020b).

Identifying the contribution sources for VOCs is also critical for developing strategies to control and reduce ozone and secondary organic aerosols. Vehicle exhaust (specified as diesel and gasoline vehicles) was the primary source of VOCs in Beijing (accounting for 33.8% annually; Liu et al., 2020), Chengdu (accounting for 31.0% annually; Simayi et al., 2020), Xi’an (accounting for 30.6%–48.4% in summer; Song et al., 2021; Sun et al., 2021), and Nanjing (accounting for 23.0% in summer; Fan et al., 2021); however, they only contributed 7.7% of the total VOCs at a rural site in the North China Plain (Zhang et al., 2020a). Natural gas (NG)/liquefied petroleum gas (LPG) usage, solvent usage in painting/coating, and vehicle exhaust were the dominant VOC sources in Wuhan in spring (Hui et al., 2020).

A few studies have reviewed VOC characteristics in China. Wang et al. (2021) published an overview of the current non-methane VOC pollution in China by focusing on analytical methods, spatiotemporal variations, photochemical characteristics, and sources of non-methane VOCs in urban areas. Zhang et al. (2017) presented the characteristics and sources of ambient VOCs in 23 major Chinese cities; moreover, their calculations of the ozone formation potential (OFP) and secondary organic aerosol formation potential (SOAFP) of various VOCs indicated high OFPs for aromatics and alkenes and high SOAFP for aromatics. Li et al. (2017) summarized the distribution and source apportionment of VOCs in six major Chinese cities and provided a detailed discussion on the effects of VOCs on OFP and SOAFP.

The spatiotemporal distribution of characteristics and sources of VOCs across China may vary greatly due to industrial structure, geographical and meteorological conditions, and seasonal and diurnal differences among regions. However, limited information is available on the spatiotemporal distribution of these characteristics. Understanding ambient VOC pollution is key in formulating policies. Therefore, it is crucial to investigate the spatiotemporal distribution of ambient VOCs in China for the effective control of VOC pollution. In this study, we examined the ambient VOC concentrations across China based on data from 33 sites in 32 major cities. The spatiotemporal distribution of the characteristics and sources of VOCs are discussed.

### 2 METHODS

#### 2.1 Sampling Sites

Characteristics and sources of VOCs from 33 sites in 32 major cities across China were compiled from field measurements and literature. Linyi (117.40°E–119.18°E, 34.37°N–36.22°N), which is in the eastern part of the North China Plain and is the largest and most populous city in Shandong Province, was selected for field measurements in this study. The Atmospheric Environment Super Station (AESS) site located in the Lanshan district of Linyi was selected for online VOC monitoring (Fig. S1). The sampling site was placed on the rooftop of the AESS (10 m above ground level). The AESS site was surrounded by commercial, cultural, and residential areas to the north and...
industrial and rural areas to the east and west; the Yi River was nearby to the east, west, and south. Roads with moderate traffic were located 270 and 100 m away on the east and north side of the sampling site, respectively. Therefore, although there was only one monitoring site, it was representative of the mixed urban environment in Linyi, with educational activities, traffic, and residential areas. Table S1 provides detailed information of sampling sites, including measurement time, number of VOC compositions, and sampling methods.

2.2 Sampling and Analysis Methods
Detailed information on the sampling methods used for each site (#1–#33) is available in Table S1. In the present study, the VOC samples were collected using an online monitoring system with a time resolution of 1 h and subsequently measured using gas chromatography–mass spectrometry (GC–MS) (Shanghai Panhe Scientific Instrument Co., Ltd, Shanghai, China) in Linyi. A chromatographic column (30 m × 0.25 mm × 1.4 µm) was used in the GC system. For GC analysis, the initial temperatures were held at 35 °C for 0.5 min, increased to 110°C at a rate of 20°C min⁻¹, and then increased to 210°C at a rate of 35°C min⁻¹. The ion source temperature and transmission line temperature were both 250°C in the MS system. The monitoring mode is full scan ranging from is 29 to 300 amu. In total, 55 chemical compositions for VOCs were determined with the method detection limit of 0.01 ppb, including 29 alkanes, 10 alkenes, 15 aromatics, and 1 alkyne (Table S2). The sampling period ran from January to December 2020, in which March to May represented spring, June to August represented summer, September to November represented autumn, and December to February represented winter, respectively.

2.3 Quality Assurance and Quality Control
A blank experiment was also performed. The canisters were pre-cleaned and vacuumed in ultra-pure nitrogen (N₂, 99.999%) to remove water vapor, air, and any organic compounds. The canisters were then filled with ultra-pure nitrogen, and an online test was conducted. The online monitoring system was used to confirm the lack of adsorption or pollution through a blank analysis. Photochemical assessment monitoring station (PAMS)–certified gasses (Spectra, USA) were used to create a calibration curve for each target compound at four different concentrations (0, 5, 10, and 20 ppbv). The correlation coefficients were all above 0.99, which showed good linearity in the relationship between the integral areas of the peaks and the corresponding concentrations. The stability of the online monitoring system for long-term operation was verified using a VOC standard gas with a 2.0 ppbv concentration at 0:00 every day. If the ratio of the quantitative result of the target component to the theoretical value ranged from 0.7 to 1.3, the monitoring system was considered stable and reliable and was used for sample analysis. Otherwise, it was necessary to calibrate the system and establish a new calibration curve.

3 RESULTS AND DISCUSSION

3.1 VOCs Concentrations in China
3.1.1 Average annual VOC Concentrations exhibited spatial variation
Based on our field measurements and recent publications, we summarized VOC concentrations measured over 29 urban sites and four background sites (Changbai Mountain in Baishan, the Ecological Protection Zone in Shenzhen, a plateau in Lhasa, and Gongga Mountain in the Tibetan Autonomous Prefecture of Garzê) in China from 2007 to 2020, to analyze the spatial variations in VOC pollution (Table S1). We divided the study area into the following eight regions based on geographical location: northern China (NC), northeastern China (NEC), eastern China (EC), central and southern China (CSC), southwestern China (SWC), northwestern China (NWC), Hong Kong, and the background regions (Table S3 and Fig. 1). A total of 5, 2, 10, 6, 3, 2, 1, and 4 sites were collected in NC, NEC, EC, CSC, SWC, NWC, Hong Kong, and the background regions, respectively (Fig. 1).

The annual average VOC concentrations across all sites ranged from 6.7 to 75.0 ppbv; this is approximately an 11.2-fold difference across the nation, indicating the extent of spatial variations in VOCs (Fig. 1). The average annual VOC concentration was high in the NWC region and ranged 50.3–75.0 ppbv (with an average of 62.7 ± 17.5 ppbv) (Fig. 2). As one of the major cities in the
Fig. 1. VOC concentration and major chemical composition in China. Note: The label corresponding to of each pie chart presents the location, average annual VOC concentration, and study period. The color of the line indicates different VOC concentration ranges, < 10, 10–20, 20–30, 30–40, 40–50, > 50 ppbv. The gray line indicates that the unit of VOC concentration is μg m⁻³. Site #25 (background site in Shenzhen) is not included in Fig. 1 because only VOC concentration was reported (9.3 ppbv) without chemical composition information. Areas with different colors on the map represent different regions. The references for different sites and cities are as follows: Beijing (Liu et al., 2020), Tianjin (Liu et al., 2016), Handan (Liu, 2020), Langfang (Song et al., 2019a), Taiyuan (Zhang, 2016), Harbin (Xuan et al., 2011), Changbai Mountain (Wu et al., 2016), Qingdao (Xue et al., 2015), Jinan (Liu et al., 2014), Linyi (in this study), Shanghai (Ye, 2020), Nanjing (An et al., 2014), Wuhu (Gao et al., 2020), Hefei (Zhang, 2016), Ningbo (Zheng et al., 2014), Hangzhou (Jing et al., 2020), Fuzhou (Qiu, 2019), Wuhan (Hui et al., 2018), Changsha (Zhang, 2016), Guangzhou (Zou et al., 2015), Shenzhen (Zhu et al., 2012; Yun et al., 2021), Foshan (Deng et al., 2021), Heshan (Zhang et al., 2020e), Chengdu (Song et al., 2018), Chongqing (Zhai et al., 2013), Kunming (Zhang, 2016), Uhasa (Zhang, 2016), Gongga Mountain (Zhang et al., 2012), Xi’an (Li, 2015), Kuitun (Guo et al., 2019), and Hong Kong (Li et al., 2020).

NWC region, vehicle exhaust and industrial emissions were the dominant VOC contributors in Xi’an, indicating an urgent need to control VOC pollution (Song et al., 2021). The VOC concentrations ranged from 32.0 to 36.0 in the NC, CSC, and SWC regions; the concentration in the EC region was slightly lower (23.0 ± 10.9 ppbv). The VOC concentrations in the NEC region varied greatly, ranging from 84.8 to 150.4 μg m⁻³. Relatively low concentrations were common in the background regions, with a value of 6.7 to 9.3 ppbv (average 7.6 ± 1.2 ppbv). From the perspective of specific cities, the VOC concentrations in Xi’an, Shenzhen, and Kuitun were higher than 50.0 ppbv (50.3–75.0 ppbv), while those in Handan, Nanjing, Guangzhou, Chengdu, Chongqing, and Hong Kong were only slightly lower (41.6–45.3 ppbv) (Fig. 1). The higher concentration in Xi’an was mainly due to the special basin geomorphic structure, which was not conducive to the diffusion of pollutants (Li et al., 2015). The manufacturing industry in Shenzhen is developed, among which furniture, plastic, printing, and spraying are in the leading position in China, consuming a large number of organic solvents and making a significant contribution to VOC concentrations (Zhu et al., 2012). Chemical enterprises were located around the sampling sites of Kuitun and regional transmission might contributed to the VOC concentration of this site (Guo et al., 2019). Therefore, the government faces a considerable challenge to effectively tackle the serious VOC pollution in these cities. The VOC
Fig. 2. Annual average of VOC concentrations in different regions. NWC: northwestern China, PRD: Pearl River Delta, CSC: central and southern China, BTH: Beijing-Tianjin-Hebei, YRD: Yangtze River Delta, SWC: southwestern China, NC: northern China, EC: eastern China. The references for each region are the same as in Fig. 1.

concentrations in Qingdao (9.2 ppbv) and Fuzhou (9.7 ppbv) were consistent with the background regions.

The Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), and Pearl River Delta (PRD) regions are the primary urban areas in China. The spatial variation in VOC pollution in these regions was also examined. The annual average VOC concentration in the PRD was 42.1 ± 19.9 ppbv, higher than those in the BTH and YRD regions, which had average values of 33.8 ± 8.0 and 34.6 ± 12.5 ppbv, respectively (Fig. 2). Emission properties and meteorological conditions play a non-negligible role in the variation of ambient VOCs. VOCs were mainly derived from power, industry, residential, and transportation emissions. The emission inventory of 2017 was used to analyze the spatial variation of VOC concentrations due to different measured time in each site. High VOC concentrations were observed, even though VOC emissions in the PRD were low in 2017, with a value of 2.1 million tons (http://meicmodel.org/). Higher values of 2.7 and 5.6 million tons were found in the BTH and YRD, respectively. Therefore, we speculate that the higher VOC concentration in the PRD region might due to relatively high consumption of ambient VOCs in the BTH and YRD regions, resulting from photochemical reactions.

3.1.2 Concentration exhibited different seasonal variation across China

VOC concentrations exhibited different seasonal variations across China, influenced by the local climate, emission distributions, transport, and chemical transformation. In Handan, Hangzhou, Wuhan, Shenzhen, Foshan, Chengdu, and Xi’an, average VOC concentrations were highest in winter and 1.5–2.4 times lower in summer when they were the lowest. In Beijing, Tianjin, Harbin, Qingdao, and Wuhu, average VOC concentrations were highest in autumn and 1.3–2.4 times lower in spring at their lowest. In Linyi, Fuzhou, and Guangzhou, average VOC concentrations were highest in winter and 1.3–1.5 times lower in spring when they were lowest (Table S1 and Fig. S2). However, the VOC concentration in winter in Guangzhou (47.8 ppbv) was much lower than the results of Luo et al. (2020) with 352.5 ppbv in industrial, 129.2 ppbv in urban, and 75.1 ppbv in rural using Proton-Transfer-Reaction Time-of-Flight Mass Spectrometry (PTR-ToF-MS). This might be the difference of emission sources around the sampling sites.

The high concentrations in winter and autumn, especially for cities in North China (e.g., Handan, Xi’an, Beijing, Tianjin, Harbin, and Qingdao), can be attributed to the combined contributions of meteorological conditions and heating activities. Although lower ambient temperatures in winter and autumn limit the volatilization of VOCs from solvent usage and fuel evaporation sources, they also cause lower photochemical reaction intensity. Moreover, unfavorable meteorological conditions, specifically low wind speed and low boundary layer height, weaken the atmospheric
turbulent exchange and hinder the diffusion of VOCs in the vertical direction (Shen et al., 2018; Wang et al., 2018; Zhang et al., 2013; Zhao et al., 2020b). Additionally, during winter and autumn nights, temperature inversion often occurs, which is conducive to the accumulation of VOCs. More importantly, heating activities, such as coal, gas, and biomass combustion, increased in these cities, leading to higher VOC concentrations. The total amount of urban heat supply, which refers to the total amount of steam and hot water delivered to the city by thermal power plants, thermal power companies, and centralized heating boilers, in major provinces and cities in NC was 91.5 times higher than that in Southern China (Table S4). The sum of VOC emissions in winter and autumn throughout China was 9.2% higher than that in spring and summer (Table S5). Moreover, the abundant precipitation in summer and spring can greatly alleviate VOC pollution. However, in Shanghai and Heshan, the highest VOC concentrations were found in spring due to the location of the sampling sites and local climate, indicating that long-term observation is needed to further clarify seasonal variations.

3.2 Chemical Compositions of VOCs in China

3.2.1 Chemical compositions dominated by alkanes across China

Although several studies have conducted field observations of ambient VOCs during specific seasons (Hui et al., 2020; Song et al., 2021; Zhan et al., 2021; Maji et al., 2020; Zhang et al., 2020a), the characteristics of ambient VOCs vary among different seasons. In addition, the type of VOC chemicals considered varies among studies, including alkanes, alkenes, aromatics, alkynes, OVOCs, and halocarbons. Therefore, only studies that carried out VOC measurements in all four seasons were used, and only alkanes, alkenes, aromatics, and alkynes were analyzed in this study to make the data comparable.

The annual average chemical composition of ambient VOCs was dominated by alkanes across China (36.5%–69.3%, average ± standard deviation of 55.7 ± 7.9%), followed by aromatics (7.7%–47.8%, 21.0 ± 9.5%), alkenes (6.9%–53.4%, 17.7 ± 9.0%), and alkynes (0.7%–15.0%, 7.8 ± 4.8%); however, there were differences in VOC concentrations among regions (Figs. 1 and 3). The NC, EC, NWC, and Hong Kong regions had higher alkane content (45.1%–69.3%) compared to other regions; alkanes are generally emitted from fuel evaporation (e.g., oil and gas development) and fossil-fuel combustion in industry (e.g., petroleum refining and petrochemical production) (Watson et al., 2001) (Fig. 3). The SWC, EC, and NWC regions had higher alkene content (9.2%–53.4%). The NE region had higher aromatic content (31.5%–37.1%). The NC region had higher alkylene content (10.9%–15.0%). Compared to other cities and sites, the alkylene content in Chongqing and Gongga Mountain was lower, at 37.0% and 36.5%, respectively (Fig. 1). Ambient VOCs were dominated by alkenes and aromatics in Chongqing (53.4%) and Gongga Mountain (47.8%), respectively. We also found that the alkenes presented obvious variation in different cities (e.g., Tianjin, Qingdao, Wuhu and Foshan). Ethyne is the most important composition of alkylene, which can be used for lighting, welding, and cutting metal (oxyacetylene flame) in industry. It is also the basic raw material for manufacturing acetaldehyde, acetic acid, benzene, synthetic rubber, and synthetic fiber. We speculated that the variation of alkylene content at different sampling sites was caused by the difference of industrial emissions. We also found that the ambient VOCs was dominated by alkanes in urban area of Guangzhou (59.9%), followed by aromatics (23.2%). Han et al. (2019) reported that the dominant composition was aromatics in vehicle detection station of Guangzhou (41.4%), indicating the impacts of sampling sites on chemical compositions.

We also compared the chemical compositions of the VOCs in the three urban areas mentioned above. In the BTH region, the VOC concentration was ranked in the following order: alkanes (49.2%–66.2%) > alkenes (11.1%–26.3%) > aromatics (7.7%–18.5%) > alkynes (10.9%–15.0%). Alkanes are the main composition of automotive emissions and the dominant proportion of alkanes were attributed to the increasing demand of fuel combustion in the BTH region (Guo et al., 2017; Song et al., 2019a), while the solvent-related emissions were relatively lower (Liu et al., 2020). C2–C5 alkanes were the abundant species in alkanes (e.g., propane, ethane, and n-butane), and ethylene, 1-butene, cis-2-pentene, and propy-lene were main species in alkenes (Li et al., 2022). In the YRD and PRD regions, they ranked as follows: alkanes (45.1%–63.2%) > aromatics (19.8%–35.0%) > alkenes (6.9%–25.3%) > alkynes (5.4%–7.3%). The concentrations of ethane, propane, and n-butane were also the abundant species in alkanes, while the aromatic were mainly dominated by benzene,
toluene, and ethylbenzene, emitted from automotive emissions and solvent volatilization (An et al., 2014; Zhu et al., 2012). Guo et al. (2017) and Wu and Xie (2017) also reported that aromatics contributed substantially (~30%) to the total VOCs in the YRD and PRD regions.

Compared to domestic cities, the content of alkanes in foreign cities (e.g., Mexico, Calgary, and Seoul) is relatively high (63.0%–75.3%); however, the content of alkenes with high OFP is relatively low (7.8%–14.8%, Garzon et al., 2015; Bari et al., 2018; Song et al., 2019b), indicating that there is still much room for improvement in VOC control in China.

3.2.2 Chemical compositions exhibited different seasonal variations across China

The chemical composition of VOCs exhibited different seasonal variations across China. High temperatures are conducive to the volatilization of VOCs. Therefore, biogenic emissions and anthropogenic sources (e.g., solvent usage) increase the concentrations of alkenes and aromatics. However, highly active alkenes and aromatics are also easily converted into ozone by atmospheric photochemical reactions under high temperatures and strong radiation (Qin et al., 2021; Zhang et al., 2017). Thus, the relative content of alkenes and aromatics are closely related to emissions and consumption.

We found that VOCs were also dominated by alkanes in different seasons across China (Fig. 4). The alkene content was higher in autumn and winter, especially in NC (e.g., Handan and Beijing), which might be due to open straw burning and residential heating, as biomass combustion was one of the main sources of alkenes (He et al., 2011). Solvent usage (e.g., building decoration materials, furniture, and adhesive), fuel evaporation, and vehicles are the main sources of aromatic (Gao et al., 2020; Xuan et al., 2021). High ambient temperature in summer increased the volatilization of aromatics from solvent usage and fuel evaporation, such as in Tianjin, Hangzhou,
Fig. 4. Seasonal variation of chemical compositions in different cities. The bars for each city, from left to right, represent spring, summer, autumn, and winter. The references for each city are the same as in Fig. 1.

and Guangzhou. However, the aromatic content was highest in winter in Jinan, Linyi, and Shanghai. As one of the main source of aromatics, the temperature of vehicle engine is low under low ambient temperature in winter, resulting in insufficient fuel combustion and increased emissions. George et al. (2014) conducted vehicle emissions testing on a chassis dynamometer at two ambient temperatures (–7 and 22°C) operating over cold start and warm start. The results indicated that cold temperature and cold start conditions caused dramatic enhancements in VOCs emissions (e.g., benzene and toluene) compared to the warmer temperature. The seasonal variation in alkyne concentration was relatively small, with a deviation of 3.7% among the cities during for all four seasons, indicating that the emissions of alkynes were stable and hardly changed with meteorological conditions, especially in Jinan and Wuhan.

3.3 Vehicle Exhaust, NG/LPG, Fuel Combustion, and Solvent Usage Were the Dominant VOC Contributors

Eight VOC sources were identified across China, specifically solvent usage, vehicle exhaust, fuel evaporation, NG/LPG, industrial, fuel combustion, biogenic, and others (e.g., secondary generation), using positive matrix factorization (PMF) and principal component analysis (PCA) models (Table S1).

Vehicle exhaust, NG/LPG, fuel combustion, and solvent usage were the dominant VOC contributors across China, with annual contributions of 12.0%–70.7%, 12.6%–45.0%, 8.3%–49.0%, and 4.7%–42.4%, respectively, followed by industrial (10.0%–21.5%), fuel evaporation (7.9%–17.0%), and biogenic (1.7%–9.0%) (Fig. 5). The dominant contributors should be given more attention to control their emissions.

The contribution of vehicle exhaust to VOCs was generally higher in Jinan (70.7%), Chengdu (45.0%), and Langfang (44.8%), with the number of vehicles ranging from 1.4 to 5.2 million (Ministry of Ecology and Environment; http://www.mee.gov.cn/). We also found that their contribution at the background site at Gongga Mountain accounted for nearly half of the VOCs (44.9%), which might have resulted from regional transmission. Vehicle exhausts were also a major source of VOCs in other cities, except for Shanghai, with contributions higher than 21.0%. Components of the NG/LPG and fuel combustion sources are also associated with vehicle emissions. Therefore, the annual contribution of vehicle exhausts might have been higher than the estimated value, indicating that the control of vehicle emissions is still of high priority for reducing VOC concentrations across China (Li et al., 2020).
Fig. 5. Annual VOC source contributions in China. The references for each city and site are the same as in Fig. 1.

The contribution from NG/LPG was generally high in Hong Kong (45.0%), Langfang (24.9%), Changbai Mountain (22.3%), and Wuhu (20.8%). The highest contribution in Hong Kong was attributed to LPG, which has been used as a major clean fuel source for most public transportation, taxis, and private light buses. Additionally, high-density traffic from LPG-fueled vehicles occurred at the sampling site. However, the contribution from NG/LPG in Hong Kong was approximately average relative to estimates from other studies (39.1%–60.0%; Huang et al., 2015; Lyu et al., 2016; Yao et al., 2019).

The contribution from fuel combustion was generally high in Beijing (49.0%), Shanghai (41.0%), Harbin (24.9%), and Hong Kong (20.0%). Fuel combustion, including biomass (26.7%) and coal burning (22.3%), was the dominant VOC contributor in Beijing. Biomass burning is an important source of ambient VOCs (Wang et al., 2014). Although biomass burning emitted only 3000 tons of VOCs in Beijing in 2014 (Zhou et al., 2017), that is much more frequent in the surrounding Hebei province, accounting for 2.8 × 10^5 tons of VOCs. Shandong Province has a high coal consumption rate and dense distribution of coal-fired plants (Xiong et al., 2016; Zhao et al., 2021), resulting in a greater release of VOCs into the atmosphere (Sun et al., 2021). Controlling regional transmission from surrounding regions, especially rural areas, might be the key factor for VOC reduction in Beijing. Vehicle exhausts were the second highest contribution source (33.8%, including 21.7% for gasoline vehicles and 12.1% for diesel vehicles) in Beijing, although the city has the most vehicles in China (5.9 million; http://www.mee.gov.cn/).

Solvent usage was higher in Foshan (42.4%), Chengdu (26.0%), Hangzhou (18.1%), and Wuhu (16.6%). The high contribution in Foshan was attributed to industrial operations around the sampling site, including coating production, plastic products processing, furniture manufacturing, and rubber solvent (naphtha) production (Deng et al., 2021).

4 OUTLOOK

4.1 Continuous Reduction of VOCs Concentrations to Alleviate Ozone Pollution

Continuous reduction of VOCs is critical for alleviating ozone pollution, as VOCs are important
precursors to the formation of photochemical smog. Studies have reported that the annual average VOC concentration in Beijing decreased from 44.0 ppbv in 2016 to 39.5 ppbv in 2018–2019, with a reduction ratio of 10.2% (Liu et al., 2020; Wei et al., 2021). In Shanghai, it decreased from 34.8 ppbv in 2009–2010 to 25.8 ppbv in 2019, with a reduction ratio of 25.9% (Ye, 2020; Wang et al., 2013). This can be attributed to the reduction of VOC emissions, with the highest reduction ratio occurring in Beijing from 2016 to 2017 (9.6%), followed by Shanghai (3.7%) (http://meicmodel.org/). Moreover, a series of emission standards were released for different industries in Beijing (e.g., wooden furniture manufacturing industry, printing industry, industrial surface coating, waterproof sheet industry, petroleum refining, and petrochemical manufacturing industry; http://sthjj.beijing.gov.cn/), and Shanghai implemented more stringent emission limits for air pollutants than the national emission standards to reduce VOC concentrations (https://sthj.sh.gov.cn/index.html). However, in Jinan, the total VOC concentrations were comparable in 2010, 2011, 2015, and 2016 (60.1–72.2 µg m⁻³) and lower in 2012, 2013, and 2014 (20.9–35.1 µg m⁻³), indicating an urgent need to control VOC pollution (Sang and Wei, 2019).

The decrease in ambient VOC concentration was likely due to emission reductions from gasoline vehicles. In Jinan, the contribution of vehicle emissions to ambient VOCs decreased from high concentrations of 15.0%–38.0% to lows of 14.0%–22.0% (Sang and Wei, 2019). Pang et al. (2015) reported that, for most compounds, a decrease in annual concentration was highly correlated with a decrease in acetylene, a marker for vehicle emissions from light-duty gasoline vehicles. Therefore, to reduce VOC concentrations, more attention should be given to vehicle emissions.

Listing VOCs as mandatory monitoring pollutants in environmental monitoring stations in China is critical for understanding of the spatiotemporal distribution of ambient VOCs. The VOCs in some sites in this study were measured in 2007 or earlier (e.g., Gongga Mountain), and thus a change in the distribution of emission sources might have influenced the changes in concentration level, chemical composition, and source contribution of VOCs since that time. Therefore, this study demonstrates that it is necessary to update this data continuously.

4.2 Developing Accurate Source Apportionment Methods to Guide Policy Making

It is difficult to accurately identify the sources of VOCs, as their contributions are affected by the emission strength of each source, meteorological conditions, and photochemical reaction processes (Hui et al., 2020). The receptor-oriented source apportionment model is a widely used method for estimating source contributions to VOCs, including chemical mass balance (CMB), PMF, and PCA (Feng et al., 2019; Yao et al., 2021a; Zhang et al., 2021; An et al., 2014). For example, the contribution of vehicle exhausts in spring in Beijing in the present study was 33.5%, which is consistent with other studies (33.8%, Zhang et al., 2020d; 32.3%, Zhang et al., 2020c; 33.4%, Yao et al., 2021a); however, the annual contribution of vehicle exhausts in Chengdu (45.0%) is much higher than that in previous studies (24.0%–27.5%, He, 2018; Simayi et al., 2020; Wang et al., 2020a). Thus, the source contributions in the same region may be quite different due to the inclusion of VOC species, location of sampling sites, and defects in the model. The technical guide for source apportionment for atmospheric particulate matter and ambient ozone was released on 14th August in 2013 and 3rd July in 2018, respectively by the Ministry of Ecology and Environment (http://www.mee.gov.cn/). Technical guidelines for source apportionment of VOCs and development of accurate source apportionment methods are urgently needed to guide policy making.

5 CONCLUSION

Improving our understanding of the spatiotemporal distributions of characteristics and sources of VOCs across China is critical for the effective control of VOC pollution. It is vital to control VOC pollution in the NWC region, which had concentrations ranging from 50.3–75.0 ppbv. No significant difference was found among the NC, CSC, and SWC regions, with values of 32.0 ± 8.1, 36.0 ± 18.1, and 34.1 ± 16.4 ppbv, respectively. Low concentrations occurred in the background regions, as well as in Qingdao and Fuzhou. VOC concentrations exhibited different seasonal variations across China. The high concentrations in winter and autumn for most cities in NC can be attributed to the combined contributions of weather and heating activities. VOC concentrations exhibited
spatial variation, ranging from 6.7 to 75.0 ppbv across China. VOC compositions were dominated by alkanes, followed by aromatics, alkenes, and alkynes, and exhibited different seasonal variations depending on the emission and consumption of VOCs. The content of alkenes with high OFP was relatively high (6.9%–53.4%) compared with foreign cities (7.8%–14.8%), indicating that there is still much room for improvement in VOC control in China. Vehicle exhaust, NG/LPG, fuel combustion, and solvent usage were the dominant VOC contributors across China. The control of vehicle emissions is still a high priority for reducing VOC concentrations across China.

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SUPPLEMENTARY MATERIAL

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