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## Review

# A review of atmospheric benzene homologues in China: Characterization, health risk assessment, source identification and countermeasures

Yuanyuan Ji<sup>1,2</sup>, Fuhong Gao<sup>1</sup>, Zhenhai Wu<sup>2</sup>, Lei Li<sup>3</sup>, Dandan Li<sup>4</sup>,  
 Hao Zhang<sup>2</sup>, Yujie Zhang<sup>2</sup>, Jian Gao<sup>5</sup>, Yingchen Bai<sup>2</sup>, Hong Li<sup>2,\*</sup>

<sup>1</sup>College of Earth Sciences, Jilin University, Changchun 130061, China

<sup>2</sup>State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

<sup>3</sup>Academy of Environmental Planning and Design, Co., Ltd, Nanjing University, Nanjing 210093, China

<sup>4</sup>Department of Urban Construction, Beijing City University, Beijing 100083, China

<sup>5</sup>College of Environmental and Resource Sciences, Zhejiang University, Hangzhou 310058, China

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## ABSTRACT

Benzene homologues are important chemical precursors to the formation of ground-level ozone and secondary organic aerosol (SOA) in the atmosphere, in addition, some toxic species are harmful to human health. Strict countermeasures have been taken to fight air pollution since 2013, and total amount control of volatile organic compounds is being promoted in China at present. Therefore, it is important to understand the pollution situation and the control status of ambient benzene homologues in China. This paper reviews research progress from published papers on pollution characteristics, atmospheric photochemical reactivity, health risk assessment and source identification of ambient benzene homologues in recent years in China, and also summarizes policies and countermeasures for the control of ambient benzene homologues and the relevant achievements. The total ambient levels of benzene, toluene, ethylbenzene and xylenes (BTEX) shows a declining tendency from 2001 to 2016 in China. The mass concentrations of BTEX are generally higher in southern regions than in northern regions, and they present vertical decreasing variation characteristics with increasing altitude within the height range of about 5500 m. Toluene has the highest ozone formation potential and SOA formation potential both in urban areas and background areas, while benzene poses an obvious carcinogenic risk to the exposed adult populations in urban areas. Source identification of ambient benzene homologues suggested that local governments should adopt differentiated control strategies for ambient benzene homologues. Several recommendations are put forward for future research and policy-making on the control of ambient benzene homologues in China.

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\* Corresponding author.

E-mail: [lihong@craes.org.cn](mailto:lihong@craes.org.cn) (H. Li).

## Introduction

According to the World Health Organization (WHO), volatile organic compounds (VOCs) are organic compounds with a boiling point of 50–260°C and a saturated vapor pressure greater than 133.32 Pa at 20°C (WHO, 2000). Benzene homologues are important species of VOCs, including monocyclic aromatic hydrocarbons such as benzene (B), toluene (T), ethylbenzene (E), *o*-xylene (*o*-X), *m*-xylene (*m*-X), *p*-xylene (*p*-X), styrene, and isopropyl benzene. In particular, benzene, toluene, ethylbenzene, and xylenes (referred to as BTEX) are representative species of benzene homologues (Lee et al., 2002), which account for 46%–91% of the total mass concentration of benzene homologues in ambient air (Bai et al., 2016; Barletta et al., 2008; Hang and Xue, 2004; Jia et al., 2016; Li et al., 2014; Tang et al., 2005; Zhang et al., 2009). Benzene homologues are highly reactive in atmospheric photochemical reactions and they are key precursors of ground-level ozone and secondary organic aerosol (SOA) (O'Malley, 1996). Some benzene homologues are directly harmful to the human respiratory system, hematologic system, reproductive system, and nervous system. Long-term exposure to carcinogenic benzene in ambient air can cause severe diseases, such as bone marrow diseases, lymphoma, and leukemia (WHO, 2000).

With rapid urbanization and industrialization, air pollution has become increasingly severe in China. At present, fine particulate matter (PM<sub>2.5</sub>) and ozone are the primary pollutants in China. The VOCs in ambient air are key precursors of ozone and SOA, so the control of VOCs has also attracted much attention in China. A series of policies were introduced in China to fight air pollution, and the implementation of these policies was beneficial to air quality improvement (MEE, 2019). To prevent VOCs pollution, the “13th Five-Year Plan for Ecological Environmental Protection” and “13th Five-Year Plan for Prevention and Control of Volatile Organic Compounds Pollution” were issued during the 13th Five-Year Plan period. In 2018, the State Council issued the “Monitor Scheme for Volatile Organic Compounds in Ambient Air in Key Regions 2018” to promote the development of monitoring systems and reduce the total amount of VOCs in ambient air, as well as providing the basis for the effective implementation of various schemes. The implementation of these policies and control measures also promoted the prevention and control of ambient benzene homologues pollution in China.

In 2013, Zhao et al. (2013) reviewed the spatial and temporal distribution, reaction characteristics, and sources of BTEX in ambient air in urban areas, and found that the proportions of toluene and *m*&*p*-xylene were high with high ozone formation potential (OFP). The BTEX in ambient air exhibited obvious seasonal and diurnal variations, and their main sources were vehicle-related emissions, paints, and solvent usage. In 2017, Zhang et al. (2017) reviewed the species, levels, sources, and spatial distribution of VOCs in China, and calculated the OFP and SOA formation potential (SOAFP). They found that toluene was the most abundant species among the benzene homologues in the ambient air in China, followed by xylenes and benzene. The mixing ratio of benzene exceeded the standard limit of European Union ( $1.43 \times 10^{-9}$ ) and Japan ( $0.93 \times 10^{-9}$ ). In addition, the species with high OFP and SOAFP were benzene homologues among the measured VOCs. In the same year, Guo et al. (2017) reviewed the main studies conducted in China about the characteristics and sources of VOCs, their relationships with ozone and SOA, and the techniques for their removal. They found that BTEX accounted for 11%–38% of the measured VOCs and contributed significantly to OFP and SOAFP (Cai et al., 2010; Cao et al., 2016; Jia et al., 2016; Zhang et al., 2009; Zhu et al., 2016).

Although some comprehensive studies on VOCs have been carried out in China, there is a lack of systematic and in-depth summaries on the research and pollution control of benzene homologues. This review focuses on the scientific research and control measures in the recent years in China, in which the pollution characteristics, atmospheric photochemical reactivity, human health risk assessment, and source identification of ambient benzene homologues are thoroughly discussed, and the preventative and control measures implemented in China are introduced and preliminarily evaluated. Some directions are emphasized for scientific research and pollution control of ambient benzene homologues in order to provide supportive information to the prevention and control of benzene homologues in the ambient air in China in the future.

## 1. Data sources and processing

### 1.1. Data sources

The data reported in this review were obtained by literature searches from China National Knowledge Infrastructure (CNKI), Web of Science, Science Direct, and PubMed in publication years ranging from 2001 to 2018. The keywords employed in the literature searches were: VOCs, volatile organic compounds, benzene homologues, BTEX, benzene, toluene, ethylbenzene, *o*-xylene, *m*-xylene, *p*-xylene, styrene, isopropylbenzene, *n*-propylbenzene, *o*-ethyltoluene, *m*-ethyltoluene, *p*-ethyltoluene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, and 1,2,3-trimethylbenzene. The selection criteria for the publications were as follows: (1) samples were obtained from ambient air, excluding air monitoring samples from indoor and pollution sources; (2) detailed descriptions were provided of the quality assurance and control processes for sample analyses, and the precision and accuracy of the analytical equipment should meet the relevant specifications; and (3) details of the sampling time and site were required, as well as the concentrations of benzene, toluene, ethylbenzene, *m*&*p*-xylene, and *o*-xylene. Based on these principles, 56 papers and 97 data sets were selected for this paper. It should be noted that there might still be some studies of benzene homologues in China that have not been included in this review although we have tried to review all the major findings.

### 1.2. Data processing

According to the selected data (Appendix A Table S1), the main research regions for ambient benzene homologues in China were divided into background areas (BGA,  $n = 15$ ) and urban areas ( $n = 82$ ) in this study. The urban areas were then divided into northern regions and southern regions. The northern regions included Northeast China (NEC,  $n = 8$ ), Beijing-Tianjin-Hebei region (BTH,  $n = 38$ ), and the Fenhe-Weihe Plain (FWP,  $n = 6$ ). The southern regions included the Yangtze River Delta region (YRD,  $n = 12$ ) and Pearl River Delta region (PRD,  $n = 18$ ). BTEX were the key species analyzed in benzene homologues. To ensure the comparability of the data, all of the mixing ratios were converted into mass concentrations ( $\mu\text{g}/\text{m}^3$ ) under standard conditions (0°C, 101.325 kPa).

As shown in Appendix A Table S1, the measurements of atmospheric benzene homologues in China were generally scattered and were mainly conducted in the eastern regions. The time series were incomplete, the analytical methods were not unified, and most of the experiments were conducted for short time periods. It should be noted that the conclusions based on these limited data may have some uncertainty. In order to obtain more general research results, long-term and system-

atic observations of atmospheric benzene homologues are urgently needed in the future in China.

### 1.2.1. Calculation of OFP

The formation of ozone in ambient air is a complex photochemical process, and VOCs and NO<sub>x</sub> are important precursors in the formation of ozone (Haagen-Smit, 1952). The formation of ozone has significant nonlinear relationships with VOCs and NO<sub>x</sub> (Tang et al., 2006b). The photochemical formation of ozone in different regions is usually divided into VOCs-limited, NO<sub>x</sub>-limited, or transitional regimes. At present, the maximum incremental reactivity (MIR) is widely used to calculate the OFP of VOCs in VOC-limited regions. Given that most urban areas are VOC-limited regions in China (Guo et al., 2017), the OFP of benzene homologues was calculated using the concentrations of benzene homologues from the literature and the latest MIR values from Carter's studies (<https://www.engr.ucr.edu/~carter/SAPRC/>) in this paper.

### 1.2.2. Calculation of SOAFP

SOA are formed from VOCs via oxidation, nucleation, and gas-particle partitioning processes (Seinfeld and Pandis, 2016). It is difficult to separate SOA from primary organic aerosol at present, so the concentrations of SOA cannot be measured directly (Koppmann, 2010). Various methods are used to study the formation of SOA from VOCs, such as smog chamber experiments, the organic tracer method, and model simulations (Kleindienst et al., 2007). Many studies have shown that benzene homologues are important precursors of SOA (Dzepina et al., 2009; Han et al., 2016; Kleindienst et al., 2007; Lane et al., 2008). The fractional aerosol coefficients (FAC) obtained from smoke chamber experiments are widely used to calculate the SOAFP for VOCs (Grosjean, 1992; Grosjean and Seinfeld, 1989). Some studies have suggested that this method does not consider the important contributions of intermediate VOCs and semi-VOCs to SOA (Robinson et al., 2007), but it can still be used to estimate the SOAFP for VOCs to some extent. Based on the data obtained from literature searches, the SOAFP of benzene homologues in different regions of China is calculated using the FAC in this study.

### 1.2.3. Health risk assessment

In this paper, the health risk assessment method based on inhalation exposure, which is recommended by the National Health Commission of the People's Republic of China (WS/T 666-2019) and the US Environmental Protection Agency (US EPA) (EPA-540-R-070-002), was applied to make the health risk assessment of benzene homologues to exposed adult populations. The relevant parameters were taken from the Integrated Risk Information System (<https://www.epa.gov/iris>) and "Chinese Population Exposure Parameter Manual: Adult Volume". The impacts of toxic VOCs in ambient air on human health can be divided into carcinogenic and non-carcinogenic effects. In this method, excess cancer risk (Risk, dimensionless) and hazard quotient (HQ) were used to represent the carcinogenic risk and non-carcinogenic risk, respectively, of a certain chemical species. Hazard index (HI, dimensionless) was the sum of HQ of several pollutants. Among the benzene homologues, benzene is related to various human hematological diseases such as leukemia, and it is categorized as a group I carcinogen by the WHO. In addition, benzene has a non-carcinogenic risk. Ethylbenzene may be carcinogenic in humans, but no relevant parameters are available. There is no evidence that toluene and xylenes are carcinogenic in humans (Li et al., 2014). Therefore, the carcinogenic and non-carcinogenic risks were analyzed for benzene, and only the non-carcinogenic risks were studied for toluene, ethylbenzene, and xylenes among the benzene homologues in this study.

### 1.2.4. Source identification

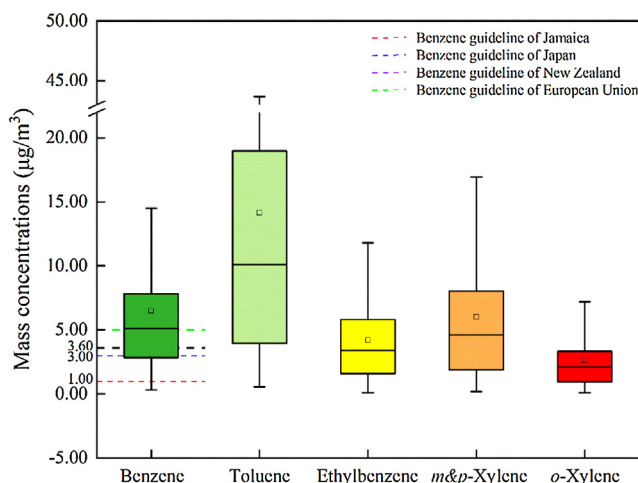
At present, the methods employed for VOCs source apportionment mainly comprise the characteristic ratio method and receptor model method. Some studies showed that the sources of VOCs could be accurately determined based on the ratios of BTEX species and the correlations of BTEX species with specific source tracers (Barletta et al., 2005, 2008; He et al., 2015; Liu et al., 2009). However, there are overlaps among toluene to ethylbenzene, toluene to xylenes, and ethylbenzene to xylenes ratios in the different source profiles, so using these ratios as indicators for source identification is not recommended. The ratio of benzene to toluene (B/T) overlaps little for different sources, and thus it has been widely used for identifying the sources of VOCs (He et al., 2015) although the characteristic ratio can be affected by photochemical ages of BTEX. To better identify the sources of VOCs, Zhang et al. (2016) plotted a ternary diagram based on the relative proportions of benzene, toluene, and ethylbenzene in the source profiles of various sources. The ternary diagram was then partitioned according to the relative positions of the three sources, which indicated that VOCs came mainly from biomass/biofuel/coal burning, vehicle-related emissions, and industrial emissions and solvent use emissions. Finally, the ambient concentrations of benzene, toluene, and ethylbenzene were plotted in the partitioned ternary diagram to identify the possible sources of VOCs. The receptor model methods include factor analysis (FA), principal component analysis (PCA), positive matrix factorization (PMF), and the chemical mass balance model (CMB). In particular, FA, PCA, and PMF can be used to analyze the sources of VOCs without a source inventory, whereas CMB requires a source inventory and it is more suitable for analyzing the sources of stable species (Song et al., 2008). The receptor model method is used rarely to directly analyze the sources of benzene homologues, and thus the characteristic ratio method was used to identify the main sources of benzene homologues in China in the present study.

## 2. Results and discussion

### 2.1. Ambient levels and variations

#### 2.1.1. Overall ambient levels

From 2001 to 2016, the mass concentrations of BTEX in the ambient air in the study areas of China ranged from 0.71 to 132.09 µg/m<sup>3</sup>, with a mean of 33.42 ± 28.68 µg/m<sup>3</sup>. The mass concentrations of benzene varied from 0.19 to 53.90 µg/m<sup>3</sup>, with a mean of 6.48 ± 7.19 µg/m<sup>3</sup>. The mean mass concentration of benzene was much lower than the standard value (110 µg/m<sup>3</sup>, 1-hr average) according to the indoor air quality standard for China, but it was seven times the guideline value specified by Jamaica (1 µg/m<sup>3</sup>, annual average), about twice the guideline values specified by Japan and New Zealand (annual averages of 3 and 3.6 µg/m<sup>3</sup>, respectively), and 1.36 times the standard value specified by the European Union (5 µg/m<sup>3</sup>, annual average). The mass concentrations of toluene varied from 0.24 to 61.61 µg/m<sup>3</sup>, with a mean of 14.15 ± 13.79 µg/m<sup>3</sup>, which was much lower than the standard 1-week average mass concentration in ambient air specified by the WHO (260 µg/m<sup>3</sup>) and the standard 1-hr average mass concentration of toluene in the indoor air quality standard for China (200 µg/m<sup>3</sup>). The mass concentrations of ethylbenzene varied from 0.04 to 14.72 µg/m<sup>3</sup>, with a mean of 4.20 ± 3.44 µg/m<sup>3</sup>, which was much lower than the guideline annual average mass concentration in ambient air specified by the WHO (22,000 µg/m<sup>3</sup>). The mass concentrations of *m* & *p*-xylene varied from 0.04 to 24.42 µg/m<sup>3</sup> with a mean of 6.02 ± 5.23 µg/m<sup>3</sup>, and the mass concentrations of *o*-xylene varied



**Fig. 1** – Mass concentrations of benzene, toluene, ethylbenzene and xylenes (BTEX) in ambient air in China. Boxes with error bars represent the 10th, 25th, 75th, and 90th percentiles for each species, respectively. The lines and small boxes in the boxes represent the median and mean values for each species, respectively. Only benzene guidelines are presented because the guideline/standard values of other species in BTEX are much higher than their maximum values.

from 0.04 to 12.45  $\mu\text{g}/\text{m}^3$  with a mean of  $2.61 \pm 2.27 \mu\text{g}/\text{m}^3$ . The mean mass concentration of xylenes was  $8.60 \pm 7.40 \mu\text{g}/\text{m}^3$ , which was much lower than the guideline 24-hr average mass concentration of xylenes specified by Jamaica (2300  $\mu\text{g}/\text{m}^3$ ), the guideline annual average mass concentration of xylenes specified by the WHO (1000  $\mu\text{g}/\text{m}^3$ ), and the standard 1-hr average mass concentration of xylenes in the indoor air quality standard for China (200  $\mu\text{g}/\text{m}^3$ ) (Fig. 1).

These analyses indicate that benzene pollution is a serious problem in China, and thus strict measures should be implemented to control the emission of benzene.

### 2.1.2. Temporal variations

During 2001 to 2016, the annual average mass concentration of BTEX exhibited a characteristic of overall decline in China, with a decrease of 35.11% from 2001 to 2016 (Fig. 2). At the same time, the major emission sources of anthropogenic VOCs in China changed from fossil fuel combustion and transportation to industrial processes, transportation and solvent utilization (Wu et al., 2016), which may lead to an increase in benzene homologues emissions. The decrease of ambient concentrations of BTEX indicated that the series of control policies issued for VOCs in China might have played a positive role in reducing the concentrations of benzene homologues. During 2001–2008, the annual average mass concentration of BTEX showed a decreasing characteristic, with a decrease of 73.60% from 2001 to 2008. During 2008–2012, the mass concentrations of BTEX increased initially and then decreased. The highest value of 54.04  $\mu\text{g}/\text{m}^3$  was reached in 2010, which was 204.52% higher compared with that in 2008. During 2013 to 2016, the annual average mass concentration of BTEX varied slightly, where it was about 42  $\mu\text{g}/\text{m}^3$  (Fig. 2).

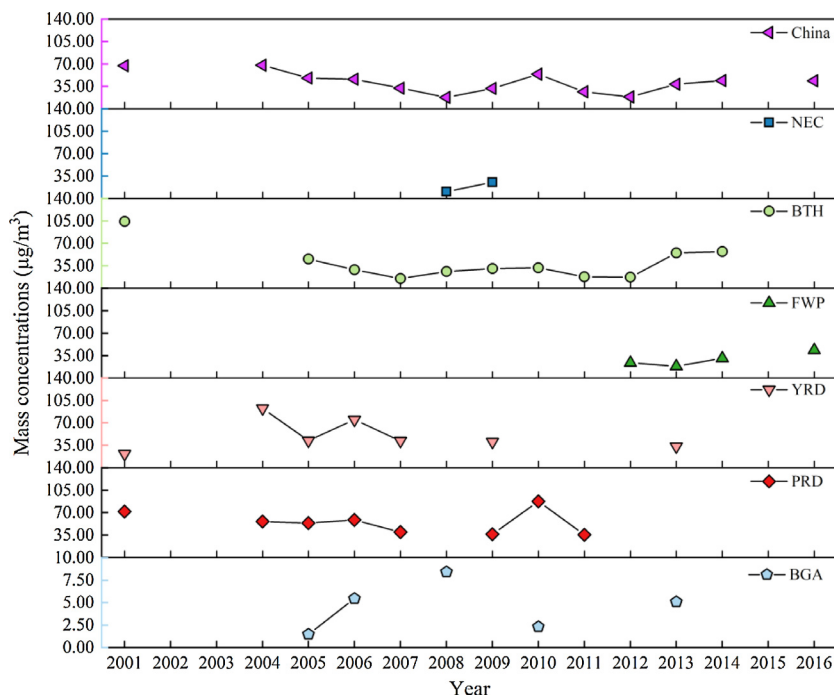
The annual average mass concentrations of BTEX in different areas exhibited different variations (Fig. 2). From 2008 to 2009, the mass concentrations of BTEX were upward in NEC, with a significant increase of 135.42%. From 2001 to 2014, the mass concentration of BTEX decreased initially in BTH,

before fluctuating slightly, and then increasing continuously. The lowest values occurred in 2007 and 2012, i.e., 15.07 and 17.10  $\mu\text{g}/\text{m}^3$ , respectively. From 2012 to 2016, the mass concentrations of BTEX in FWP decreased initially and then increased. From 2001 to 2013, the mass concentrations of BTEX in YRD increased and then decreased. From 2001 to 2011, the mass concentrations of BTEX in PRD decreased initially, before increasing and finally decreasing. These different variation characteristics may have been related to the diverse industrial structures in these regions, the change in VOCs emissions in various industries, and differences in the BTEX chemical consumption under different weather conditions. From 2005 to 2013, the overall mass concentrations of BTEX were upward in BGA.

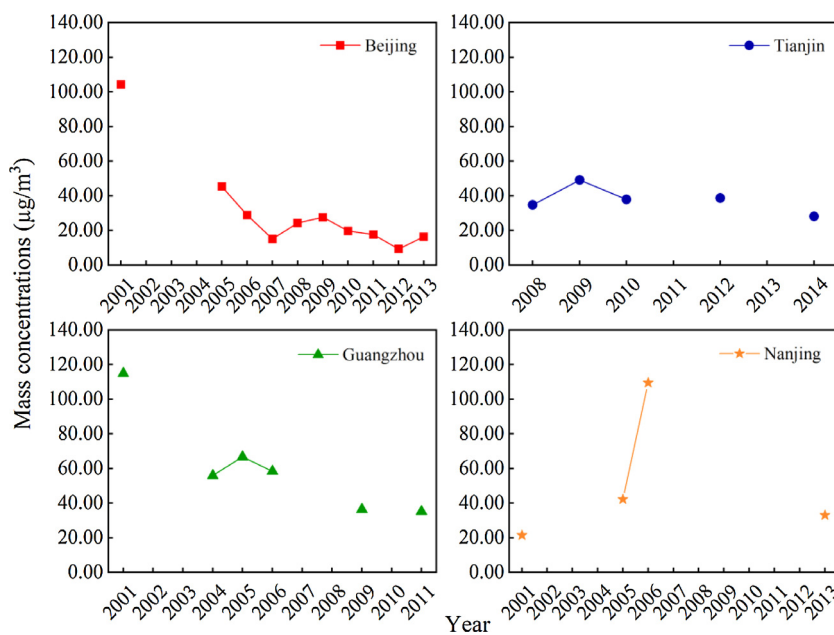
The variation characteristics of the annual average mass concentrations of BTEX in typical cities varied among the different regions (Fig. 3). From 2001 to 2013, the mass concentration of BTEX in Beijing exhibited a decline, where it was 84.30% lower in 2013 than that in 2001. From 2008 to 2014, the mass concentrations of BTEX varied slightly in Tianjin, where it was 19.11% lower in 2014 than that in 2008. The mass concentrations of BTEX were downward after 2010 in both cities, which may have been related to the implementation of the first VOCs control policy "Guidelines on Promoting Joint Prevention and Control of Air Pollution to Improve Regional Air Quality", which was issued in 2010. From 2001 to 2011, the mass concentration of BTEX exhibited a characteristic of decline in Guangzhou, where it was 69.38% lower in 2011 than that in 2001. From 2001 to 2013, the mass concentrations of BTEX varied significantly in Nanjing, where it increased initially and then decreased. The highest value occurred in 2006, i.e., 109.50  $\mu\text{g}/\text{m}^3$ .

The seasonal variations in the concentrations of benzene homologues varied among the different regions. In addition, the concentrations of benzene homologues in the same region varied among different sampling sites. The studies by Zhou et al. (2011) and Zhang et al. (2012b) of ambient air in Beijing showed that the BTEX concentrations were higher in the autumn and winter and lower in the spring and summer, where the highest concentration occurred in the winter and the lowest in the summer. The BTEX concentrations were higher in the autumn and winter because the low temperature was not conducive to the diffusion of pollutants, elimination due to photochemical reactions was limited, and emissions from coal combustion increased. The BTEX concentrations were lower in the spring and summer because the temperature is higher and the meteorological conditions was favorable for photochemical reactions to increase the consumption of BTEX. Sun et al. (2011) studied the seasonal variations in BTEX in Beijing and showed that the BTEX concentrations were higher in the spring and summer and lower in the autumn, mainly due to the variations in seasonal emission sources. In addition, weather conditions such as the temperature and wind had specific impacts on the BTEX concentration. Ning et al. (2012) found that there were obvious seasonal variations in the concentrations of BTEX in Tianjin, where the overall characteristics comprised highest level in the winter, lowest level in the spring, and similar intermediate levels in the summer and autumn. The higher BTEX concentration in the winter may have been related to the local atmospheric stability, meteorological conditions, and emissions from coal combustion during the heating period. Hang and Xue (2004) investigated the seasonal variations in benzene homologues in Nanjing and found that the concentrations in the summer and autumn were higher than those in the spring and winter.

Studies by Li et al. (2014), Wang et al. (2011), Xia et al. (2014), Zhang et al. (2012a) and Zhu et al. (2016) showed that the con-



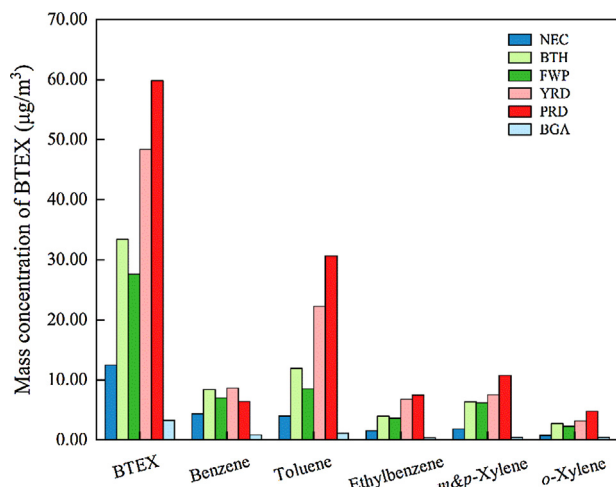
**Fig. 2 – Annual variations in mass concentrations of BTEX in different regions in China. NEC: Northeast China; BTH: Beijing-Tianjin-Hebei region; FWP: Fenhe-Weihe Plain; YRD: Yangtze River Delta region; PRD: Pearl River Delta region; BGA: background areas.**



**Fig. 3 – Annual variations in mass concentrations of BTEX in Beijing, Tianjin, Guangzhou, and Nanjing in China.**

centrations of BTEX were higher in the morning and evening, but lower at noon and in the afternoon, where the lowest value occurred in the afternoon (13:00–15:00). These results indicate that vehicle exhaust emissions during the rush hours in the morning and evening might have been an important source of BTEX in the ambient air. In addition, photochemical reactions were enhanced at noon and in the afternoon, thereby increasing the consumption of BTEX. The meteorological conditions

were also important factors that affected the diurnal variations in benzene homologues. Temperature inversion and the low boundary layer in the morning and evening were conducive to the accumulation of pollutants, whereas the boundary layer rose at noon and in the afternoon, which was conducive to the rapid diffusion of pollutants. Therefore, the main factors that affected the diurnal variations in BTEX comprised the pollution sources and atmospheric stability.



**Fig. 4 – Concentrations of BTEX in ambient air in different regions of China.**

The mass concentrations of benzene homologues in the ambient air are generally higher at night than in the daytime. Yu et al. (2009) studied VOCs in the ambient air in a forest and found that the activities of aromatic hydrocarbons were lower than those of alkenes and terpenes, and the concentrations were higher at night and lower in the daytime. This diurnal variation was mainly due to the balance between plant emissions and physical and chemical consumption, and similar results were obtained by Sun et al. (2011). However, some studies found that the mass concentrations of benzene homologues might be higher in the daytime than at night. Tang et al. (2006a) measured benzene homologues in ambient air and found that the mass concentrations of benzene homologues were higher in the daytime than that at night. This was mainly because vehicle-related emissions were higher in the daytime than those at night and the amount of benzene homologues derived from solvent volatilization was much higher in the daytime than at night because of the higher temperatures in the daytime, and the consumption of benzene homologues was lower than their emissions in the daytime.

### 2.1.3. Spatial variations

The mean mass concentrations of BTEX were  $38.94 \pm 27.83 \mu\text{g}/\text{m}^3$  (range: 6.40–132.09  $\mu\text{g}/\text{m}^3$ ) and  $3.25 \pm 2.35 \mu\text{g}/\text{m}^3$  (0.71–8.43  $\mu\text{g}/\text{m}^3$ ) in the urban areas and BGA in China, respectively. The mean mass concentration in the urban areas was 12.04 times that in BGA, and the mass concentration of each BTEX species was higher in the urban areas than those in BGA (Fig. 4). These findings indicate that human activities have greatly affected the amount of BTEX in ambient air. In the urban areas, the mass concentration of BTEX was higher in southern regions than in the northern regions, which may be due to the higher VOC emissions in the southern regions (Wu et al., 2016). The mass concentrations of BTEX from high to low in different regions followed the order of: PRD > YRD > BTH > FWP > NEC, with mean values of  $59.83 \pm 26.83$  (23.44–114.94)  $\mu\text{g}/\text{m}^3$ ,  $48.39 \pm 28.18$  (21.44–109.50)  $\mu\text{g}/\text{m}^3$ ,  $33.42 \pm 25.78$  (7.45–132.09)  $\mu\text{g}/\text{m}^3$ ,  $27.62 \pm 9.94$  (15.32–43.61)  $\mu\text{g}/\text{m}^3$ , and  $12.50 \pm 5.75$  (6.40–25.16)  $\mu\text{g}/\text{m}^3$ , respectively, and all were much higher than those in BGA (Fig. 4). The mass concentrations of all BTEX species excluding benzene were highest in PRD and lowest in NEC, which was consistent with the distribution of the total mass concentrations of BTEX. The mass concentration of benzene in YRD ( $8.64 \pm 3.85 \mu\text{g}/\text{m}^3$ ) was comparable to that in BTH ( $8.40 \pm 10.22 \mu\text{g}/\text{m}^3$ ), but

higher than those in FWP and PRD, and the lowest occurred in NEC ( $4.37 \pm 1.20 \mu\text{g}/\text{m}^3$ ), which might have been related to the different background concentrations, industrial structures, and intensity of the control policies in each region.

There were some researches about vertical distributions of VOCs on a larger scale by using meteorological tower, aircraft or tethered balloon. The concentrations of benzene homologues varied with the height above the ground. Mao et al. (2005) investigated the characteristics of VOCs at heights of 8, 32, 140, and 320 m, and found that the mass concentrations of benzene homologues increased initially and then decreased as the height increased, with the highest levels at 32 m. Wu et al. (2015) studied the characteristics of VOCs at heights of 1.5, 18.5, 24.5, 48.5, 66.5, 72.5, and 84.5 m, and found that the trends in the concentrations of toluene, ethylbenzene, and xylenes were similar with an overall fluctuating trend, where relatively higher mass concentrations occurred at 24.5 and 84.5 m. The trends in the concentration of benzene differed slightly compared the trends determined for toluene, ethylbenzene, and xylenes, where a relatively higher mass concentration of benzene occurred at 48.5 m. Yin et al. (2015) researched the characteristics of VOCs at 0, 21, 42, and 78 m above the ground and found two peaks in the mass concentration of benzene homologues at 21 and 28 m, and similar results were reported by Wu et al. (2015). These findings indicate that there were two peaks in the mass concentrations of benzene homologues at certain heights, possibly because the benzene homologues accumulated at a certain height and then decreased due to their consumption in photochemical reactions. After their upward transmission, they gradually accumulated at a certain height due to the presence of the temperature inversion and the concentration then increased again.

Wang et al. (2009) studied the characteristics of VOCs at ground level and other three height level of 900–1000, 1100–1300 and 2000–2300 m by an aircraft study in Beijing in 2007, and found that the mix ratios of each BTEX species decreased with increasing height, and similar study result was reported by Liu et al. (2013) in Beijing. Xue et al. (2011) investigated the vertical distributions of non-methane hydrocarbons in an altitude range of 0.5–5.5 km by an intensive aircraft study in Jilin Province. They found that the profile of benzene showed a peak at the surface, which then decreased with altitude. This pattern can be explained by the interplay among surface emissions, turbulent mixing, and atmospheric processing of air masses. Zhang et al. (2018) carried out the measurements of VOCs at three height level of 50–350, 350–700, and 700–1000 m by using tethered balloon in Shanghai, and found that the mix ratios of benzene, toluene, ethylbenzene and m&p-xylene decreased with increasing height. They also found that 14 benzene homologues attributed 46.76% of the 56 VOCs at ground level, but it decreased to only 19.89% at 350 m.

## 2.2. Compositions

The compositions of the BTEX species in ambient air were similar in the urban areas and BGA in China, where toluene accounted for the largest proportion, followed by benzene (Fig. 5). These two species accounted for 36.55%–88.76% of the total, and ethylbenzene and xylenes accounted for 11.24%–63.45%. In urban areas, the relative proportions from the highest to the lowest followed the order of: toluene (41.42%) > benzene (19.93%) > m&p-xylene (18.32%) > ethylbenzene (12.57%) > o-xylene (7.76%), where the proportion of toluene was 2.08 times that of benzene and o-xylene had the lowest proportion. By contrast, in BGA, the relative proportions from the highest to the lowest followed the order of: toluene (34.60%) > benzene (26.41%) > o-xylene (14.19%) > m&p-xylene (13.81%) > ethylbenzene (10.9%), where the proportion of toluene was

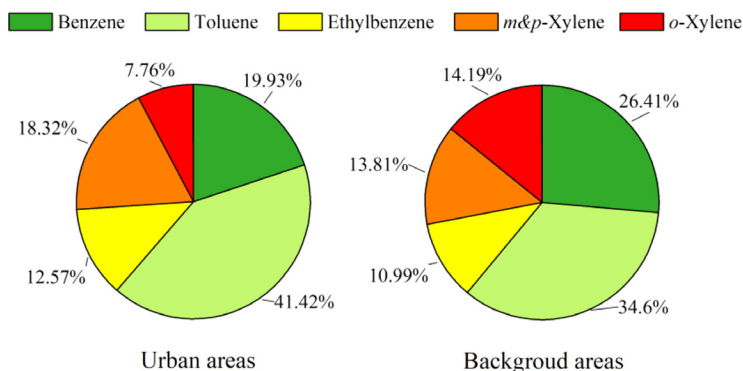


Fig. 5 – Compositions of BTEX in ambient air in urban areas and background areas of China.

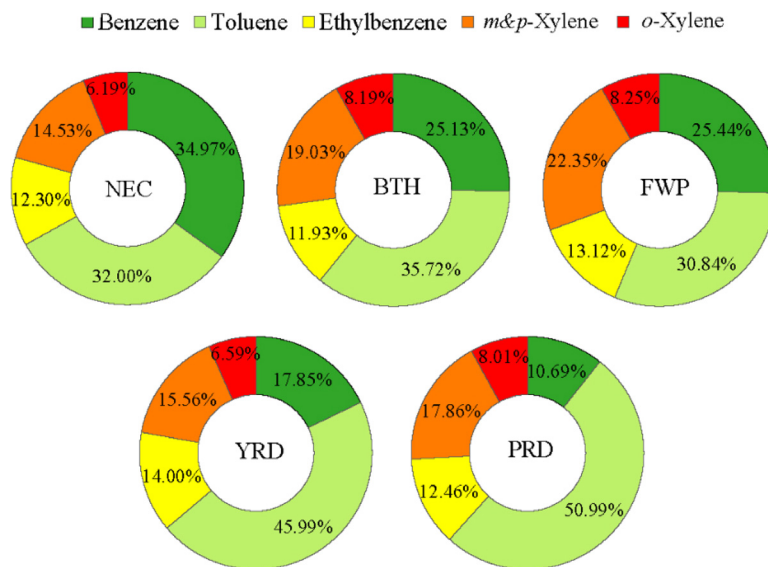


Fig. 6 – Compositions of BTEX in ambient air in typical urban areas of China.

1.31 times that of benzene, and ethylbenzene had the lowest proportion (Fig. 5).

In urban areas, the relative proportions of BTEX species in FWP, BTH, and YRD were consistent with the overall characteristics in urban areas, where the proportions of toluene, benzene, *m&p*-xylene, ethylbenzene, and *o*-xylene were 30.84%–45.99%, 17.85%–25.44%, 15.56%–22.35%, 11.93%–14.00%, and 6.59%–8.25%, respectively (Fig. 6). The relative proportions of BTEX species in NEC and PRD differed from the overall characteristics, mainly because the relative proportion of benzene varied greatly. In NEC, the proportion of benzene (34.97%) was higher than that of toluene (32.00%). In PRD, the proportion of benzene (10.69%) was lower than those of *m&p*-xylene (17.86%) and ethylbenzene (12.46%), and only higher than that of *o*-xylene (8.01%) (Fig. 6).

In general, the relative proportions of the BTEX species varied significantly in the northern and southern urban areas. The proportion of toluene was higher in the southern urban areas (more than 45%) than in the northern urban areas (about 30%), and it reached up to 50.99% in PRD. The proportion of benzene was higher in the northern urban areas (25.13%–34.97%) than in the southern urban areas (10.69%–17.85%). The proportions of ethylbenzene and xylenes differed little in the northern and southern urban areas.

### 2.3. Contributions to ozone formation

Wei et al. (2014) investigated 51 VOCs using the MIR method in Beijing in BTH and found that the mass concentration of BTEX accounted for 17.9% of the total VOCs whereas their OFP accounted for 24.7%. Ran et al. (2009) studied 109 VOCs in YRD and found that the OFP for toluene, ethylbenzene, and xylenes accounted for 45.7%, and toluene (14.7%) contributed most to the OFP of 109 VOCs in this region. Barletta et al. (2008) found that toluene was one of the species that contributed most to the OFP among 48 species in PRD. Studies in these three regions showed that the mass concentration proportion of BTEX among the total VOCs was low, but their OFP was large among the total VOCs, thereby indicating that they were important precursors for the ozone formation.

The OFP of BTEX was much higher in urban areas than in BGA in China (Fig. 7a). The OFP for BTEX in different regions from lowest to highest followed the order of: NEC < FWP < BTH < YRD < PRD, with OFP of 56.50, 144.28, 171.50, 252.27, and 338.75  $\mu\text{g}/\text{m}^3$ , respectively, thereby indicating that the OFP were lower in northern regions than in southern regions (Fig. 7a). In these five regions, the OFP proportion of benzene (2.71%–11.06%) was smallest, and thus benzene made a small contribution to the OFP of BTEX in these regions. The

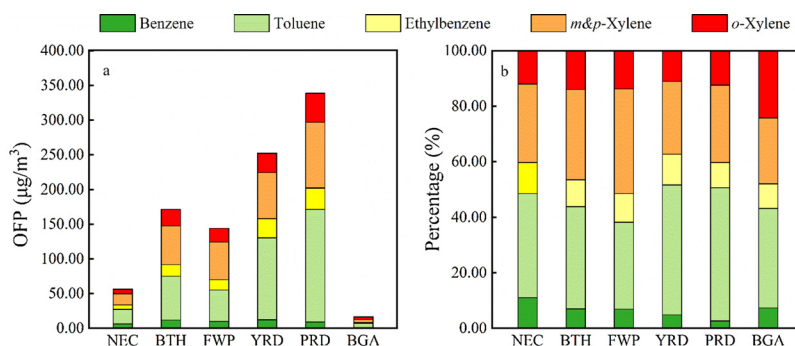


Fig. 7 – (a) Ozone formation potential (OFP) of BTEX species in ambient air and (b) the proportions of each species in different regions of China.

OFP for toluene and *m&p*-xylene were over 65% and up to 75.78%, so toluene and *m&p*-xylene made greater contributions to the OFP of BTEX. In NEC, BTH, YRD, and PRD, the BTEX species with the highest OFP proportion was toluene, where it accounted for 37.51%, 36.88%, 46.76%, and 47.94%, respectively, whereas the BTEX species with the highest OFP in FWP was *m&p*-xylene, with 37.61% (Fig. 7b). In BGA, the BTEX species with the highest OFP was toluene, with 35.84%, and the OFP were similar for *m&p*-xylene and *o*-xylene, with 23.72% and 24.22% respectively, thereby indicating that toluene was the main contributor to the OFP in BGA (Fig. 7b).

Model simulation have been applied to determine the contribution of VOCs to ozone formation. Han et al. (2013) assessed the sensitivity of the ozone formation to VOCs in Tianjin by using the National Center for Atmospheric Research Master Mechanism (NCAR-MM) model. Aromatics were found to be the second contributors to ozone production, accounting for 35.1% of the total. Geng et al. (2008) studied the sensitivity of the ozone formation to VOCs in Shanghai by using the NCAR-MM model. The results showed that aromatics play a dominated role for the ozone production, accounting for 45% of the total ozone production, although the concentration of aromatics accounts for only 25% of the total VOC concentrations. An et al. (2015) evaluated the relative importance of VOCs to ozone in Nanjing by relative incremental reactivity (RIR) values extracted from the observation-based model (OBM), and found that aromatic made a pronounced contribution in producing ozone. Cheng et al. (2010a) assessed photochemical ozone formation at Wangqingsha, Guangzhou by using a photochemical trajectory model (PTM) coupled with the Master Chemical Mechanism (MCM), and found that alkenes and aromatics presented high reactivity. Among the aromatics, *m*-xylene and toluene were the key species for ozone formation. These results are consistent with studies which ozone-precursor relationships were examined in PRD by using an OBM (Cheng et al., 2010b; Lu et al., 2010; Xue et al., 2014; Zhang et al., 2007, 2008), and are consistent with those obtained based on observed data and MIR.

In general, toluene and *m&p*-xylene are important precursors in BTEX for ozone formation in China. Thus, efforts should focus on controlling the emissions of toluene and *m&p*-xylene in ambient air in order to prevent and control ozone pollution.

## 2.4. Contributions to SOA formation

In 2006, Lue et al. (2009) estimated the SOAFP for 31 VOCs by using FAC during the summer in Beijing in BTH and found that the SOAFP of BTEX accounted for 54.36% of the 31 VOCs. The SOAFP of toluene is highest, followed by *m&p*-xylene, ac-

counting for 19.66% and 15.66% of the 31 VOCs, respectively. In 2013, Lin et al. (2015) studied the SOAFP for 24 VOCs in Nanjing in YRD and found that the SOAFP proportion for BTEX was 77.90%, where toluene and ethylbenzene accounted for 27.28% and 26.12%, respectively. Studies in these two regions showed that the SOAFP of BTEX was high.

The SOAFP of BTEX was much higher in urban areas than in BGA in China (Fig. 8a). In urban areas, the SOAFP for BTEX from the lowest to highest in order were: NEC < FWP < BTH < YRD < PRD, with SOAFP of 0.58, 1.36, 1.66, 2.56, and 3.34  $\mu\text{g}/\text{m}^3$ , respectively, and thus they were lower in northern regions than in the southern regions (Fig. 8a). Toluene accounted for the largest SOAFP proportion in NEC, FWP, BTH, YRD and PRD, with 42.37%, 38.50%, 44.10%, 53.28%, and 56.36%, respectively, thereby indicating that toluene made the largest contribution to SOAFP of BTEX in these regions. Xylenes (22.43%–33.05%) was the second main contributor to the SOAFP of BTEX in these five regions, followed by ethylbenzene (14.26%–16.96%). Benzene contributed 4.28%–16.77% to SOAFP of BTEX in these regions (Fig. 8b). Similar to urban areas, toluene accounted for the largest SOAFP proportion in BGA, followed by xylenes, with the percentage of 42.24% and 32.19% respectively, whereas ethylbenzene (13.89%) and benzene (11.68%) made lower contributions to SOAFP of BTEX (Fig. 8b).

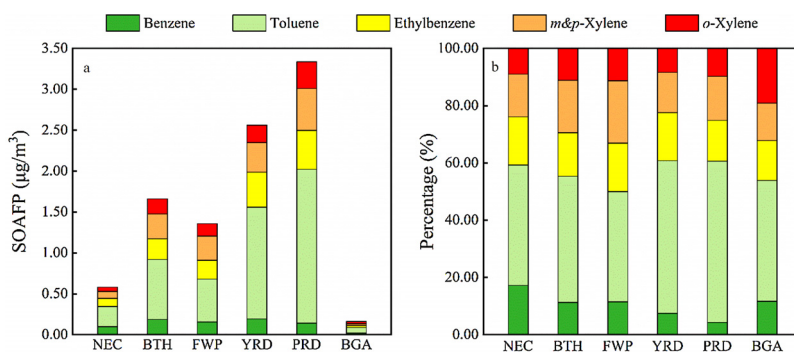
In general, toluene and xylenes are important precursors in BTEX for SOA formation in China. Therefore, efforts should focus on controlling the emissions of toluene and xylenes in ambient air in order to prevent and control pollution with  $\text{PM}_{2.5}$ .

## 2.5. Health risk assessment

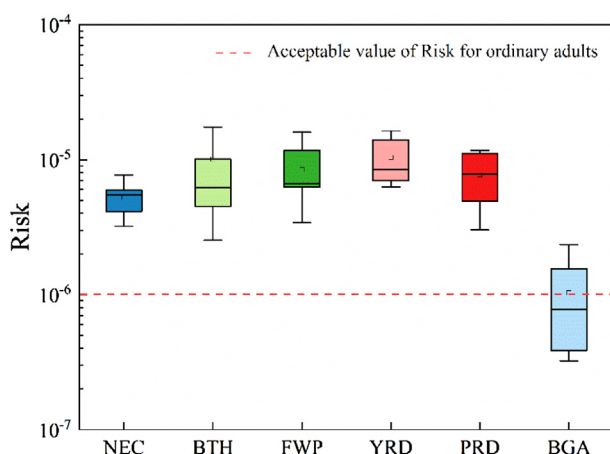
### 2.5.1. Carcinogenic risk assessment

The carcinogenic risks of benzene in ambient air ranged from  $1.36 \times 10^{-6}$  to  $6.48 \times 10^{-5}$  in urban areas in China, with a mean of  $9.03 \times 10^{-6}$  (Fig. 9). All these values exceeded the acceptable risk value for adults of  $1.00 \times 10^{-6}$  recommended by the US EPA, thereby indicating that benzene posed an obvious carcinogenic risk to exposed adult populations in urban areas. However, the values of carcinogenic risk of benzene varied in different urban areas in China. The carcinogenic risk was highest in YRD with a mean of  $1.04 \times 10^{-5}$  ( $5.21 \times 10^{-6}$ – $1.95 \times 10^{-5}$ ). The carcinogenic risk in BTH was comparable to that in YRD, with a mean of  $1.01 \times 10^{-5}$  ( $1.36 \times 10^{-6}$ – $6.48 \times 10^{-5}$ ). The mean values of carcinogenic risk in FWP and PRD were  $8.45 \times 10^{-6}$  ( $3.42 \times 10^{-6}$ – $1.60 \times 10^{-5}$ ) and  $7.72 \times 10^{-6}$  ( $2.60 \times 10^{-6}$ – $1.31 \times 10^{-5}$ ), respectively, which are lower than those in YRD and BTH. Benzene had the lowest carcinogenic risk in NEC, with a mean of  $5.25 \times 10^{-6}$  ( $3.21 \times 10^{-6}$ – $7.71 \times 10^{-5}$ ) (Fig. 9).





**Fig. 8 – (a) Secondary organic aerosol formation potential (SOAFP) for BTEX species in ambient air and (b) the proportions of each species in different regions of China.**



**Fig. 9 – Carcinogenic risk of benzene in ambient air in different regions of China. Boxes with error bars represent the 10th, 25th, 75th, and 90th percentiles for the Risk of each region, respectively. The lines and small boxes in the box represent the median and mean values for each region, respectively.**

The carcinogenic risks of benzene ranged from  $2.28 \times 10^{-7}$  to  $3.01 \times 10^{-6}$  in BGA in China, with a mean of  $1.03 \times 10^{-6}$  (Fig. 9), which was comparable to the acceptable risk value for adults. Thus, the benzene in BGA posed no significant carcinogenic risk to exposed adult populations in BGA. It should be noted that among the 15 BGA sites studied, the carcinogenic risks of benzene at Taishan in Shandong, Dezhou in Shandong, Wuyi Mountain in Fujian, and Ganzi in Sichuan exceeded the acceptable risk value for adults. It is suggested that external transport may have a great impact on the mass concentrations of benzene at these four sites.

### 2.5.2. Non-carcinogenic risk assessment

The HI values for BTEX in ambient air ranged from  $8.67 \times 10^{-3}$  to 2.05 in the urban areas of China, with a mean of  $3.58 \times 10^{-1}$ . The US EPA states that a pollutant with a HQ value less than 1 will not cause significant harm to human health. Therefore, BTEX posed no significant non-carcinogenic risk to exposed adult populations in the urban areas of China. As shown in Table 1, The highest HI values for BTEX occurred in YRD, with a mean of  $4.06 \times 10^{-1}$  ( $2.42 \times 10^{-1}$ – $7.75 \times 10^{-1}$ ). The HI values for BTEX were comparable in PRD and BTH, with means of  $3.80 \times 10^{-1}$  ( $1.81 \times 10^{-1}$ – $7.16 \times 10^{-1}$ ) and  $3.77 \times 10^{-1}$  ( $6.19 \times 10^{-2}$ –2.05), respectively, which were lower than that

in YRD. It should be noted that the maximum HI values exceeded 1 in three studies among the 38 studies conducted in BTH. The two cities involved were Beijing and Shijiazhuang, and thus BTEX might have significant non-carcinogenic risk for human health in these two cities. In FWP, the mean HI value was  $3.24 \times 10^{-1}$  ( $1.40 \times 10^{-1}$ – $6.24 \times 10^{-1}$ ). The HI value was smallest in NEC with a mean of  $1.74 \times 10^{-1}$  ( $1.04 \times 10^{-1}$ – $2.69 \times 10^{-1}$ ). Benzene had the largest HQ, followed by xylenes, and toluene had the smallest in urban areas.

The HI values for BTEX ranged from  $8.67 \times 10^{-3}$  to  $1.13 \times 10^{-1}$  in BGA in China, with a mean of  $3.83 \times 10^{-2}$ . Benzene had the largest HQ, followed by xylenes, and toluene had the smallest, with mean values of  $2.86 \times 10^{-2}$ ,  $9.10 \times 10^{-3}$ , and  $2.25 \times 10^{-4}$ , respectively, thereby indicating that BTEX posed no significant non-carcinogenic risk to exposed adult populations in BGA (Table 1).

### 2.6. Source identification

The B/T ratios vary significantly among different countries for VOCs from the same sources. Roger and Ivan (1995) showed that the B/T ratio was about 0.5 for VOCs from vehicle-related emissions in London during 1995. Lan and Binh (2012) studied the B/T ratios in Asia during 2012 and found that they ranged from 0.16 to 0.19 in Japan and Singapore, and from 0.36 to 0.77 in Malaysia and Vietnam for VOCs from vehicle-related emissions.

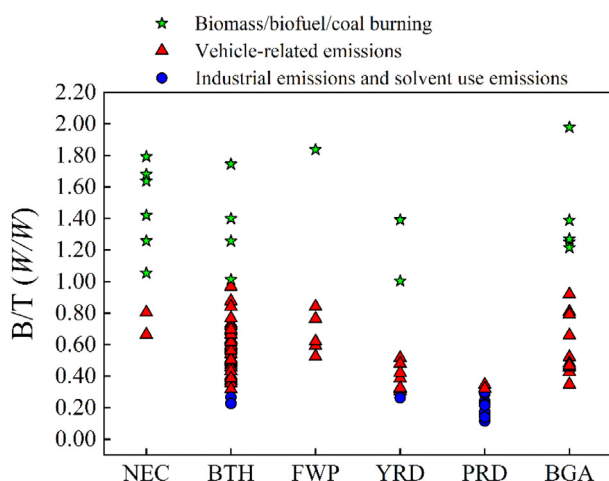
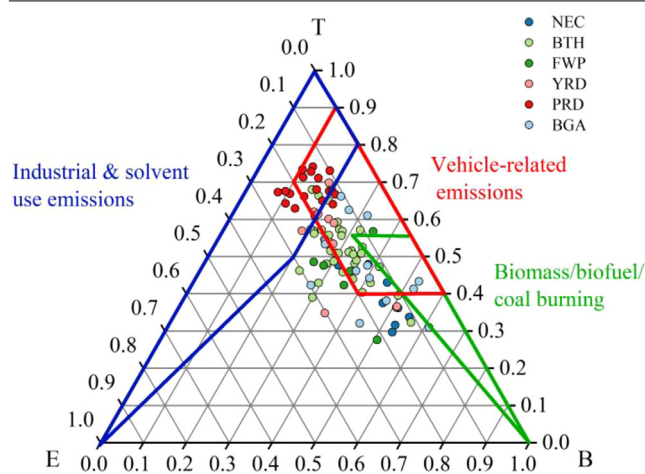
To verify whether these data could be used for identifying the sources of VOCs in China, Barletta et al. (2005, 2008) analyzed source samples from 25 cities in China during 2005. The results showed that B/T (W/W) ratio was  $0.6 \pm 0.2$  for VOCs from vehicle-related emissions, which was consistent with the results obtained by Liu et al. (2009, 2005) in Beijing (0.64, 0.61), and Tan et al. (2012) in Foshan ( $0.45 \pm 0.24$ ). The B/T ratio was greater than 1 for VOCs from biomass/biofuel/coal burning, which is consistent with the results obtained by Li et al. (2009) in Shandong ( $1.9 \pm 0.62$ ). The B/T ratio was less than 0.2 for VOCs from industrial emissions. In 2015, He et al. (2015) analyzed samples from different VOCs sources in China and found that the B/T ratios were 0.3–1.11 for VOCs from gasoline and diesel emissions during refueling, 0.01–0.3 for VOCs from solvent use emissions, and 0.005–0.016 for VOCs from industrial emissions.

Based on the results given above, the B/T ratios and corresponding VOCs sources were summarized into three categories. The B/T ratios and the relative proportions of B, T, and E were used to determine the sources of ambient benzene homologues in China in the present study. The corresponding VOCs sources are industrial and solvent use emissions, vehicle-related emissions, and biomass/biofuel/coal burning at the B/T ratios (W/W) of <0.3, 0.3–1, and >1, respectively.

**Table 1 – Non-carcinogenic risk assessment results for benzene, toluene, ethylbenzene and xylenes in ambient air in typical regions of China.**

Region	HQ					HI
	Benzene	Toluene	Ethylbenzene	<i>m</i> & <i>p</i> -Xylene	<i>o</i> -Xylene	
NEC	$1.46 \times 10^{-1}$	$8.00 \times 10^{-4}$	$1.54 \times 10^{-3}$	$1.82 \times 10^{-2}$	$7.74 \times 10^{-3}$	$1.74 \times 10^{-1}$
BTH	$2.80 \times 10^{-1}$	$2.39 \times 10^{-3}$	$3.99 \times 10^{-3}$	$6.36 \times 10^{-2}$	$2.74 \times 10^{-2}$	$3.77 \times 10^{-1}$
FWP	$2.34 \times 10^{-1}$	$1.70 \times 10^{-3}$	$3.62 \times 10^{-3}$	$6.17 \times 10^{-2}$	$2.28 \times 10^{-2}$	$3.24 \times 10^{-1}$
YRD	$2.88 \times 10^{-1}$	$4.45 \times 10^{-3}$	$6.77 \times 10^{-3}$	$7.53 \times 10^{-2}$	$3.19 \times 10^{-2}$	$4.06 \times 10^{-1}$
PRD	$2.14 \times 10^{-1}$	$6.13 \times 10^{-3}$	$7.49 \times 10^{-3}$	$1.07 \times 10^{-1}$	$4.54 \times 10^{-2}$	$3.80 \times 10^{-1}$
BGA	$2.86 \times 10^{-2}$	$2.25 \times 10^{-4}$	$3.57 \times 10^{-4}$	$4.49 \times 10^{-3}$	$4.61 \times 10^{-3}$	$3.83 \times 10^{-2}$

NEC: Northeast China; BTH: Beijing-Tianjin-Hebei region; FWP: Fenhe-Weihe Plain; YRD: Yangtze River Delta region; PRD: Pearl River Delta region; BGA: background areas; HQ: hazard quotient; HI: hazard index.

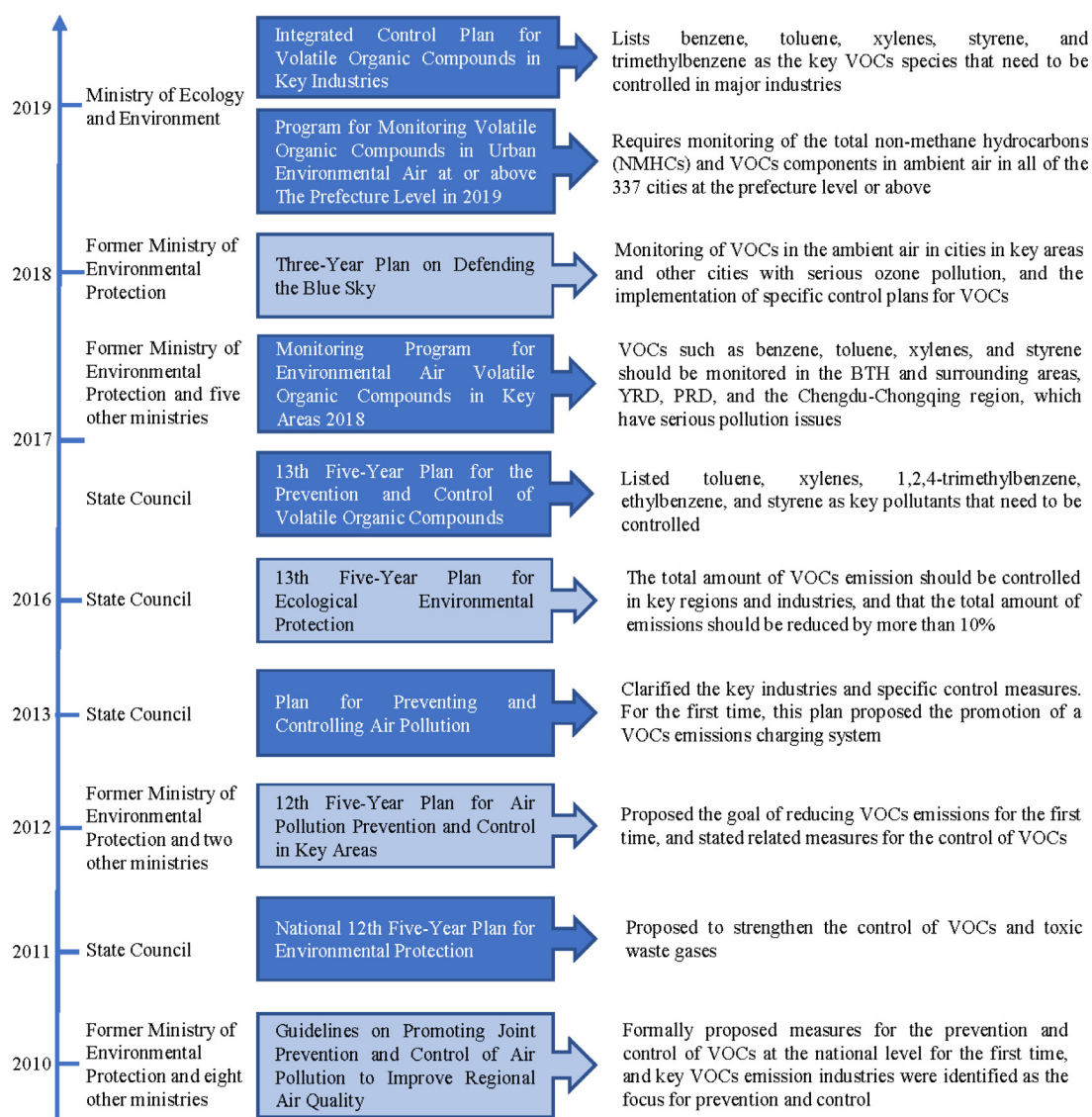
**Fig. 10 – B/T ratio distribution for benzene homologues in ambient air in different regions of China.****Fig. 11 – Relative proportions of benzene (B), toluene (T), and ethylbenzene (E) in different regions of China.**

The B/T ratios distribution and the relative proportions of B, T, and E for ambient benzene homologues in typical regions of China were shown in Figs. 10 and 11, respectively. The B/T ratios ranged from 0.66 to 1.79 in NEC (Fig. 10), the proportions of B, T, and E in NEC were scattered within the composition ranges for vehicle-related emissions, and some were scattered close to the composition ranges

for biomass/biofuel/coal burning (Fig. 11), indicating that the benzene homologues came mainly from vehicle-related emissions and biomass/biofuel/coal burning in NEC. The B/T ratios ranged from 0.23 to 1.74 in BTH, but mainly from 0.3 to 1 (Fig. 10), the proportions of B, T, and E in BTH were scattered within or close to the composition ranges for vehicle-related emissions (Fig. 11), indicating that the benzene homologues came mainly from vehicle-related emissions in BTH, and they were also influenced by industrial emissions, solvent use emissions, and biomass/biofuel/coal burning. The B/T ratios ranged from 0.52 to 1.84 in FWP, but mainly from 0.3 to 1 (Fig. 10), the proportions of B, T, and E in FWP were scattered within or close to the composition ranges for vehicle-related emissions (Fig. 11), indicating the benzene homologues came mainly from vehicle-related emissions in FWP, and they were also affected by biomass/biofuel/coal burning. The B/T ratios ranged from 0.26 to 1.39 in YRD, but mainly about 0.3 (Fig. 10), the proportions of B, T, and E in YRD were scattered within or close to the overlap composition ranges for vehicle-related emissions, industrial emissions and solvent use emissions, and some were scattered close to the composition ranges for biomass/biofuel/coal burning (Fig. 11), indicating that the benzene homologues came mainly from vehicle-related emissions, industrial emissions and solvent use emissions in YRD, and they were also affected by biomass/biofuel/coal burning. The B/T ratios ranged from 0.11 to 0.35 in PRD (Fig. 10), the proportions of B, T, and E in PRD were scattered within or close to the overlap composition ranges for industrial emissions and solvent use emissions and vehicle-related emissions (Fig. 11), indicating the benzene homologues came mainly from industrial emissions, solvent use emissions, and vehicle-related emissions in PRD. These findings show that vehicle-related emissions and biomass/biofuel/coal burning were main sources of ambient benzene homologues in northern regions of China, whereas vehicle-related emissions, industrial emissions and solvent use emissions were main sources of ambient benzene homologues in southern regions of China. In addition, the B/T ratios ranged from 0.35 to 1.98 in BGA (Fig. 10), the proportions of B, T, and E in BGA were scattered within or close to the composition ranges for vehicle-related emissions and biomass/biofuel/coal burning (Fig. 11), indicating that the benzene homologues came mainly from vehicle-related emissions and biomass/biofuel/coal burning in BGA.

## 2.7. Policies

In general, the Chinese government mainly controls ambient VOCs pollution by adopting the countermeasures in the four aspects: (1) issuing relevant laws and regulations to legally support environmental protection; (2) releasing relevant plans and clarifying the control targets for VOCs/benzene homo-



**Fig. 12 – History of major plans and programs to control atmospheric benzene homologues in China.**

logues; (3) developing relevant standards and implementing strict environmental access; and (4) Launching prevention and control technology policies, and strengthening environmental protection supervision. Usually, the Chinese Central Government, or the former Ministry of Environment Protection/the Ministry of Ecology and Environment issues national policies, whereas local governments formulate specific local policies or countermeasures within the framework of the national policies accordingly. The carry-out of a series of policies and measures related to the pollution control of ambient VOCs also accelerated the prevention and control of ambient benzene homologues pollution in China.

### 2.7.1. Laws and regulations

In order to control VOCs, the “Law of the People’s Republic of China on the Prevention and Control of Atmospheric Pollution” was amended in 2015. Compared to the 2000 version, it clearly stipulated the control and management measures for raw materials, products, and waste gas containing VOCs at the legal level for the first time. In 2018, the “Environmental Protection Tax Law of the People’s Republic of China” was implemented, which stipulated that benzene homologues in am-

bient air, such as benzene, toluene, xylenes, and styrene, are taxable pollutants. It is the first law to levy taxes on benzene homologues emissions in China and helps control emissions of benzene homologues.

### 2.7.2. Plans and programs

A series of plans/programs to control VOCs have been issued since 2010 in China (Fig. 12). These plans/programs are usually issued by the former Ministry of Environmental Protection / Ministry of Ecology and Environment or the State Council, focusing on the prevention and control of VOCs pollution in the key areas and the key industries.

In 2010, “Guidelines on Promoting Joint Prevention and Control of Air Pollution to Improve Regional Air Quality” was proposed at the national level for the first time to prevention and control of VOCs. In 2011, it was clarified that monitoring of VOCs should be carried out in key areas, and pollutant emission standards in key industries should be improved. The goal of reducing VOCs emissions was proposed in 2012 and specific measures were issued in 2013. In 2016, it was explicitly requested that the total amount of VOCs emissions should be reduced by more than 10% in the 13th Five-Year Plan period

(2016–2020). In 2017, toluene, xylenes, 1,2,4-trimethylbenzene, ethylbenzene, and styrene were identified as key pollutants. The monitoring of VOCs was started in key areas of China in 2018, and the observation range was expanded from key areas to the 337 cities at the prefecture level or above in 2019.

### 2.7.3. Standards

**Air quality standards:** Ambient air quality standards for benzenes homologues have not been issued in China so far, but air quality standards for indoor benzenes homologues have been issued. The indoor air quality standards are mainly for buildings, coaches and passenger cars, including the “Indoor Air Quality Standards” (GB/T 18883-2002), “Hygienic Standard for The Air Quality inside Long Distance Coach” (GB/T 17729-2009), “Code for Indoor Environmental Pollution Control of Civil Building Engineering” (GB 50325-2010), and “Guidelines for Air Quality Assessment of Passenger Cars” (GB/T 27630-2011). The VOCs species considered in these standards are mainly benzene homologues and formaldehyde. The “Indoor Air Quality Standard” (GB/T 18883-2002) specifies the standard 1-hr mean values for benzene, toluene, and xylenes as 0.11, 0.20, and 0.20 mg/m<sup>3</sup>, respectively.

**Industrial emissions standards: National standards.** In 1996, the “Integrated Emission Standard of Air Pollutants” (GB16297-1996) was issued, which stipulated emissions limits for benzene, toluene, and xylenes among the benzene homologues. A series of emission standards for the key VOC-related industries, such as the petrochemical industry, synthetic and artificial leather industry, and rubber industry, were also established to control the emission of benzene homologues. In addition to standards for traditional industries, new standards including the emission standards of pollutants for the pharmaceutical industry and manufacture of paint, ink and adhesive were implemented by Ministry of Ecology and Environment in 2019 (Table 2). The benzene homologues emission limits varied for different industries, but they were all more stringent than those in GB16297-1996. Some industries also set emission limits for ethylbenzene and xylenes among the benzene homologues (Table 2). In 2019, the Ministry of Ecology and Environment issued the “Standard for Fugitive Emission of Volatile Organic Compounds” (GB 37822-2019), which specified the fugitive emission limits for total non-methane hydrocarbons (NMHCs) for the first time, thereby contributing to the control and management of benzene homologues emissions.

**Industrial emissions standards: Local standards.** Some provinces and municipalities have formulated corresponding local standards under the guidance of the national standards, such as Beijing, Guangdong, Shanghai, and Jiangsu. These local emissions standards were equal to or stricter than the corresponding national standards. In addition to the local industry standards corresponding to the national standards, some provinces and municipalities have also formulated local emissions standards according to the development status of local industries. For example, emission standards for the printing industry, furniture manufacturing industry, and shoe-making industry have been implemented by Guangdong, and emission limits of benzene homologues in these standards are more stringent than those in GB16297-1996 (Table 3).

**Vehicular emissions standards: National standards.** In 2001, in order to control emissions of CO, NO<sub>x</sub>, and hydrocarbons by vehicles, the 1st phase of the vehicle emissions standards was issued and implemented, after then 2nd, 3rd, 4th, and 5th phase of the vehicle emissions standards were implemented in 2004, 2007, 2010, and 2018, respectively. Compared to the previous version, the 5th phase of the vehicle emission standards specified emissions limits for NMHCs for the first time. The 6th phase of the vehicle emissions standards was issued in 2016 and will fully implement the 6th phase-a and 6th phase-b standards nationwide on July 1, 2020 and July 1, 2023,

**Table 2 – Relevant industrial emission standards for benzene homologues in ambient air in China (National Standards).**

Standards	Maximum allowable emission concentration (mg/m <sup>3</sup> ) (1-hr mean)	Concentration limit at fugitive emission reference point (mg/m <sup>3</sup> ) (1-hr mean)
Integrated Emission Standard of Air Pollutants (GB16297-1996)	Benzene (12); toluene (40); xylenes (70)	Benzene (0.4); toluene (2.4); xylenes (1.2)
Emission Standard for Pollutants Form Synthetic Leather and Artificial Leather Industry (GB 21902-2008)	Benzene (2); toluene (30); xylenes (40)	Benzene (0.1); toluene (1.0); xylenes (1.0)
Emission Standard of Pollutants for Rubber Products Industry (GB 27632-2011)	Toluene and xylenes (15)	Toluene (2.4); xylenes (1.2)
Emission Standard of Pollutants for Coking Chemical Industry (GB 16171-2012)	Benzene (6)	Benzene (0.4)
Emission Standard of Air Pollutants for Steel Rolling Industry (GB 28665-2012)	Benzene (8); toluene (40); xylenes (40)	Benzene (0.4); toluene (2.4); xylenes (1.2)
Emission Standard of Pollutants for Petroleum Refining Industry (GB 31570-2015)	Benzene (4); toluene (15); xylenes (20)	Benzene (0.4); toluene (0.8); xylenes (0.8)
Emission Standard of Pollutants for petroleum chemistry Industry (GB 31571-2015)	Benzene (4); toluene (15); xylenes (20); ethylbenzene (100); styrene (50)	Benzene (0.4); toluene (0.8); xylenes (0.8)
Emission Standard of Pollutants for Synthetic Resin Industry (GB 31572-2015)	Benzene (4); toluene (15); ethylbenzene (100); styrene (50)	Benzene (0.4); toluene (0.8)
Emission Standard of Air Pollutants for Pharmaceutical Industry (GB 37823-2019)	Benzene (4); toluene (25); xylenes (40)	Benzene (0.4)
Emission Standards for Air Pollutants from Manufacture of Paint, Ink and Adhesive (GB 37824-2019)	Benzene (1); toluene (15); xylenes (30); styrene (20)	Benzene (0.1); toluene (0.3); xylenes (0.3); styrene (0.42)

respectively. The emissions limits for NMHCs in 6th phase-a are equal to those in 5th phase, but the limits in the 6th phase-b are more stringent than those in 6th phase-a. These vehicle emissions standards do not cover the specific emissions limits for benzene homologue species, but still can promote the control of benzene homologues.

**Vehicular emissions standards: Local standards.** Following the issue of the vehicle emissions standards by central government, some local governments will implement the latest standards in their own cities before the date specified

**Table 3 – Relevant industrial emission standards for benzene homologues in ambient air in China (Local Standards) (partial).**

Standards	Maximum allowable emission concentration (mg/m <sup>3</sup> ) (1-hr mean)	Concentration limit at fugitive emission reference point (mg/m <sup>3</sup> ) (1-hr mean)
Integrated Emission Standard of Air Pollutants (Beijing) (DB11/501-2017)	Benzene (8); toluene (25); xylenes (40)	Benzene (0.1); toluene (0.2); xylenes (0.2)
Emission Standards of Air Pollutants for Organic Chemicals Manufacturing Industry (Beijing) (DB11/1385-2017)	Benzene (1); toluene (10); xylenes (10); styrene (10)	Benzene (0.2); toluene (0.4); xylenes (0.4)
Emission Standard of Air Pollutants for Painting Process of Automobile Manufacturing Industry (Beijing) (DB11/1227-2015)	Benzene (1)	Benzene (0.5)
Emission Standard of Volatile Organic Compounds for Printing Industry (Beijing) (DB11/1201-2015)	Benzene (0.5); toluene and xylenes (15)	Benzene (0.1); toluene and xylenes (0.5)
Emission Standards of Air Pollutants for Petroleum Refining and Petrochemicals Manufacturing Industry (Beijing) (DB11/447-2015)	Benzene (8); toluene (25); xylenes (40)	Benzene (0.4); toluene (0.8); xylenes (0.8)
Emission Standards of Air Pollutants for Wood Furniture Manufacturing Industry (Beijing) (DB11/1202-2015)	Benzene (0.5)	Benzene (0.1)
Emission Standard of Volatile Organic Compounds for Furniture Manufacturing Operations (Guangdong) (DB44/814-2010)	Benzene (1); toluene and xylenes (40)	Benzene (0.1); toluene (0.6); xylenes (0.2)
Emission Standard of Volatile Organic Compounds for Printing Industry (Guangdong) (DB44/815-2010)	Benzene (1); toluene and xylenes (30)	Benzene (0.1); toluene (0.6); xylenes (0.2)
Emission Standard of Volatile Organic Compounds for Shoe-Making Industry (Guangdong) (DB44/804-2010)	Benzene (1); toluene and xylenes (30)	Benzene (0.1); toluene (0.6); xylenes (0.2)
Emission Standard of Volatile Organic Compounds for Surface Coating of Automobile Manufacturing Industry (Guangdong) (DB44/803-2010)	Benzene (1); toluene and xylenes (30); toluene, xylenes and trimethylbenzene (100)	Benzene (0.1); toluene (0.6); xylenes (0.2); trimethylbenzene (0.2)

in the national standard. Beijing, Tianjin, Henan, Shanghai, Guangzhou, and other provinces and cities will implement the 6th phase of the vehicle emissions standards in advance in 2019. Some provinces such as Henan will implement the 6th phase-a standard, and other cities such as Beijing will directly implement the stricter 6th phase-b standard. These emission standards will all help to reduce emissions of benzene homologues.

2.7.4. Other policies

In 1999, in order to protect the atmospheric environment and prevent and control pollution due to vehicle emissions, the “Technological Policy of Pollution Control for Motor Vehicles” was issued, which proposed emission control technologies and suitable combinations for the conditions in China. In 2013, the former Ministry of Environmental Protection issued the “Technological Policy of Pollution Control for Volatile Organic Compounds,” which proposed pollution prevention and control strategies and methods for the production, storage, transportation, sales, use, and consumption of VOCs materials and products containing VOCs. In 2014, the former Ministry of Environmental Protection issued the “Technical Guide for Compilation of Emission Inventory of Atmospheric Volatile Organic Compounds (trial),” which provided technical guidance for the preparation of VOCs emission inventories in China, and recommended emissions coefficients for benzene homologues from some sources. In 2015, the Ministry of Finance, the National Development and Reform Commission, and the former Ministry of Environmental Protection jointly issued the “Pilot Measures for Charging of Emission of Volatile Organic Compounds” and began charging for the emission of VOCs such as benzene, toluene, and xylenes. The progressive implementation of various prevention and control technical policies has provided strong technical support for reducing the emissions of benzene homologues, which will help to reduce the concentrations of benzene homologues in ambient air.

2.7.5. Effectiveness of the policies

At present, long-term variations of VOCs amount is usually based on the investigation of VOCs emission inventories, and can provide useful information about the effectiveness of the control policies of VOCs. Li et al. (2019) estimated the emissions trend of non-methane volatile organic compounds from 1990 to 2017 by using a bottom-up emission inventory framework and studied the main drivers behind the trends. During 1990–2017, aromatics emissions in China have shown a continuously increasing trend, which was similar to the trend of non-methane volatile organic compounds. Aromatics emissions were estimated to be 1.9 Tg in 1990, 3.2 Tg in 2000, 7.0 Tg in 2010, and 9.3 Tg in 2017, with annual growth rates of 6.7% (1990–2000), 12.1% (2000–2010) and 4.5% (2010–2017), respectively. Aromatics emission fractions increased from 19.5% in 1990 to 32.5% in 2017. Solvent usage and industry were the main drivers for the aromatics emissions increase during 1990–2017, whereas aromatics emissions from transportation and residential usage were stable and shrinking, respectively. It should be noted that after “Guidelines on Promoting Joint Prevention and Control of Air Pollution to Improve Regional Air Quality” was issued to control VOCs at the national level in 2010, annual growth rate of aromatics emissions was significantly reduced from 12.1% to 4.5% although the total amount of aromatics emissions was increasing. And with the implementation of more stringent policies, the growth rate of aromatics emissions is decreasing from 5.0% (2010–2011) to 1.2% (2016–2017). These conclusions suggest that policies for VOCs control are very effective.

### 3. Conclusions and prospects

A series of countermeasures have been issued to control atmospheric benzene homologues since 2010 in China. From 2001 to 2016, the mass concentrations of BTEX in ambient air ranged from 0.71 to 132.09  $\mu\text{g}/\text{m}^3$  in China, with a mean of 33.42  $\mu\text{g}/\text{m}^3$ , and show a general decline. Toluene accounts for the largest proportion followed by benzene both in urban areas and BGA. The mass concentrations of all BTEX species in ambient air are higher in urban areas than in BGA in China. In urban areas, the mass concentrations of BTEX are generally higher in southern regions than in northern regions. The vertical distribution of BTEX present characteristics of decreasing with increasing altitudes in the height range of about 5500 m. The seasonal variations of BTEX differ among regions. The BTEX concentrations are higher in the morning and evening but lower at noon and in the afternoon, with the lowest value in the afternoon (13:00–15:00). In general, the mass concentrations of benzene homologues are higher at night than in the daytime.

In China, the OFP and SOAFP of BTEX in ambient air are higher in urban areas than in BGA, and higher in southern urban regions than in northern urban regions. Toluene and xylenes are the key species for ozone and SOA formation. Benzene poses an obvious carcinogenic risk to exposed adult populations in urban areas, whereas BTEX poses no obvious non-carcinogenic risk for exposed adult populations in China. It is recommended that the control of toluene, benzene, and *m*&*p*-xylene pollution should be strengthened in China in order to prevent and control ozone and  $\text{PM}_{2.5}$  pollution, and protect human health.

Vehicle-related emissions and biomass/biofuel/coal burning were main sources of ambient benzene homologues in northern regions of China, whereas vehicle-related emissions, industrial emissions and solvent use emissions are main sources of ambient benzene homologues in southern regions of China. It is suggested that local governments should adopt differentiated control strategies for ambient benzene homologues.

The measurements of benzene homologues in ambient air in China are scattered in spatial distributions, discontinuous in time series and various in analytical methods, thereby limiting our scientific understanding of the benzene homologues in ambient air. It is recommended that the construction and improvement of VOCs emission inventories for key sources should be promoted in nationwide, and not only the total amount but also the active components of VOCs should be included to the emission inventories, in order to lay the foundation for accurate source apportionment of ambient benzene homologues, as well as for identification and refined control of key sources of benzene homologues. It is strongly suggested that the VOCs monitoring should be strictly and continuously carried out in China, which including promote the monitoring of benzene homologues from key areas to whole country, as well as increase the frequency of monitoring in order to obtain a deeper understanding of the pollution characteristics of benzene homologues. In order to more accurately estimate the contribution of benzene homologues to the production of ozone and SOA, not only the concentration of benzenes should be continuously monitored, but also the mechanism of their formation should be further studied. In addition, in order to protect human health and effectively reduce the human health risks caused by benzene homologues in ambient air, it is suggested that the establishment of guidelines for toxic VOCs in ambient air should be accelerated in China.

In general, in order to control the pollution of benzene homologues in ambient air more effectively and promote continuous improvements of the ambient air quality in China, it is

recommended that a comprehensive environmental management system for VOCs should be established, such as including important species of benzene homologues in the “Ambient Air Quality Standards”, as well as strengthening the supervision and management of policy implementation; it is also recommended that the feasibility and effectiveness of policies should be timely assessed so as to dynamically adjust the policies.

### Declaration of competing interest

We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

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### Appendix A. Supplementary data

Supplementary material associated with this article can be found in the online version at doi:10.1016/j.jes.2020.03.035.

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