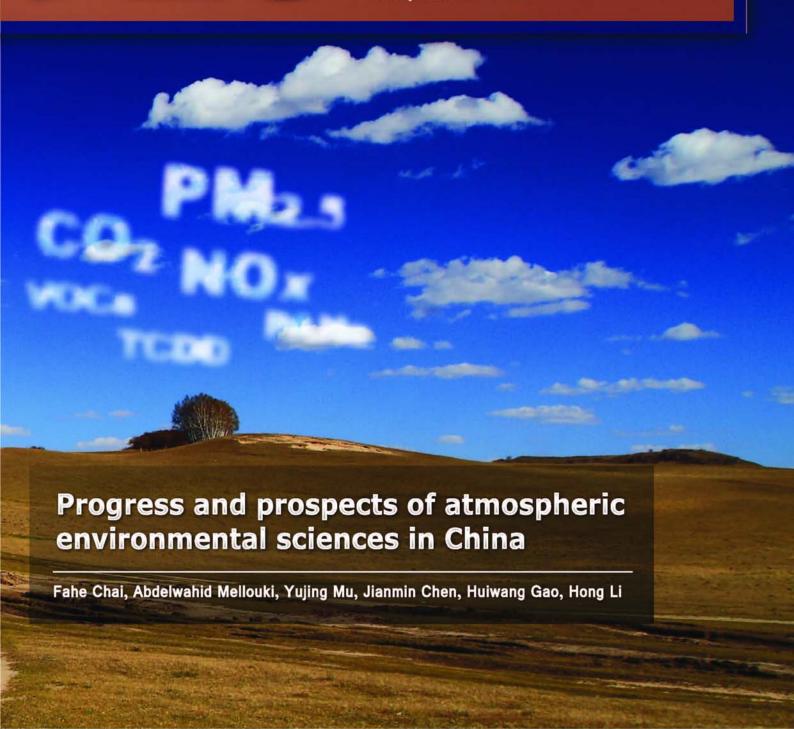
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# Pollution characteristics and health risk assessment of benzene homologues in ambient air in the northeastern urban area of Beijing, China

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#### ABSTRACT

Ambient benzene homologues were measured at a site in the northeastern urban area of Beijing, China, from August 24 to September 4, 2012 by SUMMA canister sampling followed by laboratory determination using cryogenic cold trap pre-concentration-GC-MS/FID, and their health risks were also assessed. Daily total benzene homologues ranged from 0.99 to 49.71 μg/m³ with an average of 11.98 μg/m³. Benzene homologues showed higher concentrations in the morning and evening than that at noontime. Comparison with previous studies revealed a trend of decrease for ambient benzene homologues probably due to the effective emission control in Beijing in recent years. Vehicular exhaust was the main source while volatilization of paints and solvents also made substantial contributions. Health risk assessment showed that BTEX (benzene, toluene, ethylbenzene, *o*-xylene, *m*-xylene and *p*-xylene) and styrene had no appreciable adverse non-cancer health risks for the exposed population, while benzene has potential cancer risk of 1.34E-05. Available data from cities in China all implied that benzene imposes relatively higher cancer risk on the exposed populations and therefore strict control measures should be taken to further lower ambient benzene levels in China

#### Introduction

Benzene homologues are a general term for benzene and its derivatives. In the atmospheric environment field, it usually refers to monocyclic aromatics such as BTEX (benzene, toluene, ethylbenzene, *o*-xylene, *m*-xylene and *p*-xylene), styrene and isopropyl benzene. Being as the most typical components of volatile organic compounds (VOCs) in the air, benzene homologues are also important precursors of near-ground ozone and secondary organic aerosol (Carter, 1994; Zhang et al., 1998). In the ambient

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air, most of benzene homologues are emitted from the volatilization of paints, solvents and adhesives, the release of construction and decoration materials, the incomplete combustion of fossil fuels and biomass burning, and vehicular exhaust, etc. (Liu et al., 2008). Because of their strong toxicity and carcinogenicity (Khan, 2007; Smith et al., 2011), benzene homologues pose serious risk on the respiratory system, hematopoietic system and nervous system of human being. Among the species of benzene homologues, benzene has been categorized to be in group I carcinogens by the International Agency for Research on Cancer (IARC). Long term exposure to benzene may cause bone marrow and induce genetic damage, leukemia and lymphatic diseases. According to the second edition



of the Air Quality Guidelines for Europe (World Health Organization, WHO, 2000), leukemia cancer risk in life cycle of the population exposing to the air with a benzene concentration of  $1 \mu g/m^3$  is  $6 \times 10^{-6}$ .

With the development of the economic growth and acceleration of urbanization in China, the air pollution of VOCs in urban areas, especially benzene homologues, has become a problem in urban areas. In some large and medium-sized cities in China, the concentrations of benzene homologues are high in the air, and the concentrations of benzene in some monitoring sites are higher than the corresponding values designated in the national ambient air quality standard in some countries, such as Japan and Britain (Zhang et al., 2006). At present, research on the environmental criteria of VOCs in the ambient air is still at the starting stage in China, and the national ambient air quality standard for benzene homologues has not been set. However, researchers have begun to pay attention to the ambient air levels of VOCs and toxicological characteristics of benzene homologues in the ambient air in China (Wang et al., 2011; Zhang et al., 2012; Liu et al., 2011; Fan and Zhou, 2008; Bai et al., 2012).

The environment management strategy is now transiting from "Environmental pollution control oriented" to "environmental quality improvement oriented", and will develop into "Environmental risk prevention and control oriented" in China (http://www.mep.gov.cn/zhxx/hjyw.htm). In the National 12th Five-Year Plan for Environment Protection issued in 2012 in China, the study on population exposure and health risk assessment is emphasized. In this context, it is essential to carry out the research about the pollution characteristics, exposure levels and health risk assessment of benzene homologues in the ambient air in China.

With the social and economic development, the air pollution, especially the pollution of ozone and atmospheric particulates, has become a serious problem in Beijing. Recently, multiple occurrences of the wide range of fog and haze for long duration have attracted great attention from the government and the people.

In this article, VOCs samples were collected in the ambient air in the northeastern urban area of Beijing from August 24 to September 4, 2012, and the ambient levels, characteristics and sources of benzene homologues were analyzed. The health risk of BTEX and styrene in the ambient air in the study area were also assessed to provide basis for the improvement of ambient air quality in Beijing and accumulate essential data for the research about environmental health criteria of benzene homologues in China.

#### 1 Materials and methods

# 1.1 Sampling site

Air samples were collected on the roof of the Atmospheric Environment Observation and Research Station in the Chinese Research Academy of Environmental Sciences in the northeastern urban area of Beijing (40°02′N, 116°25′E). The site is 15 m above the ground with no obvious local pollution source nearby. Since the population density in the surrounding area of the monitoring site is relatively high, the monitoring result can reflect the ambient air quality of the breathing zone of the most residents in this area. Therefore, the monitoring data obtained in this site can be used to investigate the pollution level and to make health risk assessment of benzene homologues in the ambient air of Beijing.

#### 1.2 Sampling and analysis

The samples were collected at 8:30-9:00 a.m., 1:30-2:00 p.m., 6:00-6:30 p.m. and 10:00-10:30 p.m. from August 24 to September 4, 2012. From the noontime of September 1 to the morning of September 2, the sampling had to be stopped because of the rain. Totally 43 samples were collected during the experiment. TO-15 method, the standard method to analyze toxic organics in ambient air recommended by U.S. Environmental Protection Agency (US EPA), was used as the reference sampling method. The canisters have the internal surfaces specially passivated by electric polish and silvlation treatment process, and the compressed gas in the canister is specified to have a maximum pressure of  $2.76 \times 10^5$  Pa. Among these canisters, 30 of them have the volume of 3.2 L, and 13 of them have the volume of 3.0 L. The sampling speed, controlled by flow-limiting valve, was 101.5 mL/min; and the sampling time was set to be 30 min. The sampling canisters were placed in a position 1.5 m higher than the ground of the roof and kept a certain distance away from the air outlet on the roof. During the sampling, the meteorological data in the sampling site including wind directions, wind speeds, temperatures, dew point temperatures, solar radiations, ultraviolet radiations and visibilities were determined once an hour using the meteorological measuring instrument (Vaisala Inc., the Netherlands).

According to the TO-15 method, the VOCs samples were analyzed by cryogenic cold trap preconcentration-GC-MS/FID two-dimensional chromatography. The VOCs sample with a certain volume was enriched and concentrated by passing through the Entech 7100A pre-concentration system (Entech Instruments, Inc., USA), and the water and CO<sub>2</sub> were also removed from the sample, then the components of VOCs were rapidly gasified and fed in to the GC-MS/FID system (GC, HP-7890A, Hewlett Packard Co., USA; MSD, HP-5975C, Hewlett Packard Co., USA) to be separated and quantitatively analyzed (Li et al., 2010; Yuan et al., 2010). Most of the target compounds such as alkanes, alkenes and aromatics were separated through the DB-624 chromatographic column (60 m × 0.25 mm × 1.8 μm, J&W Scientific, USA) and quantitatively analyzed

by the mass spectrometry. For the mass spectrometry analysis, the temperature and energy of the ion source were 200°C and 70 eV respectively; the scanning speed was 4 scan/sec; and the scanning scope was 35–350 amu. For the other eight components of  $C_2$ – $C_4$ , they were separated through the PLOT chromatographic column (30 m × 0.25 mm × 3.0  $\mu$ m, J&W Scientific, USA) and measured by FID. For the gas chromatographic analysis, the total running time was 47 min: the initial temperature in GC oven was kept at 30°C for 7 min, then increased to 120°C at a rate of 5°C min, kept at 120°C for 5 min, and then increased to 180°C with a rate of 6°C/min, and kept at 180°C for 7 min.

Standard compounds and internal standard method were used to build the multi-point calibration working curves. The standard gases used were multi-component standard gas including 56 VOCs (PAMS, Scott Specialty Gases Company, USA) and multi-component standard gas which was recommended in the TO-15 method (Scott Specialty Gases Company, USA). The three internal standard compounds used were bromochloromethane, *p*-difluorobenzene and 1-bromo-3-fluorobenzene (Scott Specialty Gases Company, USA). 16 species of benzene homologues were detected in all of the VOCs samples. The daily average mass concentrations of the 16 benzene homologues are listed in **Table 1**.

#### 1.3 Health risk assessment

A human health risk assessment is the process to estimate the nature and probability of adverse health effects in humans who may be exposed to chemicals in contaminated environmental media, now or in the future (http://www.epa.gov/risk/health-risk.htm). In 1983,

the U.S. National Academy of Sciences proposed a fourstep health risk assessment method, which includes hazard identification, dose-response assessment, exposure assessment and risk characterization. "Hazard identification" is to identify whether a chemical is harmful to human health, "Dose-response assessment" is to study the causal relationship between exposure and its effects on human health, "exposure assessment" is to assess and describe the methods, intensities, frequencies and durations of population or ecosystem exposing to risk factors, and "risk characterization" is to determine the probabilities of harmful effects (National Research Council, 1983). This method has become an internationally recognized health risk assessment method and has been widely adopted in many countries such as the Netherlands, France, Japan and China. Based on whether the pollutant is carcinogenic or not, the health risk assessment is divided into cancer risk assessment and non-cancer risk assessment. The US EPA proposed a health risk assessment method for inhalable pollutants for specific places in 1989 (EPA/540/1-89/002), and updated the method in 2009 (EPA-540-R-070-002). According to this new version of the methodology for health risk assessment, concentration of the pollutant in air, rather than inhalation intake of the pollutant in air based on human inhalation rate (IR) and body weight (BW) (e.g., mg/(kg·day)), should be used as the exposure metric (e.g., mg/m<sup>3</sup>). The cancer risk is expressed as the production of the inhalation unit cancer risk (or cancer intension coefficient) and life cycle average exposure concentration. The details are shown as below:

Chronic and sub-chronic exposure concentration (EC,  $\mu g/m^3$ ):

 $EC = (CA \times ET \times EF \times ED)/AT$ 

Pollutant	Mean		Ra	inge	MDL (pptV)	$r^2$
	pptV	μg/m <sup>3</sup>	pptV	μg/m <sup>3</sup>	(FF. · · )	·
Benzene	1.01	1.72	0.11–2.96	0.19–5.09	6.5	0.999
Toluene	2.70	5.41	0.17-10.96	0.35-22.25	4.3	0.998
Ethylbenzene	0.52	1.19	0.04-2.46	0.10-5.76	4.8	0.996
m/p-Xylene	0.90	2.09	0.05-4.50	0.10-10.53	12.5	0.996
o-Xylene	0.23	0.53	0.02-1.11	0.05-2.60	5.2	0.996
Styrene	0.07	0.16	0.01-0.41	0.03-0.94	10.6	0.995
i-Propylbenzene	0.02	0.04	0.01-0.05	0.01-0.13	4.3	0.994
n-Propylbenzene	0.03	0.09	0.01-0.09	0.02-0.23	1.6	0.994
1,3,5-Trimethylbenzene	0.03	0.07	0.01-0.07	0.02-0.18	6.1	0.993
1,2,4-Trimethylbenzene	0.10	0.25	0.02-0.29	0.05-0.77	9.7	0.992
1,2,3-Trimethylbenzene	0.03	0.07	0.01-0.07	0.03-0.20	9.7	0.991
<i>m</i> -Ethyltoluene	0.06	0.15	0.01-0.22	0.03-0.58	7.3	0.992
p-Ethyltoluene	0.04	0.10	0.01-0.14	0.03-0.36	8.4	0.994
o-Ethyltoluene	0.03	0.09	0.01-0.07	0.02-0.25	4.3	0.993
<i>m</i> -Diethylbenzene	0.01	0.03	0.01-0.09	0.01-0.24	5.2	0.990
Benzene homologues	5.78	11.98	0.48-23.50	0.99 -49.71		

MDL: method detection limit.



where, CA (mg/m<sup>3</sup>) is the contaminant concentration in air, ET (24 hr/day) is the exposure time, EF (365 days/yr) is the exposure frequency, ED (70 yr) is the exposure duration, AT  $(70 \times 365 \times 24 \text{ hr})$  is the averaging time.

Non-cancer risk hazard quotient (HQ):

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

where, RfC  $(mg/m^3)$  is the inhalation reference concentration.

Hazard index (the sum of hazard quotients of several pollutants, HI, dimensionless):

$$HI = \sum_{i=1}^{n} HQ_i$$

where,  $HQ_i$  is the non-cancer risk hazard quotient of the ith chemical.

Life cycle cancer risk (*R*, dimensionless):

$$R = EC \times IUR$$

where, IUR (m<sup>3</sup>/µg) is the inhalation unit risk.

#### 2 Results and discussion

#### 2.1 Ambient level and composition characteristics

The mean and range of the total mass concentrations of the 16 benzene homologues in the ambient air in the study area were  $11.98 \,\mu\text{g/m}^3$  and  $0.99\text{-}49.71 \,\mu\text{g/m}^3$ , respectively.

The daily average mass concentration of benzene, toluene, ethylbenzene, m/p-xylene, o-xylene and styrene were 1.72  $\mu g/m^3$ , 5.41  $\mu g/m^3$ , 1.19  $\mu g/m^3$ , 2.09  $\mu g/m^3$ , 0.53  $\mu g/m^3$  and 0.16  $\mu g/m^3$ , respectively. The daily average mass concentrations of the other 9 benzene homologues were relatively low, which were lower than 0.10  $\mu g/m^3$  except those of 1,2,4-trmethyl-benzene, m-ethyltoluene and p-enthyltoluene. The sum of the daily mass concentrations of BTEX species accounted for 91.3% of the total mass concentrations of the 16 benzene homologues, therefore, BTEX were the main components of benzene homologues. The concentration of toluene was the highest, followed by m/p-xylene (**Table 1**).

The relative composition of benzene homologues in the four sampling periods is shown in **Fig. 1**. The sum of daily mass concentrations of BTEX species accounted for about 90% of the total mass concentrations of the 16 benzene homologues, with the percentage of 91.6%, 91.8%, 89.9% and 92.0% in the four durations, respectively. Therefore, from the point of view of relative composition, BTEX were the main components of benzene homologues. In each sampling period, the mass concentrations of toluene were the highest among the 16 benzene homologues. The mass concentrations of benzene were higher than those of *m/p*-xylene in the sampling period of 1:30 and 2:00 p.m., while lower than those of *m/p*-xylene in the other three sampling periods.

The concentrations of BTEX species in the ambient air in Beijing and other domestic and foreign cities were compared as shown in **Table 2**. The sum of the mass

Table 2 Comparison of the mass concentrations of BTEX in ambient air among this study and the related study results (unit: μg/m³)							
City	Sampling time	Benzene	Toluene	Ethylbenzene	Xylene	BTEX	Reference
Beijing	2005/08	5.43	11.14	4.08	12.45	33.1	Song et al., 2008
Beijing	2006/08	8.37	11.41	5.14	13.35	38.27	Duan et al., 2008
Beijing	2008/10-2009/10	8.9	12.4	4	6.2	31.5	Sun et al., 2011
Beijing	2009	4.43	7.03	2.27	6.24	19.97	Wang et al., 2011
Beijing	Summer, 2010	2.1	5.9	2.3	5.1	15.4	Zhang et al., 2012
Beijing	2012/08-2012/09	1.72	5.41	1.19	2.62	10.94	This work
Guangzhou	Summer and winter, 2002	8.87	40.33	7.32	10.82	67.34	Wang et al., 2008
Guangzhou	2007/10-2007/12	2.5	18.04	6.17	12.92	39.63	Ling et al., 2011
Shanghai	2004/12	14.4	55.6	16.1	32.94	119.04	Zhang et al., 2006
Shanghai	2004/12-2005/02	13.23	43.66	13.5	22.01	92.4	Tang et al., 2006
Nanjing	2005/09	7.69	25.44	8.27	14.99	56.39	Lu, 2006
Nanjing	2006/04-2007/01	6.4	19.8	2.9	5.5	34.6	Wang and Zhao, 2008
Zhengzhou	2003	17.23	16.34	3.79	10.67	48.03	Wang et al., 2008
Lianyungang	2006/01-2006/10	16.95	19.36	3.86	16.69	56.86	Li and Zhang, 2008
Shenyang	2008/04-2009/07	5.6	5.8	1.4	11.4	24.2	Liu et al., 2011
Ashiya (Japan)	2005-2009	1.6	13	8.1	8.7	31.4	Okada et al., 2012
Sumoto (Japan)	2005-2009	2	12	8.1	8.7	30.8	Okada et al., 2012
Yangsan (Korea)	2006/02-2006/12	0.67	5.09	4.39	7.6	17.75	Song et al., 2009
Pamplona (Spain)	2006/06-2007/06	2.84	13.26	2.15	6.01	24.26	Parra et al., 2009
Delhi (India)	2001/10-2002/09	48	85	7	45	185	Hoque et al., 2008
Camden (USA)	2004/06-2006/07	1.35	2.48	0.40	1.68	5.91	Wu et al., 2012

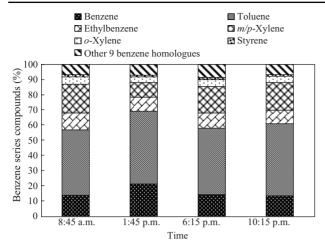


Fig. 1 Relative composition of benzene homologues.

concentration of BTEX species in the study area was lower than that of BTEX in other cities in China and higher than that of BTEX in Camden, USA (Wu et al., 2012); the mass concentration of benzene was lower than that of benzene in other cities in China, in Delhi, India (Hoque et al., 2008) and in Pamplona, Spain (Parra et al., 2009), and equal to that in Ashiya, Japan (Okada et al., 2012), in Sumoto, Japan (Okada et al., 2012) and in Camden, USA (Wu et al., 2012), and higher than that of benzene in Yangsan, Korea (Song et al., 2009); the mass concentration of toluene was lower than that of toluene in some cities in China, in Ashiya, Japan (Okada et al., 2012), in Sumoto, Japan (Okada et al., 2012), in Delhi, India (Hoque et al., 2008) and in Pamplona, Spain (Parra et al., 2009), equal to that of toluene in Shenyang, China (Liu et al., 2011) and in Yangsan, Korea (Song et al., 2009), and higher than that of toluene in Camden, USA (Wu et al., 2012); and the mass concentration of toluene was lower than the daily average exposure concentration limit of toluene designated in WHO documents (8.21  $\mu$ g/m<sup>3</sup>) (Kourtidis et al., 2002). According to the comparison of mass concentrations of BTEX species in the ambient air in Beijing since 2005, it was indicated that the sum of the mass concentrations of BTEX species, the mass concentration of benzene and that of toluene in the summer of 2012 decreased by 53.4%, 68.3% and 51.4%, respectively, and the sum of the mass concentrations of BTEX species, the mass concentration of benzene showed the decreasing trend in the summer season. This might be related to the fact that Beijing has stepped up efforts to control vehicle exhaust pollution in recent years. In general, the air pollution of BTEX in the urban area in China was more serious than the cities in other countries, and the mass concentrations of toxic benzene and toluene in most cities in China were higher than those of benzene and toluene in the cities in other countries. However, the deterioration trend of pollution status of benzene homologues has been controlled to some extent due to the effective control countermeasures taken in Beijing in recent years.

#### 2.2 Variation characteristics

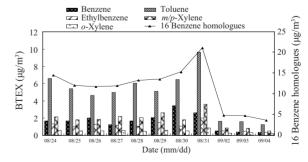
#### 2.2.1 Daily variation

The daily variations of the mass concentrations of benzene homologues in the ambient air are shown in Fig. 2. The daily variation trend of the sum of the mass concentrations of BTEX species was quite similar to that of the total mass concentrations of the 16 benzene homologues. From August 24 to August 26, the total mass concentrations of the 16 benzene homologues showed downward trend, while from August 26 to August 31, the total mass concentrations of the 16 benzene homologues increased from 11.07 to 20.09 μg/m<sup>3</sup>. The analysis of the observation meteorological conditions during the sampling period showed that the weather conditions were stable from August 26 to August 30 with sunny days and the average ambient temperature being 26.94-31.10°C, thus the total mass concentrations of the 16 benzene homologues showed little variations in these days. The total mass concentrations of the 16 benzene homologues (20.09 µg/m<sup>3</sup>) was obviously higher on August 31 than those in the period of August 26 to August 30 since it was cloudy on August 31, which means that the benzene homologues pollutants accumulated in the air due to the weak photochemical reaction with lower temperature and low light intensity.

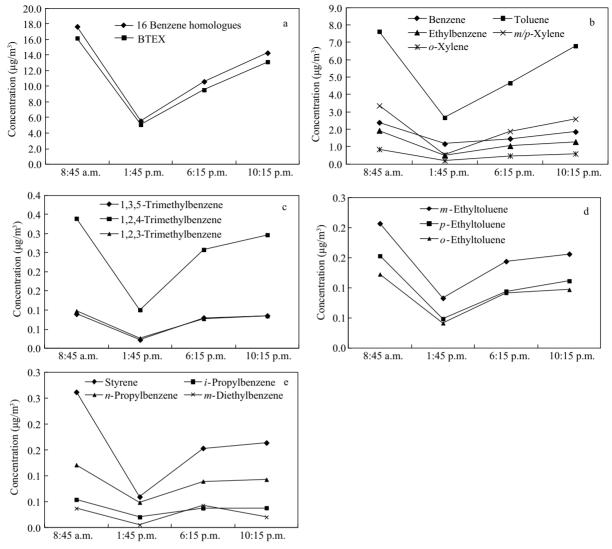
From September 2 to September 4, the total mass concentrations of 16 benzene homologues and the sum of the mass concentrations of BTEX species reduced obviously. The range of the total mass concentrations of 16 benzene homologues was 3.15– $4.32 \,\mu g/m^3$ , which reduced by 78.5%–84.3% compared to that on August 31. The average ambient temperature was 21.94–23.92°C in the period of September 2 to September 4, and it was raining on September 1, so it can be speculated that the rainfall had obviously removed the benzene homologues from the air. Therefore, the meteorological conditions and intensity of photochemical reaction have significant effect on the mass concentrations of benzene homologues in the ambient air.

#### 2.2.2 Diurnal variation

The diurnal variations of the mass concentrations of benzene homologues in ambient air in the study area are shown in Fig. 3. The total mass concentrations of the 16



**Fig. 2** Daily variations of the mass concentrations of benzene homologues.



 $\textbf{Fig. 3} \quad \text{Diurnal variations of the mass concentrations of benzene homologues}.$ 

benzene homologues, the sum of the mass concentrations of BTEX species and each species of benzene homologues were all higher in the morning and in the evening, and lower at noontime, and the minimums appeared at the period of 1:30-2:00 p.m. The total mass concentrations of the 16 benzene homologues decreased by 68.5% from the period of 8:45-9:00 a.m. to the period of 1:30-2:00 p.m., and increased 1.57 times during the period of 1:30-2:00 p.m. to 10:00-10:30 p.m.; while the mass concentration of benzene reduced by 51.3% from the period of 8:45-9:00 a.m. to the period of 1:30-2:00 p.m., and increased 0.6 times from the period of 1:30-2:00 p.m. to the period of 10:00-10:30 p.m. The total mass concentrations of the 16 benzene homologues and the sum of the mass concentrations of BTEX species increased more quickly from the period of 1:30-2:00 p.m. to the period of 6:00-6:30 p.m. than from the period of 6:00-6:30 p.m. to the period of 10:00-10:30 p.m.

The diurnal variation characteristics of BTEX obtained

in this study are in consistent with the study results in the references. Sun et al. (2011) studied the concentration variation of BTEX in ambient air in Beijing and pointed out that the diurnal variation trend of BTEX has a double peak changing pattern, with peak values appearing in the periods of 8:00–10:00 a.m. and 6:00–9:00 p.m., and minimum value appearing at about 2:00 p.m. The research made by Velasco et al. (2007) showed that the minimum concentration of BTEX in the urban ambient air in Mexico also appeared at about 2:00 p.m.

The diurnal variation trend of the mass concentrations of benzene homologues is mainly influenced by the variation of emission sources, photochemical reaction intensity, and meteorological diffusion conditions. During the traffic rush in the morning and in the evening, the exhaust from benzene homologues emission sources increase and it is difficult for benzene homologues to spread due to the low temperature and atmospheric boundary layer height, thus the mass concentrations of benzene homologues are

higher in the morning and in the evening. Whereas at noontime, the mass concentrations of benzene homologues are obviously reduced by the rapid photochemical reaction and high diffusion speed of benzene homologues induced by the strong light intensity, high temperature and atmospheric boundary layer height, so the minimum concentrations of benzene homologues appeared at the period of 1:30–2:00 p.m. in this study.

#### 2.3 Sources identification

Previous studies has shown that the potential emission sources of BTEX can be identified based on the correlation of each BTEX species in ambient air and the specific concentration ratio of benzene to toluene (B/T) (Atkinson and Arey, 2003; Barletta et al., 2005). It is generally believed that a value of 0.5 for B/T (wt./wt.) implies the vehicular exhaust is the main emission source of benzene homologues in the urban ambient air (Barletta et al., 2005); higher B/T value implies that the mass concentration of benzene homologues may be influenced by the petrochemical industry and the combustion of fossil fuels; while lower B/T value indicates that the concentration of benzene homologues may be impacted by the volatilization of paints and solvents besides the influence from traffic exhaust. As shown in Table 3, there are close correlation between the BTEX species in ambient air in the study area, thus the BTEX species have good homology with each other. Except for benzene and xylene whose correlation coefficients (R) were less than 0.8, the correlation coefficients of other BTEX species were all larger than 0.8, and some even reach 0.98. The low correlation between benzene and xylene may be due to the photochemical consumption intensities difference between them caused by the their photochemical activity difference since among the BTEX, the photochemical activity of benzene is the weakest and that of the *m/p*-xylene is the strongest (Atkinson, 2007). In this study, the B/T value was  $0.36 \pm 0.15$ , which means that the vehicular exhaust was the main emission source of benzene homologues in the urban ambient air in the study area, and the volatilization of paints and solvents was also an important source.

#### 2.4 Health risk assessment

Based on the above analysis of the pollution characteristics of benzene homologues, the health risk of the 6 species of

BTEX and styrene (that means totally 7 species of benzene homologues), whose dose-response data can be obtained in the Integrated Risk Information System database of US EPA, on the adults in the study area were assessed by adopting the health risk exposure assessment methodology of US EPA. It is worth noting that carcinogen benzene also has non-cancer risk and its non-cancer risk should be assessed; for ethylbenzene and styrene, they are possible carcinogen to humans, while there are no relating parameters for cancer risk assessment at present, so cancer risk assessment cannot be made for ethylbenzene and styrene; for the other 4 benzene homologues, they are not carcinogens, so only non-cancer risk is needed to be assessed for these 4 benzene homologues species. The calculated parameters for non-cancer risk assessment and cancer risk assessment of the 7 benzene homologues are shown in **Table 4**. For 7 benzene homologues EC varied in the range of  $0.16-5.41 \mu g/m^3$ , and HQ changed from 1.60E-04 to 5.73E-02 and HI was 8.59E-02. Since the values of HO of benzene and xylene are both higher than those of the other 5 species of benzene homologues, the non-cancer risk of benzene and xylene are bigger than that of the other 5 species of benzene homologues in the air. The value of risk of benzene was 1.34E-05. At present, there is no health risk assessment standard in China, whereas according to US EPA document "EPA-540-R-070-002", if the value of HQ of a specific pollutant is lower than 1, the pollutant has no obvious non-cancer risk to human health, and the acceptable value of Risk for ordinary adult is 1E-06. By comparison with the corresponding values designated in the US EPA document, it can be seen that the values of HI of the 7 benzene homologues in ambient air in the study area were all less than 1, so the 7 benzene homologues have no obvious non-cancer risk to the general population in the study area; while the value of Risk of benzene was larger than 1E-06, which indicated that there was potential cancer risk to the populations from benzene if the population suffer long-term benzene exposure in the study area.

Systematic health risk assessment and management system have not been established in China up to now, and the studies on the exposure and health risk assessment of benzene homologues in urban ambient air are few and scattered. In this study, the results of health risk assessment on toxic and harmful VOCs in the urban ambient air in China were summarized to further analyze the health risk of benzene homologues in urban ambient air in China

Table 3 Correlation coefficients between BTEX							
Pollutant	Benzene	Toluene	Ethylbenzene	m/p-Xylene	o-Xylene		
Benzene	1						
Toluene	0.847	1					
Ethylbenzene	0.805	0.906	1				
m/p-Xylene	0.775	0.905	0.985	1			
o-Xylene	0.786	0.915	0.987	0.988	1		

Pollutant	$EC$ $(\mu g/m^3)$	RfC* (mg/m <sup>3</sup> )	$IUR^* \\ (m^3/\mu g)$	HQ	Risk
Benzene	1.72	0.03	7.80E-06	5.73E-02	1.34E-05
Toluene	5.41	5	_	1.08E-03	-
Ethylbenzene	1.19	1.0	_	1.19E-03	_
m/p-Xylene	2.09	0.1	_	2.09E-02	-
o-Xylene	0.53	0.1	_	5.32E-03	_
Styrene	0.16	1	_	1.60E-04	_

EC: exposure concentration, Rfc: inhalation reference concentration, IURL: inhalation unit risk, HQ: hazard quotient, HI: hazard index. \*The values of RfC and IUR are taken from IRIS, U.S. EPA (http://www.epa.gov/iris).

(Table 5). Although researches about the exposure and health risk of benzene homologues have been conducted in China, the number of case studies and the concerned cities are limited, and the target benzene homologues species are mainly BTEX and styrene. In the listed results, the values of HQ of the 7 benzene homologues in ambient air varied from 1.60E-04 to 1.58E-01 and the value of HI of the 7 benzene homologues in each study case were less than 1, which means that there might be no obvious non-cancer risk in the urban air from BTEX and styrene. The value of HQ of benzene was larger than that of the other 6 benzene homologues species, respectively, which indicated that the non-cancer risk from benzene is larger than the other 6 benzene homologues species in the ambient air. Except for Nanjing, the values of Risk of benzene in ambient air of the other cities were all bigger than 1E-06, which showed that benzene has cancer risk to the exposed populations in ambient air in these cities.

#### 3 Conclusions

In this study, VOCs samples were collected by SUM-MA canisters in northeastern urban area in Beijing, and 16 benzene homologues species were analyzed by using the cryogenic cold trap pre-concentration-GC-MS/FID

chromatographic analysis method. The ambient levels, variation characteristics and emission sources of benzene homologues were studied, and the health risk of BTEX species and styrene in ambient air in the study area were assessed. The results showed that the mean and the range of the daily total mass concentrations of 16 benzene homologues were 11.98 µg/m<sup>3</sup> and 0.99-49.71 μg/m<sup>3</sup>, respectively. The species with the highest mass concentration among the 16 benzene homologues was benzene, followed by m/p-xylene. BTEX were the main components of benzene homologues in the study area and the mean of the sum of the daily mass concentration of BTEX species was 10.94 µg/m<sup>3</sup>. The diurnal variation of the mass concentrations of benzene homologues showed a changing trend of higher in the morning and in the evening, and lower at noontime, and the minimums appeared at the period of 1:30-2:00 p.m. Comparative study showed that the air pollution of BTEX in the urban area in China was more serious than the cities in other countries; however, the deterioration trend of pollution status of benzene homologues has been controlled to some extent due to the effective control countermeasures taken in Beijing in recent years. Emission sources identification analysis showed that vehicular exhaust was the main source of benzene homologues with the volatilization of paints and solvents being the important emission sources. Health risk assessment showed that the non-cancer HO of BTEX and styrene were between 1.60E-04 and 5.73E-02, which means that BTEX and styrene have no appreciable risk of adverse non-cancer health effect on the exposed population. The cancer risk of benzene was 1.34E-05, which indicates that benzene has potential cancer risk to exposed population. By comparing the corresponding data about health risk assessment of benzene series compounds in some cities in China in the references and the data obtained in this study, up to now, it is concluded that benzene can impose relatively high cancer risk to the exposed populations in the ambient air of cities in China. Therefore, strict counter measures should be taken to further control ambient air levels of benzene, and it is imperative to start the related studies and develop the environmental health criteria and national ambient air quality standard for benzene in China. However, due to the limitation of

Table 5	Table 5 Comparison of the results of health risk assessment among this study and the related study results in China								
City	Ben	zene	Toluene	Ethylbenzene	m/p-Xylene	o-Xylene	Styrene	Reference	
	Risk	HQ	HQ	HQ	HQ	HQ	HQ		
Beijing	1.34E-05	5.73E-02	1.08E-03	1.19E-03	2.09E-02	5.32E-03	1.60E-04	This work	
Beijing	2.21E-05	1.58E-01	_	1.69E-02	7.76E-03		1.06E-02	Zhou et al., 2011	
Beijing	4.19E-05	1.57E-01	2.39E-02	3.29E-03	8.06E-03	3.53E-03	_	Zhang et al., 2012	
Tianjin	2.18E-05	_	_	_	_	_	_	Zhou et al., 2011	
Hangzhou	9.43E-06	9.43E-02	2.30E-02	2.0E-03	5.61E-02	_	4.0E-04	Zhang, 2007	
Nanjing	1E-07	-	-	_	-	-	-	Hu et al., 2010	

-: no data.

<sup>-:</sup> no data.

the number and the representative of the air samples, the pollution status, sources and health risk assessment of benzene homologues were only preliminarily discussed in this study; further research is needed in the future.

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