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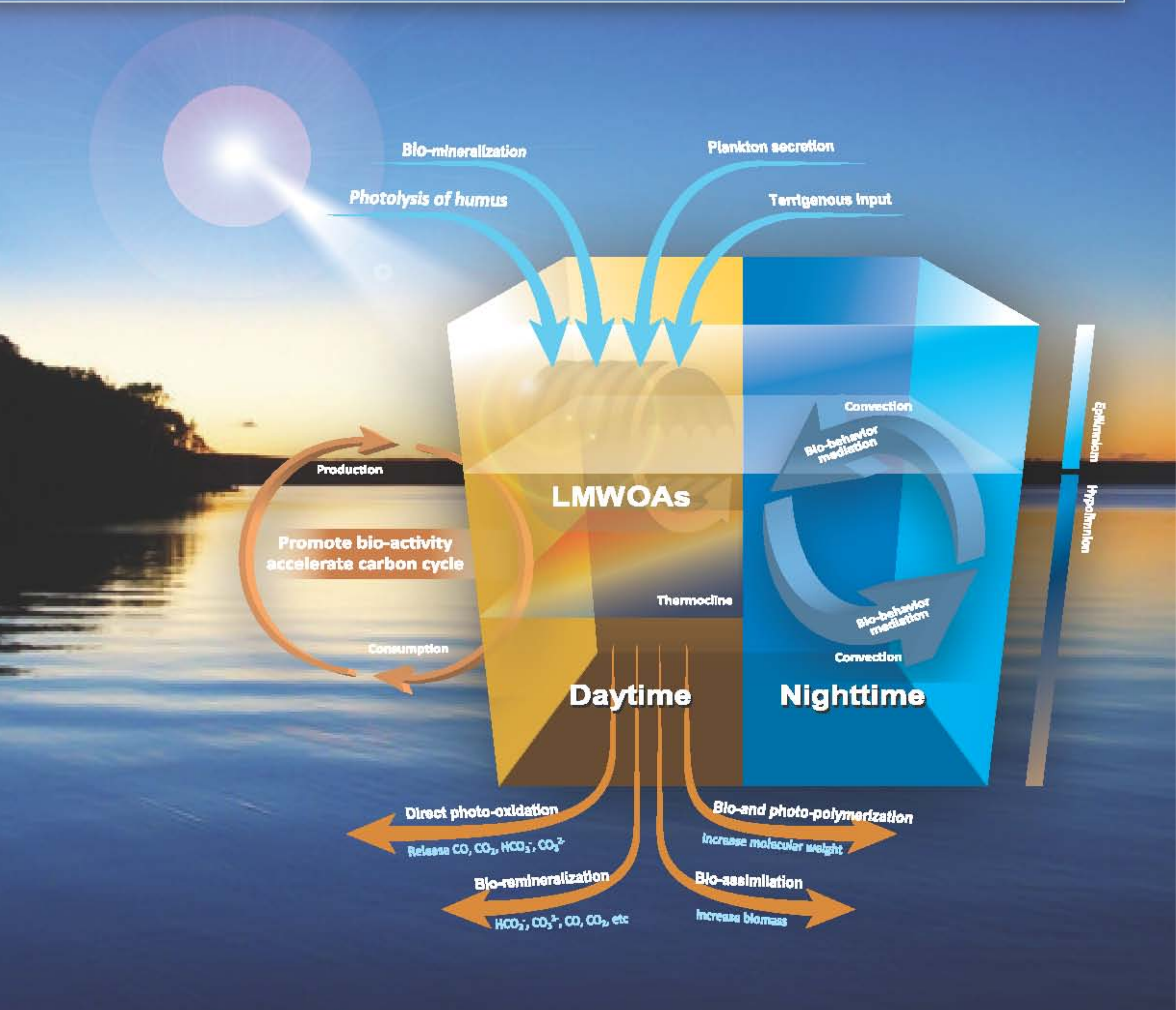

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BTEX pollution caused by motorcycles in the megacity of HoChiMinh

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Abstract

Monitoring of benzene, toluene and xylenes (BTEX) was conducted along with traffic counts at 17 roadside sites in urban areas of HoChiMinh. Toluene was the most abundant substance, followed by *p,m*-xylenes, benzene, *o*-xylene and ethylbenzene. The maximum observed hour-average benzene concentration was 254 μg/m^3. Motorcycles contributed to 91% of the traffic fleet. High correlations among BTEX species, between BTEX concentrations and the volume of on-road motorcycles, and between inter-species ratios in air and in gasoline indicate the motorcycle-exhaust origin of BTEX species. Daily concentrations of benzene, toluene, ethylbenzene, *p,m*-xylenes and *o*-xylene were 56, 121, 21, 64 and 23 μg/m^3, respectively. *p,m*-xylenes possess the highest ozone formation potential among the BTEX family.

Key words: air pollution; benzene; BTEX; HoChiMinh; motorcycle exhaust

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Introduction

Rapid global industrialisation in the last half-century has resulted in a rapid increase in the urban population and the formation of megacities with high population and population density. This phenomenon has been most dramatic on the least-urbanised Asian and African continents. Currently, Asia alone has more than 10 megacities, including Shanghai, China (23 million, 2011), Beijing, China (19.6 million, 2011), Dhaka, Bangladesh (16 million, 2011), Tokyo, Japan (13 million, 2011), Karachi, Pakistan (13 million), Delhi, India (12.8 million, 2011), Guangzhou, China (12.7 million, 2011), Mumbai, India (12.5 million, 2011), Manila, the Philippines (11.5 million, 2009), Seoul, Korea (10.6 million, 2011) and Jakarta, Indonesia (10.2 million, 2011). Most of the megacities in developing countries are currently experiencing serious air pollution by particulate matter and volatile organic carbons (VOCs) due to the uncontrolled expansion of private means of transportation. Among the VOC pollutants, special attention has been paid to BTEX species, especially to benzene, due to their adverse effects on human health. A main source of BTEX in urban areas is gasoline evaporation and vehicle emission. It is accepted that benzene is a human carcinogen (US EPA, 2012). The risk of leukaemia by lifetime exposure to benzene at 17, 1.7 and 0.17 μg/m^3 is, respectively, 10^{-4}, 10^{-5} and 10^{-6} (WHO, 2000). Additionally, benzene and other aromatic hydrocarbons also contribute to the formation of ground ozone, photochemical smog and toxic peroxyacylnitrates through atmospheric photochemical processes. Benzene is highly toxic; therefore, the WHO and US EPA do not specify any save level for benzene exposure. The Vietnamese National Air Quality Standards (NAQS) for time-weighted average hour and annual ambient concentrations of benzene are 22 and 10 μg/m^3, respectively.

Benzene pollution is an important issue for the megacities of developing countries. The annual average roadside benzene concentration in Delhi, India was (86.47 ± 53.24) μg/m^3 in 2001–2002, and has increased continuously despite the full implementation of compressed natural gas (CNG) in a the public transportation system in December 2002. Average benzene concentrations of the pre- and post-CNG periods at two traffic intersections in Delhi were (116.32 ± 51.65) and (187.49 ± 22.50) μg/m^3, respectively. The reason for these enhanced concentrations has been solely attributed to the increase in the vehicular population from 3.5 million in 2001–2002 to 5.1 million in 2007 (Khillare et al., 2008). Benzene concentrations have been found to be 67 μg/m^3 in Cairo, Egypt (Khoder, 2007), 14.7 μg/m^3 in Mumbai, India (Gauri et al., 2011), 11.8 μg/m^3 in Manila; the Philippines (Balanay and Lungu, 2008); 51.5 μg/m^3 in Guangzhou, China (Wang et al., 2002) and 27 μg/m^3 in Algiers, Algeria

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(Rabah et al., 2006).

HoChiMinh has a population of around 8.5 million, and is the financial, industrial and commercial centre of Vietnam. Urban transportation in HoChiMinh, like in any urban area in Vietnam, depends on motorcycles. The motorcycle and automobile populations in the city in March 2008 were 3,444,868 and 346,355, respectively; these numbers increased to 3.9 million and 386,000, respectively, in June 2009. The estimated number of motorcycles in the middle of 2012 is around 5 million. A large number of vehicles employ old technology. Traffic volume is extremely high and traffic jams are frequent; as a consequence, pollution by VOC and particulate matter has resulted in a dramatic decline in air quality. VOC monitoring is not obligatory in Vietnam, hence VOCs have been desultorily monitored in Hanoi and HoChiMinh through some short-term programmes carried out by the EPA or university research groups. A study on 73 hydrocarbons and chlorinated hydrocarbons at nine sites in HoChiMinh in 2002 using Compendium Method TO-15 (US EPA, 1999) revealed a high total VOC concentration of 1262 ppbv and a benzene concentration of 63 ppbV (Lan et al., 2006). The weekly average benzene concentrations measured by the HoChiMinh EPA using Radiello passive samplers on six streets in HoChiMinh in July 2008–June 2009 were 7.9–72.9 μg/m^3 (HoChiMinh EPA, 2009), additionally, benzene concentrations at all sites in the first half of 2009 were 1.07–1.46 times higher than those in the first half of 2008. Hanoi is the capital city of Vietnam with a population of 3.5 million. Truc and Oanh (2007) reported a roadside benzene level of 65 μg/m^3 on a busy street in Hanoi. The EURO II standard came into effect in Vietnam in July 2008.

This study aimed to investigate roadside BTEX in urban areas in HoChiMinh and the impact of different means of transportation on BTEX pollution.

1 Materials and methods

1.1 Sampling sites

Samples were collected at 17 sites (S1–S17) located on main roads in nine residential districts (**Fig. 1**). All sites have wide pavement for pedestrians. Sites S13–S17 are located in the central district of HoChiMinh.

1.2 Sampling and analyses

The NIOSH 1501 method (NIOSH, 2003) using active sampling and solvent extraction was applied for air sampling. Sample tubes containing 200 mg of activated charcoal were purchased from Sibata (Japan). A programmable minipump (MP∑30, Sibata, Japan) was calibrated using a bubble flow meter. A breakthrough experiment was conducted by pumping air at a flow of 130 and 150 mL/min in 56 min through two sample tubes connected in series. Sampling was done during rush hour

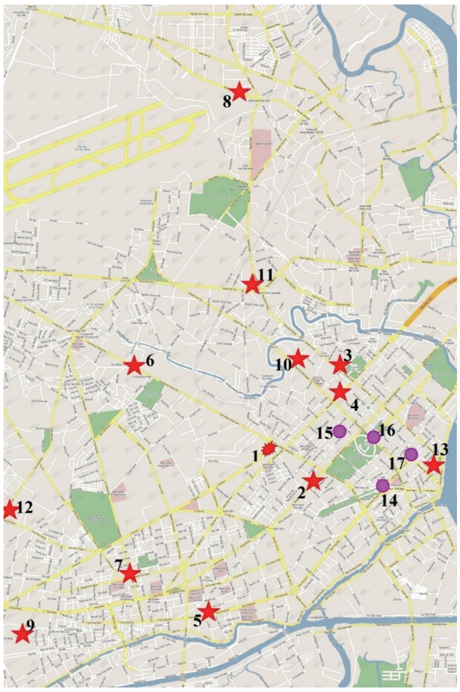

Fig. 1 Locations of sampling sites (S1–S17).

at the entrance of a university motorcycle park. Sample tubes were sealed with polypropylene caps and sent to the laboratory. The charcoal was transferred into GC vials and extracted with 1 mL of benzene-free carbon disulphide containing fluorobenzene and chlorobenzene as the internal standards. The transfer was performed in a glove box filled with zero air. Vials were shaken occasionally for 45 min, and left for 1 hr in a refrigerator for the charcoal to settle. Aliquots were quantified using a Hewlett Packard HP 5890 II gas chromatograph equipped with a flame ionisation detector, an autosampler and an HP-5MS 30.0 m × 0.25 mm × 0.25 μm column. An amount of any BTEX species in the second tube was not more than 0.9% of that in the first tube, confirming the applicability of the sampling procedure.

Sampling at sites was performed on work days in the fourth quarter of 2009. Sampling was performed at a flow rate of 130 mL/min in 56 min. The total sampling volume was 7.41–7.42 L. Sampling was carried out each hour consecutively for 24 hr at site S1; during rush hour (7:00–8:00 and 17:00–18:00) and non-rush hour (12:00–13:00) at sites S2–S13; and from 8:00–9:00 at the remaining sites. The inlet of sample tube was placed 1.7 m above the ground and 1.5 m away from the roadside. In total, 64 samples were collected. A blank was transported to the sampling sites and back to the laboratory. Samples were sealed with polypropylene caps, kept in an air-tight polypropylene tube packed in a aluminium zippered laminar envelope and cold-stored until analysis.

Samples were analysed within one week of sampling. Analysis was carried out as reported above. The concentration of a pollutant in air (C, μg/m^3) was evaluated using the following equation:

$$C = \frac{W_s - W_b}{V \times DE} \times 1000$$

where, W_s (μg) and W_b (μg) are the collected and the blank amount of the pollutant by analysis, V (L) is sampling volume and DE is the desorption efficiency. DE was given by the manufacturer for each lot of activated charcoal and was 98% in this study.

Two gasoline types marketed in Vietnam, RON92 and RON95, were analysed. Gasoline was diluted in benzene-free carbon disulphide containing internal standards and injected into the gas chromatograph for quantification.

1.3 Traffic observation

Traffic was recorded by a video camera. Recorded videos were replayed for traffic counts. Means of transportation were divided into the following groups: motorcycles, under 9-seat cars, 12–24-seat passenger cars, above 25-seat passenger cars, light-duty trucks and heavy-duty trucks.

2 Results and discussion

2.1 Hour-average BTEX concentrations

Toluene was the most abundant species, followed by *p,m*-xylenes (*p,m*-X), benzene (B), *o*-xylene (*o*-X) and ethylbenzene (E) in all samples. **Figure 2** shows the diurnal variation in hour-average BTEX concentrations at site S1. Pollutant levels were high in the daytime and low at night. Peaks were observed during rush hour at 7:00–9:00 and 17:00–18:00. The benzene concentration was lower than the NAQS only for a short period of time at night from 0:00–5:00. Daily average concentrations of benzene, toluene, ethylbenzene, *p,m*-xylenes and *o*-xylene at site S1 were, respectively, 56, 121, 21, 64 and 23 μg/m^3.

The BTEX concentrations at 17 sites are reported in **Fig. 3**. Commonly, BTEX concentrations were low at midday and high during rush hour, and they were higher in the afternoon than in the morning. High BTEX concentrations were observed at sites S1, S6, S7, S8, S9 and S11 located on narrow roads with dense traffic fleets and high buildings. All observed benzene concentrations during the day were higher than the NAQS. Maximum hour-average concentrations of benzene, toluene, ethylbenzene, *p,m*-xylenes and *o*-xylene were 254, 619, 95, 263 and 116 μg/m^3, respectively.

2.2 Relationships between benzene and C_1,C_2-benzene

The inter-species ratios of BTEX pollutants depend on fuel composition, sources, climatic conditions as well as the age of air parcels since BTEX species sink at different rates under sunlight due to their different photochemical activities. Inter-species ratios are an important indicator of sources. A main source of BTEX species in urban areas is vehicle emission. It is accepted that benzene originates from traffic emissions and gasoline evaporation only, while other BTEX species may be airborne from other additional sources like industry or construction. C_1,C_2-benzene/benzene (C_1,C_2-B/B), especially toluence/benzene (T/B) ratios, are often used to identify sources. T/B values below 3 have been found to be characteristic of traffic emissions worldwide, including in Vietnam and China (Perry and Gee, 1995; Brocco et al., 1997; Heeb et al., 2000b; Monod et al., 2001; Chan et al., 2002; Hiesh et al., 2006; Kumar and Tyagi, 2006; Khoder, 2007; Truc and Oanh, 2007; Hoque et al., 2008; Liu et al., 2009; Matysik et al., 2010). T/B values of 1.5–4.3 are considered an indicator of traffic emissions, as reported by Hoque et al. (2008) and Liu et al. (2009). For T/B values > –4.3, solvent source impacts are likely. A specific B/T ratio below 0.20 has been proposed and used as an indicator of samples strongly affected by industrial emissions in Dongguan, China (Barletta et al., 2008), while a ratio of 0.4–1.0 has been used as an indicator of air propelled by vehicular exhaust in Beijing (Wang et al., 2012). T/B > 4.3 was used to identify sources

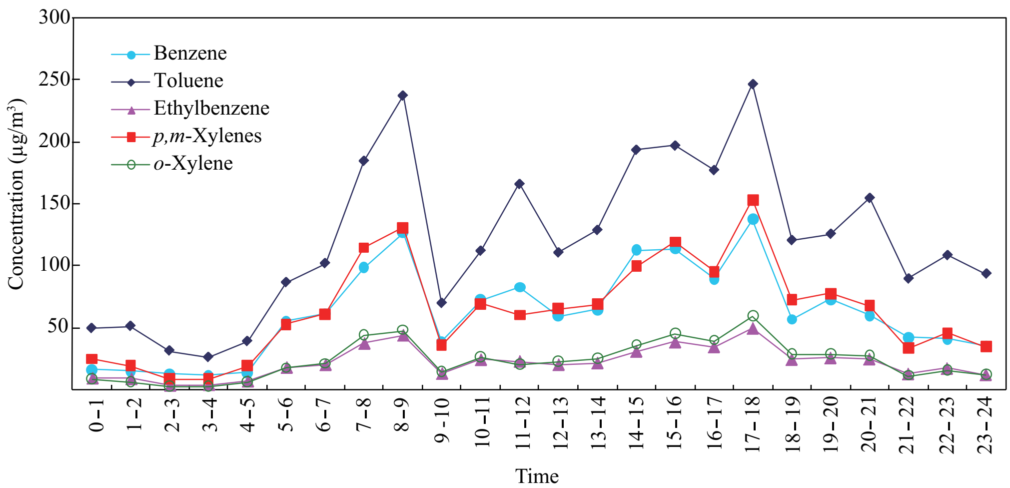

Fig. 2 Diurnal variation of BTEX concentrations at roadside in HoChiMinh City.

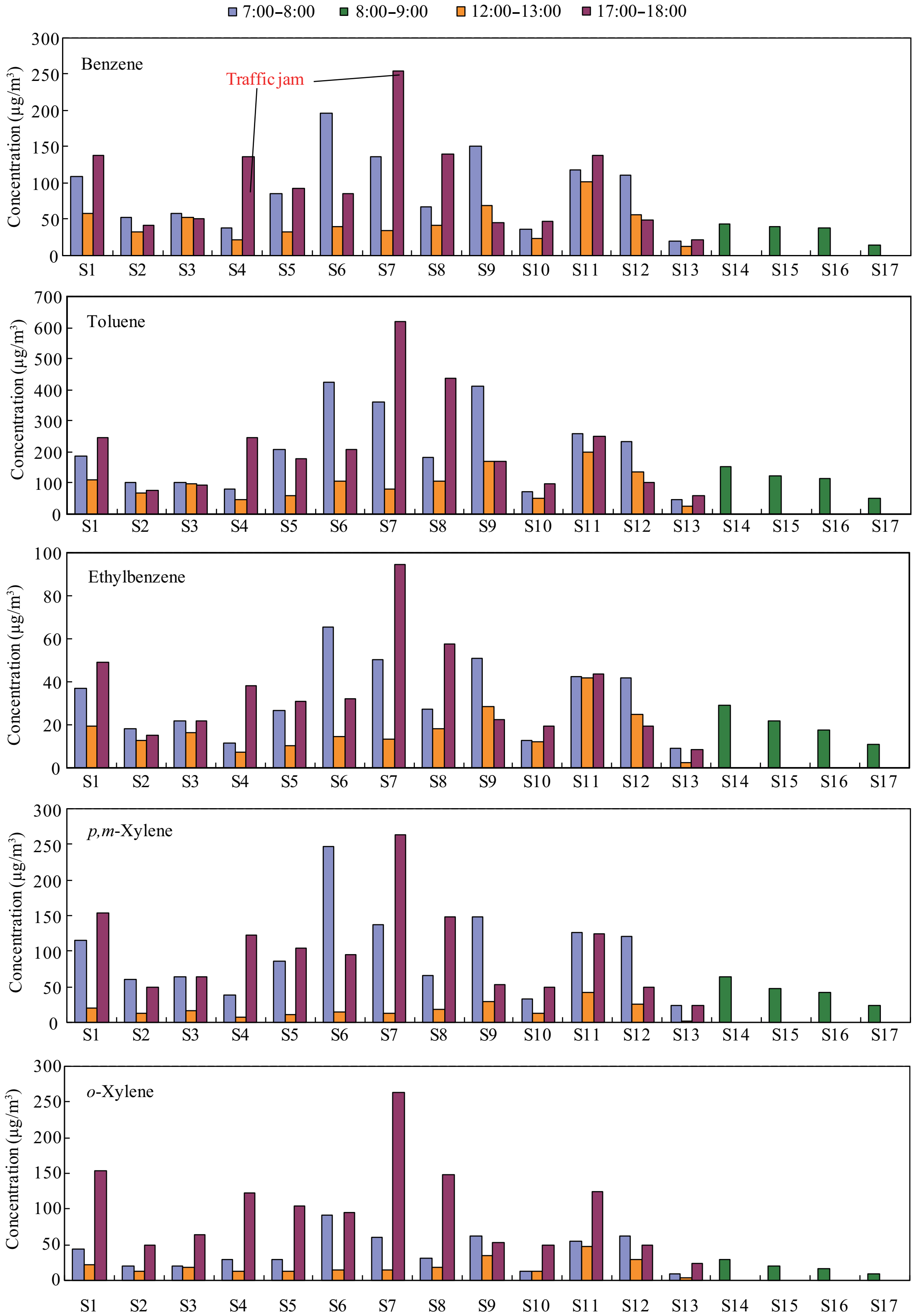

Fig. 3 BTEX concentrations at the sampling sites during different sampling campaigns.

influenced by solvent use in Windsor, Ontario, Canada (Xu et al., 2010). A high T/B ratio (8.6) in a neighbourhood of an industrial park in Taiwan suggested large additional sources of toluene from industry (Hiesh et al., 2006). An overview of inter-species ratios between BTEX species in different environments in Asia, Europe and South America was provided by Monod et al. (2001). A T/B value of 2.3 (R^2 = 0.91) and a B/E value of 2.16 (R^2 = 0.87, E/B = 0.46)

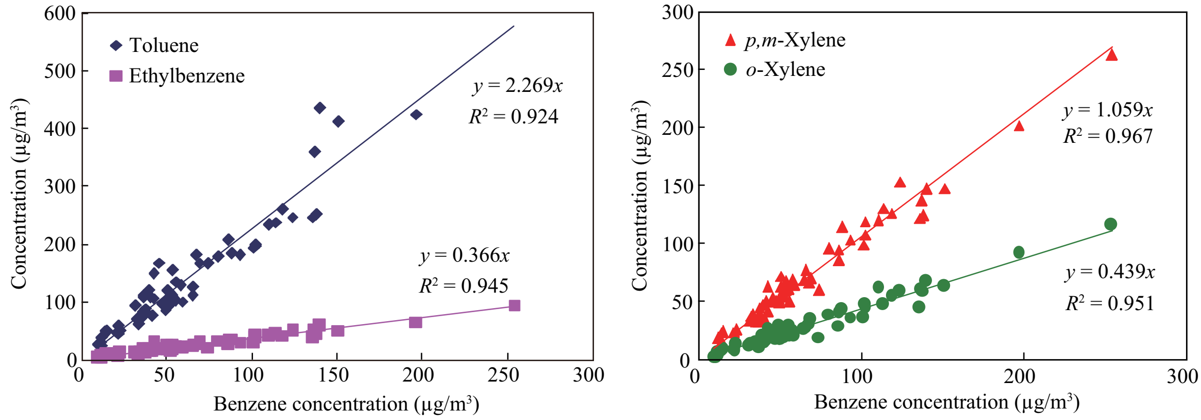

Fig. 4 The correlation between benzene and other BTEX species.

were reported for a traffic microenvironment.

A good linear correlation between concentrations of benzene and other BTEX species at all sites (**Fig. 4**) indicates a same main source of BTEX species. The contents of benzene, toluene, ethylbenzene, *p,m*-xylenes and *o*-xylene in RON92 and RON95 gasoline were 1.81, 4.55, 0.30, 1.21, 0.45 and 1.85, 4.20, 0.54, 1.70 and 0.77 wt.%, respectively. Thus, the T/B, E/B, *p,m*-X/B and *o*-X/B ratios in A92 and A95 gasoline were 2.5, 0.17, 0.7 and 0.25 and 2.3, 0.29, 0.9 and 0.42, respectively. The C_1,C_2-B/B values obtained in roadside air were very close to those in A95 gasoline. All these findings confirm that traffic emission is the main source of BTEX alongside roads in HoChiMinh.

2.3 Diurnal variation of inter-species ratios

The diurnal variation in inter-species ratios is shown in **Fig. 5**. Roadside BTEX originates from fresh vehicle emissions, so their inter-species ratios should be close to those in vehicle emissions. The observed diurnal variation in inter-species ratios was possibly related to a variation in the constituents of a traffic fleet. The *p,m*-X/E ratio was almost unchanged throughout the day, while the *o*-X/E and *p,m*-X/*o*-X ratios increased at night. A clear increase in the C_1,C_2-B/B and T/C_2-B ratios at night was due to the increased contribution of trucks in the traffic fleet as reported below. Another reason is likely an increase in the contribution of toluene released from construction painting or solvent use due to low traffic volume at night. The elevated T/C_2-B ratio from 11:00 –12:00 at site S1 was possibly due to nearby biomass burning in household cooking. According to Monod et al. (2001), T/B and T/E ratios in biomass burning environment are 0.45 and 9.41. The T/*p,m*-X and T/*o*-X ratios for biomass burning have not been reported, but they can be easy evaluated from the *m*-X/*p*-X, *m*-X/*o*-X, *m*-X/E and T/E ratios. The evaluated T/*p,m*-X and T/*o*-X ratios for biomass burning are 8.9 and 8.5, much higher than for traffic emission. Thus, a contribution from biomass burning would result in an increase in the T/C_2-B ratio.

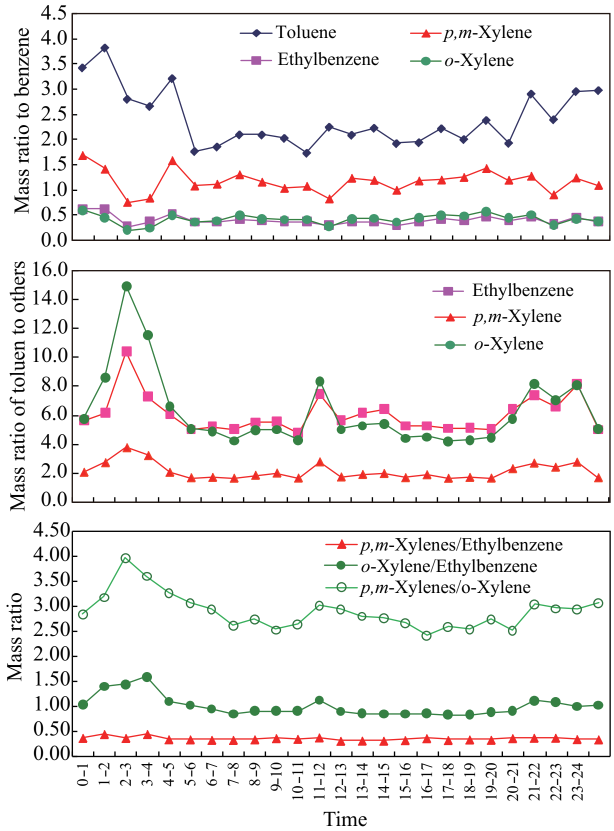

Fig. 5 Diurnal variation in the inter-species ratios.

2.4 Relationship between BTEX and traffic

Figure 6 shows the BTEX concentrations and traffic volumes at site S1. Traffic volume was extremely high at 10,500–22,300 vehicles/hr during the day, and decreased to 780–16,600 vehicles/hr at night. It was low at 0:00–5:00. Motorcycles contributed to 74%–97% of the traffic fleet, with an average of 92.5%. The constituents of the traffic fleet varied during the day. Heavy trucks were only seen at night since they are allowable in the city only from 20:00–6:00, according to local traffic regulations.

Traffic volumes at other sites were 880–22,876 vehicles/hr. Traffic jams were seen at sites S4 and S7 in

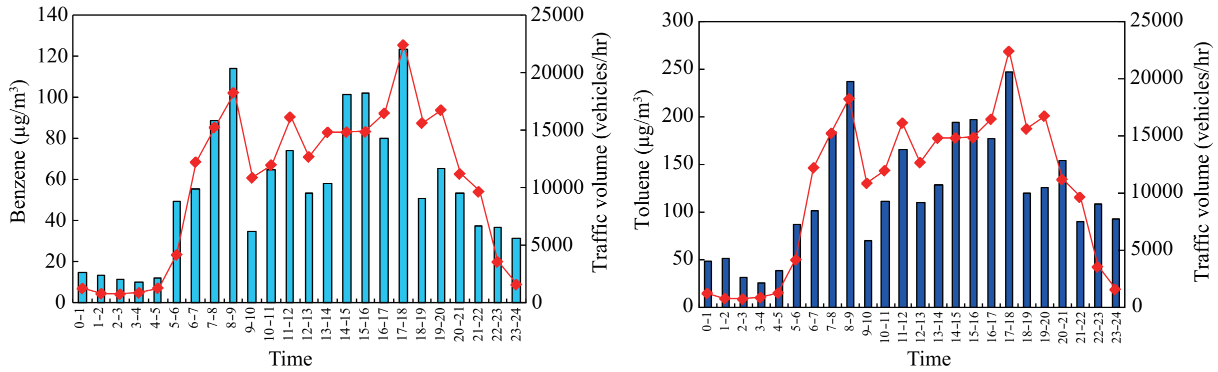

Fig. 6 BTEX concentrations and traffic volume at Site 1.

the afternoon rush hours. Motorcycles were always the predominant means of transportation. The contribution of motorcycles was the lowest (58%–86%) at site S13, located in the city centre and surrounded by luxury hotels and shopping malls. The contribution of motorcycles at other sites was 83%–98%. The average contribution of motorcycles in the traffic fleet in the whole city was 91%. This is the same as the motorcycle population within the total vehicle population in the city.

The correlation between benzene and toluene concentrations and traffic volume at site S1 is clearly seen in **Fig. 6**. The correlation coefficients (R^2) for the linear regression of BTEX concentration versus number of motorcycles in the traffic fleet at all sites were 0.595, 0.455, 0.559, 0.574 and 0.548, respectively, for benzene, toluene, ethylbenzene, *p,m*-xylenes and *o*-xylene. The impact of different means of transportation on BTEX pollution are summarised in **Table 1**. These relationships between the total concentration of BTEX species and the number of motorcycles point out that the biggest contributor to roadside BTEX in HoChiMinh is motorcycle exhaust.

2.5 Ozone formation potential of BTEX species

Ozone formation potential can be evaluated using Carter's maximum incremental reactivity (MIR). Unitless MIR is the amount of ozone formed when one gram of VOC is added to an initial VOC-NO*x* mixture under relatively high NO*x* conditions (Carter, 1990, 1994). It was impossible to evaluate the daily-average BTEX concentration for the city; therefore, ozone formation potentials were evaluated from the daily-average concentrations of benzene, toluene, ethylbenzene, *p,m*-xylenes and *o*-xylene at site S1 and are given in **Table 2**. Among the BTEX species, *p,m*-xylenes were the biggest contributors to ozone formation followed by toluene, while benzene was the lowest contributor.

2.6 Comparison of BTEX pollution in HoChiMinh and in other cities

Benzene is monitored and controlled throughout the world. **Table 3** summarises roadside benzene concentrations reported in the literature. Antwerp (Belgium), Melbourne (Australia) and Tokyo (Japan) are considered clean. Guangzhou (China), HoChiMinh, Hanoi (Vietnam), Cairo (Egypt) and Delhi (India) are badly polluted with benzene. Vehicle exhaust is the main source of BTEX in the above mentioned polluted cities, except in Guangzhou. Traffic in HoChiMinh, Hanoi, Cairo and Delhi is characterised by very high traffic volume and a high number of motorcycles in the traffic fleet. The contribution of motorcycles in the traffic fleet has been found to be 91% in HoChiMinh (this study), 94%–96% in Hanoi (Truc and Oanh, 2007) and 63% in Delhi (Hoque et al., 2008). Motorcycles are an important means of transportation in Cairo, with a motorcycle population of 300,000. Most motorcycles in Delhi and Cairo are two-stroke engines (Hoque et al., 2008; CAIP, 2000), while over 95% of motorcycles in

Table 1 Correlation coefficient (R^2) between BTEX concentrations and traffic volumes

	Motorcycles	4–9-seat cars	12–24-seat cars	25–50-seat cars	Light trucks	Heavy trucks
For site S1	0.783	0.551	0.347	0.441	0.034	0.171
For all sites	0.529	0.011	0.001	0.095	0.002	0.004

Table 2 Ozone formation potential

Hydrocarbon	Benzene	Toluene	Ethyl benzene	*m,p*-Xylenes	*o*-Xylene
Daily-average concentration at site S1 (μg/m^3)	56	121	21	64	23
MIR	0.42	2.70	2.70	8.20	6.50
O_3 formation potential[a] (μg/m^3)	24	327	57	525	150

[a]VOC (μg/m^3) × MIR.

Table 3 Benzene concentrations in some cities in the world

City	Benzene	Reference
Antwerp, Belgium	2.5	Buczynska et al., 2009
Melbourne, Australia	2.8–3.6 (0.85–1.1 ppb)	EPA Victoria, 2006
Tokyo, Japan	3–7	Laowagul et al., 2009
UK	Up to 6.3 (1.9 ppb)	UK DEFRA, 2012
Nanjing, China	6.4	Wang and Zhao, 2008
Christchurch, New Zealand	5.65 –9.10	Myles, 2005
Kathmandu, Nepan	13–20	Chiranjibi, 2004
Nanhai, China	20.0	Wang et al., 2002
Hongkong, China	26.7	Chan et al., 2002
Bangkok, Thailand	35	Leong et al., 2002
Macau, China	34.9	Wang et al., 2002
Guangzhou, China	51.5	Wang et al., 2002
HoChiMinh, Vietnam	56	This study
Hanoi, Vietnam	65	Truc and Oanh, 2007
Cairo, Egipt	67	Khoder, 2007
Dehli, India	87	Hoque et al., 2008

Hanoi and HoChiMinh are four-stroke engines. There are numerous reasons for the high BTEX levels in the above mentioned urban areas: high traffic volume, a high number of motorcycles in the traffic fleet, high benzene content in gasoline and a high emission factor for motorcycles. The current benzene content in gasoline is ca. 1.8 wt.% in Vietnam, almost double the 1% in the US, Europe and Japan. Motorcycles do not have any exhaust gas treatment system; hence, a motorcycle emits much more benzene than a gasoline-powered car equipped with a converter. Our research on 23 in-use motorcycles in HoChiMinh showed average emission factors of benzene, toluene, ethylbenzene and xylenes at 105, 195, 32 and 137 mg/km with geometric values of 61, 118, 16, 49 and 18 mg/km and median values of 54, 142, 21, 66 and 23 mg/km (Lan et al., 2010). The benzene emission factor (BEF) was much higher than the BEFs of 3.8 mg/km, 5.9–17 mg/km and 12.2 mg/km found for catalytic-converter cars in Europe, the US and Japan, respectively; almost the same BEF range of 71–96 mg/km has been found for pre-catalyst cars in the US (Heeb et al., 2000a; Dasch and Williams, 1991; Kaga et al., 2004).

Inter-species ratios of BTEX species for traffic-related sites are similar to those in vehicle exhaust due to a short spatial and temporal distance from sources. They depend mainly on fuel composition and vehicle technology. The average T/B ratio was about 2.3 in this study, 1.8–2.54 in Delhi (Hoque et al., 2008), 0.7–1.3 in Hanoi (Truc and Oanh, 2007) and 1.29–2.45 in Cairo (Khoder, 2007). The above mentioned values were lower than values commonly found in developed countries such as 3.7 in Naples (Pasquale et al., 2009), 3.8–4.4 in Antwerp (Buczynska et al., 2008), 6.4–8.5 in Tokyo (Hoshi et al., 2008) and 9.2–11.5 in Hong Kong (Ho et al., 2004). The E/B values in this study were around 0.38, in the same range of 0.23–0.43 found in Hanoi (Truc and Oanh, 2007). They were remarkably lower than the E/B values of 1.2 and 1.0 found for roadsides in Tokyo (Hoshi et al., 2008) and in Bangkok (Laowagul et al., 2008), respectively, but higher than 0.15–0.21 found in Delhi (Hoque et al., 2008). The same phenomenon was observed for the *p,m*-X/B and *o*-X/B ratios. The *p,m*-X/B and *o*-X/B values in HoChiMinh were 1.06 and 0.44, respectively; these values were much lower than 2.1 and 0.8 found in Tokyo (Hoshi et al., 2008), but higher than 0.64–0.87 and 0.31–0.47 found in Delhi (Hoque et al., 2008).

In this study, the interspecies ratios for roadside air were similar to those in gasoline. The low C_1,C_2-benzene/benzene ratios in HoChiMinh, Hanoi, Delhi and Cairo are possibly related to the composition of gasoline available locally. One possible reason that could lower the C_1,C_2-benzene/benzene ratio is evaporation from the gasoline tanks of cheaply made motorcycles with the high atmospheric temperatures in hot tropical regions. Benzene vapour pressure is higher than that of other BTEX species, so according to Raoul's law, its content in a vapour is richer than in a liquid mixture.

From the discussion above, it is clear that the inter-species ratios in both the gas phase and liquid fuel are important for the assessment of sources.

3 Conclusions

(1) Roadside BTEX levels were monitored at 17 urban sites in HoChiMinh. All the observed hour-average benzene concentrations during the day were much higher than the NAQS of 22 μg/m^3. The maximum observed benzene level was 254 μg/m^3.

(2) Traffic emission is the main source of roadside BTEX in HoChiMinh. Motorcycles are the biggest contributor to BTEX pollution in HoChiMinh.

(3) Daily concentrations of benzene, toluene, ethylbenzene, *p,m*-xylene and *o*-xylene in HoChiMinh were 56, 121, 21, 64 and 23 μg/m^3, respectively.

(4) BTEX pollution in Vietnam is characterised by a high benzene level and low C_1,C_2-benzene/benzene ratios. The inter-species ratios of BTEX species in roadside air and in gasoline are similar.

Acknowledgments

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