




Ambient BTEX levels over urban, suburban and rural areas in Malaysia

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Abstract

Benzene, toluene, ethylbenzene and xylene isomers (BTEX) are volatile organic air pollutants of concern which arise from natural and anthropogenic sources. This study aims to determine and evaluate the BTEX levels in ambient air in selected areas of Malaysia, namely: Kuala Lumpur (KL-urban), Penang (PG-urban), Bangi (BG-suburban), Langkawi (LGK-suburban) and the Danum Valley (DV-rural). Active sampling with sorbent tubes was applied in this study and samples were analysed using thermal desorption (TD) coupled with gas chromatography-mass spectrometry (GC-MS). The results show that the urban area of KL had the highest Σ BTEX ($40.36 \pm 6.99 \mu\text{g}/\text{m}^3$) followed by PG ($30.82 \pm 8.06 \mu\text{g}/\text{m}^3$). Σ BTEX concentrations in the suburban areas of LGK and BG were measured as $20.22 \pm 11.42 \mu\text{g}/\text{m}^3$ and $12.36 \pm 2.26 \mu\text{g}/\text{m}^3$, respectively. The rural area of DV had the lowest concentration of Σ BTEX ($5.55 \pm 2.54 \mu\text{g}/\text{m}^3$). The average toluene to benzene (T:B) ratio at KL, PG and LG were found to be within the range of 2.00–5.00 thus indicating vehicle emissions as the main source. The benzene level at both KL ($7.43 \mu\text{g}/\text{m}^3$) and PG ($5.12 \mu\text{g}/\text{m}^3$) were found to be slightly higher than the annual benchmark of $5 \mu\text{g}/\text{m}^3$ as suggested by the European Union (EU). The results of health risk assessments found that the cancer risk (CR) based on benzene concentrations in urban, suburban and rural areas was $> 10^{-6}$ thus indicating a link between human cancer risks and ambient benzene exposure.

Keywords VOCs · Active sampling · Gas chromatography · BTEX ratio · Cancer risk

Introduction

A class of volatile organic compounds (VOCs), namely benzene, toluene, ethylbenzene and xylene (BTEX) isomers are

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well-known toxic air pollutants. Recently, there has been increasing concern over BTEX levels, particularly in urban and industrial areas due to the adverse effects of these compounds on human health (Cerón-Bretón et al. 2014; Maré et al. 2014; Miri et al. 2016; Dehghani et al. 2018). BTEX compounds can cause adverse health effects on the respiratory and nervous system, even at low concentrations (Amini et al. 2017; Heibati et al. 2018). The World Health Organization (WHO 1996) and the International Agency for Research on Cancer (IARC 2012) have classified benzene as the most toxic of the BTEX compounds and one which is highly carcinogenic to humans. Based on the estimation by WHO (1996), lifetime exposure to benzene concentrations of $1.0 \mu\text{g}/\text{m}^3$ in urban environments causes six cases of leukaemia per 1 million inhabitants (Buczynska et al. 2009; Zhang et al. 2012). The European Union (EU) directive 2000/69/EC sets the permissible limits of benzene in ambient air at an annual average of $5 \mu\text{g}/\text{m}^3$ (European Commission 2000; Walgraeve et al. 2011).

Growing urbanisation, which leads to growth in transportation as well as industrial activities is largely responsible for the production and emissions of BTEX in ambient air (Maré et al. 2016; Jiang et al. 2017; Hajizadeh et al. 2018; Liu et al. 2018). Many researchers suggest that the dominant sources of

BTEX emissions from urban areas into the atmosphere are the combustion engine-led movement of motor vehicles, the evaporation of fuels, automobile service stations, solvent usage, industrial emissions, oil refining, and individual domestic heating systems which use low-quality coal (Okada et al. 2012; Maré et al. 2016; Masih et al. 2016; Dehghani et al. 2018). Interspecies BTEX ratios can be used for the identification of BTEX emission sources and the photochemical age of the air parcel in the area (Tiwari et al. 2010; Kumar et al. 2018). The toluene to benzene (T:B) ratio is commonly used as an indicator of vehicular emissions (Miri et al. 2016; Jiang et al. 2017). The worldwide ratio of T:B, however, differs depending on the type of gasoline formulation used by vehicles (Cerón-Bretón et al. 2014). Meanwhile, the xylene to benzene (X:B) and mp-xylene to ethylbenzene (mp-X:EB) ratios can provide information on the photochemical age of an air mass within a given area (Alghamdi et al. 2014; Maré et al. 2014).

As reviewed by Han and Naeher (2006), many urban areas in developing countries are challenged by traffic-related VOC pollutants including BTEX. There are several related studies on BTEX, many of which are focused on urban areas of South East Asian (SEA) countries. Gee and Sollars (1998), Tunsaringkarn et al. (2014) and Wong et al. (2013) found that the mean levels of benzene in Bangkok, Thailand; Manila, Philippines and Singapore were $18.2 \mu\text{g}/\text{m}^3$, $12.6 \mu\text{g}/\text{m}^3$ and $3.46 \mu\text{g}/\text{m}^3$, respectively. While Lan and Binh (2012) determined the levels of benzene at urban roadsides locations in many urban SEA areas: Ho Chi Minh City ($87 \mu\text{g}/\text{m}^3$), Hanoi ($52 \mu\text{g}/\text{m}^3$), Kuala Lumpur ($48 \mu\text{g}/\text{m}^3$) and Singapore ($6.9 \mu\text{g}/\text{m}^3$). Their study found that motor vehicle emissions seriously impacted air quality through the production of BTEX pollution in urban areas, such as Kuala Lumpur and those in Vietnam.

As there is a lack of detailed studies on BTEX in different environments in Malaysia, this study aims to determine the concentrations of BTEX at a variety of Malaysian sites. The measurements of BTEX will primarily be focused on Kuala Lumpur's urban environment with comparisons then made with other urban and suburban areas and also rural areas to assess background BTEX concentration levels. The BTEX ratio will be used to determine potential sources. This study also aims to evaluate the potential health risks of BTEX exposure based on available data collected.

Material and methods

Sampling sites

Sampling was conducted at five sites, namely: Danum Valley (DV, $n = 9$); Kuala Lumpur (KL, $n = 7$); Bangi (BG, $n = 7$); Penang (PG, $n = 7$) and Langkawi (LGK, $n = 8$) from 31 July to 5 August, 1 to 2 and 18 to 19 September, 21 to 24 September, 4 to 7 November and 3 to 6 December 2015,

respectively (Fig. 1, Supplementary 1). KL and PG represent the urban areas in the middle and northern areas of the Malaysian Peninsula. The KL sampling site is located near the Chow Kit Road area which is the most crowded and traffic-heavy area in KL (Jamhari et al. 2014). The volume of vehicles on the road in KL in the year 2015 was reported as being over 4.8 million which is the highest for all Malaysian states (www.data.gov.my). KL is also known as the most developed area within Malaysia comprising a mix of commercial areas, hotels, shopping malls and tourist attractions. The sampling site at PG was located in the George Town area which is the capital city of Penang state. This area is also heavily developed, particularly with commercial and residential areas. Georgetown is the second largest city in Malaysia, located on Penang Island and connected to the mainland via the 13.5-km-long Penang Bridge which crosses the Penang Strait. The sampling site for this location was close to a road that was especially busy during peak hours. The nearest industrial areas to this site were in Perai (~20 km) and Bayan Lepas (~25 km), both of which are popular with electrical and engineering manufacturers. The sampling site Bangi (BG) was located at Universiti Kebangsaan Malaysia (UKM), Bangi. The BG site was in a well-developed semi-urban town in Selangor state which predominantly comprises residential and industrial areas (Wahid et al. 2014). Langkawi (LGK), another sampling site, represented a suburban study area. LGK was located in Kedah state, which is in the north of the Malaysian Peninsula and is well-known in Malaysia for being a tourist island. The sampling site was located in Kuah Town, which is the largest town on Langkawi Island. This area has a high density of commercial areas, such as shopping malls, hotels and restaurants. The LGK site is congested with traffic, especially during school holidays (December). In contrast, DV represents a rural and remote site on the island of Borneo. The location is assigned as a regional Global Atmospheric Watch (GAW) station, is surrounded by lowland tropical forest and has very limited human activity (Sumari et al. 2010). The classification of urban and suburban areas was based on population, non-agricultural activities and the development of the gazetted areas (Hasan and Nair 2014). Detailed descriptions of the study areas and the meteorological conditions (temperature and humidity) for each during the sampling period are described in Supplementary 1.

Sampling for BTEX

Samples for each location were collected during the daytime between 10 and 11 a.m. and between 4 and 5 p.m. over a period of 4 to 5 days. These specific times were selected and used consistently to represent BTEX concentrations in ambient air outside peak hours. The sampling times relate to mid-range level of the solar radiation period. Solar radiation will affect the abundance of BTEX compounds through

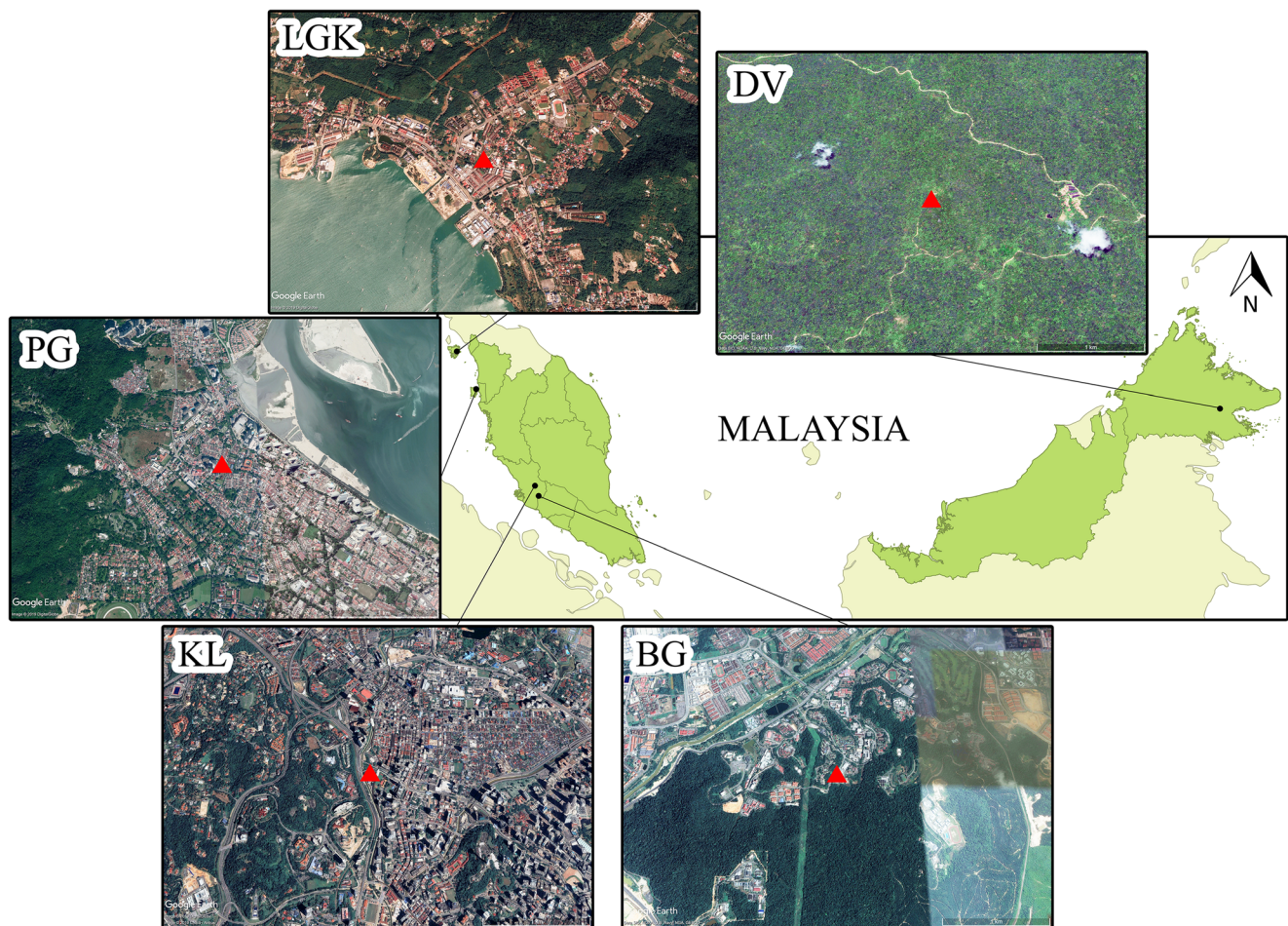


Fig. 1 Sampling sites for BTEX determination in urban, suburban and rural area in Malaysia

photochemical reactions. Therefore, consistent sampling time is important to represent the concentration of BTEX at different locations. The active sampling method was performed using a stainless steel sorbent tube (89 mm length \times 6.4 mm internal diameter) packed with 350 mg multi-sorbent Tenax® TA/Carbograph 1TD (Markes, UK) absorbent. A low flow pump (Supelco, PAS, USA) and dual flow pump (Gemini, UK) were calibrated with an air flow calibrator (Sensidyne, Go-Cal, USA) to operate at a flow rate of 50–100 mL/min for 60 min of sampling and collect approximately 3–6 L of air. All absorbent tubes were positioned at least 3 m above the ground. Prepared samples in sorbent tubes were sealed with brass screw caps combined with PTFE ferrules and then stored at $-4\text{ }^{\circ}\text{C}$ in a refrigerator. All samples were analysed within 1 week.

Analysis for BTEX

Air samples collected in the Tenax® TA/Carbograph 1TD tubes were analysed using a combination of thermal desorption (TD), gas chromatography (GC), and mass spectrometry (MS). TD was undertaken using a Unity-2 and Ultra-TD

sampler (Markes, UK). The desorbed analytes from the samples were refocused on an electrically cooled adsorbent trap (Air Toxic Analyser Trap U-T15ATA-2S, (Markes, UK) to concentrate the BTEX before further desorption was undertaken by the GC-MS systems. In the above analysis, the separation of gases was performed using GC (Agilent 6890N, USA) and detected via Mass Spectrometry (Agilent 5975C, USA). A capillary column (DB-624, J&W Scientific, USA) with a particular specification (30 m length, 0.32 mm i.d, 1.80 μm film thickness) was used for the separation process. The optimised parameters for TD and GC-MS systems were modified from the previous studies by Ramírez et al. (2010) and Ribes et al. (2007) as shown in Supplementary 2.

Quality assurance and quality control

In order to quantify the concentrations of BTEX, multiple point calibrations of a BTEX standard were prepared using the mixed 62 compounds (including BTEX) of the 100 ppb VOC gas standard (SCOTTYY, USA) in nitrogen. TD tubes were reconditioned prior to sampling to remove available BTEX. The concentrations of BTEX in the blank tube were

also determined to ensure the interference did not affect the BTEX measurement calculations. Gas standards were then transferred onto Tenax® TA/Carbograph 1TD tubes at different volumes via the calibration sampling loading rig (CSLR). The flow rate was controlled at 100 mL/min to provide only the specific volume required. The range of 30–400 ng for individual BTEX mass was selected for multiple calibration points. A good correlation, $r^2 > 0.986$, was found for all compounds as is shown in Supplementary 3. The breakthrough value (BTV) is an important parameter to evaluate the loss of analyte during sampling. Sorbent tubes were arranged in parallel series and 100 mL of 100 ppb VOC gas standard was pumped in with a 100 mL/min flow rate. Both sorbent tubes were analysed and the mass volume was calculated. The BTV value was calculated as the percentage of VOC mass found in the back tube relative to the total mass in both tubes (Ribes et al. 2007). BTV values for individual BTEX were found to be < 5%. A certified Reference Standard (Markes, UK) containing 100 ng of benzene, toluene, and xylenes in Tenax® sorbent tubes were tested to evaluate the method performance.

All sampling tubes were reconditioned for 30 min at 310 °C prior to the sampling task. A blank value for each tube was recorded for further calculations. Additionally, field blanks (not exposed to air) were transported to the site during sampling. The limit of detection (LOD) for BTEX was calculated based on seven replicates of the blank values. Quality assurances (QAs) of the sampling process were maintained by routinely checking the calibrated sampling pump flow rate and artefacts/carryover blank checks from the sampling tubes. BTEX quantitation was established by comparing the targeted integrated BTEX peak areas with the calibration curve. The mass (ng) of the compounds collected in the sorbent tubes was converted to concentrations (ppbv) using the formula:

$$\text{Concentration (ppbv)} = \frac{\text{Mass (ng)}}{\text{Molecular Weight (g/mol)}} \times \frac{\text{Molar Volume} \left(\frac{\text{mol}}{\text{L}} \right)}{\text{Volume (L)}}$$

The concentration of BTEX (ppbv) was then converted to micrograms per cubic metres.

Estimation of health risk

Human exposure to BTEX is largely through the inhalation pathway. Benzene and ethylbenzene are categorised as carcinogenic pollutants, while toluene, mp-xylene and o-xylene are classified as non-carcinogenic but still hazardous to human health (Masih et al. 2016). The health risk from BTEX exposure in ambient air due to inhalation was estimated based on cancer risk (CR) and non-cancer risk, as hazard quotient (HQ).

The chronic effects of the compounds were calculated by following the USEPA guidance (USEPA 2009) and applied in several other studies undertaken by Li et al. (2014); Bari and Kindzierski (2017). In this study, the estimation of CR and HQ are shown in the following equations:

$$\text{Cancer risk (CR)} = \text{EC} \times \text{IUR}$$

$$\text{Non-cancer risk or hazard quotient (HQ)} = \frac{\text{EC}}{\text{RfC} \times 1000}$$

where EC is the exposure concentration ($\mu\text{g}/\text{m}^3$), IUR is the inhalation unit risk ($\mu\text{g}/\text{m}^3$)⁻¹ and RfC is the reference concentration (mg/m^3).

The IUR and RfC values were derived from the Integrated Risk Information System (IRIS) database. The toxicity value (IUR) for benzene, as reported by website USEPA (2015), is $7.8 \times 10^{-6} \mu\text{g}/\text{m}^3$ as a maximum value. Meanwhile, the ethylbenzene IUR value, as reported by the Office of Environmental Health Hazard Assessment (OEHHA), California Environmental Protection Agency (CalEPA 2007), is $2.5 \times 10^{-6} \mu\text{g}/\text{m}^3$. The non-cancer risk values for RfCs as given for benzene, toluene, ethylbenzene and xylene according to USEPA (2015) were 0.03, 5, 1 and 0.1 mg/m^3 , respectively.

The EC calculation was based on chronic exposure via inhalation by adult non-workers for the residential receptor. ECs can be estimated using the following equation;

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

where

EC ($\mu\text{g}/\text{m}^3$)	the exposure concentration;
CA ($\mu\text{g}/\text{m}^3$)	the contaminant concentration in air;
ET (h/day)	the exposure time (4 h for non-workers);
EF (days/year)	the exposure frequency (350 days);
ED (years)	the exposure duration (24 years) for HQ and 70 years for CR
AT (ED in years \times 365 days/year \times 24 h/day)	average time

Results and discussion

Level of BTEXs in ambient air

The concentration levels of BTEX in the ambient air of Kuala Lumpur (KL), Penang (PG), Langkawi (LGK), Bangi (BG) and Danum Valley (DV) during the sampling period are shown in Table 1. The average of the total BTEX levels was found to be the highest in KL ($40.36 \pm 6.99 \mu\text{g}/\text{m}^3$) followed

Table 1 Daytime BTEX concentrations ($\mu\text{g}/\text{m}^3$) at urban, suburban and rural sampling areas

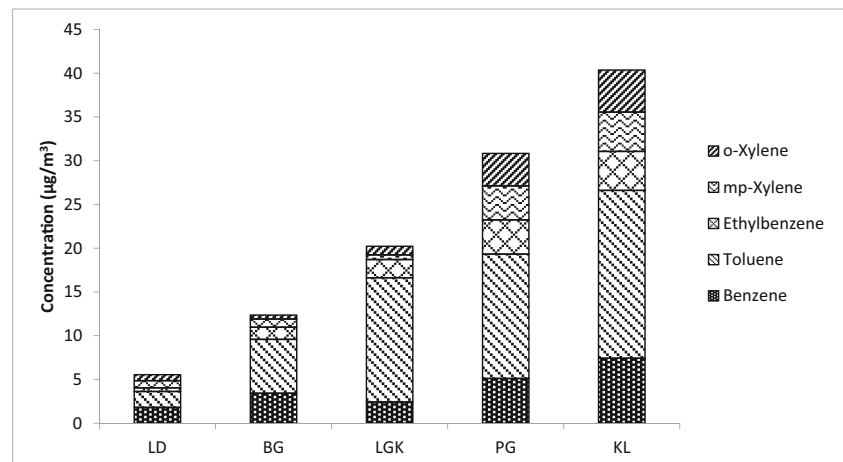
Location	DV - (rural)		KL - (urban)		BG - (suburban)		PG - (urban)		LGK - (suburban)	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
VOCs concentration ($\mu\text{g}/\text{m}^3$)										
Benzene	1.81	0.84–2.68	7.43	5.93–10.73	3.43	2.78–4.43	5.12	3.68–8.64	2.43	1.47–3.34
Toluene	1.82	1.13–2.84	19.18	14.09–26.46	6.15	5.10–8.44	14.20	5.82–20.24	14.20	7.06–23.33
Ethylbenzene	0.41	0.33–0.49	4.45	3.63–5.43	1.39	1.02–1.88	3.92	2.45–4.61	2.08	1.88–2.37
mp-Xylene	0.82	0.29–2.28	4.49	2.45–5.79	0.94	0.73–1.31	3.88	1.75–6.12	0.53	0.20–0.86
o-Xylene	0.69	0.41–0.82	4.81	2.49–6.04	0.45	0.29–0.73	3.71	1.55–5.95	0.98	0.65–1.43
Total BTEXs	5.55		40.36		12.36		30.82		20.22	

by PG ($30.82 \pm 8.06 \mu\text{g}/\text{m}^3$), LGK ($20.22 \pm 11.42 \mu\text{g}/\text{m}^3$), BG ($12.36 \pm 2.26 \mu\text{g}/\text{m}^3$) and DV ($5.55 \pm 2.54 \mu\text{g}/\text{m}^3$). Comparison analyses between sites using analyses of variance (one-way ANOVA) showed that all BTEX compositions were significantly different ($p < 0.05$). Whilst all compounds were dominant in urban areas, KL experienced the highest individual compound with benzene ($10.73 \mu\text{g}/\text{m}^3$), toluene ($26.46 \mu\text{g}/\text{m}^3$), ethylbenzene ($5.43 \mu\text{g}/\text{m}^3$) and o-xylene ($6.04 \mu\text{g}/\text{m}^3$). Meanwhile, the highest mp-xylene level was detected at PG at $6.12 \mu\text{g}/\text{m}^3$, demonstrating that the PG area was also exposed to air pollution in the form of industrial emissions, particularly from the metal, electronic and chemical industries (Ismail et al. 2017). The lowest BTEX was observed at rural DV with benzene ($0.84 \mu\text{g}/\text{m}^3$), toluene ($1.13 \mu\text{g}/\text{m}^3$), ethylbenzene ($0.33 \mu\text{g}/\text{m}^3$) and o-xylene ($0.41 \mu\text{g}/\text{m}^3$). The lowest mp-xylene detected was at LGK at $0.20 \mu\text{g}/\text{m}^3$. Figure 2 clearly shows that the BTEX level was higher in ambient air in urban (KL, PG) > suburban (LGK, BG) > rural area (DV).

The average benzene concentration in ambient air at KL ($7.43 \pm 1.90 \mu\text{g}/\text{m}^3$) and PG ($5.12 \pm 0.52 \mu\text{g}/\text{m}^3$) exceeded the yearly average permissible limit of $5 \mu\text{g}/\text{m}^3$ as set by the EU directive 2000/69/EC for 2010 (European Commission 2000). Furthermore, Norbäck et al. (2017) observed that the level of benzene in outdoor urban areas in Johor Baharu, Malaysia

was $5.1 \mu\text{g}/\text{m}^3$. However, the benzene level at KL in this study was found to be lower than the roadside value ($48 \mu\text{g}/\text{m}^3$) reported by Lan and Binh (2012). These differences present clear evidence that heavy traffic is a prominent source of benzene. According to Lan and Binh (2012), the high level of benzene in Malaysia was due to the considerable number of motorcycles on the roads and their high emissions. Another reason for such significant benzene levels in urban locations such as Vietnam and Kuala Lumpur is due to the high benzene content in the fuel used (Lan and Binh 2012). Conversely, the average benzene level was lower at BG ($3.43 \pm 0.56 \mu\text{g}/\text{m}^3$), LGK ($2.43 \pm 0.84 \mu\text{g}/\text{m}^3$) and DV ($1.84 \pm 0.19 \mu\text{g}/\text{m}^3$). In these locations, it was below the tolerance limit.

The relative abundance for BTEX in urban areas was in the following order: toluene > benzene > mp-xylene/ethylbenzene/o-xylene. While in the suburban areas of BG and LGK, abundance followed the order: toluene > benzene > ethylbenzene > mp-xylene/o-xylene. Similarly, urban and suburban areas all over Asia were reported by Ho et al. (2004); Laowagul et al. (2008) and Niu et al. (2012) as showing a dominance of toluene in relation to the other compounds. DV demonstrated the lowest BTEX, with the following composition: benzene/toluene > mp-xylene > o-xylene > ethylbenzene. Interestingly, benzene and also toluene were found at higher levels than the other compounds at DV. This pattern

Fig. 2 Distribution and level of BTEX in different study areas

resembled one from studies undertaken by Villanueva et al. (2014) in a rural national park. It is quite possible that this area was affected by air from a nearby urban site which had undergone the chemical oxidation process and degraded the alkylated benzene to benzene (Atkinson 1990). The highest average concentrations at KL and PG revealed that these cities were heavily polluted by BTEX. The main source of which was vehicular emissions. BG ambient air was also polluted by BTEX, which was likely due to an increasing volume of automobiles used in BG (Wahid et al. 2014) as well as the industrial sources located near Bangi and Nilai (Fujii et al. 2016). DV, as a rural area sampling site, was exposed with to the lowest average of BTEX, indicating trans-boundary distribution to an otherwise pristine region. MacKenzie et al. (2011) also reported a lower BTEX level (< 0.5 ppbv) at DV using photon transfer reaction–mass spectrometry (PTR-MS). However, the BTEX levels in the forested DV area was found to be higher when compared to BTEX levels in the Amazon rainforest, as reported by Paralovo et al. (2016). However, it should be noted that small amounts of BTEX may arise from biogenic emissions. As such, since DV is situated in a forest area, some BTEX such as toluene, could well be emitted by plants (Yu et al. 2008).

The BTEX levels in major cities of South East Asia (SEA) are tabulated in Table 2. The occurrence of the BTEX compounds in all major cities indicates that the ambient air was polluted by anthropogenic sources. The average BTEX results for KL and PG in this study were found to be within the range detected in other SEA cities. The highest Σ BTEX results were reported in Manila, followed by Hanoi, Bangkok, Ho Chi Minh City, Kuala Lumpur, Penang and Singapore. Compared with other SEA cities, the average levels of BTEX at KL and PG were lower than all the other locations studied apart from Singapore. Lan and Binh (2012) also found that roadside benzene levels in Singapore were lower when compared to other SEA cities. The levels of BTEX at KL and PG were identical to those in Hanoi, as reported by Phuc and Kim Oanh (2018). Since SEA cities generally have similar climatic and meteorological conditions, the variations in BTEX distribution could be the result of traffic density, industrial activities, fuel composition and combustion, solvent usage in industrial processes, the intensity of human activities, land use patterns and the chemical removal of BTEX from the atmosphere (Alghamdi et al. 2014). Besides, several local conditions including wind speed and direction, pressure, temperature, topography, seasonality, location and magnitude of emission sources have also been shown to influence BTEX in ambient air (Słomińska et al. 2014).

BTEX correlation and ratio analysis

Table 3 shows the Pearson correlations between BTEX compounds in the study areas. BTEX correlations were evaluated for each site. The overall data is in normal distribution based

Table 2 BTEX comparison in ambient air ($\mu\text{g}/\text{m}^3$) with other urban cities in South East Asia (SEA)

City	Sampling period	Sampling type	Detection	Benzene	Toluene	Ethyl benzene	mp-Xylene	o-Xylene	Reference
Kuala Lumpur, Malaysia	September 2015	Tenax TA/Carbograph 1TD	TD-GCMS	7.43	19.18	4.45	4.49	4.81	This study
Penang, Malaysia	November 2015	Tenax TA/Carbograph 1TD	TD-GCMS	5.12	14.20	3.92	3.88	3.71	This study
Bangkok, Thailand	February–June 2013	Charcoal tube	GC-FID	18.59	77.41	4.33	4.13	3.16	Tunsaringkam et al. (2014)
Singapore	February 2012	Tenax/CarbopackX	TD-GCMS	3.46	20.18	4.36	3.33	2.36	Wong et al. (2013)
Hanoi, Vietnam	January–February 2015 October–November 2015	SKC Charcoal tube	GC-FID	6–9	7–12	2–4	5–6	3–5	Hong Phuc and Kim Oanh (2018)
Ho Chi Minh City, Vietnam	December 2017–January 2018	SKC Charcoal tube	GC-FID	15.4	54.2	6.6	16.1	5.9	Huong Giang and Kim Oanh (2014)
Manila, Philippines	1995–1996	Carbopack/Carbosieve	TD-GC-FID	12.6	168	21.9	55.8	16.8	Gee and Sollars (1998)

Table 3 BTEX Pearson correlation matrix (*r*) in different study areas

	B	T	EB	mp-X	o-X
KL (<i>n</i> = 7)					
B	1.00				
T	0.66	1.00			
EB	0.76*	0.94***	1.00		
mp-X	0.51	0.90**	0.72*	1.00	
o-X	0.82*	0.90**	0.89*	0.72*	1.00
PG (<i>n</i> = 7)					
B	1.00				
T	0.15	1.00			
EB	0.86**	0.53	1.00		
mp-X	0.96***	0.35	0.94***	1.00	
o-X	0.96***	0.34	0.95***	0.99***	1.00
BG (<i>n</i> = 7)					
B	1.00				
T	0.70	1.00			
EB	0.86*	0.89**	1.00		
mp-X	0.71	0.98***	0.94**	1.00	
o-X	−0.08	−0.64	−0.57	−0.72	1.00
LGK (<i>n</i> = 8)					
B	1.00				
T	−0.13	1.00			
EB	−0.28	0.96***	1.00		
mp-X	−0.14	0.99***	0.96***	1.00	
o-X	−0.10	0.94***	0.98***	0.95***	1.00
DV (<i>n</i> = 9)					
B	1.00				
T	0.73*	1.00			
EB	0.76**	0.24	1.00		
mp-X	0.75**	0.94***	0.33	1.00	
o-X	0.70*	0.73*	0.63*	0.85**	1.00

Significant at *** $P < 0.01$; ** $P < 0.05$; * $P < 0.10$

on probability plots (P-P and Q-Q plots). A significant correlation was found at KL for toluene to other compounds ($p < 0.05$). As shown in Table 3, correlations between benzene, ethyl benzene and xylenes were good ($r > 0.86$). At the same time, these three molecules did not have good correlations with toluene. Correlations (*r* values) of benzene, ethylbenzene and xylenes with toluene are only 0.15, 0.53 and 0.34, respectively. The low correlation of toluene with other molecules was possibly due to multiple sources of this compound within the PG areas especially, from point sources such as industry activities. PG is also known as industrial area, and besides the emission from vehicles, other sources might be nearby industries and nearby fuel station. Solvents used in industries may possibly increase the level of toluene at PG. A high correlation of BTEX species at KL and PG clearly showed that these pollutants were likely to have derived from

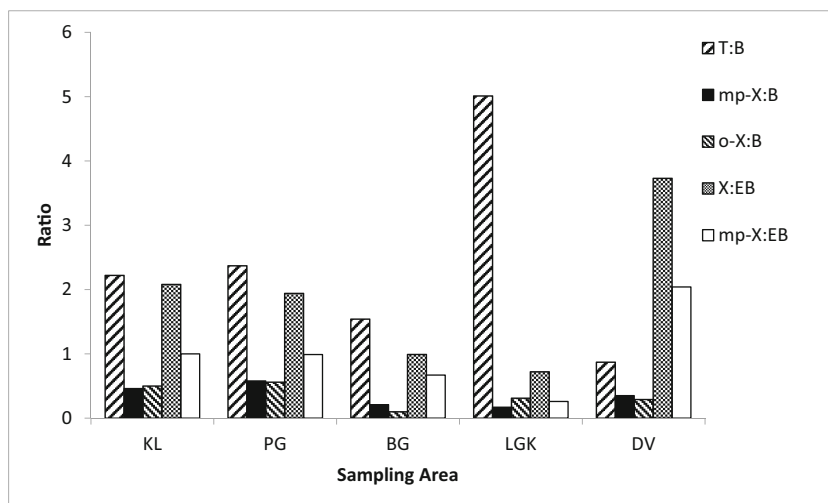
the same source. Meanwhile, the correlation of BTEX at BG showed a negative correlation for o-xylene with other compounds. As xylene essentially originates mainly from industrial and motor vehicle sources (ATSDR 2007) this indicates that the area is unlikely to be influenced by o-xylene. A negative correlation for benzene with other compounds at LGK may be due to a mix of potential sources of BTEX at this site, which is in an area where diesel-engine vessels are frequently used for tourism. Benzene at DV also showed a significant correlation ($p < 0.05$) with the other compounds even though BTEX predominantly originated from anthropogenic sources. A lower concentration of BTEX might have been emitted from the vehicles used by the researchers at the nearby field centre. The presence of BTEX in the rural DV area could also be the result of trans-boundary anthropogenic pollutants as explained by Ashfold et al. (2015).

The average observed BTEX ratios are shown in Fig. 3. The toluene to benzene (T:B) ratios for LGK, PG and KL were 5.01, 2.37 and 2.22, respectively, thus indicating that the study areas were polluted by vehicular emissions. Results of the T:B ratios for KL and PG also clearly revealed that vehicular emissions were the main source. Al Madhoun et al. (2010) reported that in Penang, the levels of benzene were relative to traffic volume. As expected, even though LGK is only a suburban island, tourist activities leading to a high usage of motor vehicles have led to the area being affected by BTEX.

The T:B ratio for KL (2.22) was similarly reported by Lan and Binh (2012). According to Niu et al. (2012), The T:B ratio of 2 indicates vehicular emissions as the main source and a higher ratio of T:B suggests additional sources. Nevertheless, many South East Asian (SEA) cities reported a T:B ratio greater than 2, for example, Bangkok (8.7) and Manila (13.4), as noted by (Gee and Sollars 1998). The differences in T:B ratios within urban areas were mainly due to variations in vehicular fleets, urban infrastructure, fuel type and climatic influences (Alghamdi et al. 2014).

The ratios for mp-xylene to benzene (mp-X:B) and o-xylene to benzene (o-X:B) for all sites were relatively low (< 1) thus indicating that xylene was actively involved in photochemical reactions with OH radicals to produce ozone at those locations (Bauri et al. 2016). It also indicates the age of the air mass (Tiwari et al. 2010). Interestingly, both xylene to ethylbenzene (X:EB) and mp-xylene to ethylbenzene (mp-X:EB) ratios at DV recorded higher values of 3.73 and 2.04, respectively. The ratios were greater than those in the observed urban sites of PG (1.94 and 0.99) and KL (2.08 and 1.00). It might be due to a lower level of ethylbenzene at DV since the ethylbenzene sources were mainly derived from vehicular emissions. Miri et al. (2016) also suggested that a ratio of 1.5–4.3 indicated vehicular emissions as a source. According to the data from other literature, higher values of the mp-X:EB concentration ratio (> 3.28) imply a fresh air mass and a fresh emission source (Miller et al. 2011; Marć et al. 2014).

Fig. 3 BTEX interspecies ratio in sampling areas



Health risk assessment

Cancer risk (CR) and non-cancer risk (HQ) were evaluated for BTEX in ambient air based on an average of 1 h daytime sampling within the studied areas. $CR > 1 \times 10^{-6}$ is considered

to be an unacceptable limit and can pose a cancer risk to humans (USEPA 2009). Meanwhile, the $HQ > 1$ is considered to be an unacceptable limit for non-cancer risk. The results for the health risk estimations are presented in Table 4. The average benzene level in urban, suburban and rural areas clearly

Table 4 BTEX health risk estimation for cancer risk (CR) and hazard quotient (HQ) at studied areas

Compound	Sites	Average concentration ($\mu\text{g}/\text{m}^3$)	Exposure concentration (EC)	Cancer risk (CR)	Hazard quotient (HQ)
Benzene	KL	7.43	1.19	9.26E-06	3.96E-02
	PG	5.12	0.82	6.38E-06	2.73E-02
	BG	3.43	0.55	4.28E-06	1.83E-02
	LGK	2.43	0.39	3.03E-06	1.30E-02
	DV	1.81	0.29	2.26E-06	9.64E-03
Toluene	KL	19.18	3.07		6.13E-04
	PG	14.20	2.27		4.54E-04
	BG	6.15	0.98		1.97E-04
	LGK	14.20	2.27		4.54E-04
	DV	1.82	0.29		5.82E-05
Ethylbenzene	KL	4.45	0.71	1.78E-06	7.11E-04
	PG	3.92	0.63	1.56E-06	6.26E-04
	BG	1.39	0.22	5.54E-07	2.22E-04
	LGK	2.08	0.33	8.31E-07	3.33E-04
	DV	0.41	0.07	1.63E-07	6.52E-05
mp-Xylene	KL	4.49	0.72		7.17E-03
	PG	3.88	0.62		6.19E-03
	BG	0.94	0.15		1.50E-03
	LGK	0.53	0.08		8.48E-04
	DV	0.82	0.13		1.30E-03
o-Xylene	KL	4.81	0.77		7.69E-03
	PG	3.71	0.59		5.93E-03
	BG	0.45	0.07		7.17E-04
	LGK	0.98	0.16		1.56E-03
	DV	0.69	0.11		1.11E-03

indicated a potential cancer risk as the CR value for benzene $> 10^{-6}$. The highest benzene CR observed was at the urban KL site (9.26×10^{-6}) and the lowest at DV (2.26×10^{-6}). In a previous study, Hamid et al. (2017) estimated the CR from benzene in outdoor suburban areas of Malaysia at 4.35×10^{-6} . Additionally, the CR for ethylbenzene at urban KL and PG was calculated as 1.78×10^{-6} and 1.56×10^{-6} , respectively. Meanwhile, the CR for ethylbenzene in suburban and rural areas was calculated $< 10^{-6}$. Clearly, the cancer risk in urban areas due to benzene and ethylbenzene was significantly higher and exceeded the recommended safe limit. The results suggested that a cancer risk could potentially occur in any of the study areas, but more so in the urban ones.

In addition, the HQ for other compounds was found to be < 1 , which indicated fewer non-cancer risks within the studied areas. The HQ values for benzene and xylene for all sampling locations was found to be higher compared to other compounds, specifically in urban areas, indicating that a non-cancer risk from benzene and xylene is possible. Based on the results of the BTEX health risk estimation, both the cancer and non-cancer risks observed posed a greater and more hazardous impact on humans in urban $>$ suburban $>$ rural areas.

Conclusion

BTEX compounds were successfully detected at all sites using a sorbent tube with active sampling, followed by TD-GCMS analysis. The total observed average BTEX was in the following order for urban KL ($40.36 \pm 6.99 \mu\text{g}/\text{m}^3$) and urban PG ($30.82 \pm 8.06 \mu\text{g}/\text{m}^3$), followed by suburban LGK ($20.22 \pm 11.42 \mu\text{g}/\text{m}^3$) and BG ($12.36 \pm 2.26 \mu\text{g}/\text{m}^3$) and lastly, the local site, DV ($5.55 \pm 2.54 \mu\text{g}/\text{m}^3$). The results clearly showed BTEX due to anthropogenic sources following the order urban $>$ suburban $>$ rural areas. Even though the level of BTEX in the rural and forest areas of DV was the lowest, a chronic cancer risk was still viable. Estimation of the health risks established that urban, suburban and rural areas all had a benzene CR $> 10^{-6}$ which means there is a potential for benzene to be harmful to humans. The ethylbenzene CR for urban sites KL and PG was found to be $> 10^{-6}$ suggesting a high health risk due to this compound. The toluene to benzene ratio (T:B) indicated that the KL, PG and LGK areas were all affected by vehicular or traffic emissions. Since BTEX levels in urban and suburban areas were found to be relatively high and the number of vehicles on the road is ever increasing, there is a real need for the relevant authorities to implement a control strategy in order to reduce traffic, particularly in urban areas. Continuous monitoring of BTEX and other hazardous air pollutants in these urban areas is also essential if a better understanding of the pollutants and their potential public health risks are to be discerned. In future work, BTEX profiles with continuous sampling are suggested during the daytime and

night time for more accurate investigation. The BTEX content of local gasoline and diesel fuel also needs to be study thoroughly for source identification. Detailed studies on the effects of these parameters need to be conducted on a regular basis, particularly in locations which are close to roadsides and/or industrial areas.

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