STUDY ON FORMATION OF STANDARD CONCENTRATION BTEX TOXIC POLLUTANT IN AIR

FINAL REPORT

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ABBREVIATIONS

AT	averaging time for non-carcinogenic risk calculation
BTEX	Benzene, Toluene, Ethylbenzene and Xylene
BW	Body weight
CA	contaminant concentration in air
CDI	chronic daily intake
CF	conversion factor
E	Daily exposure
EC	Exposure Concentration
ED	exposure duration
EF	exposure frequency
ET	exposure time
GC	Gas Chromatography
HQ	Health Quotient
IARC	International Agency for Research on Cancer
IR	inhalation rate for an adult
KIPC	Kerteh Integrated Petrochemical Complex
LOD	Limit of detection
LTCR	Life-time Cancer Risk
MS	Mass Spectrometer
NEPC	National Environment Council (Australia)
NEPM	National Environment Protection (Air Toxics) Measure (Australia)
PPBF	Pusat Penyelidikan Bukit Fraser
QC	Quality Control
RfC	chronic inhalation reference concentration
RfD	chronic inhalation reference concentration x 20
TD	Thermal Desorption
UKMKL	Universiti Kebangsaan Malaysia Kuala Lumpur
UMS	Universiti Malaysia Sabah
UMT	Universiti Malaysia Terengganu
US EPA	United States Environmental Protection Agency
US NIOSH	United States National Institute for Occupational Safety and Health
USA	United Stated of America
VOCs	Volatile organic compounds
WHO	World Health Organization

1 Project background

BTEX is the acronym for a group of volatile organic compounds (VOCs) i.e. benzene, toluene, ethylbenzene and xylene. In urban areas, BTEX accounts for about 60% of total volatile organic carbons (VOCs) [1]. It has been long known that VOCs including BTEX are harmful to human health, as long term exposure may lead to respiratory and cardiovascular illnesses [2, 3]. Human exposure to BTEX is of particular concern due to its high toxicity [4]. All of the BTEX compounds can cause neurological impairment while exposure to benzene can additionally cause haematological effects including aplastic anaemia and acute myelogenous leukaemia [5].

Anthropogenic emissions of BTEX in the environment are from automobile emission, petrochemical industry, use of solvents and paints as well as petrochemical fuels derivatives such as petrol or gasoline [6, 7]. Due to its volatile nature (these organic compounds can vaporise at temperatures above 20°C), fugitive emission of BTEX can occur even during storage and transportation of petroleum based fuels. Given their widespread use, BTEX may also be released into the ambient environment via evaporation during fuel tank filling, oil and gas operations, solvent usage and spills from accidents [8].

This research attempts to ascertain the current ambient concentration of the toxic pollutant BTEX in different areas (urban, industrial, sub-urban and background) in Malaysia and the potential sources of these BTEX emissions. Based on the results, health risk assessment of BTEX exposure was conducted. Lastly, suggestions for ambient standards and plan for BTEX management were suggested.

1.1 Project description

1.1.1 Objectives

The objectives of this research are as follows:

a) identify the sources of the toxic pollutant BTEX and its precursors in order to suggest control/mitigation measures

b) determine suitable ambient standard and criteria for maximum allowable exposure to the toxic pollutant BTEX that is safe based on local conditions with reference to specification by World Health Organization (WHO) and subsequently outline the guideline for ambient BTEX standard.

c) design an action plan for prevention and reduction of the toxic pollutant BTEX in order to assist policy makers in drafting policy and mitigation strategy for overcoming air pollution; and

d) assess health impact to the public due to the toxic pollutant BTEX and suggest control measures to reduce the impact.

1.1.2 Research scope

The scopes of this research are to:

a) determine methodology that will be used in designing standard for the toxic pollutant BTEX.

b) carry out sampling for the toxic pollutant BTEX at selected locations (based on the following categories: industrial, urban, sub-urban and background) to obtain the concentration for the toxic pollutant BTEX.

c) analyse collected sample data for the pollutant BTEX and determine pollution concentration pattern and possible relationship with meteorological parameters.

d) ascertain mitigation measures and monitoring of activities that contribute to the formation of the toxic pollutant BTEX, and

e) develop an action plan for the reduction of the toxic pollutant BTEX

1.1.3 Conceptual framework

The conceptual framework for project implementation is shown in Figure 1. Components essential to achieve the goals of the project include a comprehensive literature review and two BTEX sampling methodologies. The sampling results are then used for determining BTEX distribution, potential sources and relationship with meteorological parameters. All the gathered information will then be used to identify mitigation measures for reducing ambient BTEX levels.

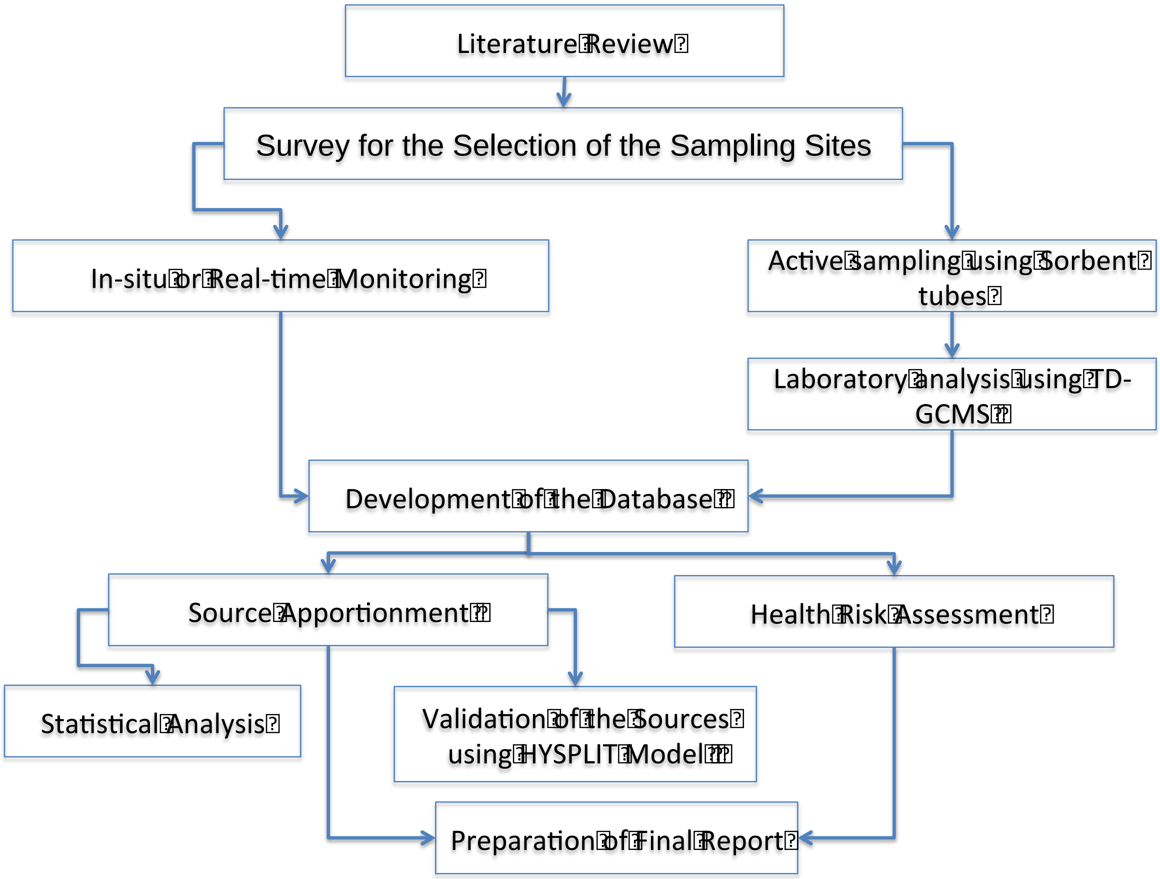


Figure 1 : Research framework for BTEX measurement

1.1.4 Research output

The expected outputs of this research are as follows:

a) pollutant concentration pattern of the toxic pollutant BTEX and its possible relationship with meteorological parameters.

b) air pollution status due to the toxic pollutant BTEX and information on main sources that contribute to the formation of the toxic pollutant BTEX around sampling location.

c) suggestion for ambient standard for the toxic pollutant BTEX.

d) suggestion for maximum allowable exposure to the toxic pollutant BTEX that is safe based on local suitability and with reference to specification by WHO (World Health Organization)

e) action plan for reducing the toxic pollutant BTEX and measures to implement improvement to air quality particularly at problem areas; and

f) documented guideline regarding information on the toxic pollutant BTEX and the ambient air quality standard for the toxic pollutant BTEX.

2 Literature review

2.1 BTEX composition, sources and health impact

Volatile organic compound (VOC) is a term that is used to encompass reactive compounds that contain carbon in its chemical structure. Benzene, toluene, ethylene and xylene (BTEX) are part of VOCs. Figure 2 shows the chemical structure of BTEX including the three structural isomers of xylene (distinguished by the designations *ortho*–, *meta*–, and *para*–). The common characteristic of each compound is the presence of the aromatic hydrocarbon ring. BTEX are colourless and due to their organic content, are not very water soluble. However, they evaporate relatively easily [9].

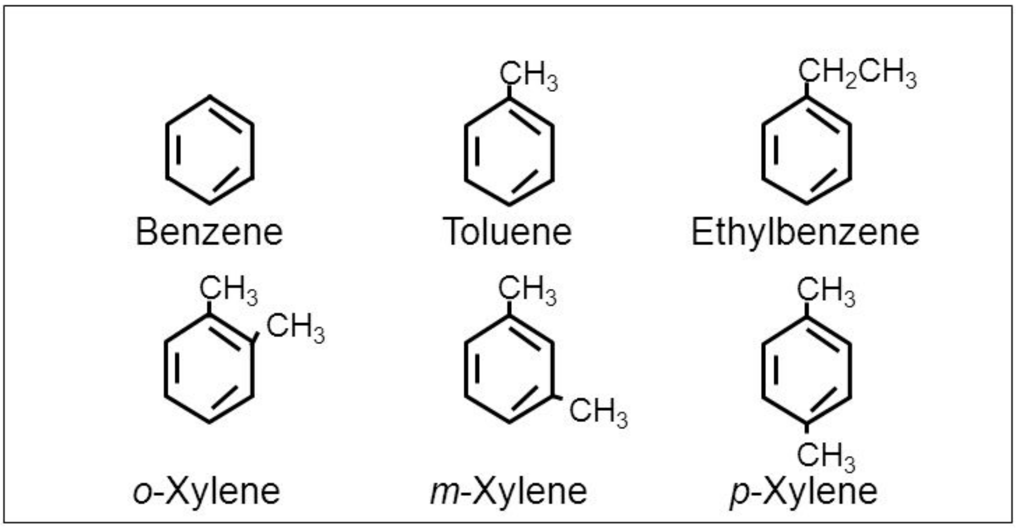


Figure 2 : Chemical structure of BTEX compounds [10]

Physico-chemical properties of BTEX are shown in Table 1. Overall, the molecular weight of BTEX range between 78 to 106 g mole^{-1}. They have log K_{ow} values in the range of 2 – 3.20 and their water-solubility is in the medium range. BTEX are also relatively volatile having Henry´s law constants in the range of 0.50 – 0.80 kPa·m^3/mole. Among BTEX molecules, benzene is expected to be the most easily evaporated due to its low molecular weight, low boiling point and high vapour pressure. Benzene is also more easily soluble in water [11].

Table 1: Physio-chemical properties of BTEX

Compound	Molecular weight (g mole^{-1})	Density (g ml^{-1})	Boiling point (°C)	Water solubility (mg L^{-1})	Vapor pressure (mmHg)	Log K_{ow}
Benzene	78	0.88	80.1	1780	76	2.13
Toluene	92	0.87	110.8	535	22	2.69
o-Xylene	106	0.88	144.4	175	5	2.77
m-Xylene	106	0.86	139	135	6	3.20
p-Xylene	106	0.86	138.4	198	6.5	3.15
Ethylbenzene	106	0.87	136.2	152	7	3.15

Source: EUGRIS 2017 [11]

BTEX are naturally present in crude oil. Hence it can be found in sea water close to petroleum deposits. Other natural sources for BTEX include gaseous emissions from volcanoes. Anthropogenic sources of BTEX are primarily from the combustion and fugitive emission of fossil fuels. Around 18% of gasoline is BTEX and the largest fraction of BTEX present in gasoline is xylene (Figure 3). Figure 4 shows the fraction of each component in diesel fuel. Benzene fraction is not included in the diesel composition as its concentration was below the detection limit of 1.58 µg/kg in the study conducted by Mariano [12].

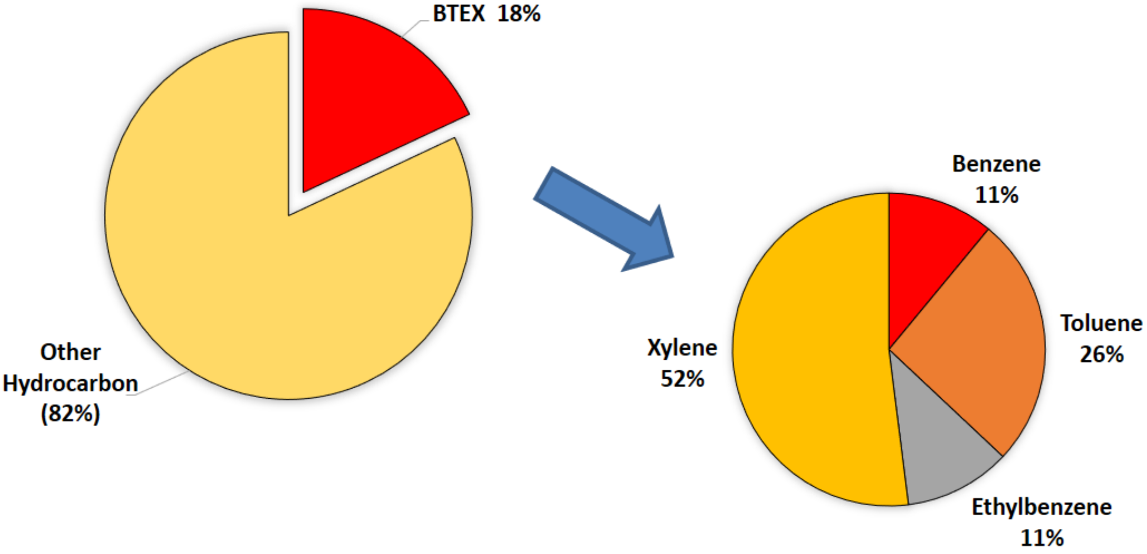


Figure 3: BTEX components in gasoline (petrol)

Source: Adapted from Ohio Department of Health (2014) [13]

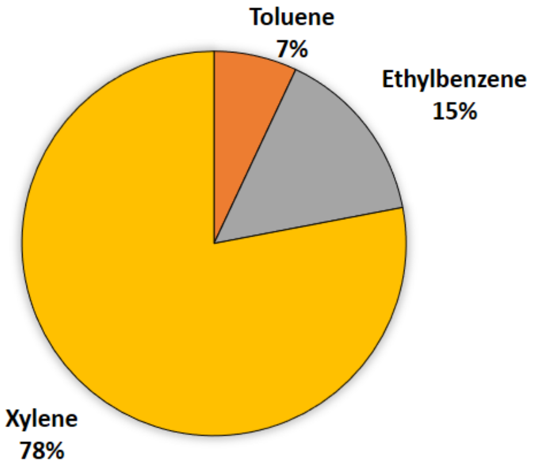


Figure 4: BTEX components in diesel

Source: Adapted from Mariano et. al. 2008 [12]

Exposure to BTEX particularly in the long term has been associated with damage to the central nervous system, liver and kidney damage as well as increased cancer risk. Studies have also shown that even at concentrations much lower than safe daily exposure level (United States Environmental Protection Agency (US EPA)), health effects have been observed. Studies on exposure to benzene indicate adverse effects on foetal development, male reproductive function, respiratory system, metabolic function and immune system. Toluene, ethylbenzene and xylene also adversely affects development, immune system, and causes respiratory symptoms. In addition, all three have been shown to increase risk for cardiovascular disease. However, how benzene, toluene, ethylbenzene and xylene actually affects human health is still not well understood. Studies specifically focusing on combined exposure to BTEX have indicated developmental effects in foetal brain growth and birth size and increased cases of physician diagnosed asthma in adults. The results of the study were presented in a review paper and are summarised in Table 2 [14].

Table 2: Health effects of ambient BTEX combined exposure

Health outcome		Exposure concentration
Development	Biparietal diameter	2.27−30.31 µg/m^3
	Birth weight	14.49 µg/m^3
Respiratory function	Asthma	B, 1.21 µg/m^3; T, 14.33 µg/m^3; E, 2.55 µg/m^3; o-X, 2.16 µg/ m^3; m-+p-X,5.97 µg/m^3
	Wheezing attacks	B, 1.21 µg/ m^3; T, 14.33 µg/ m^3; E, 2.55 µg/ m^3; o-X, 2.16 µg/m^3; m-+p-X, 5.97 µg/ m^3

B, benzene; T, toluene; E, ethylbenzene; m-+p-X,m-+p- xylene; o-X,o-xylene [14]

There are many studies trying to correlate the level of BTEX to noncarcinogenic and carcinogenic health risk. The estimation of noncarcinogenic and carcinogenic health risk in these studies were determined based on hazard quotient (HQ) and inhalation lifetime cancer risk (LTCR) values. A study in Iran found the concentration of benzene, m,p-xylene, o-xylene, toluene, and ethylbenzene to be 1.056, 2.929, 1.044, 4.291, and 0.837 ppb (or 3.444, 12.734, 4.53, 16.25 and 3.63 µg/m^3) respectively [15]. Further investigation found that the HQ for BTEX was within the acceptable value for HQ (<1) and the LTCR for benzene was below 1 x 10^{-6} (i.e. 1 person per million). In northern India however, the integrated LTCR was found to be higher than acceptable value (1 x 10^{-6}). The cancer risk for benzene (at all study sites) and ethylbenzene (at roadsides) ranged between 1.0 x 10^{-5} and 7.6 x 10 $^{-6}$ [16]. The mean concentration of benzene and ethylbenzene recorded were 15.9 and 3.9 µg/m^3 respectively. A study in Brazil that focused on ambient health risk at gas stations found that ambient benzene concentrations were higher than 3.2 µg/m^3 limit set by US NIOSH [17]. The cancer risk was also very high for workers (exposed to benzene for 30 years) as it was within the range of 4.06× 10^{-5} - 3.78× 10^{-4} (mean of 1.82× 10^{-4}) at 10 selected gas stations. The noncarcinogenic risk was also high for benzene 1.84 (acceptable limit <1) showing that among the BTEX, benzene exposure posed the highest health risk to workers at gas stations [17].

2.2 BTEX concentrations worldwide

Concentrations of benzene in ambient air in a rural environment has been observed within the range of 0.2 – 16 µg/m^3. The concentration range for toluene and ethylbenzene in rural environment has been reported within the range of 0.5 – 260 µg/m^3 and 0.2 – 1.6 µg/m^3 respectively. Xylene typically records the lowest concentration at < 0.1 – 3 µg/m^3. In industrial areas with high traffic density however, benzene, toluene, ethylbenzene and xylene concentrations can reach up to 349 µg/m^3, 1310 µg/m^3, 360 µg/m^3 and 775 µg/m^3 respectively [9]. In Europe, mean benzene concentration in ambient air are about 1 µg/m^3 and 5–20 µg/m^3 in rural and urban areas respectively [18]. The average daily weekday concentration of BTEX at a suburb in Caserta, Italy is 8.6 µg/m^3 benzene, 25.2 µg/m^3 toluene, 6.3 µg/m^3 ethylbenzene, 14.0 µg/m^3 m,p-xylene, and 11.7 µg/m^3 o-xylene. The weekend concentrations were lower by about 1.6 times [19]. In Iran, the annual median for benzene was found to be higher than the European Union air quality standard of 5 µg/m^3 at most of the sites studied [20]. The study also presented results of measurements near gas stations and airports. The annual median at gas station was 9.1 µg/m^3 within a 300 m buffer region. Concentrations outside this buffer region was lower. The study however found that annual total BTEX concentration was not affected at a location within 1 km of the airport. Another study that looked at total BTEX concentration in Gorakhpur, India found that the total contribution by sites decreased in order of industrial (68.8 µg /m^3) > roadside (65.6 µg /m^3) > agricultural (35.9 µg /m^3) > residential (27.8 µg /m^3) sites [16]. The highest concentration among the BTEX was contributed by toluene, followed by benzene, ethylbenzene and xylene.

Daily roadside air measurements for BTEX were taken at several Asian cities including Malaysia and the results showed high daily benzene concentrations of 87, 52, 32, 23, 13, 12 and 48 µg/m^3 at Ho Chi Minh City (HCM), Hanoi (HN), Cantho, Danang, Hue, Vung Tau (Vietnam), and Kuala Lumpur (Malaysia), respectively (Figure 5) [21]. BTEX is relatively high in Kuala Lumpur and the study suggested motorcycles as possible cause. A summary of BTEX concentration in ambient air in major cities in the world as well results from some studies related to seasonal variability are presented in Table 3 [22-25].

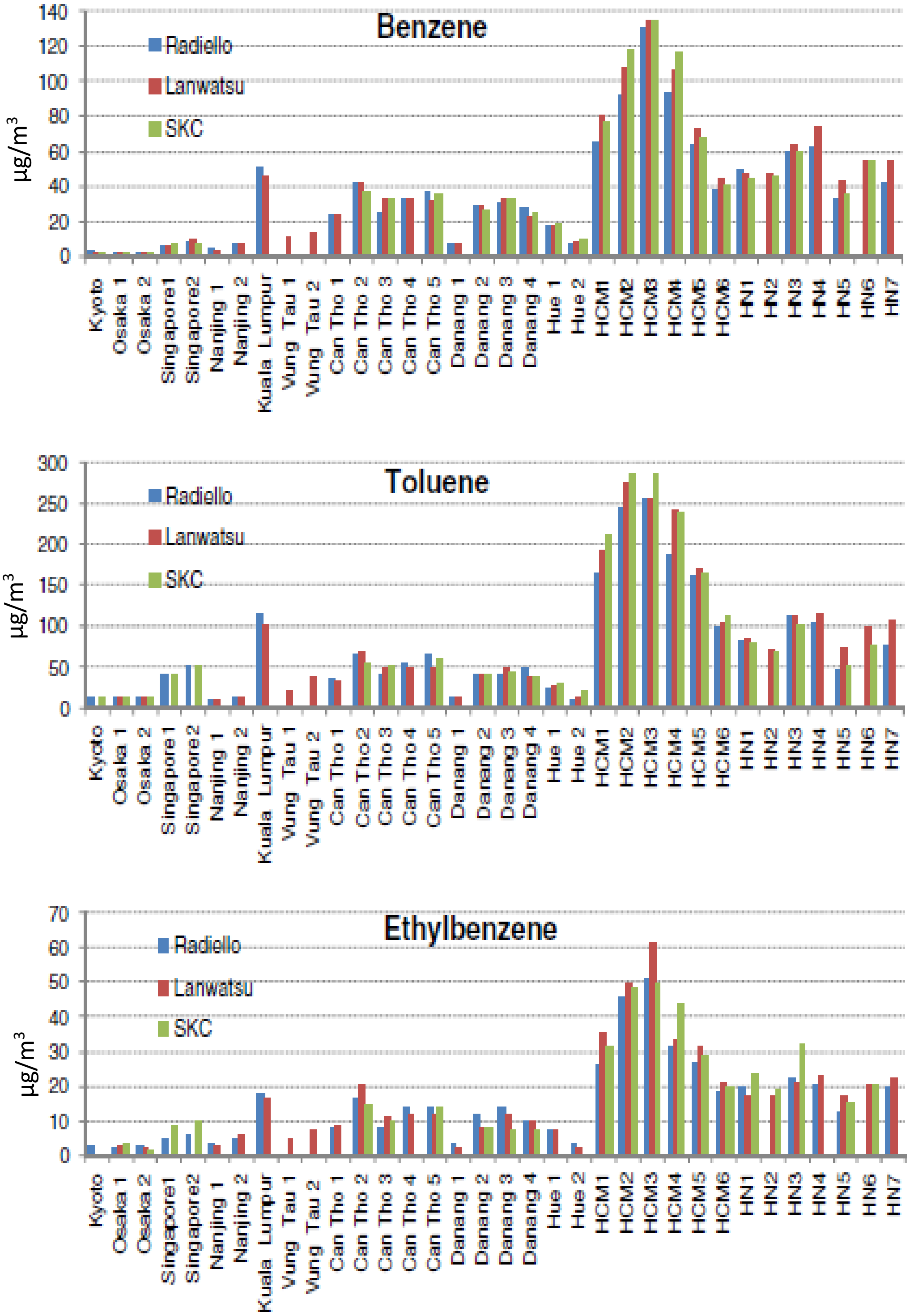


Figure 5: Daily BTEX concentrations in East Asia measured by the Radiello, SKC Ultra I and Lanwatsu passive samplers [21]

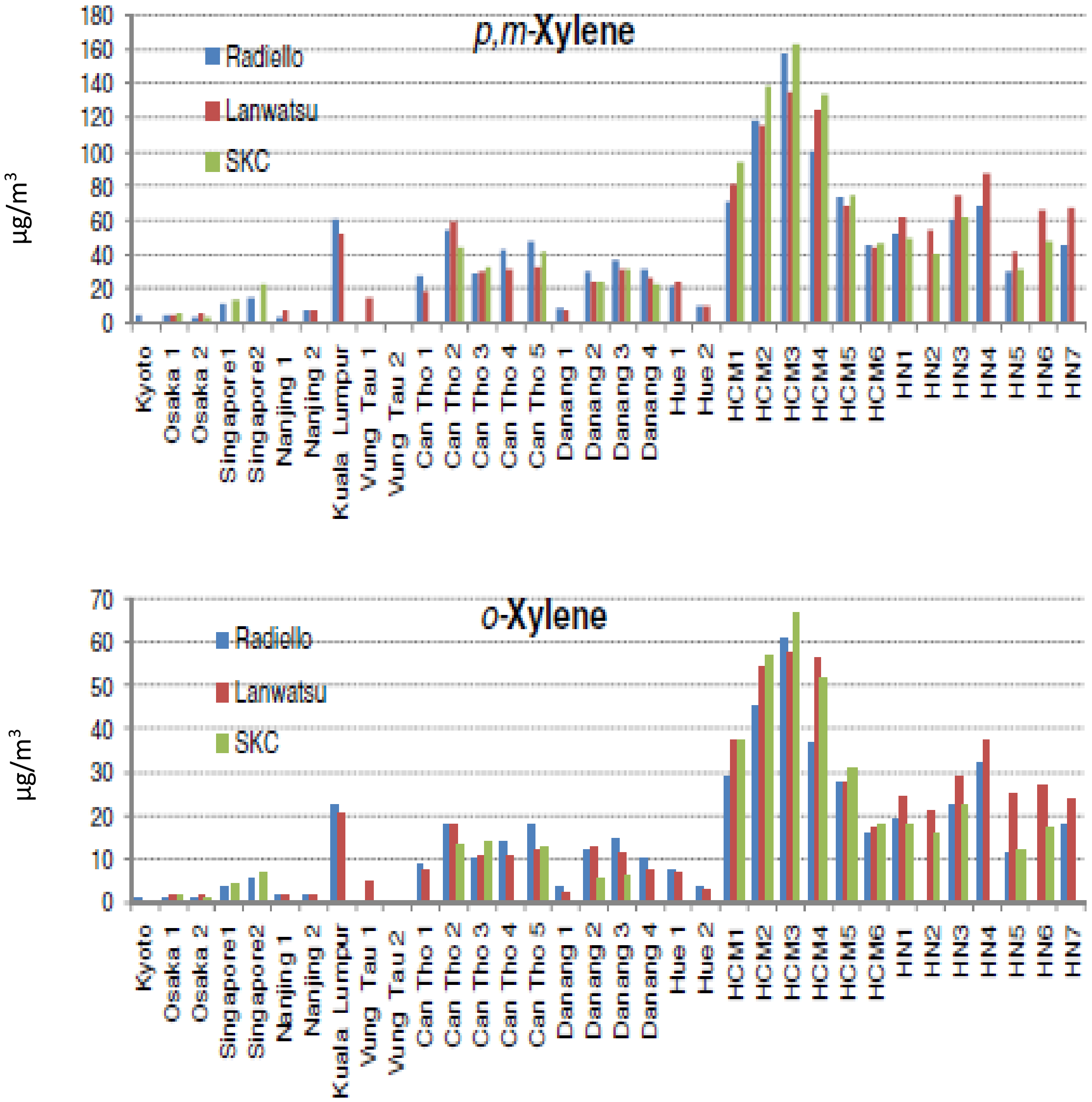


Figure 5: continued

Table 3: Comparison of average level and seasonal trend with studies in other urban area

City, Country	Reported value (site characteristic		Comments
	Benzene ($\mu g/m^3$)	Toluene ($\mu g/m^3$)	
Bangkok, Thailand	5.8 (ambient)	36.1 (ambient)	The average concentrations of ethylbenzene *m*-, *p*-xylene and *o*-xylene were 4.1, 11.0 and 3.7 $\mu g/m^3$, respectively. The pollutants were monitored in ten sites which covered roadside, residential and background areas during April-August 2007.
Hanoi, vietnam	123 (roadside)	87 (roadside)	The high traffic volume and the vicinity of the industrial estate may be the cause for the higher pollution at this roadside
Hong Kong, China	4.9 (roadside)	28.8 (roadside)	Different sources for total BTEX in different seasons are indicated.
Pearl River Delta, China	15.4-67.3 (urban-roadside)	28.6-106.9 (urban-roadside)	Autumn (November) BTEX level is 26-56% more than their summer (July) level. Meteorological conditions such as source and characteristics of air mass are the reason for such massive seasonal variation rather than variation in source input or photochemistry.
Delhi, India	12-55 (urban-residential) 80-550 (urban-traffic crossing)	10-80 (urban-residential) 18-55 (urban-traffic crossing)	Winter VOC level is distinctly higher than summer or monsoon level in general, B/T ratio ranged from 1.04-2.05.
Shizuoka, Japan	0.478 in summer 0.946 in winter	4.339 in summer 6.403 in winter	BTEX level in winter was higher than summer. Pollutants were more homogeneously distributed in winter.

City, Country	**Reported value (site characteristic**		**Comments**
	Benzene (µg/m^3)	Toluene (µg/m^3)	
	(industrial urban)	(industrial urban)	Seasonal variation was also influenced by emission sources.
Yazd, Iran	21±18 (ambient urban)	38±42 (ambient urban)	Toluene and xylene had the highest average concentration compared to benzene and ethylbenzene
Algiers city, Algeria	27.1 (roadside) 9.6 (urban)	39.2 (roadside) 15.2 (urban)	Minor seasonal variation with winter concentration 10% higher than summer. T/B is 1.5-2.1.
Hong Kong, China	417 pptv (urban) (≈1.3 µg/m^3)	2765 pptv (urban) (≈10.4 µg/m^3)	Winter levels of toluene and other VOCs were significantly higher than the summer. A strong local/regional source during winter is suggested. The VOC levels are affected by Asian monsoon circulation; the clean maritime inflow air dilutes (reduces) them significantly during summer also resulting low annually averaged VOC level.
United States*	1.03 (Urban)	2.38 (Urban)	Higher concentration in cooler season for BTEX. Changes in emission activity, removal rates, or dispersion/dilution/transport may explain the seasonal variation
Delhi, India	48-110 (metropolitan-urban)	85-204 (metropolitan-Urban)	Winter levels are higher than summer. Meteorology, variation in source strength and availability of OH radical were identified to be the controlling factors. T/B ratio varied between 1.8-2.5
São Paulo, Brazil	1.30-11.31 (metropolitan-urban)	2.05-16.92 (metropolitan-urban)	Higher VOC concentration in winter (August) than summer (December). Variation in sources is attributed to be the probable cause.

City, Country	Reported value (site characteristic		Comments
	Benzene ($\mu g/m^3$)	Toluene ($\mu g/m^3$)	
Kolkata, India	29.2 (metropolitan -urban)	45.4 (metropolitan - urban)	Significantly higher winter level due to meteorological factors like lowered mixing height and lesser dispersion and also enhanced photochemical removal of TEX in summer; insignificantly lower summer level for benzene due to less photochemical reactivity, only meteorological factors increases the winter level. T/B ratio ranged from 1.3 to 2.2.

*multi-city

Source: Adapted from Majumdar et. al. 2011 [22], Laowagul et al. 2008 [24], Truc and Kim Oanh, 2007 [23], and Hajizadeh et al., 2017 [25]

2.3 Guideline and Standard for BTEX in different countries

Currently guidelines or standards suggested in different countries are typically drafted for annual or 24 hour averaging periods. Table 4 shows ambient guidelines for benzene in several countries including those in the South East Asian region [21]. The United States of America Environmental Protection Agency (US EPA) guidelines and regulations for benzene primarily focus on benzene content in water and occupational exposure (1 part of benzene per million parts of air (1 ppm) in the workplace during an 8-hour workday, 40-hour workweek). Benzene is included in the US 1990 Clean Air Act Amendments and regulated from more than 170 industrial source categories. The Massachusetts Department of Environmental Protection however does provide a more comprehensive guideline for ambient BTEX. European Commission benzene ambient air guideline was at 5 $\mu g/m^3$ per year in 2000 with a target reduction of 1 $\mu g/m^3$ each year (this target is yet to be achieved). The Queensland Environmental Protection (Air) Policy 2008 Australia ambient air quality objective also provides a more comprehensive objective for BTX management (ethylbenzene is not included) [26]. The World Health Organisation (WHO) does not specify ambient standards for BTEX in air. WHO also states that no safe level of exposure can be recommended for indoor air quality.

According to WHO, the estimated excess lifetime risk of leukaemia due to benzene exposure of 1 µg/m^3 in air is 6×10^{-6} [18, 27]. As shown in the table, several countries in Asia which also have their own standard or guideline for Benzene concentration. Korea and India adopted 5 µg/m^3, while Japan, Nepal, Thailand and Vietnam adopt 3 µg/m^3, 20 µg/m^3, 1.7 µg/m^3 and 10 µg/m^3, respectively. These countries however, do not have standards for toluene, ethylbenzene and xylene in ambient air.

Table 4: The 24 hour and annual average ambient guidelines for BTEX used in different countries and organizations

	Benzene		**Toluene**		**Ethylbenzene**		**Xylene**	
Region, Country or Organisation	24 hour	1 year	24 hour	1 year	24 hour	1 year	24 hour	1 year
					(µg/m^3)			
United Kingdom	-	16	-	-	-	-	-	-
European Union	-	5	-	-	-	-	-	-
EPA-Ireland		5						
India	-	5	-	-	-	-	-	-
Japan	-	3	-	-	-	-	-	-
Korea	-	5	-	-	-	-	-	-
Massachusetts (USA)	0.6	0.1	80	20	300	300	11.8	11.8
Nepal	-	20	-	-	-	-	-	-
Queensland (Australia)	-	10	4100	410			1200	950
Thailand	-	1.7	-	-	-	-	-	-
Vietnam	-	10	-	-	-	-	-	-
WHO	No safe level	No safe level	-	-	-	-	-	-

3 Methodology

3.1 Sampling strategy and sampling sites

Two sampling strategies for BTEX determination in ambient air will be applied: continuous sampling and active sampling. Continuous monitoring sites will monitor BTEX for a longer period at selected stations in Peninsular Malaysia and Sabah. Active sampling will be conducted at selected hotspot areas such as petrochemical industries, petrol stations and roadsides with a high density of motor vehicle traffic. Active sampling will also be conducted at sites selected for continuous monitoring. These areas have been recognised to record high concentration of BTEX based on previous researches. The list of locations for continuous and active sampling are presented in Table 5 and Table 6 respectively. The pictures and locations (map) of selected sampling stations for both continuous and active sampling are also presented in Appendix 3 and Appendix 4.

Table 5: Selected location and duration for continuous sampling

Continuous monitoring	Station type	Sampling Date
1) UKMKL, Kuala Lumpur	Urban	6/1/2017 – 29/3/2017
2) UMT, Kuala Terengganu, Terengganu	Sub-urban	8/2/2017 – 9/3/2017
3) UMS , Kota Kinabalu, Sabah	Sub-urban	15/2/2017 – 28/2/2017
4) Pusat Penyelidikan Bukit Frasers (PPBF), Raub, Pahang	Background	16/1/2017 – 9/3/2017

Table 6: Detailed information on active sampling site locations

Location	Remarks	Date	Time	Additional remark
UKMKL UKM, Kuala Lumpur N3.168133°, E101.700433°	Urban	9/8/16 11/8/16 14/8/16 31/8/16	1.30 pm 8.00 am 9.00 am 11.00am	Heavy traffic
PPBF Pusat Penyelidikan Bukit Frasers N3.726472°, E101.713806°	Background	17/1/17 18/1/17	9.00am, 2.00pm, 5.00pm	Background
UMT Universiti Malaysia Terengganu, Kuala Terengganu N5.412267°, E103.085050°	Sub- Urban	18/8/16 8/2/17 9/2/17	10.40am 9.00am, 2.00pm, 5.00pm	
UMS Universiti Malaysia Sabah, Kota Kinabalu N6.032878°, E116.120687°	Sub-urban	16/2/17 17/2/17	9.00am, 1.00pm, 4.00pm	Suburban
ROADSIDE Chow Kit, Kuala Lumpur N3.167361°, E101.698472°	Roadside	9/8/16 11/8/16 14/8/16	12.00 pm 9.00 am 11.00 am	Normal Heavy traffic Weekend
INDUSTRIAL Kerteh Integrated Petrochemical Complex (KIPC) N4.579383°, E103.449917°	Industrial site	19/8/16 19/8/16 19/8/16 20/8/16 18/8/16	9.45 am 4.00 pm 10.00 pm 5.00 am 6.00 pm	Petrochemical
PETROL STATION Bandar Baru Bangi: Seksyen 15 N2.934419°, E101.768479° Seksyen 7 N2.967465°, E101.7743316°	Petrol Station	16/5/17 23/5/17	3.00pm	
AIRPORT Kuala Lumpur International Airport (KLIA2), Sepang N2.731433°, E101.680017°	Airport	11/5/17 25/5/17	11.00am	Outside the airport/runway

3.2 BTEX Sampling and Analyses

3.2.1 Continuous monitoring of BTEX

Continuous monitoring for BTEX in ambient air was performed by using the AMA Online Gas Chromatography GC 5000 BTX (AMA Instruments, Germany). The system was equipped with capillary column for separation and flame ionization detector (FID) for the detection of BTEX. This instrument measures BTEX for 30 min or 1 hour resolution. This instrument consists of an in-built low flow meter, Tenax® column and a compact Gas Chromatography (GC) system. A certain volume of air (about 30-50 mL) is to be pre-concentrated in the Tenax® column and then, desorbed into the GC system to detect the BTEX in a full spectrum. The instrument displays the concentration of each BTEX after went through a regular calibration on basis of standard gas mixtures. The AMA 5000GC BTX FID setup parameters are as follows:

a) Column type: AMAsep 1 (30 m, 0.32 mm ID, 1.5 um film)
b) GC temp program: 50°C 3 min, 8^{o}C/min, 130^{o}C 5 min
c) Carrier gas pressure: (0.45-0.52 Bar H_2)
d) Std Dev for 5 ppb benzene (5 replicates) : < 1.5%
e) Signal to noise : < 600 µV (1 min)

3.2.2 Active sampling

This section provides the sampling stations and methodology for the determination of benzene, toluene, ethylbenzene, m,p-xylene, and o-xylene analysis in ambient air. Generally, the procedure involves an active sampling method of pumping a known volume of air onto the sorbent tubes followed with Thermal Desorption (TD) hyphenated with Gas Chromatography (GC) analysis. 200 mg Tenax GR® sorbent with mesh 60/80 was pre-packed onto 89mm (L) x 6.5 mm (OD) stainless steel tube was used for this BTEXs analysis.

Sampling pump Escort ELF®, USA and Twin port sampler Gemini ®, USA were calibrated with an air flow calibrator Sensidyene®, USA. Ambient air was pumped at flow rate 50ml/min and 100ml/min for each sampling port to collect 2 L and 4 L of air. All sample tubes were stored at 2°C in a freezer and analysed within one week with Thermal Desorption System (TDS Unity-1 and Ultra Series 2 (Markes, UK) couple to Gas Chromatography (GC)- Mass Spectrometer

(MS) (Agilent, USA) (Figure 6). Analysis of sorbent tubes samples using TD-GCMS was developed for VOCs determination including BTEXs. All parameters were set as Table 7.

Figure 6 Thermal Desorption (TD) System coupled with Gas Chromatography-Mass Spectrometer (GC-MC) method

Table 7: Thermal desorption and GC-MS experimental condition for this study

Thermal Desorber		GC Condition		MS Condition	
Desorb temperature	280°C	Initial temperature	36°C	Mass range	35-300 amu
Desorb Time	30 min	Ramp 1	5°C/ min	Ionization	Electron Ionisation
Desorb Flow	30 ml/min	Temperature 2	250°C	Source temperature	230°C
Valve Temperature	270°C	Column	HP-624 60 m x 320 µm x 1.80 µm	Transfer line temperature	280°C
Trap low	25°C	Column flow	2.0 ml/min	-	-
Trap high	280°C	Inlet temperature	200°C	-	-

GC-MS result and chromatogram were evaluated with Chemstation and MS NIST 2.0 library softwares. BTEXs compounds were calculated in mass (ng) of sorbent retained in tubes. A further calculation to convert the results in concentration (ppb) was as follows:

$$Concentration\ (Fraction) = \frac{Mass\ (g)}{Molar\ mass\ (\ g\ mol^{-1})} \times \frac{25}{Volume\ pumped\ onto\ tube\ (L)}$$

The results are then converted to unit of µg/m^3. Summary of the method work flow of this experimental work is visualized as Figure 7. Quality control measures (QC) are described in detail in Section 3.3.

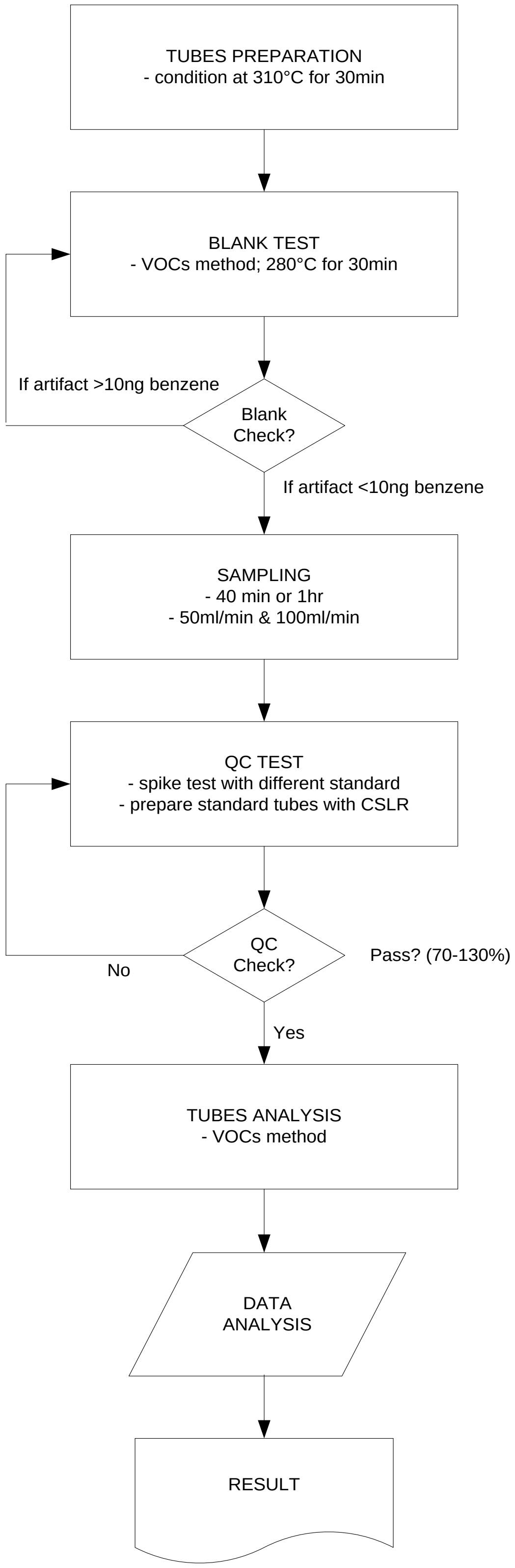


Figure 7: BTEXs analysis work flow

3.3 Quality Control and Quality Assurance (QA/QC)

3.3.1 Continuous measurement of BTEX

QC Check for BTEX continuous monitoring with AMA 5000 GC was performed by using BTEX 10 ppm standard gas (MESA, USA) diluted to 10 ppb with purified nitrogen in 10 L Tedlar bag with an aid of calibration solution loading rig (CSLR). The QC check result is shown in Table 8.

Table 8: QC check dates for BTEXs continuous monitoring with AMA 5000 GC

Site Location	QC check (%) n=3
UKMKL, Kuala Lumpur	B:102, T:89, EB:52, mp-X: 84, o-X: 54
PPBF, Pahang	B:127, T:104, EB:76,mp-X: 61, o-X: 63
UMT, Terengganu	B:98, T:102, EB:86,mp-X: 83, o-X: 72

B, benzene; T, toluene; E, ethylbenzene; m,p-X, m-+p- xylene; o-X,o-xylene

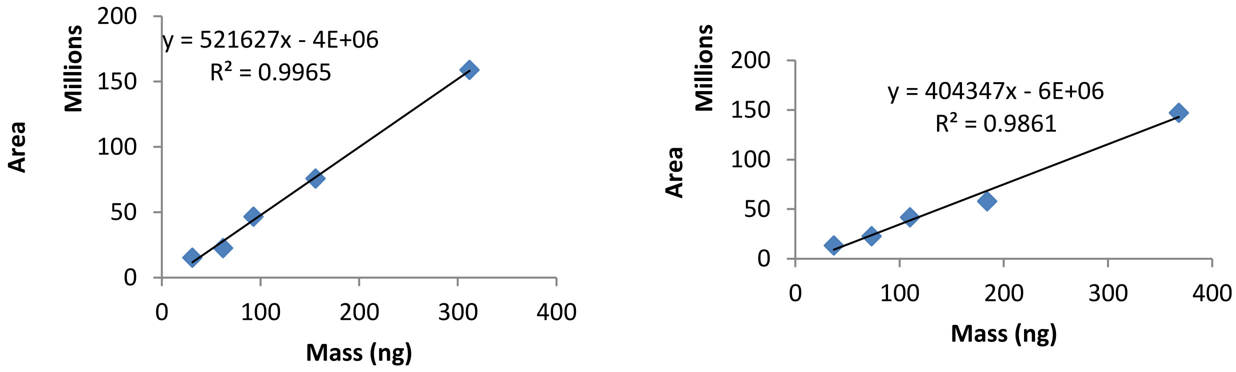
3.3.2 Active sampling

During the measurement of BTEX using the active sampling procedure, all necessary quality control (QC) measures were taken into consideration such as blank tubes, breakthrough, and QC recovery. In this thermal desorption method, US EPA recommended TO-17 method will be considered as part of the QA/QC. An external calibration with 5 points (30-400 ng mass) was followed by using CSLR (Markes, UK). 10 ppm of BTEXs gas standard (MESA, USA) was used for the calibration. For the purpose of routine QC check, spike of 100 ppb VOCs standard gas (SCOTTY, USA) containing BTEX was used for this study. Validation of calibration curves was performed with a certified reference sample (CRS) (Markes, UK) which contained 100 ng of Benzene, Toluene and o-Xylene in Tenax ® tubes. Limit of detection (LOD) was determined with the analysis of ten replicates of blank tubes by the TD-GC-MS system. Results of QCs measurement and calibration curves are shown in Table 9 and Figure 8.

Table 9: Quality Control check results for BTEX analysis

Compound Name	Formula	Correlation coefficient R_2	Spike QC Rec (%)	CRS QC Rec (%)	LOD (ppb)
Benzene	y=521627x-4E+06	0.9965	89.04	91.61	0.08
Toluene	y= 404347x-6E+06	0.9861	91.18	102.80	0.07
Ethylbenzene	y=323622x-1E+06	0.9947	75.51	NA	0.02*
m,p-Xylene	y=652512x-107871	0.9998	67.56	NA	0.02*
o-Xylene	y=266305x-6E+06	0.9936	109.71	119.71	0.02*

* Represent similar type of standard



(a) Benzene (b) Toluene

Figure 8: Calibration curves for (a) Benzene (31.2 -312 ng) and (b) Toluene (36.8-368 ng)

3.4 Backward Trajectories

The analysis of the 72 hour backward trajectories for wind direction to specific sampling location was undertaken using the Hybrid-Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model. This method will determine simulations of BTEX transport in the atmosphere from its possible sources. Hybrid Single Particle Langragian Integrated Trajectory model (HYSPLIT version 4.9) developed by the National Oceanic and Atmospheric Administration (NOAA)’s Air Resource Laboratory (ARL) was employed to calculate 72 hour back trajectories to determine origin of air mass arriving at the monitoring stations. [28].

3.5 Statistical Analysis

Statistical analysis has been employed to analyse all data collected. Several analyses, such as descriptive statistics and Pearson correlation, will be carried to indicate the variations of BTEXs data and its correlation.

3.6 Health Effect of BTEX Exposure

3.6.1 Health Risk Assessment

The health risk assessment of BTEX via inhalation route was performed using US EPA recommended method. The chronic daily intake (CDI) of BTEX was estimated as follows:

$$CDI = (CA \times CF \times IR \times ET \times EF \times ED)/BW \times AT$$

Where,

CDI = chronic daily intake ($\mu g/m^3$)
CA = contaminant concentration in air ($\mu g/m^3$)
IR = inhalation rate (m^3/h) for an adult (0.83) (Hazrati et. al. 2015 [29]; US EPA 2005[30])
CF = conversion factor (0.001 mg/µg)
ET = exposure time (24 hours/day) (US EPA 2005[30])
EF = exposure frequency (350 days/year) (US EPA 2005 [30])
ED = exposure duration, (24 years for adult) (US EPA 2005 [30])
BW = Body weight (70 kg for adults)
AT = averaging time (ED in years x 365 days/year) for non-carcinogenic risk calculation (US EPA 2009 [31])
AT = averaging time (70 years x 365 day/year) for carcinogenic risk calculation (US EPA 2005 [30])

The hazard quotient (HQ) for non-cancer risk was estimated as

$$HQ = \frac{CDI}{RfD}$$

Where,

RfC = chronic inhalation reference concentration (mg/m^3) obtained from https://rais.ornl.gov/cgi-bin/tools/TOX [32].
RfD = RfC (inhalation reference concentration mg/m^3) × 20 (assumed adult inhalation rate m^3 /day) × 1/BW (kg); based on RfCs for USEPA, IRIS (benzene=0.03 mg/m^3 , toluene=5 mg/m^3 , ethylbenzene=1 mg/m^3 , and xylenes=0.1 mg/m^3). The RfD values of benzene, toluene, ethylbenzene and xylenes were 0.00855, 1.4, 0.286 and 0.029 mg/kg/day, respectively (USEPA 2005 [30]; Hazrati et. al. 2015 [29]

3.6.2 Life-time cancer risk (LTCR)

Similarly, the life-time cancer risk (LTCR) for benzene and ethylbenzene was estimated using the US EPA suggested formula (USEPA 2005 [30]).

The LTCR was calculated using the following equation:

$$LTCR = CDI \ \times SF$$

Where
SF = carcinogenic slope factor for a pollutant to be exposed via inhalation route.
The SF value of benzene and toluene were obtained based on the unit risk of benzene and ethylbenzene and the inhalation rate for an adult per day (20 m^3/day) from http://www.epa.gov/iris [33]. Thus, the SF values of benzene and ethylbenzene used were 0.0273 and 0.00385 mg/kg/day^{-1}, respectively [34]

4. Results and discussion

The results are presented in several sections. Section 4.1 describes the results at continuous monitoring sites as stated in Table 5. Section 4.2 presents the results of active sampling conducted in selected sampling sites stated in Table 6. Correlations analysis and source apportionment of BTEX is presented in Section 4.3 and 4.4 respectively. Section 4.5 presents the composition of BTEX in fuel while Section 4.6 presents the results of backward trajectory analysis. The final section (Section 4.7) presents results for analysis related to health risk.

4.1 Baseline results of BTEX from the continuous sampling

The continuous monitoring system consisting of compact GC (AMA 5000 GC, Germany) successfully detected individual BTEX concentrations (benzene, toluene, ethylbenzene, m,p-xylene and o-xylene) at the four fixed sites in UKMKL, UMT, UMS and PPBF. The summary statistics of the results for the sampling period in ppb and µg/m^3 are presented in Table 10 and Table 11, respectively.

As reported in Table 10 and Table 11, the benzene concentration was highest at the UKMKL site (1.95 ppb or 6.20 µg/m^3) as compared to the other sites. The average benzene concentration at UMT (Terengganu) was 0.51 ppb or 1.63 µg/m^3 and in UMS (Sabah) the average was 0.71 ppb or 2.25 µg/m^3. UKMKL is located near roadside. Benzene is released in the ambient air mainly from the transportation sector and industrial emission sources. Benzene is used as an additive to fuel in the transportation sector. On the other hand, UMT and UMS are considered as sub-urban sites. The benzene concentration recorded in UKMKL exceeds the concentration of 5.0 µg/m^3, the maximum value recommended by European Union for annual average. The benzene concentration at the background site in PPBF was the lowest (0.22 ppb or 0.69 µg/m^3) among all four stations. This site is located in a rural area and experiences the least influence from the nearby emission sources. The low traffic volume in the background site shows noticeable influence on the low BTEX concentrations compared to the urban and sub-urban sites.

Table 10: Descriptive statistic of the results from continuous monitoring at selected locations (in ppb)

Location	Pollutant	Mean	Min	Max	Median	Std. Dev.
UKMKL	Benzene	1.95	0.18	9.31	1.73	1.11
	Toluene	5.97	0.96	34.48	5.28	3.08
	Ethylbenzene	1.05	0.02	6.13	0.96	0.52
	m,p-Xylene	2.68	0.02	7.11	2.53	1.05
	o-Xylene	1.15	0.03	2.96	1.10	0.43
UMT	Benzene	0.51	0.10	7.96	0.40	0.48
	Toluene	2.15	0.11	28.59	0.89	3.45
	Ethylbenzene	0.39	0.10	27.41	0.30	0.82
	m,p-Xylene	0.58	0.10	2.42	0.52	0.36
	o-Xylene	0.39	0.10	9.69	0.32	0.40
UMS	Benzene	0.71	0.10	2.90	0.53	0.59
	Toluene	1.72	0.10	7.72	1.47	1.20
	Ethylbenzene	0.34	0.10	2.86	0.26	0.28
	m,p-Xylene	0.68	0.10	3.71	0.56	0.51
	o-Xylene	0.32	0.10	1.81	0.23	0.27
PPBF	Benzene	0.22	0.01	1.00	0.19	0.14
	Toluene	0.74	0.01	5.38	0.62	0.55
	Ethylbenzene	0.08	0.01	0.66	0.06	0.08
	m,p-Xylene	0.29	0.01	1.54	0.25	0.21
	o-Xylene	0.26	0.05	1.12	0.20	0.19

Table 11: Descriptive statistic of the results from continuous monitoring at selected locations (in µg/m^3)

Location	Pollutant	Mean	Min	Max	Median	Std. Dev.
UKMKL	Benzene	6.20	0.57	29.51	5.48	3.51
	Toluene	22.31	3.59	128.96	19.75	11.54
	Ethylbenzene	4.52	0.09	26.42	4.14	2.25
	m,p-Xylene	11.57	0.09	30.64	10.90	4.54
	o-Xylene	4.96	0.13	12.76	4.74	1.87
UMT	Benzene	1.63	0.32	25.23	1.27	1.51
	Toluene	8.06	0.41	106.93	3.33	12.92
	Ethylbenzene	1.68	0.43	118.14	1.29	3.54
	m,p-Xylene	2.51	0.43	10.43	2.24	1.56
	o-Xylene	1.69	0.43	41.76	1.38	1.73
UMS	Benzene	2.25	0.32	9.19	1.68	1.88
	Toluene	6.44	0.37	28.87	5.48	4.50
	Ethylbenzene	1.48	0.43	12.33	1.12	1.21
	m,p-Xylene	2.94	0.43	15.99	2.41	2.18
	o-Xylene	1.37	0.43	7.80	0.99	1.14
PPBF	Benzene	0.69	0.03	3.17	0.60	0.45
	Toluene	2.76	0.04	20.12	2.32	2.05
	Ethylbenzene	0.36	0.04	2.84	0.26	0.36
	m,p-Xylene	1.27	0.04	6.64	1.08	0.91
	o-Xylene	1.10	0.22	4.83	0.86	0.83

The concentration of toluene was the highest (5.97 ppb) at the urban site (UKMKL). The averaged toluene concentration was 2.15 and 1.72 ppb at UMT and UMS sites, respectively. At the background PPBF site, toluene recorded the lowest concentration (0.74 ppb). Toluene is predominantly emitted in ambient air from the traffic combustion, solvent use and industrial emission sources. Thus, the influence from the local source is pronounced at the UKMKL site. Ethylbenzene concentrations observed were 1.05, 0.39, 0.34 and 0.08 ppb at UKMKL, UMT, UMS and PPBF sites, respectively. Xylenes showed a similar pattern. Thus, the concentration of BTEX was the highest at the UKMKL site which is heavily influenced by the transportation sources.

Figure 9 shows the day wise variability of benzene, toluene, ethylbenzene, m,p-xylene, and o-xylene concentrations from Monday to Sunday. Benzene was the most abundant BTEX each day and the concentration of benzene was nearly similar with a small difference on Thursday, where the benzene increased by about 0.5 ppb at UKMKL site. At UMS site, the highest concentration of benzene was observed on Wednesday. However, UMS had a relatively short sampling duration compared to other stations and this may be a contributing factor to the observed variability. The concentration of benzene was relatively lower at UMT and PPBF sites. At PPBF background site, benzene concentration was less than 0.5 ppb throughout the week. Toluene also shows a similar temporal variability to benzene. However, toluene was the most abundant BTEX at all the sampling sites. At PPBF site, toluene showed very little variability and recorded the lowest concentration amongst the stations. Thus, a negligible impact from the nearby sources was observed at the PPBF site. The observations also showed similar day to day variations for ethylbenzene, m,p-xylene and o-xylene.

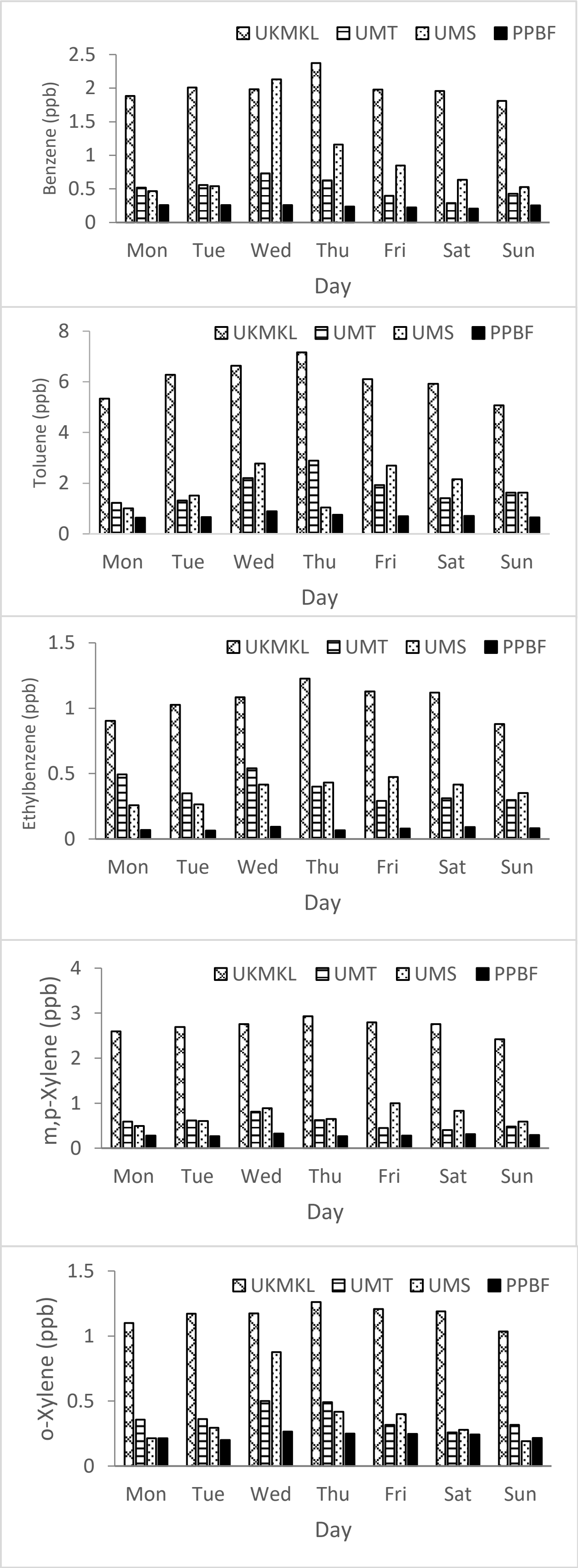


Figure 9: Day-wise concentration of BTEX at UKM KL, UMT, UMS and PPBF sites.

A diurnal line and box plots of the individual BTEX concentrations are presented in Figures 10-14 for the UKMKL, UMT, UMS and PPBF sites. Diurnal variation of BTEX provides insight to the impact of transportation sources to BTEX, photochemical degradation of BTEX and the expansion or shrinking of the planetary boundary layer (PBL). At UKMKL site, there was a clear depiction of the time dependent distribution of BTEX in the diurnal pattern as BTEX was most abundant at this urban site. Hour 8:00 to 10:00 can be categorized as a morning rush hour. The number of traffic usually peaked during this period. As traffic is a predominant emitter of BTEX, the high concentrations of benzene, toluene, ethylbenzene, m,p-xylene and o-xylene were observed at this site during the morning rush hour. A second episode of BTEX was observed during 23:00 – 24:00 hour. At midnight, the shrinking of PBL occurs due to the stable atmospheric conditions and thus, an accumulation of the BTEX compounds can be observed during this period.

UKMKL site recorded the lowest concentration of BTEX during 13:00-17:00 hour. Meteorological conditions are key factors in the degradation of BTEX via ambient photolysis reactions. The precursors of the said photolysis reactions are water vapour, hydroxyl radical (OH) and solar radiation. As this site is located at the centre of the Kuala Lumpur City, the heterogeneous reactions involved in generating OH are favourable at this site compared to sub-urban and background remote site. High temperature during mid-day can influence the expansion of PBL and thus, this process also helps to reduce the concentration of BTEX during this period.

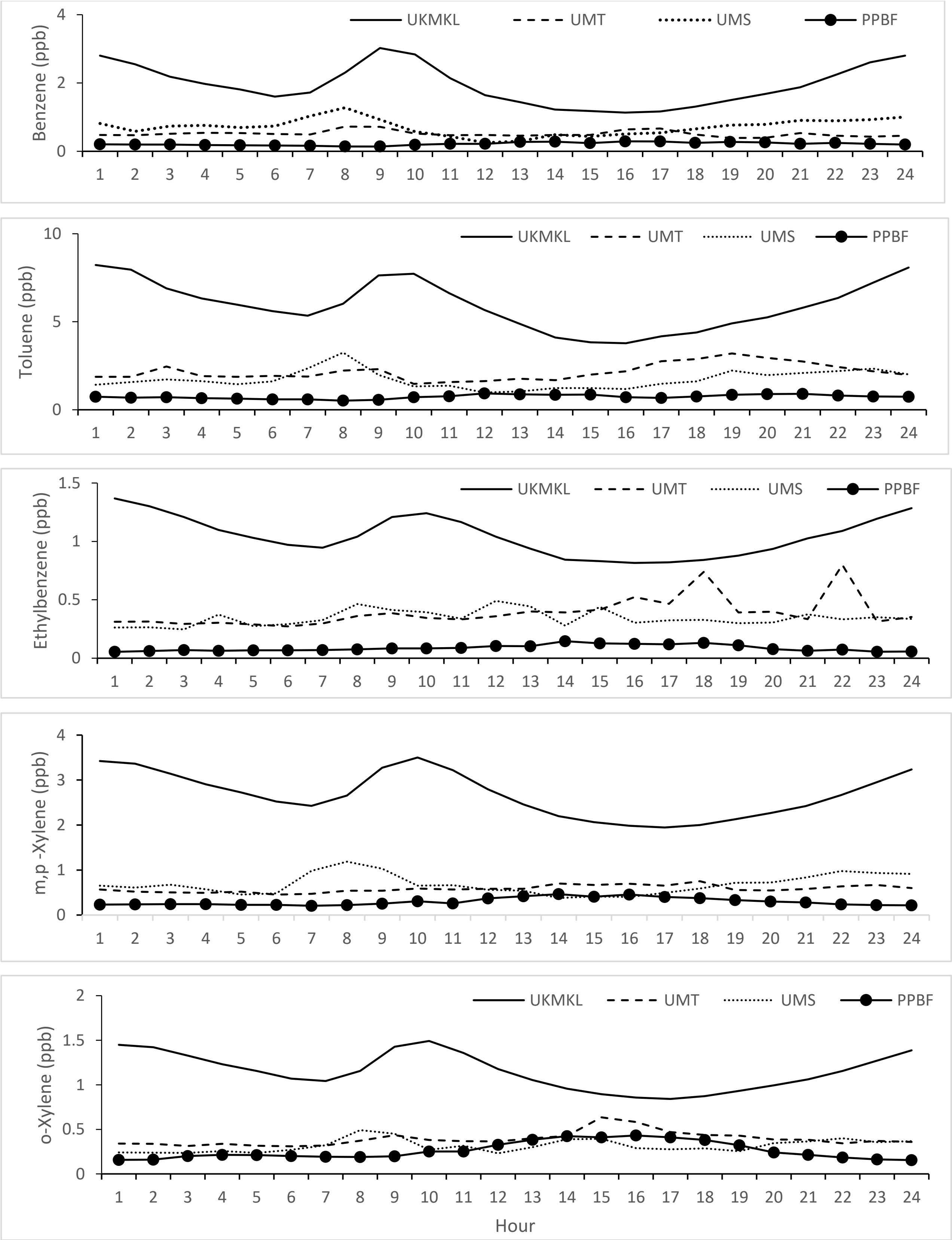


Figure 10: **Diurnal concentration of BTEX at UKM KL, UMT, UMS and PPBF sites.**

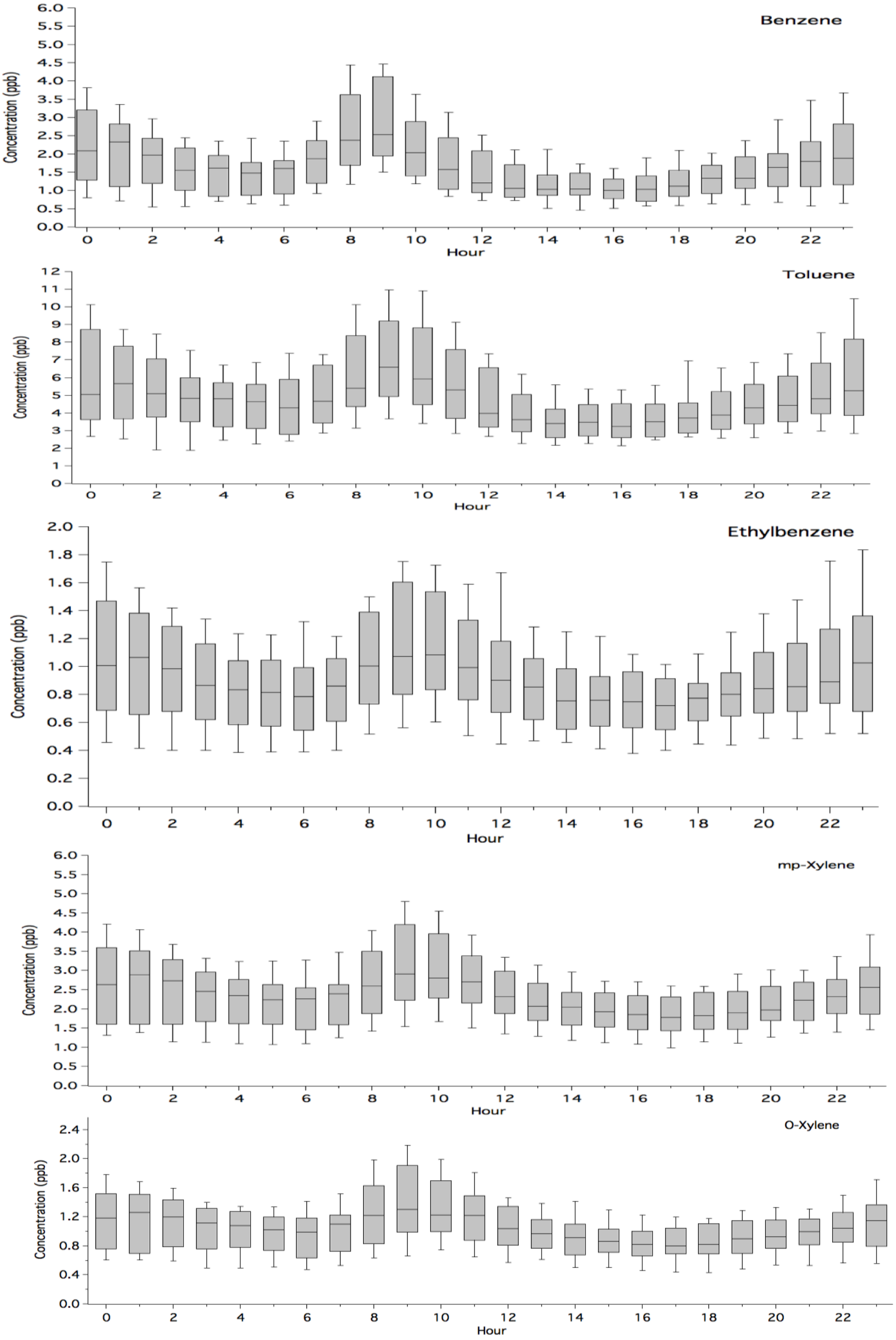


Figure 11: Diurnal box-plots of BTEX at UKMKL site

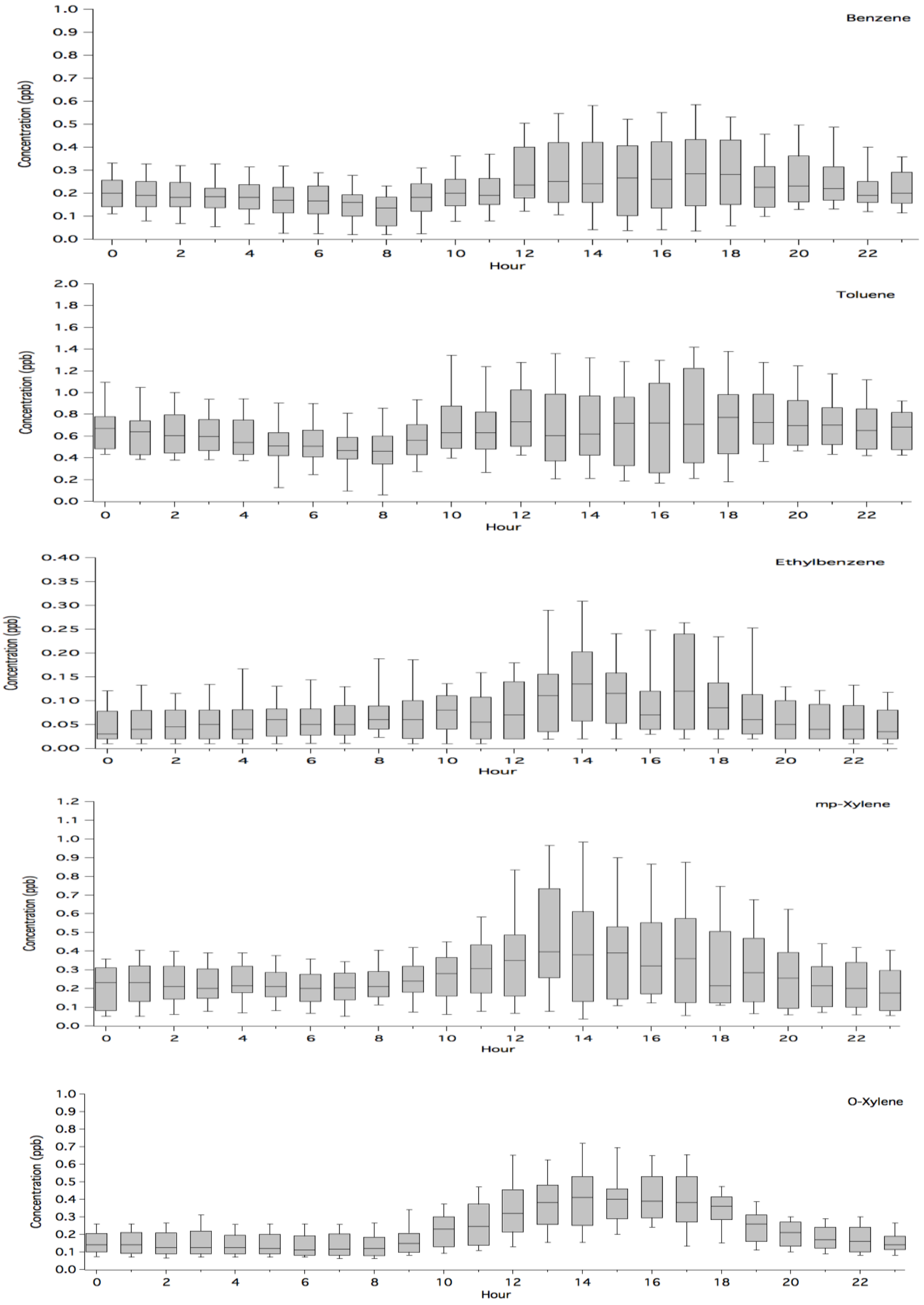


Figure 12: Diurnal box-plots of BTEX at PPBF site

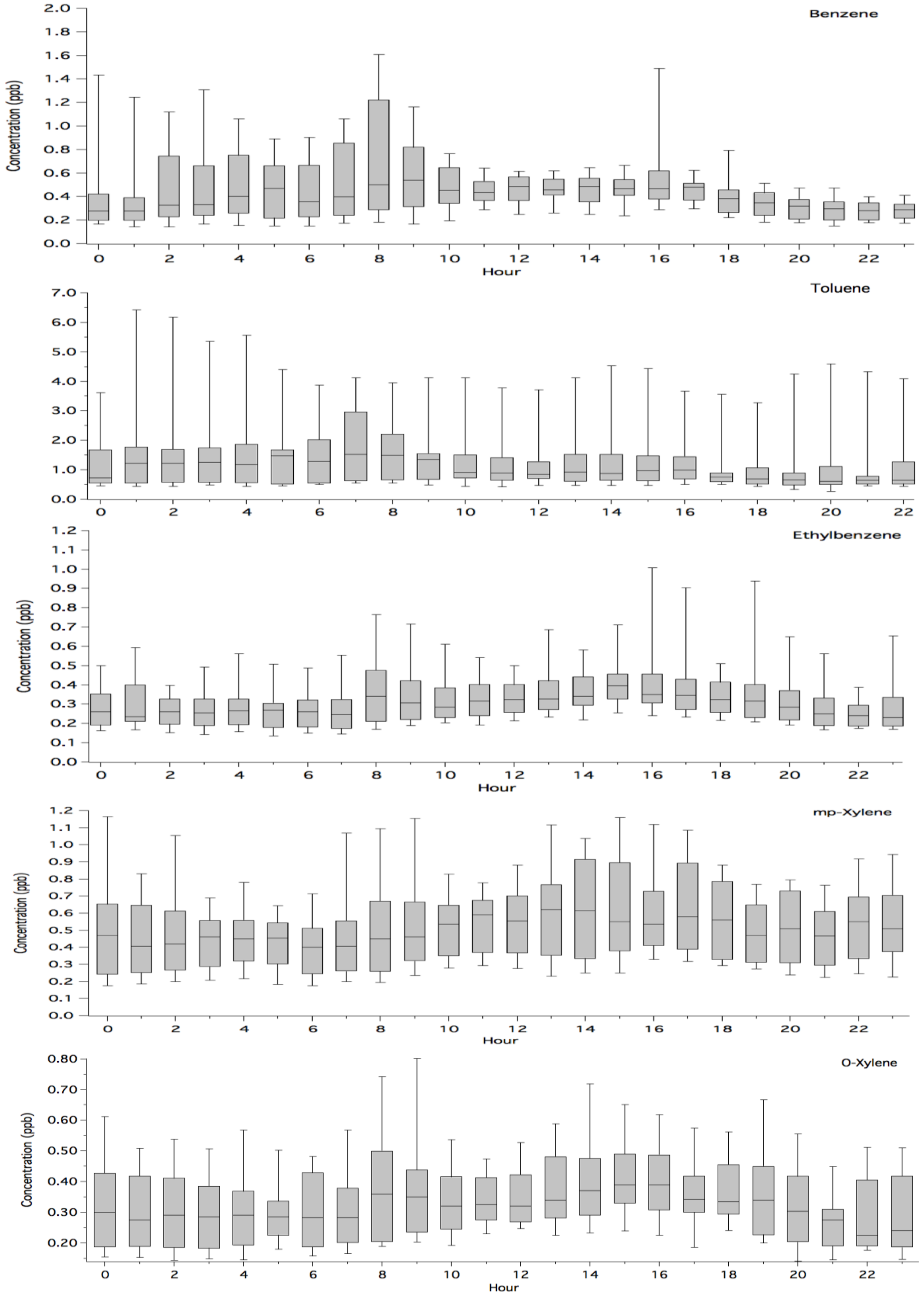


Figure 13: **Diurnal box-plots of BTEX at UMT site**

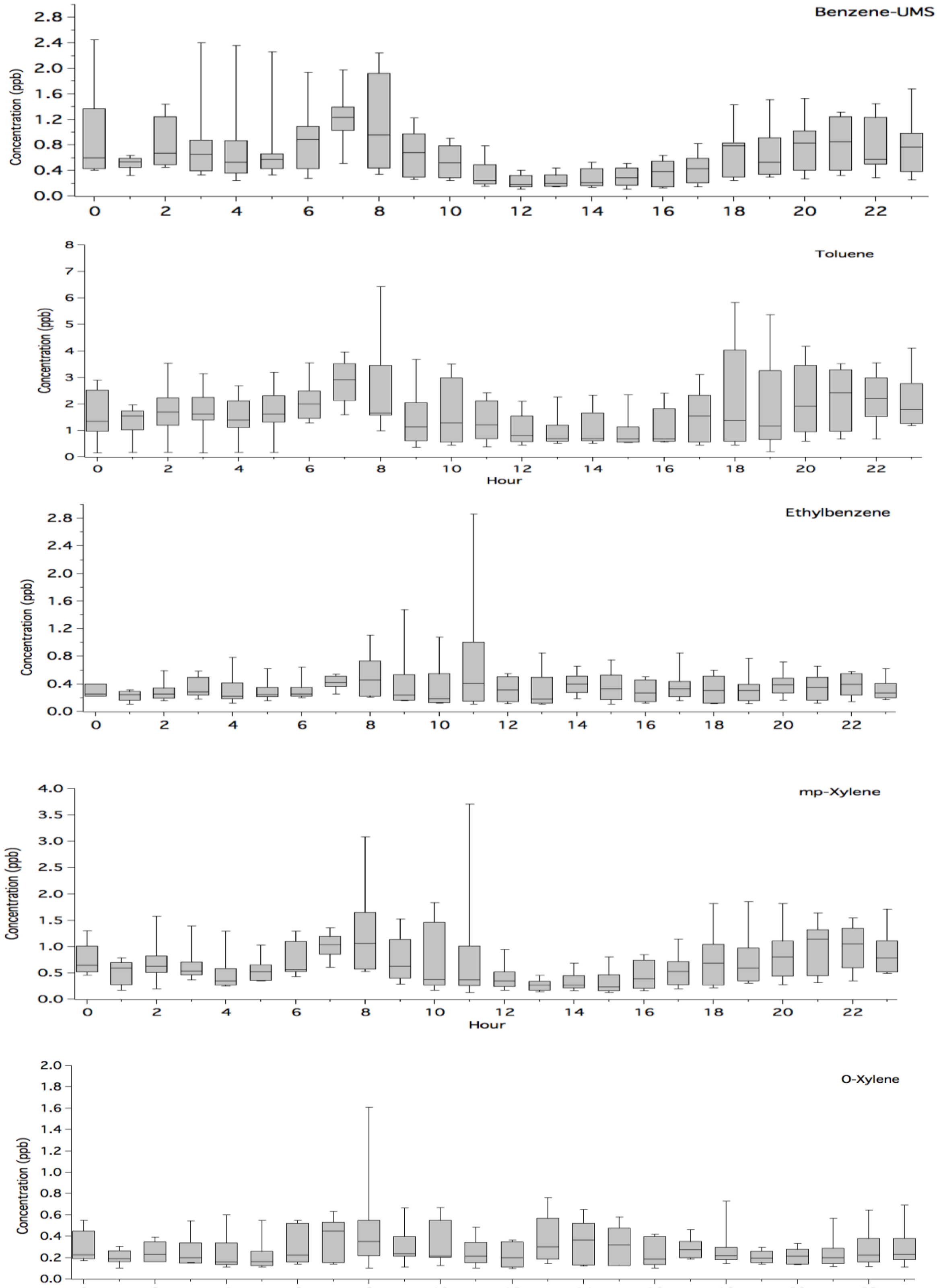


Figure 14: **Diurnal box-plots of BTEX at UMS site**

4.2 Description of BTEX determined using sorbent tube (active sampling) at fixed sites and hotspots

Active sampling of ambient air was conducted at the fixed continuous monitoring stations in UKMKL, UMT, UMS and PPBF sites. The results of the active sampling are reported in Table 12. These results are shown to provide comparison and validation with the BTEX results obtained from the continuous measurement at the similar sites. At the same time, sampling using sorbent tube was also conducted at hotspots such as petrol stations, urban roadside (Chow Kit, KL), Kuala Lumpur International Airport, and at the vicinity of an industrial area (KIPC, Kerteh). The results of the BTEX sampling at the hotspots sites are shown in Table 13. The concentration of total BTEX follow the order of roadside>petrol station > industrial site > airport. The concentration of benzene was higher at the urban roadside compared to other hotspots sites i.e. petrol station, industry and airport. The concentration of benzene at roadside is mainly contributed by combustion of fossil fuel by vehicles and the evaporation from nearby petrol filling station. As reported in the previous section for the guideline by WHO, there is no safe level of benzene in the outdoor ambient air. Benzene is a recognized genotoxic carcinogenic in human as 1 $\mu g\ m^{-3}$ of benzene can cause the excess lifetime risk of leukaemia for 6×10^{-6} people i.e. 6 in 1 million people. Comparing the observation to the guideline by the European Union, the concentrations of benzene at the three sites exceed the threshold of 5 $\mu g/m^3$. However, it should be noted that the guideline is for annual average concentration of benzene. Benzene concentrations in urban and roadside were relatively lower than the neighbouring countries (Ho Chi Minh City, Vietnam; Delhi, India; Hong Kong, China) and other continental locations (Algiers city, Algeria).

Table 12: Concentrations of BTEX at sampling site in units of ppbv and µg/m^3 (in parenthesis) from active sampling.

Compound	**Ambient UKM KL (n=13)**				**Ambient UMT (n=12)**				**Ambient UMS (n=8)**				**Ambient PPBF (n=14)**			
	Avg	Min	Max	SD	Avg	Min	Max	SD	Avg	Min	Max	SD	Avg	Min	Max	SD
Benzene	2.36	0.94	4.06	1.74	0.61	0.11	1.27	0.40	0.38	0.03	1.22	0.41	0.12	0.02	0.41	0.14
	(7.48)	(2.98)	(12.86)	(5.51)	(1.93)	(0.35)	(4.02)	(1.52)	(1.20)	(0.10)	(3.87)	(1.30)	(0.38)	(0.06)	(1.30)	(0.44)
Toluene	7.52	4.73	12.29	4.66	2.48	1.23	4.45	1.07	2.48	1.57	4.27	1.13	1.24	0.60	1.51	0.29
	(28.10)	(17.68)	(45.93)	(17.43)	(9.27)	(4.60)	(16.63)	(4.00)	(9.27)	(5.87)	(15.96)	(4.22)	(4.63)	(2.24)	(5.64)	(1.08)
Ethylbenzene	2.23	1.33	4.55	1.06	0.56	0.32	0.81	0.17	0.51	0.33	0.82	0.19	0.31	0.19	0.37	0.07
	(9.60)	(5.73)	(19.59)	(4.44)	(2.41)	(1.38)	(3.49)	(0.73)	(2.20)	(1.42)	(3.53)	(0.82)	(1.33)	(0.82)	(1.59)	(0.30)
m,p-Xylene	3.22	1.36	8.34	2.38	0.53	0.14	1.18	0.41	0.42	0.11	0.90	0.32	0.09	0.03	0.14	0.04
	(13.87)	(5.86)	(35.91)	(10.25)	(2.28)	(0.60)	(5.08)	(1.77)	(1.81)	(0.47)	(3.88)	(1.38)	(0.39)	(0.13)	(0.60)	(0.17)
o-Xylene	2.19	1.28	3.87	1.07	2.61	1.90	3.14	0.48	2.23	1.84	2.80	0.36	1.69	0.95	1.96	0.39
	(9.43)	(5.51)	(16.66)	(4.61)	(11.24)	(8.18)	(13.52)	(2.07)	(9.60)	(7.92)	(12.06)	(1.55)	(7.28)	(4.09)	(8.44)	(1.68)
Total BTEX	17.52	9.64	33.11	10.91	6.79	3.70	10.85	2.53	6.02	3.88	10.01	2.41	3.45	1.79	4.39	0.93
	(68.48)	(37.76)	(130.95)	(42.24)	(27.13)	(15.11)	(42.74)	(10.09)	(24.08)	(15.78)	(39.30)	(9.27)	(14.01)	(7.34)	(15.57)	(3.67)

Table 13: Concentrations of BTEX at sampling site in units of ppbv and µg/m^3 (in parenthesis) :Hotspots (active sampling)

Compound	Petrol Station (n=8)				Airport (n=6)				Roadside (n=11)				Industrial (n=13)			
	Avg	Min	Max	SD	Avg	Min	Max	SD	Avg	Min	Max	SD	Avg	Min	Max	SD
Benzene	3.37 (10.68)	1.14 (3.61)	5.99 (18.98)	1.82 (5.77)	0.63 (2.00)	0.09 (0.29)	0.89 (2.82)	0.28 (0.89)	5.06 (16.03)	2.08 (6.59)	10.38 (32.89)	3.22 (10.20)	3.10 (9.82)	2.45 (7.76)	3.60 (11.41)	0.70 (2.22)
Toluene	8.39 (31.36)	4.21 (15.73)	14.62 (54.64)	4.32 (16.15)	2.11 (7.89)	1.21 (4.52)	2.51 (9.38)	0.48 (1.79)	27.98 (104.57)	17.29 (64.62)	43.79 (163.66)	13.79 (51.54)	2.24 (8.37)	1.04 (3.89)	5.32 (19.88)	1.26 (4.71)
Ethylbenzene	1.21 (5.21)	0.75 (3.23)	1.74 (7.49)	0.35 (1.51)	0.52 (2.24)	0.28 (1.21)	0.59 (2.54)	0.12 (0.52)	4.64 (19.98)	1.46 (6.29)	9.97 (42.93)	3.01 (12.96)	0.62 (2.67)	0.40 (1.72)	1.10 (4.74)	0.23 (0.99)
m,p-Xylene	1.32 (5.68)	0.43 (1.85)	2.72 (11.71)	0.90 (3.88)	0.15 (0.65)	0.07 (0.30)	0.28 (1.21)	0.07 (0.30)	8.79 (37.85)	2.49 (10.72)	20.45 (88.06)	6.51 (28.03)	0.86 (3.70)	0.11 (0.47)	3.29 (14.17)	0.38 (1.64)
o-Xylene	4.68 (20.15)	3.89 (16.75)	5.79 (24.93)	0.72 (3.10)	2.91 (12.53)	1.69 (7.28)	3.43 (14.77)	0.62 (2.67)	3.41 (14.68)	1.20 (5.17)	6.84 (29.45)	2.75 (11.84)	1.75 (7.54)	1.43 (6.16)	2.39 (10.29)	0.83 (3.57)
Total BTEX	18.97 (73.08)	10.42 (41.18)	30.86 (117.76)	8.11 (30.41)	6.32 (25.30)	3.34 (13.59)	7.70 (30.72)	1.57 (6.17)	49.88 (193.11)	24.52 (93.39)	91.43 (356.99)	29.28 (114.57)	8.57 (32.10)	5.43 (20.00)	15.70 (60.49)	3.40 (13.13)

4.3 Correlation analysis

Correlation analysis was run on the BTEX database obtained from the continuous monitoring sites at UKMKL, UMT, UMS and PPBF (Tables 14-17). The results showed that the pair-wise BTEX were strongly correlated ($r \geq 0.7$) at UKMKL site (Table 14). This site is located near roadside and thus, influence of the transportation sources was pronounced. At the urban site, transportation was recognized as a predominant source of BTEX. At UMS, a sub-urban site, toluene and m,p-xylene showed a strong correlation (Table 15). The evaporation from the solvent use from the nearby sources might cause to the contribution of this pair in ambient air. However, the variables were poorly correlated at PPBF site.

Table 14: Correlation matrix for BTEX at UKMKL (continuous monitoring)

Variables	Benzene	Toluene	Ethylbenzene	m,p-Xylene	o-Xylene
Benzene	**1**				
Toluene	**0.871**	**1**			
Ethylbenzene	**0.665**	**0.694**	**1**		
m,p-Xylene	**0.866**	**0.847**	**0.832**	**1**	
o-Xylene	**0.878**	**0.845**	**0.817**	**0.978**	**1**

Values in bold are significant with $p < 0.01$

Table 15: Correlation matrix for BTEX at UMT (continuous monitoring)

Variables	Benzene	Toluene	Ethylbenzene	m,p-Xylene	o-Xylene
Benzene	**1**				
Toluene	**0.262**	**1**			
Ethylbenzene	**0.090**	**0.111**	**1**		
m,p-Xylene	**0.402**	**0.367**	**0.216**	**1**	
o-Xylene	**0.288**	**0.257**	**0.140**	**0.453**	**1**

Values in bold are significant with $p < 0.01$

Table 16: Correlation matrix for BTEX at UMS (continuous monitoring)

Variables	Benzene	Toluene	Ethylbenzene	m,p-Xylene	o-Xylene
Benzene	**1**				
Toluene	**0.535**	**1**			
Ethylbenzene	**0.233**	**0.327**	**1**		
m,p-Xylene	**0.498**	**0.732**	**0.543**	**1**	
o-Xylene	**0.552**	**0.328**	**0.295**	**0.336**	**1**

Values in bold are significant with $p < 0.01$

Table 17: Correlation matrix for BTEX at PPBF (continuous monitoring)

Variables	Benzene	Toluene	Ethylbenzene	m,p-Xylene	o-Xylene
Benzene	**1**				
Toluene	**0.171**	**1**			
Ethylbenzene	-0.063	**0.128**	**1**		
m,p-Xylene	**0.265**	**0.296**	**0.558**	**1**	
o-Xylene	**0.138**	**0.417**	**0.489**	**0.642**	**1**

Values in bold are significant with $p < 0.01$

4.4 Source apportionment of BTEX using ratio analysis

The interspecies emission ratios can help to identify the dominant sources as well as to distinguish the aged and fresh emission of BTEX. Table 18 represents the ratios of T/B, (mp-X)/B, o-X/B, and (mp-X)/E for UKMKL, UMT, UMS and PPBF sites. The ratio of toluene to benzene (T/B) at UKMKL, an urban site, was 3.26 with a range of 1.74 to 17.28 while at UMT, a suburban site, the mean ratio was 4.51 with a range of 0.19 to 66.49. At UMS, a sub-urban site, the mean ratio of T/B was 3.29 ranging from 0.06 to 36.0 while at the background site (PPBF), the mean ratio value was 5.41 ranging from 0.09 to 251.50. The T/B value can be used to evaluate the influence of traffic related emission at a specific region. As reported in the literatures, the T/B ratios were 2.9 and 3.4 in Paris, France; 2.2 in Copenhagen, Denmark; 2.4 in Cairo, Egypt; 2.0 in Bari, Italy; 1.3 to 3.0 in Windsor, Canada; 1.8 to 2.54 in Delhi, India and 2.36 to 3.16 in Texas, USA. The range of the ratio (T/B) was 2.2 to 4.4 or >2 reported at the sites with heavy traffic influence. However, the high value of the T/B ratio might be caused by the emission from industries. Industries as point sources emit a large amount of toluene. The aging effect can also be an important consideration of BTEX with varied T/B ratio values. The decomposition or oxidation of BTEX mainly depends on the OH radical in the ambient air which is usually formed from the photolysis of ozone in presence of water vapour and sunlight. The oxidation rate of toluene is faster in ambient air than benzene. Thus, a complex heterogeneous reaction plays an important role in the variation of T/B ratios.

Table 18: Ratios of T/B, (mp-X)/B, o-X/B, and (mp-X)/E for UKM KL, UMT, UMS and PPBF sites

Sites	BTEX	Valid N	Mean	Median	Minimum	Maximum	Std.Dev.
UKMKL	T/B	3081	3.26	3.05	1.74	17.28	1.02
	mp-X/B	3081	1.56	1.44	0.02	7.64	0.58
	o-X/B	3081	0.67	0.62	0.05	3.06	0.25
	mp-X/E	3081	2.73	2.68	0.04	144.50	2.94
UMT	T/B	1406	4.51	2.48	0.19	66.49	6.30
	mp-X/B	1358	1.44	1.24	0.04	14.17	1.09
	o-X/B	1275	1.01	0.83	0.07	24.65	1.13
	mp-X/E	1290	1.81	1.47	0.03	10.36	1.03
UMS	T/B	460	3.29	2.75	0.06	36.00	2.77
	mp-X/B	459	1.27	1.15	0.08	8.24	0.96
	o-X/B	369	0.59	0.44	0.08	4.64	0.54
	mp-X/E	459	1.27	1.15	0.08	8.24	0.96
PPBF	T/B	1033	5.41	3.05	0.09	251.50	12.02
	mp-X/B	1035	2.51	1.27	0.03	36.00	4.11
	o-X/B	1020	2.21	0.87	0.20	28.00	3.69
	mp-X/E	954	6.33	3.75	0.11	60.00	6.97
Petrol station	T/B	-	2.49	-	-	-	-
	mp-X/B	-	0.39	-	-	-	-
	o-X/B	-	1.39	-	-	-	-
	mp-X/E	-	1.09	-	-	-	-
Airport	T/B	-	3.35	-	-	-	-
	mp-X/B	-	0.24	-	-	-	-
	o-X/B	-	4.62	-	-	-	-
	mp-X/E	-	0.29	-	-	-	-
Roadside	T/B	-	5.53	-	-	-	-
	mp-X/B	-	1.74	-	-	-	-
	o-X/B	-	0.67	-	-	-	-
	mp-X/E	-	1.89	-	-	-	-
Industry	T/B	-	0.71	-	-	-	-
	mp-X/B	-	0.26	-	-	-	-
	o-X/B	-	0.55	-	-	-	-
	mp-X/E	-	1.33	-	-	-	-

The ratio of mp-X/E was estimated at the four fixed sites i.e. UKMKL, UMT, UMS and PPBF and the mean values were 2.73, 1.81, 1.27, and 6.33, respectively. The commonly reported ratios for the sites with high influence of traffic were between 2.8 and 3.6. However, the higher ratio might have been influenced by evaporative emission from point sources. At PPBF site, the BTEX concentration may have emitted from a non-traffic origin. Additionally, we have determined the pair-wise ratios of T/B, mp-X/B, o-X/B and mp-X/E for four hotspots sites i.e. petrol station, airport, roadside and industrial sites. The ratios at these hotspots can be helpful in evaluating the ratios of the pairs of BTEX determined at the urban, suburban and remote background locations. The T/B at the roadside (5.53) corresponds well to the ratio of this pair (3.26) at the urban site in UKMKL.

4.5 Composition of BTEX in Petrol and Diesel in Malaysia

The composition of BTEX in different types of petrol and diesel from available in the Malaysian market were analyzed to determine the percentage composition of BTEX. The results in Figure 15 shows the percentage of BTEX in Petrol-95 are dominated by xylene followed by toluene > benzene and ethylbenzene. The percentage of benzene in Petrol-97 (27%) is higher compare to benzene in Petrol-95 (21%). On the other hand, the BTEX percentage in both Diesel-5 and Diesel 2M are dominated by xylene > ethylbenzene > toluene. There was no benzene detected in diesel. Ethylbenzene in Diesel-5 (33%) was found higher compared to Diesel 2M (28%). Results from this study indicates that the fraction of benzene in petrol collected from petrol stations in Malaysia is higher compared to the amount of benzene in petrol presented by the Ohio Department of Health [13]. The fraction of another potential carcinogenic molecule, ethylbenzene, was also recorded higher compared to ethylbenzene fraction in diesel recorded by Mariano et al. [12]. However, larger sample sizes would be needed to verify the results of this study.

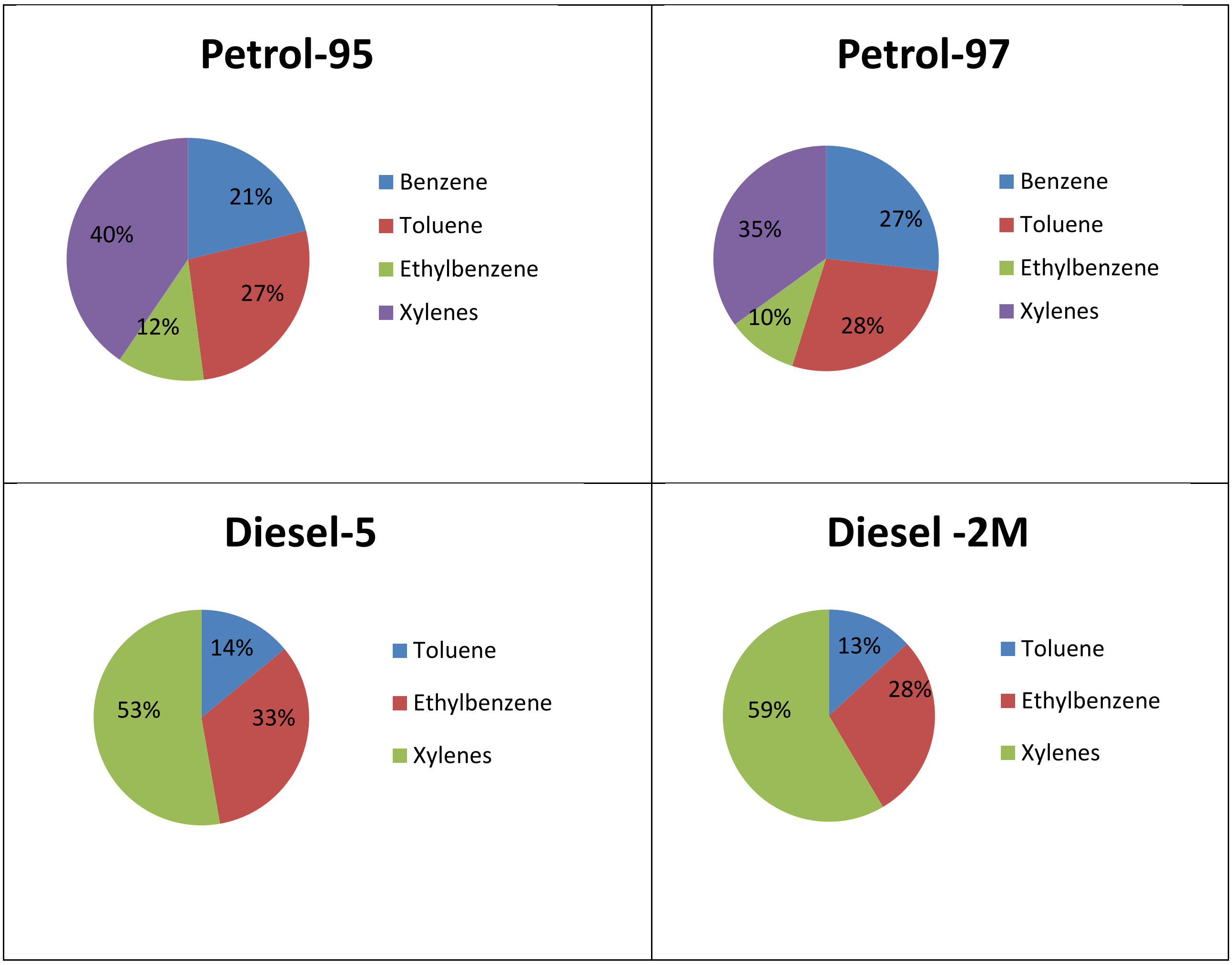


Figure 15: The composition of BTEX in petrol and diesel collected in a petrol station in

4.6 Validation of the sources or source regions

The travel pathways of the backward trajectories (BTs) over UMT, UKMKL, UMS and PPBF were plotted (Figure 16). The BTs were calculated using HYSPLIT 4.9 model. A release height of about 500 m for 72 h back trajectories with 6 h intervals was chosen to identify the origin of the air masses at the receptor point of interest in this study. Trajectory start time was chosen as 00:00 (UTC).

The period of the BT runs were from 6/1/2017 to 15/3/2017 over UKM-KL, from 8/2/2017 to 15/3/2017 over UMT, from 18/1/2017 to 8/3/2017 over PPBF and the month of February over UMS. The BTs at all sampling sites mainly originated from the north-east (NE) direction. The origin of air mass shown via clustered back trajectories at all sampling stations do not indicate any long distance influence to the concentration of BTEX in the study areas. Thus, the concentration of BTEX determined in this study is considered to be predominantly influenced by local sources.

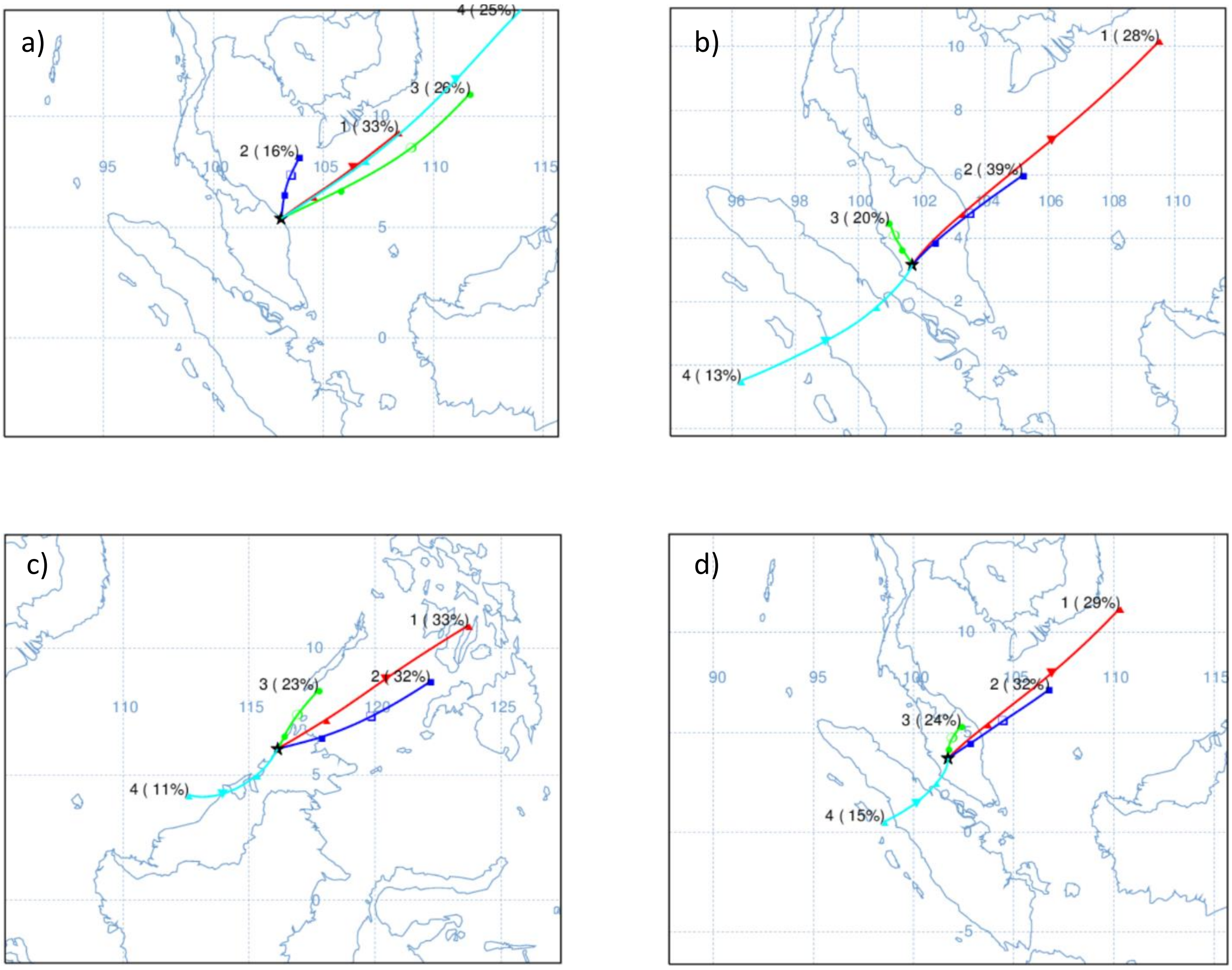


Figure 16: Backward trajectories transport of air masses in the boundary layer during continuous BTEX measurement over a) UMT b) UKMKL, c) UMS and d) PPBF

4.7 Effect of BTEX Exposure

4.7.1 Non-carcinogenic Health Risk Assessment

The results of HQ for BTEX showed that the values were <1 for benzene, m,p-xylene and o-xylene at all sites (Table 19). The HQ of BTEX was estimated for the samples collected at the hotspots sites in petrol station, roadside, airport and the industrial areas. The HQ values were 0.5116, 0.0204, 0.0191, 0.3562 and 0.1381 for benzene, toluene, ethylbenzene, m,p-xylene and o-xylene, respectively at the roadside. The results of HQ at roadside is comparable to that at UKMKL site. Therefore, it clearly shows that the BTEX at urban sites still were lower than the hazardous limit (HQ<1).

Table 19:Value of CDI and HQ determined at different sampling stations

CDI (mg/kg/day)								
BTEX	**UKMKL**	**UMT**	**UMS**	**PPBF**	**Petrol**	**Airport**	**Roadside**	**Industry**
Benzene	0.0017	0.0004	0.0006	0.0002	0.0029	0.0005	0.0044	0.0027
Toluene	0.0061	0.0022	0.0018	0.0008	0.0086	0.0022	0.0285	0.0023
Ethylbenzene	0.0012	0.0005	0.0004	0.0001	0.0014	0.0006	0.0055	0.0007
mp-Xylene	0.0032	0.0007	0.0008	0.0003	0.0015	0.0002	0.0103	0.0010
O-Xylene	0.0014	0.0005	0.0004	0.0003	0.0055	0.0034	0.0040	0.0021
HQ								
BTEX	**UKMKL**	**UMT**	**UMS**	**PPBF**	**Petrol**	**Airport**	**Roadside**	**Industry**
Benzene	0.1979	0.0520	0.0718	0.0220	0.3409	0.0638	0.5116	0.3134
Toluene	0.0043	0.0016	0.0013	0.0005	0.0061	0.0015	0.0204	0.0016
Ethylbenzene	0.0043	0.0016	0.0014	0.0003	0.0050	0.0021	0.0191	0.0025
mp-Xylene	0.1089	0.0236	0.0277	0.0120	0.0534	0.0061	0.3562	0.0348
O-Xylene	0.0467	0.0159	0.0129	0.0104	0.1896	0.1179	0.1381	0.0709

4.7.2 Carcinogenic Health Risk Assessment

The estimated LTCR of BTEX at the four sites are reported in Table 20. The LTCR was estimated for benzene and ethylbenzene due to their carcinogenic effect on human health. As per the proposed Guidelines for Carcinogen Risk Assessment by US EPA 2005 [30], benzene is characterized as a known human carcinogen, category A, for all routes of exposure based upon convincing evidence from studies on humans as well as supporting evidence from animal studies. International Agency for Research on Cancer (IARC) also listed benzene as carcinogen to human (group 1). Ethylbenzene was classified as group 2B by IARC (possibly carcinogenic to human health). Toluene and xylenes were included in group 3 as not classifiable. From the results shown in Table 20, the concentration of benzene can pose carcinogenic impact on about 1 to 4 person in 100,000 populations at UKMKL, roadside, petrol station and industrial site. Thus, the LTCR values of benzene at the urban roadside, petrol station and industrial site exceeded the US EPA recommended guideline in which the acceptable level is 1 in 1 million people [17]. On the other hand, the LTCR of benzene was in the range of acceptable level at background PPBF site. The LTCR of ethylbenzene showed acceptable level at UMS, PPBF and UMT sites. However, LTCR level was exceeded at UKMKL, petrol station and roadside.

Table 20: Life-time cancer risk (LTCR) of BTEX at UKM KL, UMT, UMS and PPBF sites

	CDI (mg/kg/day)							
BTEX	**UKMKL**	**UMT**	**UMS**	**PPBF**	**Petrol**	**Airport**	**Roadside**	**Industry**
Benzene	0.000580058	0.000152499	0.000210505	6.45548E-05	0.000999197	0.000187115	0.00149973	0.000918737
Ethylbenzene	0.000422881	0.000157177	0.000138465	3.36808E-05	0.000487436	0.000209569	0.001869283	0.000249799
	LTCR							
	UKMKL	**UMT**	**UMS**	**PPBF**	**Petrol**	**Airport**	**Roadside**	**Industry**
Benzene	1.59×10^{-5}	4.16×10^{-6}	5.75×10^{-6}	1.77×10^{-6}	2.73×10^{-5}	5.11×10^{-6}	4.10×10^{-5}	2.51×10^{-5}
Ethylbenzene	1.63×10^{-6}	6.05×10^{-7}	5.33×10^{-7}	1.30×10^{-7}	1.88×10^{-6}	8.07×10^{-7}	7.20×10^{-6}	9.62×10^{-7}

5. Suggestion for BTEX Standard

This study suggests the adoption of annual average of 5 µg/m^3 for benzene concentration to represent BTEX standard for Malaysia. Benzene is the most toxic and carcinogenic compound among BTEX. The concentrations of other BTEX compounds usually have good correlations with benzene at sites with anthropogenic sources (discussion in Section 4.3).

The value of 5 µg/m^3 for 1-year average has been adopted by many countries in the world including Korea, India and European Union. This value is higher compare to the standard suggested by Thailand (1.7 µg/m^3) and Japan (3 µg/m^3) but lower compare with the standard suggested by United Kingdom (16 µg/m^3 or 5 ppb), Vietnam (10 µg/m^3), Nepal (20 µg/m^3) and Australia (Queensland) (10 µg/m^3) (Table 3).

Based on this study, benzene concentration of 5 µg/m^3 has been exceeded in hotspot areas such as road side, petrol station and petrochemical industries. This value can be used as a warning or indicator for us to reduce the amount of BTEX emission from motor vehicles and industrial activities. Based on the calculation of LTCR, the concentration above 0.5 µg/m^3 has exceed the limit of 1 x10^{-6} as suggested by US EPA (Table 21). This is estimated based on the fact that long term exposure of benzene can contribute to cancer risk. Nevertheless, the exposure of 5 µg/m^3 does not exceed the limit of HQ (HQ<1) as suggested by US EPA health index. Based on the effect of benzene exposure to human health, the standard based on yearly average is reasonable to be implemented in Malaysia as a standard or first interim standard.

Table 21: HQ and LCR calculation based on different concentration of benzene

Benzene Concentration (µg/m^3)	RfD (mg/kg/day)	CDI (mg/kg/day)	HQ
5	0.00855	0.001364384	0.160
4		0.001091507	0.128
3		0.00081863	0.096
2		0.000545753	0.064
1		0.000272877	0.032
0.5		0.000136438	0.016
Benzene Concentration (µg/m^3)	**SF (mg/kg/day)$^{-1}$**	**CDI (mg/kg/day)**	**LTCR**
5	0.0273	0.000467789	1.27706E-05
4		0.000374231	1.02165E-05
3		0.000280673	7.66238E-06
2		0.000187115	5.10825E-06
1		9.35577E-05	2.55413E-06
0.5		4.67789E-05	1.27706E-06

6. Suggestion for Mitigation Procedures and Action Plan for reducing BTEX

Since BTEX has the potential to effect human health, mitigation procedures need to be implemented to reduce the BTEX exposure to human being. Among the mitigation procedures that can be implemented are:-

- Continuous monitoring of BTEX concentration at the hotspots areas such as roadside, petrol station, airport and industrial areas.
- Reduce number of motor vehicles on the roads especially in the city centre
- Minimise BTEX emissions from vehicle exhausts by improved design and regular monitoring of engine settings.
- Minimise exposure of BTEX from motor vehicles and industries by creating buffer zone between roadsides and industrial areas from the residential areas.
- Reduce exposure at petrol filling stations as far as possible by following best practices in location, design and extraction.
- Develop and implement policies and legislation to reduce or remove BTEX from consumer products.
- Promote the use of alternative solvents in industrial processes
- Raise public awareness regarding sources of exposure to BTEX especially benzene and risk mitigation measures.

The action plan should include these mitigation procedures which includes components of monitoring, reduction and prevention. The suggested action plan is shown in Table 22.

Table 22: Suggested action plan for reducing BTEX in ambient air

Category	Activity	Output
Monitoring	Continuous monitoring of BTEX at hotspots	Identify high risk areas and predominant sources
	Set-up standard for Benzene	Assessment of intensity and frequency of non-compliance
Reduction	Reduce number of motor vehicle (in tandem with other action plans related to reduction of motor vehicle numbers on road)	Reduction of emission source
	Disseminate information to public and encourage best practices during refuelling of vehicle.	
	Incentive to industry for reducing emissions via increased production, reducing fugitive emission or use of emission control technology (covers mobile sources, point sources and fugitive emission during storage and transport)	
	Creation of buffer zone	Minimize exposure to public
Prevention	Environmental Assessment Report needs to include information on potential benzene emission risk across all industries. High risk industries/manufacturers (e.g. petrol stations) should be identified and requested to monitor and report ambient benzene concentration at proposed development site	Minimize risk of ambient concentrations of benzene exceeding standard at locations that already has concentrations close to or above suggested ambient standard for benzene.

7. Conclusion

The preliminary results showed that the average concentration of benzene from continuous monitoring records at UKMKL has exceed the guideline proposed by the European Union for 1-year average (5 µg/m^3). Measurement at selected potential BTEX hotspots show that the average concentration of BTEX follow the sequence of roadside > petrol station > industrial site > airport. Motor vehicles are expected to contribute to high concentration of BTEX in ambient air. This is clearly seen from the higher concentration of BTEX particularly benzene during peak hours in Kuala Lumpur city centre and BTEX ratios analysis. The concentration of BTEX recorded in Kuala Lumpur was found to be lower compared to BTEX concentration recorded in Kalkota and Delhi, India; Pearl River Delta, China; Algiers, Algeria. The BTEX concentration in Kuala Lumpur is however, higher compared to cities in developed countries such as Hong Kong, United States of America and Japan.

Based on the HQ results, it can be concluded that BTEX at urban sites can pose larger hazardous effect on human health compared to the sub-urban and background sites. The concentration of benzene recorded in this study is estimated to have carcinogenic impact on about 1 to 4 person in 100,000 at UKMKL, roadside, petrol station and industrial site. The LTCR values of benzene at the urban and two sub-urban sites exceeded the US EPA recommended guideline in which the acceptable level is 1 in 1 million people.

Due to the high concentration of BTEX in ambient air particularly due to the traffic emission in the city centre, this study suggests the government to take several proactive measures to reduce emission from motor vehicles to ambient air. The ambient air standard for benzene needs to be implemented. This study suggests the Department of Environment Malaysia to adopt European standard of 5 µg/m^3 for 1-year average as the standard for benzene in Malaysia. This value is recommended based on the comparison of benzene standard in countries within this region and the effect of BTEX, especially benzene to human health.

Appendix 1: Gantt Chart for this project

No	**Activity**	**2016**								**2017**									
		May	**Jun**	**Jul**	**Aug**	**Sep**	**Oct**	**Nov**	**Dec**	**Jan**	**Feb**	Mar	**Apr**	**May**	**Jun**	**Jul**	**Aug**	**Sep**	**Oct**
1	Literature review	■	■	■	■	■	■	■	■	■	■	■	■	■	■	■	■	■	■
3	Coordination of field	■	■	■	■														
4	Active sampling				■	■	■	■	■	■									
7	Continuous monitoring								■	■									
8	Data analysis				■	■	■	■	■	■	■	■	■	■	■	■			
2	Inception Report	■	■	A															
6	Interim Report 1				■	■	■	B											
9	Interim Report 2								■	■	■	C							
10	Report Draft												■	■	■	■	D		
11	Final Report																	■	E

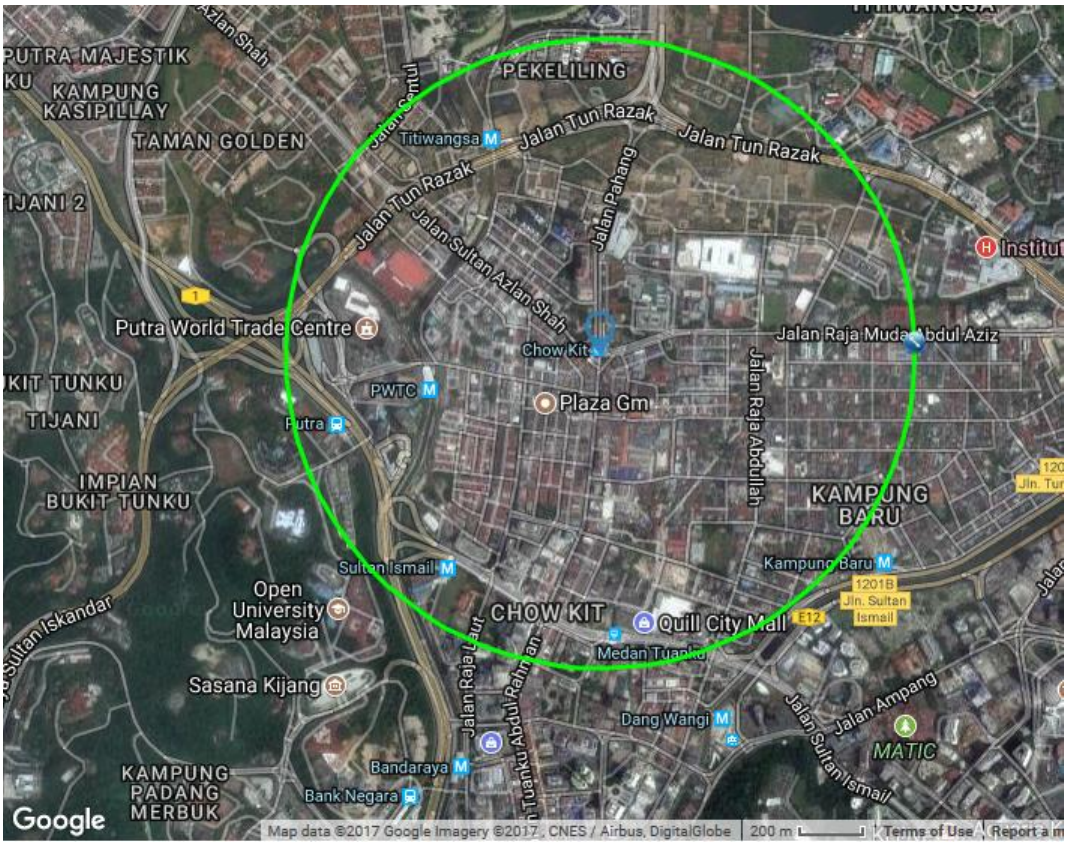
A, B, C, D and E are expected deadlines for report submission

Appendix 2: UKMP and consultant team members

No	Name	Position	Expertise
1	Mr Mohd Zin Abd Rahman	Project Director	Management
2	Ms Intan Suraya Razak	Project Manager	Environmental Science
3	Mr Azri Azuar	Project Executive	Civil Engineering
4	Prof Dr Mohd Talib Latif	Lead Consultant	Air Quality
5	Dr Mohd Shahrul Mohd Nadzir	Consultant	Air Quality
6	Dr Md Firoz Khan	Consultant	Air Quality
7	Mr Haris Hafizal Abd Hamid	Consultant	Air Quality
8	Dr Fatimah PK Ahamad	Research Assistant	Air Quality
9	Mrs Puteri Nurafidah Hosaini	Research Assistant	Air Quality

Appendix 3 : Active sampling

Jalan Chow Kit, Kuala Lumpur



https://www.freemaptools.com/radius-around-point.htm

KIPC, Kertih, Terengganu: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Map data ©2017 Google Imagery ©2017 CNES / Airbus, DigitalGlobe
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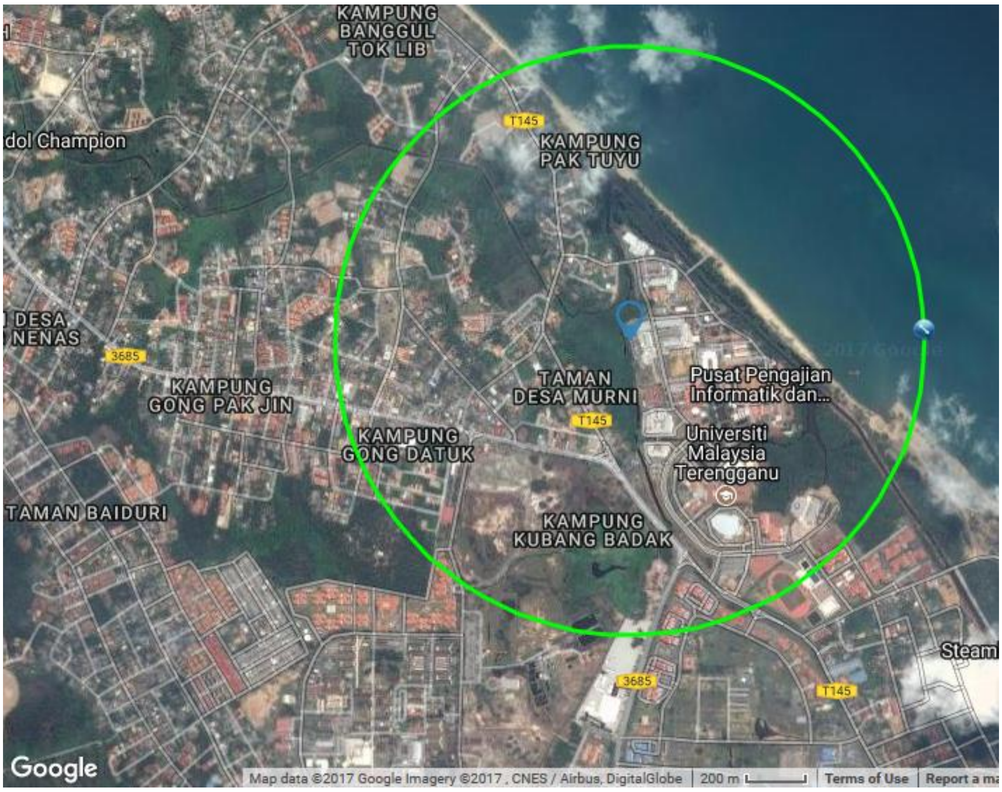
Appendix 4: Continuous monitoring

Universiti Malaysia Terengganu (UMT), Kuala Terengganu



https://www.freemaptools.com/radius-around-point.htm

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