DETERMINATION OF BTEX CONCENTRATION USING PASSIVE SAMPLERS IN A HEAVY TRAFFIC AREA OF JAKARTA, INDONESIA

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ABSTRACT: Benzene, Toluene, Ethylbenzene, and Xylene (BTEX) are hazardous air pollutants mostly emitted from transportation activity. Seeing the high motorized vehicle population and fuel specification, BTEX pollution potentially occurs in Indonesian urban areas. However, there is very limited study on atmospheric BTEX. This paper is a preliminary investigation of urban ambient BTEX in Jakarta, Indonesia, aimed to report the application of passive measurement method and initial analysis of ambient BTEX concentration in a busy-traffic area. Passive method is chosen as it is practical and suitable for measuring average ambient BTEX concentration in longer duration. The passive method utilizes activated charcoal as the adsorbent. Passive samplers of 1-week sampling duration were used to collect ambient VOC samples. Ten sets of sample were collected during July 2018 to February 2019. The 95% CI weekly average concentrations found for benzene, toluene, ethylbenzene, and xylene were 21.9 (± 4.550); 73.4 (± 25.419); 23.9 (±9.888); and 17.1 (±6.510) µg/m³, respectively. BTEX are not listed yet in NAAQS. The values reported here were to explore the potential of estimate provision to be used in a further study on long-term effect assessment. Hence, the results are not to be compared with the standard value. Nevertheless, these findings highlight the importance of HC monitoring in urban area of Indonesia, which can be used as input for setting the future ambient air quality standard and further studies to investigate how BTEX are emitted and estimated as well as to determine their control strategy in air quality management.

Keywords: Hazardous air pollutants, Ozone precursor, Toxic emission, Urban pollution, UDARA

1. INTRODUCTION

Benzene, Toluene, Ethylbenzene, and Xylene volatile organic (BTEX) are compounds categorized as Hazardous Air Pollutants (HAPs) by the US Environmental Protection Agency [1]. Benzene has been recognized as carcinogenic compounds to human, thus classified into group 1 by the International Agency for Research on Cancer (IARC) Monographs [2]. Toluene, ethylbenzene, and xylene are irritants to human and very reactive as they are precursors for secondary air pollutants, such as ozone, carbonyl, and aldehyde [3-5]

As the compounds potentially present serious effects when exposed to human and environment, BTEX have been studied in various countries. Previous studies measured BTEX compounds in the ambient air to find spatial and temporal variability [6-9]. Other studies predicted their exposure, potential health effects, and ozone potential formation [10-16]. These studies pointed out that hydrocarbon (HC), including BTEX, are harmful chemicals in urban environment. However, HC are rarely monitored in Indonesia. Only Jakarta routinely measures HC as part of its ambient air quality monitoring. The measurements are not yet specific to the toxic HC, but rather as CH_4 (methane) and NMHC (non-methane HC). Specific HC speciation measurements such as BTEX are even rare.

The most common source of BTEX in urban area reported in previous studies was transportation activity [17-19], particularly during incomplete combustion. Therefore, it is expected that BTEX concentration will be elevated in congested traffic areas. BTEX emissions, hence would be higher on vehicles with idle condition or on very low speeds [20]

Considering the population size and still limited provision of public transport, Indonesia is highly relying on private vehicles. In total, 15 million passenger cars and 113 million motorcycles were registered in 2017 [21]. Some of vehicles run on fuels with Research Octane Number (RON) less than 91. These might become the potential sources of benzene. The maximum permissible content in fuel sold in Indonesia for benzene is 5% and aromatic is 50% [22]. The levels are higher than the values recommended by World-Wide Fuel Chartered (2.5% benzene and 40% aromatic) [23]. The implication to that is the possibly high occurrence of BTEX in Indonesian urban atmosphere.

This paper reported the preliminary results of ambient air pollution measurements as part of Urban Hybrid Model for Air Pollution Exposure Assessment (UDARA) project, which aimed for investigating air pollution and health effects in Indonesia. UDARA focuses on long-term (annual) pollutant exposure estimates. For this reason, passive sampler method was used.

2. METHODS

2.1 Study Location

Sampling point was located near Bunderan Hotel Indonesia (Bunderan HI), Central Jakarta, Indonesia. This area is utilized as a commercial and business area with heavy traffic day and night. There are also many intersections equipped with stop signals that make vehicles move slower or being in idle condition.



Fig.1 Sampling point location (source: OpenStreetMap).

2.2 BTEX Sampling

Air sampling was carried out passively using adsorption method. Sets of SKC 575 - 001, 3.5 cm round badge passive sampler with 350 mg activated charcoal adsorbent were exposed for seven days to obtain weekly average BTEX concentrations. Ten data sets were collected from July 2018 to February 2019. On each passive sampler installation and removal, the date and time were recorded, so that sampling duration could be calculated.

After the sampling period, exposed samplers were immediately transported to the laboratory with cooler box and blue ice to maintain temperature of 4° C and then stored at 4° C for 1 to 4 week before analyses.

2.3 Laboratory Analysis

VOCs absorbed on the activated charcoal from exposed sampler was desorbed using 2 ml of methylene chloride and then sonicated for 30 minutes for maximizing the desorption process of BTEX from charcoal pores. To separate the liquid from charcoal particles, syringe filter was used.

BTEX concentration analysis was carried out using gas chromatography-mass spectroscopy (GC-MS), Agilent 7890a, at Laboratorium Terpadu Politeknik Kesehatan, Bandung. Agilent HP-5MS 5% Phenyl Methyl Silox (29.81 m x 250 μ m x 0.25 μ m) column was utilized to separate BTEX compounds. To obtain valid concentration measurement, GC-MS was calibrated by MISA Group 17 liquid as the certified reference material. Field and laboratory blanks were provided to detect any contamination.

BTEX on liquid concentration obtained from the GC-MS analysis was then converted to ambient concentration by Eq. (1).

$$C = \frac{SW \times 24.45 \times 10^6}{DE \times MW \times SR \times MIN \times PT}$$
(1)

Where:

C = concentration (ppm)

SW = B/T/E/X mass from GC analysis (mg)

DE = desorption efficiency

MW = B/T/E/X molecular weight

SR = sampling rate

- MIN = sampling duration (minute)
- PT = conversion factor due to temperature and pressure

DE and SR were obtained from passive sampler user guide. Concentrations are expressed in normal temperature and pressure (NTP at 25°C, 1 atm). As the field temperature and pressure do not always at NTP, a conversion factor (PT) to normalized field temperature and pressure was needed and calculated using Equation (2) below:

$$PT = \left(\frac{T_1}{T_2}\right)^{1.5} \times \left(\frac{P_2}{P_1}\right) \tag{2}$$

Where:

$$\Gamma_1$$
 = sampling site temperature (K)

 $T_2 = 298 K$

 P_1 = sampling site pressure (mmHg)

 $P_2 = 760 \text{ mmHg}$

В	Т	E	m+p - X	o-X	Logation	Dof
	(ug/m ³)					Kei
29.2	45.4	13.1	32.9	29.2	Kolkata, India	[24]
3.4	16.2	3.6	12.7	4.5	Tehran, Iran	[25]
					Ho Chi Minh,	[26]
6.0 - 53.0	18.0 - 170.0	3.0 - 24.0	5.0 - 59.0	2.0-21.0	Vietnam	
0.8 - 2.7	0.9 – 7.6	0.2 –2.3	0.8 - 8.5	0.3 – 3.3	Bari, Italy	[27]
21.9	73.4	23.9	17.1 (+6.510)		Jakarta Indonasia	This study
(±4.550)	(±25.419)	(±9.888)	$17.1(\pm 0.310)$		Jakana, muonesia	This study

Table 1 Average concentration of BTEX

3. RESULT AND DISCUSSION

3.1 BTEX Concentration

The 95% CI weekly average benzene, toluene, ethylbenzene, and xylene concentrations were 21.9 (±4.550); 73.4 (±25.419); 23.9 (±9.888); and 17.1 (±6.510) $\mu g/m^3$, respectively. Similar to measurement in Hanoi [28] toluene was the most abundant compound compared to benzene, ethylbenzene, and xylene in the sampling locations (Fig.2 and Table 1), This conformed traffic emissions as BTEX sources. Many factors could affect the BTEX ambient composition, e.g. emissions quality that is affected by fuel specification and its aromatic contents; traffic variations; emissions from other sources; as well as the dynamic of atmospheric chemistry processes at the location during days and nights of the measurement.



Benzene
■Toluene
Settylbenzene
Xylene

Fig.2 Average concentration of BTEX

While various methods were used in measuring BTEX in other studies, the concentrations reported in this study were within the ranges of concentration measured in other East Asian megacities, such as Kolkata, India [24] and Ho Chi Minh City, Vietnam [25]. Other contrasts were shown from measurement in Tehran, Iran [26] and Bari, Italy [27], see Table 1.

Table 2 Climate and Population Characteristics

	Toloowto	Ho Chi	Kolka	Teh-	Dari
Annual Ave. Temp. (°C) ^(a)	Jakarta 28	<u>Minh</u> 27	-ta 24.5	14.5	<u>Barı</u> 11
Annual Ave Precip. (a)	154.6	155.7	144.6	18.3	47.3
Pop* ×1000 (b)	10517	13595	7100	8667	614
Vehicle * ×1000	16837 (c)	3300 (d)	911 (e)	1400 (f)	80 (g)
Vehicle per 1000 pop.	1601	243	128	162	130

*at the time of the study

Source: a [29], b[30], c[31], d[25], e[32], f[33],g[27]

Jakarta, Kolkata, Ho Chi Minh City and Teheran have comparable population of over 5 million with typical transportation mode of private vehicles, consisting of 2-wheel and 4-wheel vehicles, and some proportion of 3-wheelers particularly in Kolkata and in limited areas of Jakarta. Population of Bari was relatively small compared to the other cities; however, the ratios of vehicle number to 1000 population were similar, except for Jakarta. Jakarta has exceptionally high proportion of vehicle to population, compared to the other cities, where on average there were 1.6 registered motor vehicles per person. This might be caused by one person could own both a motorcycle and a car for his convenience (Table 2). The quality of exhaust emissions are affected by engine technology. During this study, according to the Regulation of Minister of Environment number 141/2003 and number 23/2012, Indonesia used EURO II and EURO III vehicle technology. In India, vehicle engine mandatory specification was Bharat Stage II technology (equivalent to EURO II)[24]. In Vietnam, when the study was conducted, most of vehicles were motorcycles (with pre-Euro technology), and the rests were EURO II and EURO III passenger cars[25]. In contrast, the latest EURO technology was presumed to be adopted in Italy. For Tehran [26], however, there is no information on engine technology or fuel quality available.

In terms of measurement methods, Kolkata and Ho Chi Minh City also used BTEX activated carbon tubes. However, air samples were drew by active sampling, which allows shorter sampling duration while needed. In Bari, similar to our study, measurement was conducted in 1-week duration, using Radiello type passive sampler. Real-time measurement was done in Tehran, where BTEX was obtained by continuous VOC monitoring device model VOC71M-PID. There were 152 BTEX samples taken in Kolkata during dry season from December 2003 to February 2005. The study in Ho Chi Minh City resulted in 284 hourly samples, providing ample data to be analyzed by PCA to identify the sources.

Nevertheless, all previous studies have

decreased in winter [25]. It is suggested that tropical climate in Indonesia and local traffic condition might influence the levels of BTEX concentrations (see Table 2). Although no detailed information on the fuel quality, other factors of vehicle emissions such as traffic condition, engine and vehicle air pollution control technology were more or less similar to Ho Chi Minh City and Kolkata. The similarities might explain the concentration levels within the same range of magnitude of the measurements in these cities.

3.2 Concentration Variation

Meteorological condition and human activity near the sampling site had been found to affect temporal variation of BTEX concentration [9], [16]. However, no distinctive pattern or trend was observed in this study. This might be due characteristics of the sampling site, which is a busy traffic city center with mixed areas of residential, commercial, office and leisure activities for almost 24-hour during weekdays and weekends. Hence the urban activities might result in relatively constant emissions throughout the day.

Other potential causes of temporal variation are the effects of photochemical reactions, particularly for TEX, which are more potent ozone precursors compared to benzene. However, photochemical reaction would be seen as diurnal variation. This, therefore, could not be observed in this study



Fig.3 Temporal concentration variation of BTEX

corroborated our results that the main sources of BTEX were traffic/vehicular emissions. PCA results in Vietnam showed significant correlation of BTEX with gasoline fueled vehicles [25]. In Bari it was found that BTEX concentrations were affected by traffic activity and distance from the roadside [27]. In addition to traffic, in Iran industry also was found as the significant sources of BTEX [26].

Study in Iran indicated that BTEX levels were affected by climate, where BTEX concentrations

because passive sampling produces accumulative concentration for the duration of sampling (1week). However, there were tendencies that concentrations were higher in the wet season than in the dry season. This might be suggested as due to less solar radiation because cloud covers were higher during the wet season, hindering the photochemical processes, hence less consumption of the precursors.

Fig.3 shows the temporal variations of TEX during the sampling period with some similarities

to each other, while benzene had slightly different pattern. The lowest and the highest concentrations of benzene were measured in September 11th to 16th 2018 and December 30th 2018 to Jan 6th 2019, respectively. While the maximum concentrations of toluene, ethylbenzene, and xylene were found in January 13th to 19th 2019, August 19th to 26th 2018, December 3rd to 11th 2018, respectively.

The minimum concentrations of TEX were found in the sampling from December 30th, 2018 to Jan 6th, 2019. The low concentrations were assumed to relate to lower emissions as this was a new year holiday period, when business activities in this area were lower and Jakarta inhabitants tended to choose to spend the holiday time out of town. However, for the unknown reason this was not shown by benzene, which might be due to limited data.

To examine the role of local meteorology to concentration variation, correlation analysis was done to BTEX concentrations with meteorological factors (temperature, solar radiation and relative humidity). The meteorological data were obtained from automatic monitoring of local environmental agency (DLH DKI Jakarta) at the same location. The data from the local agency were averaged to the same sampling duration, resulting in 10-week pairs of data. Due to the limited number of data, non-parametric correlation analysis was used. The analysis results are shown in Table 3.

Humidity had been found to be significantly positively correlated (α =0.05) with ethylbenzene and xylene. There were also positive associations with benzene and toluene, but they were not significant. All four parameters were negatively correlated with temperature, indicating the tendency of lower concentration when the temperature was higher. This could explain the higher concentrations observed during the wet season. Solar radiation was negatively correlated with TEX, but not with benzene. This also supports the aforementioned argument on the effects of photochemical reactions on TEX and their roles as the reactants of the chemical processes.

Table3Spearman-rhocorrelationofmeteorological condition and BTEX

	В	Т	E	Х
Temp	-0.167	-0.025	-0.343	-0.268
p – value	0.667	0.949	0.366	0.486
Solar				
Rad	0.367	-0.267	-0.467	-0.400
p – value	0.332	0.488	0.205	0.286
Humidity	0.467	0.400	0.733	0.700
p – value	0.205	0.286	0.025	0.036

Reactivity in photochemical reactions might play roles in the correlation results; benzene is less reactive, whereas TEX have high reactivity to form photochemical oxidants [34]. A previous study found that the higher the relative humidity, the lower the rate of photochemical reaction, thus BTEX consumption as precursors in reaction was lower, hence the photochemical concentrations were higher [35]. The data in this paper were gathered during the wet season, supporting the suggestion that relative humidity, which is higher in the wet season, might inhibit photochemical reactions that consumed ethylbenzene and xylene as ozone precursors.

Temperature and solar radiation are usually strongly correlated, as solar radiation is the main source of temperature. On the other hand, ambient temperature is the resultant of energy dissipated from solar radiation, thermal radiation from the Earth surface and the reflected energy from air molecules to the ground. Usually temperature will speed up photochemical reaction, but it should involve photolysis reaction that requires energy from solar radiation. This explains the results where temperature appeared to affect negatively to BTEX, resulting in lower concentrations. On the other hand, solar radiation would encourage photochemical radiation, hence reduce TEX concentrations, but has no similar effect to benzene as it is less reactive compare to the other three compounds. However, more data might be needed to confirm this.

3.3 Interspecies Correlation

To further investigate the above results, correlation analysis was also applied to each component of BTEX. The results show that benzene was positively but not significantly correlated with TEX, while TEX had significant strong correlations to each other (α = 0.01 for X to T and E; and α =0.05 for T to E), as can be seen in Table 4. This could be explained by the difference in chemical properties of benzene compared to TEX (Table 5).

Table 4 S	pearman-rho	correlation	among	BTEX
			0	

	В	Т	Е	Х
В		0.267	0.233	0.350
p - value		0.488	0.546	0.356
Т			0.733	0.833
p - value			0.025	0.005
Ē				0.900
p - value				0.001
Х				
p - value				

Table 5 shows that toluene, ethylbenzene, and xylene have more similarity in properties compared to benzene. These facts are coherent with the correlation results in Table 4.

Table 5 Chemicals properties of BTEX

Properties	В	Т	E	Х
Molecular Weight (g/mol) ^a	78	92	106	106
Vapor Pressure (Pa) ^a	12672.2	3769.3	1276.7	1074.0
Half-life (h) ^a	141.8	57.1	47.0	23.3
Maximum Incremental Reactivity (MIR) (grO ₃ /gr VOC) ^b	0.42	2.7	2.7	6.5

Source : ^a [24], ^b [34]

Table 5 shows that benzene's vapour pressure and half-life are much higher than TEX [24]. Benzene reactivity also is much lower than TEX [34]. As the analysis found no correlation between benzene with TEX, it is suggested that there might be other sources of benzene near/around the sampling point or that benzene reacts much slower that TEX in photochemical reactions.

The second possibility is more likely, where BTEX could come from the same sources, but then underwent different chemical reaction paths, reaction rates and response differently to the meteorological factors in the atmosphere, as were shown in the inter correlation results in Table 4.

4. CONCLUSIONS

This initial investigation shows the occurrence of BTEX in considerable levels in the ambient air of Jakarta. The 95% weekly-average CI of benzene toluene ethylbenzene and xylene (n=10) were 21.9 (\pm 4.550) µg/m³, 73.4 (\pm 25.419) µg/m³, 23.9 (\pm 9.888) µg/m³, and 17.1 (\pm 6.510) µg/m³, respectively. The average concentration levels were of the similar ranges found in other megacities in Asian countries such as India and Vietnam. The findings raise the importance of BTEX monitoring and other toxic pollutants in urban area of Indonesia.

This result can be used for future study to investigate how BTEX is emitted and estimated, and for evaluating its effect towards the receptors as well as to determine control strategy in air quality management.

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