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Exposure levels and health risk assessment of ambient BTX at urban and rural environments of a terai region of northern India*

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Abstract

Benzene, toluene and xylene (BTX) belong to an important group of aromatic volatile organic compounds (VOCs) that are usually emitted from various sources. BTX play a vital role in the tropospheric chemistry as well as pose health hazard to human beings. Thus, an investigation of ambient benzene, toluene and xylene (BTX) was conducted at urban and rural sites of Gorakhpur for a span of one year in order to ascertain the contamination levels. The sampling of BTX was performed by using a low-flow SKC Model 220 sampling pump equipped with activated coconut shell charcoal tubes with a flow rate of 250 ml/min for 20–24 h. The analysis was in accordance with NIOSH method 1501. The efficiency of pump was checked weekly using regulated rotameters with an accuracy of $\pm 1\%$. The samples were extracted with CS_2 with occasional agitation and analyzed by GC-FID. The total BTX concentration ranged from $3.4 \mu\text{g m}^{-3}$ to $45.4 \mu\text{g m}^{-3}$ with mean value $30.95 \mu\text{g m}^{-3}$ and median $24.8 \mu\text{g m}^{-3}$. The mean concentration of total BTX was maximum during winter ($39.3 \mu\text{g m}^{-3}$), followed by summer ($28.4 \mu\text{g m}^{-3}$) and monsoon season ($25.1 \mu\text{g m}^{-3}$). The mean concentration of BTX at urban site ($11.8 \mu\text{g m}^{-3}$) was higher than that at rural site ($8.8 \mu\text{g m}^{-3}$). At both the sites, T/B and X/B ratios were highest in monsoon and lowest in winters. Toluene against benzene plot shows R^2 value of 0.96 and 0.49 at urban and rural sites respectively. Higher R^2 value at urban site clearly indicates similar sources of emission for benzene and toluene. At both the sites, the estimated integrated lifetime cancer risk (ILTCR) for benzene exceeded the threshold value of $1\text{E-}06$ whereas the individual hazard quotients (HQ) for BTX did not exceed unity at any of the sites.

Keywords

BTX; Urban & rural environments; ILTCR; HQ; Terai zone

1. Introduction

Exposure to air pollution has become a major concern for human health. Volatile organic compounds (VOCs) constitute an important class of organic contaminants whose vapor

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pressure is at least 10 Pa at 20 °C (Wang & Zhao, 2008). Because of their high volatility, these compounds have the ability to easily diffuse in the environment (Khoder, 2007). VOCs when released into the atmosphere, not only affect the human health but also cause various environmental complications such as ozone layer depletion, deterioration of crops and vegetation (Srivastava et al., 2005), as well as formation of secondary organic aerosol (SOA) and ground level ozone.

Exposure to VOCs can be deleterious to human health, causing headache, nausea, weakness, lack of concentration, loss of appetite and fatigue. At higher concentrations, it can even cause cancer (Cometto-Muniz et al., 2004; Bernstein et al., 2008). Benzene, toluene and xylene isomers (BTX) are an important group of VOCs which are found ubiquitously in air (Ilgen et al., 2001a; b; Guo et al., 2003). In urban areas, the potential sources of ambient BTX are vehicular emissions due to incomplete combustion of gasoline and diesel in motor engines (de Blas et al., 2012; Moolla et al., 2015), emissions from petrochemical industries and evaporative loss of solvents during various industrial processes (Gallego et al., 2008; de Blas et al., 2012). Since, the monocyclic aromatic compounds such as benzene, toluene and xylene are often used as additives to increase the octane quality of unleaded fuels; the major contribution of BTX is undoubtedly from the transportation sector (Sergio et al., 2012). Similarly in rural areas, the emissions from diesel gen-sets, irrigation pumps, incineration activities and biomass burning may contribute to ambient levels of BTX (Fan et al., 2014).

Monitoring of BTX is necessary, because even at trace concentrations these compounds can cause adverse effects to human health (Badjagbo et al., 2010). Benzene has been classified as a group1 carcinogen by the International Agency for Research on cancer (IARC, 2002). Its exposure has been linked with occurrence of a number of blood diseases such as aplastic anemia and a variety of cancers including the acute myeloid leukemia (Duarte-Davidson et al., 2001; Buczynska et al., 2009). According to W.H.O., benzene concentration of $1.7 \mu\text{g m}^{-3}$ is likely to cause leukemia to 1 in 100,000 individuals (World Health Organisation, 2010). Toluene is a known teratogen and causes fetal abnormalities (Donald et al., 1991). Xylene acts as a skin sensitizer and can cause dryness, rupture and blistering of skin. At higher concentrations, toluene and xylene can cause weakening of nervous system, kidneys and liver (USEPA, 2004).

In India, the majority of BTX exposure studies have been conducted mainly in metropolitan cities like Delhi (Khairwal et al., 2006; Hoque et al., 2008; Singh et al., 2012; Kumar et al., 2014), Mumbai (Srivastava et al., 2006), Kolkata (Chattopadhyay et al., 1996; Mukherjee et al., 2003; Majumdar et al., 2008, 2011) and Agra (Singla et al., 2012; Chaudhary & Kumar, 2012; Bhardwaj et al., 2017). However, most of these studies focus only on the exposure levels observed in urban areas. Therefore, regardless of numerous harmful effects of BTX, there has been a shortage of studies which investigate the exposure levels observed in both rural and urban areas, especially in this specific region of north India. Monitoring of BTX in India has become more important because this country is in developing state and in most parts of the country, the dwellings are closely located with adjacent roads having moderate to heavy traffic and frequent incineration activities. Therefore the purpose of the present study is to determine the concentration of gaseous pollutants, with focus on the ambient

exposure levels of benzene, toluene and xylene at Gorakhpur - a terai region of northern India, so as to ascertain their probable health effects.

2. Materials and methods

2.1. Site description

Gorakhpur ($26^{\circ}45'32''\text{N}$ $83^{\circ}22'11''\text{E}$) is located in the foothills of the Shiwalik Himalayas, near the border of Nepal, in the terai region of eastern Uttar Pradesh, in northern India. It is connected to the capital city Lucknow through the National Highway (NH-28) and is located approximately 265 kms east of it. It covers a geographical area of almost 3483.8 Sq. Km with population of about 4,440,895 (Masih et al., 2017). The average temperature during winter season is about 18.6°C with a range of 3.5°C – 29.5°C and 71% humidity. During summers, the temperature ranges from 20.5°C to 48.5°C with an average of about 35.5°C and 82% humidity. Similarly in monsoon season the average temperature lies around 30.8°C with a range of 18.2°C – 43.8°C and 89% humidity (Masih et al., 2016).

2.2. Sampling and analysis

Air sampling was performed at urban (*Maitripuram, Bhichiya*) and rural site (*Haiderganj, Jungle Dhoosan*) simultaneously for a span of one year (Nov. 2014 to Oct. 2015). Fig. 1 shows the map of Gorakhpur pointing the sampling sites. Monitoring was performed for 20–24 h once a week in a scheduled manner and a number of 48 samples from each site and a total of 96 samples were collected from both the sites. BTX were sampled and analyzed using a methodology based on National Institute for Occupational Safety and Health (NIOSH) method 1501 (NIOSH, 1994; BIS, 2006). BTX were sampled by drawing air through activated coconut shell charcoal tubes (CSC, 8 mm $\times$ 110 mm, 600 mg) containing two sections (main section 400 mg, second section 200 mg) separated by a 2 mm urethane foam (SKC Inc.), using a low-flow SKC Model 220 sampling pump (SKC, USA) at the flow rate of 250 ml/min for 20–24 h. The air suction rate was verified every week using calibrated rotameters with an accuracy of $\pm 1\%$. Charcoal beds in the sorbent tubes were transferred to 2 ml vials and extracted by adding 1.0 ml of carbon disulphide (CS_2) with occasional agitation for 30 min. Sampled air was then analyzed with an HP 6890 gas chromatography/flame ionization detector (NIOSH, 1994). The cold trap operating temperature was -30°C and was raised to 250°C for 3 min. The carrier gas was nitrogen and an Agilent 19091J-413/E column (30 m $\times$ 0.32 mm i.d. $\times$ 0.25 mm) was used. The column temperature was held at 40°C for 5 min, after which it was raised to 100°C for 5 min and then increased to 200°C , at which it was held for 2 min. For each compound, two ions (one target and one qualifier) were monitored. Compounds were identified based on their relative retention times and ion ratios. Identified compounds were quantified using internal calibration procedure, with five levels of calibration as follows: 0.1; 1; 5; 25; 100 $\mu\text{g/ml}$ in CS_2 in all solutions.

3. Results and discussion

3.1. BTX concentrations at urban and rural sites

The mean concentration of total BTX at Gorakhpur was $30.95 \mu\text{g m}^{-3}$ for both the sites (urban & rural) together. Table 1 shows that the average concentrations of benzene, toluene

and xylene were 12.1, 22.1 and 1.3 $\mu\text{g m}^{-3}$ at urban site, whereas 7.4, 17.9 and 1.1 $\mu\text{g m}^{-3}$ at the rural site. The results of Student's t-test and Mann Whitney U test indicate that at urban site, the benzene and toluene levels were significantly higher ($p < 0.05$) than that at rural site. However, the xylene levels at both the sites were not significantly different. It was also observed that the mean concentration of BTX at urban site (11.8 $\mu\text{g m}^{-3}$) was higher than that at rural site (8.8 $\mu\text{g m}^{-3}$). Due to proximity of the urban site to the high traffic roads, this may be attributed to the vehicular emissions (Som et al., 2007; Gallego et al., 2008) as well as other combustion activities like use of gen-sets which were used as an alternative source of electricity. Concerning the BTX mass distribution, the study shows that at both the sites, toluene had the highest contribution followed by benzene, having 68% & 28% at rural site and 62% & 34% at urban site respectively. Toluene and benzene represent about 96% of the total BTX at both the sites (Fig. 2). Toluene and xylenes are largely used as solvents and, therefore, their existence may come from sources other than traffic too. In addition, photochemical degradation of toluene and xylenes is faster than that of benzene. (Kerchich and Kerbach, 2012).

3.2. Seasonal variation and interspecies ratios

The climate of Gorakhpur is generally classified into three seasons, winter (November–February), summer (March–June) and monsoon (July–October). Fig. 3 depicts the seasonal BTX pattern in ambient air at urban as well as rural sites of Gorakhpur. As evident from the figure, the trend of seasonal variation of BTX was almost similar at both the sites with maximum BTX concentration during winters followed by summer and monsoon seasons. At urban site, during winter, summer and monsoon seasons, the concentrations of benzene were 18.4, 9.7 & 8.2 $\mu\text{g m}^{-3}$ respectively. Similarly toluene and xylene levels were 24.3, 22.4 & 19.6 $\mu\text{g m}^{-3}$ and 1.7, 1.2 & 1.0 $\mu\text{g m}^{-3}$ respectively. On the other hand, at rural site, the concentrations of benzene during winter, summer and monsoon seasons were 12.7, 5.2 & 4.3 $\mu\text{g m}^{-3}$ respectively. This is supported by the fact that BTX undergoes photochemical depletion in air mainly due to reactions with OH radicals (Miller et al., 2011). During summers, high temperature and solar radiation leads to increased concentration of OH radicals invoking further atmospheric degradation of BTX, resulting in their lower ambient concentration (Kuntasal et al., 2005; Singla et al., 2012). However, higher BTX levels are generally observed during winters due to reduced photochemical degradation. Lowest concentration of BTX during monsoon season can be justified by washing out of atmospheric BTX through rain showers and thunderstorms (Masih et al., 2016). The seasonal ratios for mean concentration of BTX from winter to summer (W/S) and winter to monsoon (W/M) were 1.3 & 1.5 and 1.5 & 1.6 at urban and rural sites respectively, indicating significant seasonal variations of BTX.

The individual BTX compounds undergo photochemical degradation with varying reaction rates, thus the BTX interspecies ratios are often used to provide information regarding the characteristics of the air as well as to compare the emission sources (Hsieh et al., 2011). Table 2 illustrates the observed ratios of toluene to benzene (T/B) and xylene to benzene (X/B) at both the sites during summer, monsoon and winter seasons respectively. Higher values of these ratios suggest emissions from a nearby source while lower values indicate photochemical aging of the air mass. It was observed that at each site, the T/B ratios were

higher than the X/B ratios. This agrees with the fact, that xylene undergoes faster degradation than toluene (Zalel et al., 2008). At both the sites *i.e.* urban and rural, the maximum T/B ratio was found during monsoon season (2.38 and 3.85) followed by summer (2.32 and 3.33) and winter seasons (1.31 and 1.61) indicating that increase in concentration from monsoon to winter and summer to winter is maximum for benzene as compared to toluene and xylene. This fact is also supported by the X/B ratio which was found to be lowest in winter season (0.09 and 0.11) and increased during summer (0.12 and 0.19) and monsoon season (0.13 and 0.21) at urban and rural sites respectively. Fig. 4 shows the plot of toluene against benzene with R^2 value 0.96 and 0.49 at urban and rural sites respectively. At urban site, a remarkably linear plot was observed suggesting similar sources for emission of benzene and toluene as a result of gasoline combustion activities like vehicular emission and use of diesel gen-sets due to erratic supply of electricity. However at rural site, the plot was considerably non-linear suggesting different sources for emission of benzene and toluene such as use of diesel gen-sets for irrigation purpose as well as incineration activities including biomass burning for cooking and heating purpose.

3.3. Cancer and non cancer risk assessment

Benzene is classified as carcinogenic substance, while toluene, m,p-xylene and o-xylene are classified as non-carcinogenic but hazardous to health substances. Thus, risk assessment has been performed in regard to estimate the brunt of hazards on human health by these mono-aromatic pollutants (Zhang et al., 2012). In the present study, the integrated lifetime cancer risk (ILTCR) and non-cancer hazard as a result of BTX exposure was calculated (Majumdar et al., 2011). The integrated life time cancer risk (ILTCR) is given by the equation:

$$ILTCR = E_L (\text{mg kg}^{-1} \text{day}^{-1}) \times CPF (\text{mg kg}^{-1} \text{day}^{-1})$$

Where, E_L is the effective lifetime exposure and CPF is the carcinogenic potency factor. The inhalation cancer potency factor for benzene was $2.73 \times 10^{-2} \text{ mg kg}^{-1} \text{day}^{-1}$ as obtained from the Risk Assessment Information System (RAIS) (RAIS, 2010). The degree of cancer risk for an individual is determined by the ILTCR value. "A carcinogenic effect of considerable concern" can be recognized by an ILTCR value $> 10^{-6}$ however, any value 10^{-6} may be considered "acceptable" (Zhang et al., 2012). The non-cancer risk assessment can be carried out using the hazard quotient (HQ) which may be defined as the ratio of yearly average daily concentration (E_Y) and the reference concentration (RfC)

$$HQ = E_Y / \text{RfC}$$

Where the inhalation reference concentration (RfC) for the non-carcinogenic compounds toluene and xylene were 5 and 0.1 mg/m^3 respectively, as retrieved from the Integrated Risk Information System (IRIS) (USEPA-IRIS, 2003, 2005). For benzene, the non-cancer risk was estimated using the RfC value 0.03 (ATSDR, 2010). The detailed information concerning cancer and non-cancer risk assessment can be found in our previous paper (Masih et al., 2016).

Table 3 displays the ILTCR and HQ values obtained for an individual assuming 15 years residing time. At both the sites the ILTCR value for benzene exceeded the threshold limit of $1\text{E-}06$, suggesting probable cancer risk due to benzene exposure. The urban site ($7.3\text{E-}06$) had comparatively higher cancer risk than rural site ($4.8\text{E-}06$). Benzene also gave the highest non-cancer HQ followed by xylene and toluene. The individual HQs for BTX at both the sites were less than unity signifying no substantial risk of non-cancer health effects (Majumdar et al., 2011). At urban site, the HQ values for benzene ($3.9\text{E-}01$), toluene ($4.2\text{E-}03$) and xylene ($1.4\text{E-}02$) were higher than those found at rural site *i.e.* $2.5\text{E-}01$, $3.7\text{E-}03$ and $1.3\text{E-}02$ for B, T and X respectively. The present study was compared with other BTX studies performed in Algeria, Egypt, Spain and United States as depicted in Table 4. The table shows that most of the studies in general reported higher BTX levels in urban areas except the study conducted in Barcelona (Spain) where the BTX concentrations in rural areas were higher than those at urban sites (Gallego et al., 2008). It is also evident from the table that in the present study, the average concentration of benzene ($12.1\text{ }\mu\text{g m}^{-3}$) was lower than those reported in Egypt ($46.5\text{ }\mu\text{g m}^{-3}$) and United States ($18\text{ }\mu\text{g m}^{-3}$) while the average toluene level ($22.1\text{ }\mu\text{g m}^{-3}$) was lower than those found in Egypt ($110.2\text{ }\mu\text{g m}^{-3}$), Spain ($64.8\text{ }\mu\text{g m}^{-3}$) and United States ($48\text{ }\mu\text{g m}^{-3}$). Similarly, the mean concentration of xylene ($1.3\text{ }\mu\text{g m}^{-3}$) was lower than those reported in Egypt ($73.4\text{ }\mu\text{g m}^{-3}$), Spain ($47.6\text{ }\mu\text{g m}^{-3}$), United States ($28\text{ }\mu\text{g m}^{-3}$) as well as Algeria ($3.2\text{ }\mu\text{g m}^{-3}$). However at rural site, the average concentration of benzene ($7.4\text{ }\mu\text{g m}^{-3}$) was higher than those reported at Algeria ($6.5\text{ }\mu\text{g m}^{-3}$), Spain ($5.8\text{ }\mu\text{g m}^{-3}$), Egypt ($4.4\text{ }\mu\text{g m}^{-3}$) as well as United states ($<0.5\text{ }\mu\text{g m}^{-3}$) whereas the average toluene level ($17.9\text{ }\mu\text{g m}^{-3}$) was higher than those observed at Algeria ($13.7\text{ }\mu\text{g m}^{-3}$), Egypt ($6.4\text{ }\mu\text{g m}^{-3}$) and United States ($1.0\text{ }\mu\text{g m}^{-3}$). Though the mean concentration of xylene ($1.1\text{ }\mu\text{g m}^{-3}$) was only higher than that reported in United States ($0.5\text{ }\mu\text{g m}^{-3}$).

4. Conclusion

In order to study the BTX exposure levels and examine the related carcinogenic and non-carcinogenic risks for people residing at Gorakhpur, a monitoring study was conducted for a period of 1 year at two selected sites of Gorakhpur namely, Maitripuram (Urban Site) and Haiderganj (Rural Site). The mean levels of benzene, toluene and xylene were 12.1 , 22.1 and $1.3\text{ }\mu\text{g m}^{-3}$ at urban site whereas 7.4 , 17.9 and $1.1\text{ }\mu\text{g m}^{-3}$ at rural site respectively. At urban site, benzene and toluene levels were significantly higher ($p < 0.05$) than those at rural site. BTX levels followed a characteristic seasonal pattern. At both the sites, the concentration of BTX was highest in winters followed by summer and monsoon seasons. The interspecies ratios T/B and X/B also exhibited a seasonal variation. Both these ratios were highest in monsoon season, while lowest values were observed in winters. At urban site, the plot of toluene against benzene was substantially linear with R^2 value 0.96 , indicating similar sources for the emission of benzene and toluene. However at rural site, a considerably non-linear plot signifies diverse sources for their emission. At both the sites, the integrated lifetime cancer risk (ILTCR) for benzene was higher than the threshold value $1\text{E-}06$. However, the non-cancer hazard quotient (HQ) for BTX was less than unity.

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Fig. 1.
Map of Gorakhpur showing sampling sites.

Fig. 2.
Individual Contribution (%) of BTX at both sites.

Fig. 3.
Seasonal BTX concentrations (µg/m³) at urban and rural sites.

Fig. 4.
Plot of Toluene against Benzene at both the sites.

Table 1

Mean BTX levels ($\mu\text{g}/\text{m}^3$) at urban and rural sites of Gorakhpur.

VOC	Site		p-value	
	Urban	Rural	Student's <i>t</i> -test	Mann-Whitney U test
Benzene	12.1	7.4	0.021 *	0.036 *
Toluene	22.1	17.9	0.017 *	0.043 *
Xylene	1.3	1.1	0.315	0.309

* p-value < 0.05 indicating significant difference between sites in terms of VOC concentration.

Table 2

Average interspecies ratios (T/B and X/B) at urban and rural sites.

	Urban		Rural	
	T/B Ratio	X/B Ratio	T/B Ratio	X/B Ratio
Winter	1.31	0.09	1.61	0.11
Summer	2.32	0.12	3.33	0.19
Monsoon	2.38	0.13	3.85	0.21

Table 3

Estimate of non-cancer and cancer risk at urban and rural sites.

	Urban		Rural	
	HQ	ILTCR	HQ	ILTCR
Benzene	3.9E-01	7.3E-06	2.5E-01	4.8E-06
Toluene	4.2E-03		3.7E-03	
Xylene	1.4E-02		1.3E-02	

HQ: Hazard Quotient, ILTCR: Integrated Life Time Cancer Risk.

Table 4

Comparison of ambient BTX levels with other studies of world ($\mu\text{g}/\text{m}^3$).

Country	Benzene		Toluene		Xylene		Reference
	Urban	Rural	Urban	Rural	Urban	Rural	
United States	18.0	<0.5	48.0	1.0	28.0	0.5	Sexton & Westburg, 1984
Algiers	9.6	6.5	15.2	13.7	3.2	4.4	Kerchich and Kerbachi, 2012
Egypt	46.5	4.4	110.2	6.4	73.4	3.5	Khoder 2007
Spain	4.3	5.8	64.8	67.0	47.6	51.4	Gallego et al., 2008
India	12.1	7.4	22.1	17.9	1.3	1.1	Present Study