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ORIGINAL PAPER

Source characterization and health risks of BTEX in indoor/outdoor air during winters at a terai precinct of North India

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Abstract BTEX are the consistently found air contaminants in indoor and outdoor environments. In order to investigate the exposure levels of BTEX, the indoor and outdoor air was analyzed during winter season at homes located at four selected sites of Gorakhpur, Uttar Pradesh, India, which comprised residential, roadside, industrial and agricultural areas. BTEX were sampled with a low-flow pump (SKC model 220). Samples were extracted with CS_2 and the aromatic fraction was subjected to GC-FID. Mean indoor concentration of BTEX was highest at the agricultural (70.9 μg m^{-3}) followed by industrial (30.0 μg m^{-3}), roadside (17.5 μg m^{-3}) and residential site (11.8 μg m^{-3}). At outdoor locations, the mean BTEX levels were highest at the roadside (22.0 μg m^{-3}) followed by industrial (18.7 μg m^{-3}), agricultural (11.0 μg m^{-3}) and residential site (9.1 μg m^{-3}). The I/O ratios were greater than 1 at all the sites except roadside site, where I/O ratios for toluene, ethylbenzene and xylene were less than unity. Poor correlation between indoor and outdoor levels at each site further indicated the dominance of indoor sources. Factor analysis followed by one-way analysis of variance depicts that the presence of BTEX compounds at all the sites indicate a mixture of vehicular and combustion activities. For benzene, the ILTCR values exceeded the safe levels, whereas ethylbenzene was nearby to the recommended level 1×10^{-6}. The HQ values were above unity for agricultural (indoors) and industrial (outdoors) as an exception to all the other sites which indicted the value below unity.

Keywords BTEX · Indoor/outdoor · Source characterization · ILTCR · HQ · I/O ratio

Introduction

Urbanization and industrialization are essential for the economic growth of a nation; however, it always costs environmental degradation. The existing social lifestyle has caused degradation of air, which is the basic requirement of life (Fiore et al. 2019; Filippini et al. 2020). In recent decades, the indoor levels of volatile organic compounds (VOCs) have gained more importance because of the fact that people spend more than 80% of their time in indoor environment (Pandit et al. 2001; Son et al. 2003; Lawrence et al. 2004; Masih et al. 2010). The indoor levels of most VOCs are generally higher than that found outside (USEPA 1990; Guo et al. 2003). Benzene, toluene, ethylbenzene and isomers of xylene (BTEX) are the most abundant VOCs found in indoor and outdoor environment (Ilgen et al. 2001a,b; Guo et al. 2003). Sources of BTEX in

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indoor air can be diverse, among which the major ones include the household activities like cleaning, smoking, cooking, heating and burning. Besides these, building materials, fabrications and furnishings used, furniture foam, carpets, paints, glues and cosmetics also prove to be major sources of BTEX (Pandit et al. 2001; Schneider et al. 2001; Gallego et al. 2008). In addition to these, vehicular and industrial emissions from outdoor air may also contribute to indoor levels of BTEX (Pandit et al. 2001; Batterman et al. 2007). In ambient air, the major sources of BTEX include industrial and vehicular emissions, incomplete combustion of fuels, petrol refineries and evaporative loss during usage of solvents (Gallego et al. 2008; de Blas et al. 2012). Since toluene is more abundant in gasoline, its concentration in air is usually higher (Gallego et al. 2008; Masih et al. 2016). Exposure to VOCs is often associated with a number of health related problems such as irritation in mucous membrane, discomfort, headache, nausea, fatigue, lack of concentration and poor work efficiency (Wolkoff et al. 2006; Bernstein et al. 2008). They not only prove to be a major threat to human health, but also cause several other environmental problems like ozone layer depletion, ground level ozone formation, global warming (Som et al. 2007; Masih et al. 2016) as well as damage to crops and vegetation (Srivastava et al. 2005).

BTEX are toxic at levels found in urban air (Bono et al. 2003). Among BTEX, benzene is a proved carcinogen (USEPA 1998; IARC 2002; Dehghani et al. 2017; Abtahi et al. 2018), while ethylbenzene is a proven human carcinogen (IARC 2000). Formation of tumors and leukemia have been associated with exposure to higher levels of benzene (Duarte-Davidson et al. 2001; Hinwood et al. 2007). Toluene has been identified as human teratogen. Exposure to higher levels of BTEX can cause serious damage to the nervous system, respiratory system, liver and kidneys (ATSDR 2003; Hinwood et al. 2007). Even at concentration levels of microgram per cubic meter, BTEX can pose a significant threat to human health (Ueno et al. 2001; Badjagbo et al. 2010). Hence, monitoring of BTEX is important in outdoor as well as indoor air (Schneider et al. 2001; Pilidis et al. 2005; Khoder, 2007).

A number of studies related to outdoor levels of BTEX have been conducted in India (Hoquea et al. 2008; Saxena and Ghosh 2012; Masih et al. 2016; 2017); however, there is a shortage of comparative indoor/outdoor BTEX studies, especially in this terai region of India. Thus, the aim of present study is to describe the concentration of BTEX in indoor as well as outdoor air of homes sited at different localities of Gorakhpur region. In this study, the indoor–outdoor levels were compared at each site, in order to observe the contribution of outdoor emissions on indoor levels of BTEX. Moreover, a detailed description of BTEX emission levels due to different household activities and surrounding environment has also been given.

Materials and methods

Description of sampling sites

Gorakhpur (26°45′32″N 83°22′11″E) is situated on the basin of rivers Rapti and Rohini, exhibiting a bowl like geographical shape surrounded by other small rivers and streams from three sides. Specifically, it is located in the foothills of the Shiwalik Himalayas, near the border of Nepal in the north Indian terai region of eastern Uttar Pradesh. At present, the district of Gorakhpur is sited on the National Highway (NH-28), 265 kms east of the capital Lucknow having total population of approximately 4,440,895, and it covers a geographical area of about 3483.8 Sq. km (Masih et al. 2018). The sampling of indoor and outdoor air was performed at homes found at four different sites of Gorakhpur city, namely residential, roadside, industrial and agricultural areas. All these sites are approximately 5–10 kms apart from each other. On each site, sampling has been achieved at the nearest residence from source (ongoing activities) of that specific location to observe the actual effect. Certain activities related to that particular area were being performed within 1–10 m only of the sampling site. Information regarding the household activities of sampled homes and the lifestyle of occupants was explored with the help of interview-based questionnaires. The questionnaire included prompts to recognize the specific features of the site and the prevailing activities within the homes like cooking, smoking, painting of walls and roof, burning of mosquito coils and incense, etc. Information regarding the type of building, age and dimensions of the homes was also included. Location of all four sites is shown in a map in Fig. 1. Table 1 gives the details of sampling sites regarding the general characteristic features of homes, common indoor household activities as well as other outdoor activities that took place nearby. Taramandal was typically a residential area. Golghar was selected as a characteristic roadside area because it is situated besides a heavy traffic road. GIDA (Gorakhpur Industrial Development Area) was exclusively an industrial area because large numbers of industries are located in this area. Haiderganj was purely an agricultural/rural area.

Sampling and analysis

Sampling of air was accomplished during winter season in homes situated at the four previously mentioned sites for duration of four months (November 2015–February 2016). The indoor and outdoor air was simultaneously monitored 20–24 h at a height of 6 feet from ground to simulate the humans breathing zone, once a week in a scheduled manner. Thus, 32 samples of indoor and outdoor air (16 each) were collected from each site, and a total of 128 samples were collected from all the sites. BTEX were sampled and analyzed using a methodology based on National Institute for Occupational Safety and Health (NIOSH) method 1501 (NIOSH 1994; OSHA 2004; BIS 2006). BTEX were sampled by drawing air through activated coconut shell charcoal tubes (CSC, 8 mm × 110 mm, 600 mg) containing two sections (main sect. 400 mg and second sect. 200 mg) separated by a 2 mm urethane foam (SKC Inc.), using a low-flow SKC Model 220 sampling pump (SKC, Inc.,84, PA, USA) at the flow rate of 250 ml/min for 20–24 h. The air suction rate was verified every week using calibrated rotameter with an accuracy of ± 1%. The sample tubes wrapped in aluminum foil were put

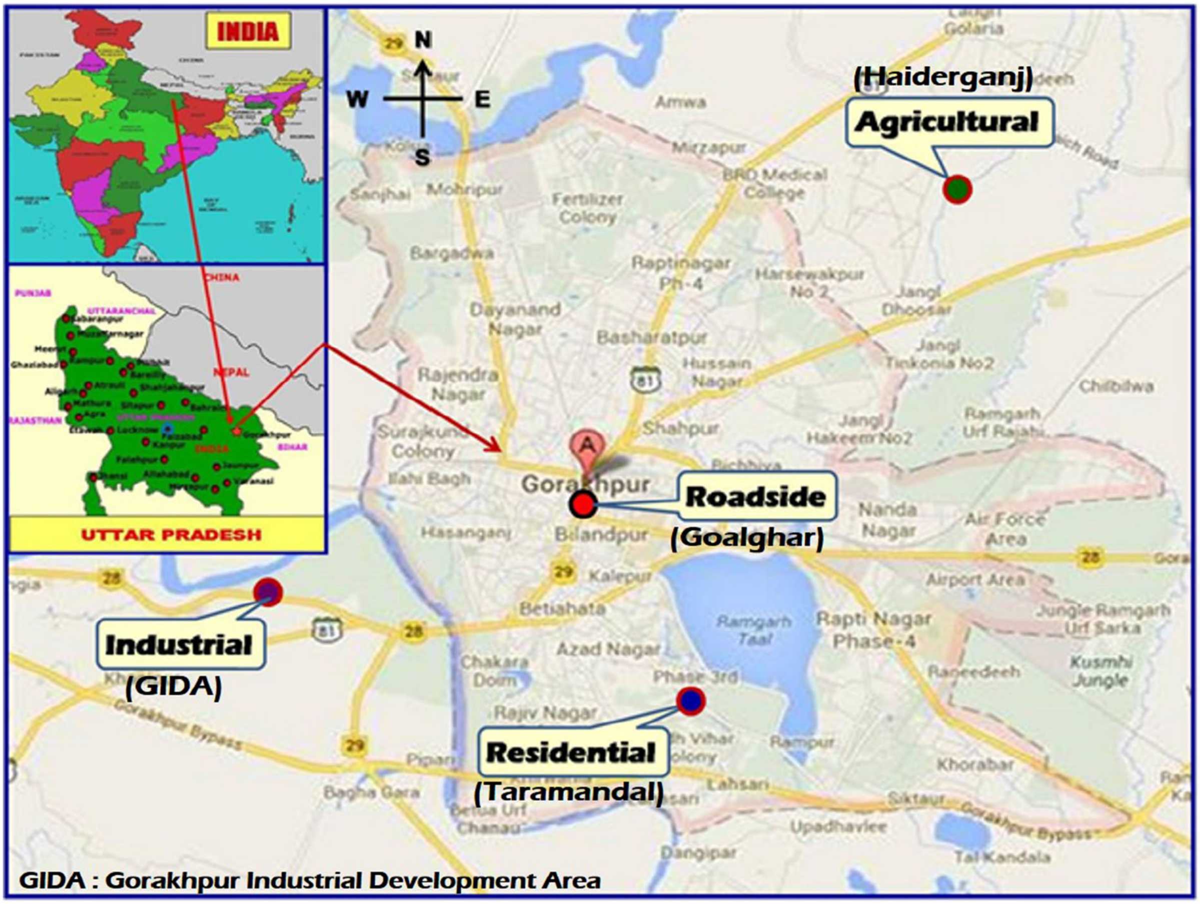

Fig. 1 Map of Gorakhpur showing sampling sites

Table 1 Description of monitoring sites and related activities at/nearby sampling sites

Sampling site [condition]	Home age and height	Living room area	Traffic load	Ventilation	Oil used for cooking	Type of fuel used for cooking	Smoking status (cigarette/bidi)	Other combustion activities	Use of diesel/kerosene gen-sets
Residential (RES) [medium population, homes made up of bricks, greenery around]	5 years and 15 ft	18–20 m^2	Almost no traffic	Use of exhaust fan, cross-ventilation and windows	Ghee, refined and mustard oil	LPG. and mixed fuel*	Less prevalent	Incense, dhoop and mosquito coil	Few of the homes using diesel/kerosene gen-sets
Roadside (RDS) [high population, homes made of bricks, shops and homes along the road, no greenery]	17 years and 15 ft	15–18 m^2	Heavy to moderate traffic with LMV	Cross-ventilation with windows	Dalda, refined and mustard oil	LPG. and mixed fuel*	Less prevalent	Incense, dhoop and mosquito coil	Few of the homes
occasionally using diesel/kerosene gen-sets									
Industrial (IND) [low population, homes made of bricks surrounded by various factories of alcohol, textile, mustard oil and pharmaceuticals]	12 years and 15 ft	10–12 m^2	Usually calm,	occasionally very less traffic with LMV and HMV	Cross-ventilation with windows	Dalda and mustard oil	LPG, mixed fuel* and cow dung	More prevalent	Incense, dhoop and mosquito coil
Few of the homes occasionally using diesel/kerosene gen-sets									
Agricultural (AGR) [low population, homes made of brick, mud, khaprail, grasses and bamboos, lots of greenery around]	More than 50 years and 15 ft	10 m^2	Almost no traffic	Poor ventilation	Dalda and mustard oil	LPG, mixed fuel* and cow dung	Highly prevalent	Incense, dhoop, ghee/kerosene oil lamp	Few of the homes using diesel/kerosene gen-sets

*Mixed fuel: wood/coal/kerosene, LPG: liquefied petroleum gas, dalda and ghee: polysaturated hydrogenated form of oils used for cooking, bidi: thin hand-rolled cigarettes (tobacco wrapped in tendu or temburni leaf), dhoop: extruded form of incense into polythene bags that were tightly closed and stored in a box in a deep freezer at −5 °C until processed. The second section of tube was analyzed in order to detect breakthrough. Charcoal beds in the sorbent tubes were transferred to 2-ml vials and extracted by adding 1.0 ml of carbon disulfide (CS_2) with occasional agitation for 30 min. Sampled air was then analyzed with an HP 6890 gas chromatography/flame ionization detector. Blank runs were performed before each sample analyses. The uncertainties of the results were calculated, using the data of the calibration curves, such as: benzene 18%, toluene 10%, ethylbenzene 21%, m, p-xylene 8% and o-xylene 12% (US EPA 1988a, b). Detailed methodology can be found in our earlier paper Masih et al. (2016). Humidity, temperature and ventilation rate at indoors were measured with the help of Young Environment System (YES-206, 205 Canada) which are indoor air quality (IAQ) monitors based on non-dispersive infrared (NDIR) technique. Table 2 shows the mean temperature, humidity, ventilation rate, sample duration, sample number and fuel/material used for prayer in indoor and outdoor environment at residential, roadside, industrial and agricultural sites.

Quality control (QAQC)

Validity of procedure for the estimation of organic pollutants was verified using synthetic standard mixtures. The efficacy of the methodology was assessed through the analysis of standard reference material (SRM). HPLC grade chemicals (MERCK, Qualigens) and glassware of borosil/corning were used during analysis.

Source characterization

Statistical analysis of the achieved results was performed with the help of IBM SPSS (version 20.0). Factor analysis was used to characterize the sources of indoor and outdoor BTEX separately. The collected data were reduced to a small number of factors which account for most of the variance in the observed variables. Varimax rotation was used to maximize the sum of variances of the square loadings so that each variable may be associated with one and only one factor. Moreover, in order to interpret the results obtained from factor analysis, at each site, different indoor–outdoor activities were analyzed for their effect on the levels of BTEX. For each activity, the data was divided into different categories. These categories were then compared using one-way analysis of variance (ANOVA) at $p<0.05$ level.

Estimation of cancer and non-cancer health risks

Risk assessment is an exclusive, fastest and premium effective tool to assess harmful effects of chemicals/pollutants. Among BTEX compounds, it is well known that benzene (IARC group 1) has been verified to be carcinogenic to humans, while ethylbenzene (IARC group 2B) has been classified as a 'possible human carcinogen' (IARC 2002). Moreover, each of benzene, toluene, ethylbenzene and xylene is identified for their various non-carcinogenic health risks to human. Therefore, in the present study, the feasible cancer and non-cancer health risks were estimated with the help of mean exposure concentrations of the pollutants using several assumptions considering the

Table 2 Average levels of temperature, humidity and air change rate in indoor and outdoor environment at different sites

Site	Mean temp. (°C)		RH* (%)		Average ACH (h^{-1})	Sampling duration (hrs)	No. of samples	Fuel used of cooking/prayers
	In	Out	In	Out				
Residential	18.6	19.3	68.8	62.4	7.2	24	32	LPG/mixed/incense/dhoop
Roadside	16.8	17.8	61.7	58.7	5.9	24	32	LPG/mixed/incense/dhoop
Industrial	18.3	19.1	69.5	65.8	4.6	24	32	LPG/mixed/CDC/incense/dhoop
Agricultural	17.1	18.8	63.9	61.3	3.8	24	32	LPG/mixed/CDC/dhoop/ghee

RH*=relative humidity, ACH=air change rate, LPG=liquefied petroleum gas, mixed: wood/coal/kerosene, CDC=cow dung cakes mean values of human body weight and rate of inhalation (USEPA 1994; Guo et al. 2004). The cancer risk assessment was based on the USEPA methodology using the potency factors given by the Risk Assessment Information System (RAIS) (RAIS 2010). The integrated life time cancer risk (ILTCR) for a particular carcinogenic air pollutant can be easily evaluated as the product of the cancer potency factor (CPF) and the effective lifetime exposure (E_L). Detailed information about the estimation of cancer and non-cancer health risks can be found in our preceding studies (Masih et al. 2016, 2018).

Results

Concentration of BTEX in indoor and outdoor air

The statistical data for BTEX concentration in indoor and outdoor air of Gorakhpur are shown in Table 3. Benzene, toluene, ethylbenzene and xylene had median concentrations 27.8, 49.3, 6.6 and 6.6 µg m^{-3}, respectively, in indoor air, whereas 20.3, 28.0, 2.3 and 2.5 µg m^{-3}, respectively, in outdoor air. At each site, toluene and benzene levels were predominantly high at indoors as well as outdoors. Table 4 depicts the trend of indoor BTEX followed the order, *i.e.,* agricultural site>industrial site>roadside site>residential site. At outdoor locations, benzene was highest at industrial site followed by roadside, agricultural and residential sites, whereas toluene, ethylbenzene and xylene was found to be higher at roadside site followed by industrial site>agricultural site>residential site. Extensive use of biomass fuel (Sinha et al. 2006; Fan et al. 2014), frequent smoking and other household activities (Brajenović et al. 2015; Hazrati et al. 2016) may probably account for higher indoor BTEX levels at agricultural and industrial sites. At outdoor places, higher BTEX levels at roadside site may be attributed to enhanced vehicular emissions (Ho et al. 2009; de Blas et al. 2012), while at industrial site, this may be on account of various factories and manufacturing plants present in this area (Hsieh et al. 2006). Lowest BTEX levels were observed at residential area in indoor as well as outdoor air.

Figure 2 displays a comparison between indoor and outdoor BTEX levels at different sites. As expected, the indoor levels of BTEX were higher than outdoors, at all the sites except roadside. At the roadside site, except for benzene, outdoor levels of TEX were higher than the indoors, which is perhaps due to close proximity of road which carries a continuous traffic of both light and heavy motor vehicles. Figure 2 shows that the highest difference between indoor and outdoor levels was observed for benzene and toluene at agricultural site. This fact directly points toward some major indoor sources of benzene and toluene at this site which may possibly be due to use of biomass fuel for cooking, smoking and other household activities as mentioned earlier.

Discussion

Indoor–outdoor ratios and correlation

In order to study the influence of ambient BTEX levels on the indoor air, the ratio of indoor to outdoor concentration (I/O) were calculated at each site. Figure 3 shows the boxplots for I/O ratios at each site. The I/O data along with the Spearman correlation coefficients between indoor and outdoor BTEX levels obtained at each site are illustrated in Table 4. It is apparent from the table that at roadside site the I/O ratios were lower than 1, except for benzene which had I/O ratio close to unity. However, the correlation between indoor–outdoor levels at this site was quite low. At residential site also, the I/O ratios were close to unity. However, at industrial and agricultural sites, the I/O ratios were higher because the indoor concentrations of BTEX were almost 2–3 times and 4–15 times higher than the outdoor levels at industrial and agricultural sites, respectively. The ventilation (air changes per hour, as in Table 2) for most of the sampled homes in these areas was poor. Thus, higher I/O ratios at industrial and agricultural homes may be attributed to poor ventilation, indoor use of biomass fuel, smoking and other indoor sources of emission. This fact is also supported by poor correlation between indoor and outdoor BTEX levels at these two sites (Table 4).

Interspecies ratios for ambient BTEX

The concentration of BTEX gradually decreases in air due to photochemical degradation predominantly caused by OH radicals (Singla et al. 2012; Masih

Table 3 Statistical data for outdoor and indoor concentrations (μg m^{-3}) of BTEX at Gorakhpur

VOC	Min	Max	Mean	S.D	Median	Percentiles					
						5%	10%	25%	75%	90%	95%
Outdoor											
B	9.9	37.2	21.8	7.8	20.3	10.9	12.1	14.8	28.5	33.3	35.4
T	18.5	56.0	32.6	11.3	28.0	19.7	20.9	23.1	41.7	50.9	52.8
E	BDL	8.1	3.0	2.6	2.3	0.0	0.0	0.3	5.2	6.8	7.6
X	1.2	8.0	3.5	2.3	2.5	1.3	1.3	1.5	5.6	7.5	7.6
Indoor											
B	10.5	181.9	55.2	56.5	27.8	12.0	14.2	18.2	93.7	160.6	169.5
T	16.5	123.7	56.8	28.4	49.3	20.2	25.6	32.1	78.9	101.0	109.4
E	0.2	29.5	10.1	9.2	6.6	0.3	0.3	0.9	17.1	24.2	26.4
X	1.4	19.0	8.1	5.7	6.6	1.6	1.8	2.5	13.0	16.2	17.7

BDL Below detection limit

Table 4 Site wise indoor and outdoor BTEX levels (μg m^{-3})

Site	VOC	Indoor		Outdoor		I/O ratio	I/O range	I–O correlation
		Median	S.D	Median	S.D			
RES	B	14.8	2.5	13.3	1.7	1.1	0.8–1.3	0.428
	T	30.2	7.7	22.2	2.0	1.4	0.7–2.4	0.559
	E	0.3	0.1	BDL	–	–	–	-
	X	1.9	0.3	1.5	0.2	1.2	0.9–1.5	0.285
RDS	B	26.1	4.7	25.2	2.7	1.0	0.9–1.4	0.455
	T	36.6	9.0	49.1	4.2	0.7	0.4–0.9	0.659
	E	3.5	0.6	6.7	0.8	0.5	0.3–0.8	0.143
	X	4.0	0.6	7.2	0.6	0.6	0.4–0.7	0.307
IND	B	30.1	4.5	32.3	3.3	0.9	0.7–1.1	0.508
	T	65.5	11.5	35.3	3.9	1.9	1.2–2.9	0.422
	E	13.9	2.5	3.8	0.5	3.7	2.6–5.1	0.515
	X	10.8	1.4	3.7	0.3	2.9	2.3–3.8	0.260
AGR	B	151.0	21.8	17.1	1.6	8.8	6.9–9.9	0.019
	T	95.6	16.0	23.9	2.2	4.0	3.0–6.0	0.164
	E	22.6	4.2	1.5	0.2	15.1	9.7–18.7	0.053
	X	15.8	1.8	1.6	0.2	10.0	6.9–11.8	0.326

et al. 2016). The atmospheric lifetimes for benzene, toluene and ethylbenzene are 9.4, 1.9 and 1.6 days, respectively (Zalel and Yuval 2008). However, the atmospheric degradation of xylene is relatively faster and it has a lifetime of only 15.6 h (Zalel and Yuval 2008). Due to difference in rates of atmospheric degradation, the ratios between BTEX species are frequently used to indicate the proximity of an emission source as well as the photochemical aging of the air (Bruno et al. 2006; Caselli et al. 2010; Miller et al. 2011). Table 5 displays the average interspecies ratios for ambient BTEX observed at different sites of Gorakhpur. Dominance of vehicular emissions is evident by benzene on toluene (B/T)

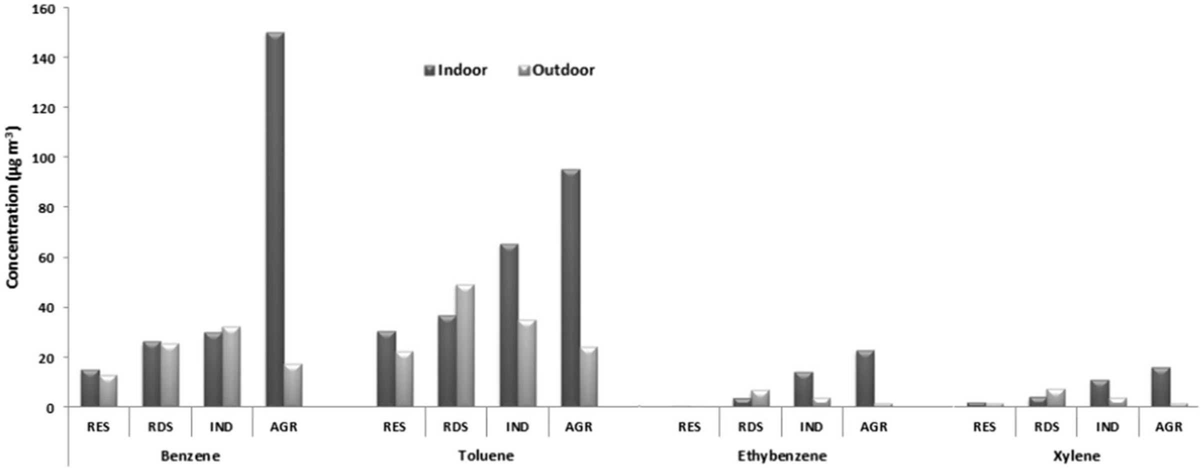

Fig. 2 Comparison between indoor and outdoor BTEX levels at different sites of Gorakhpur

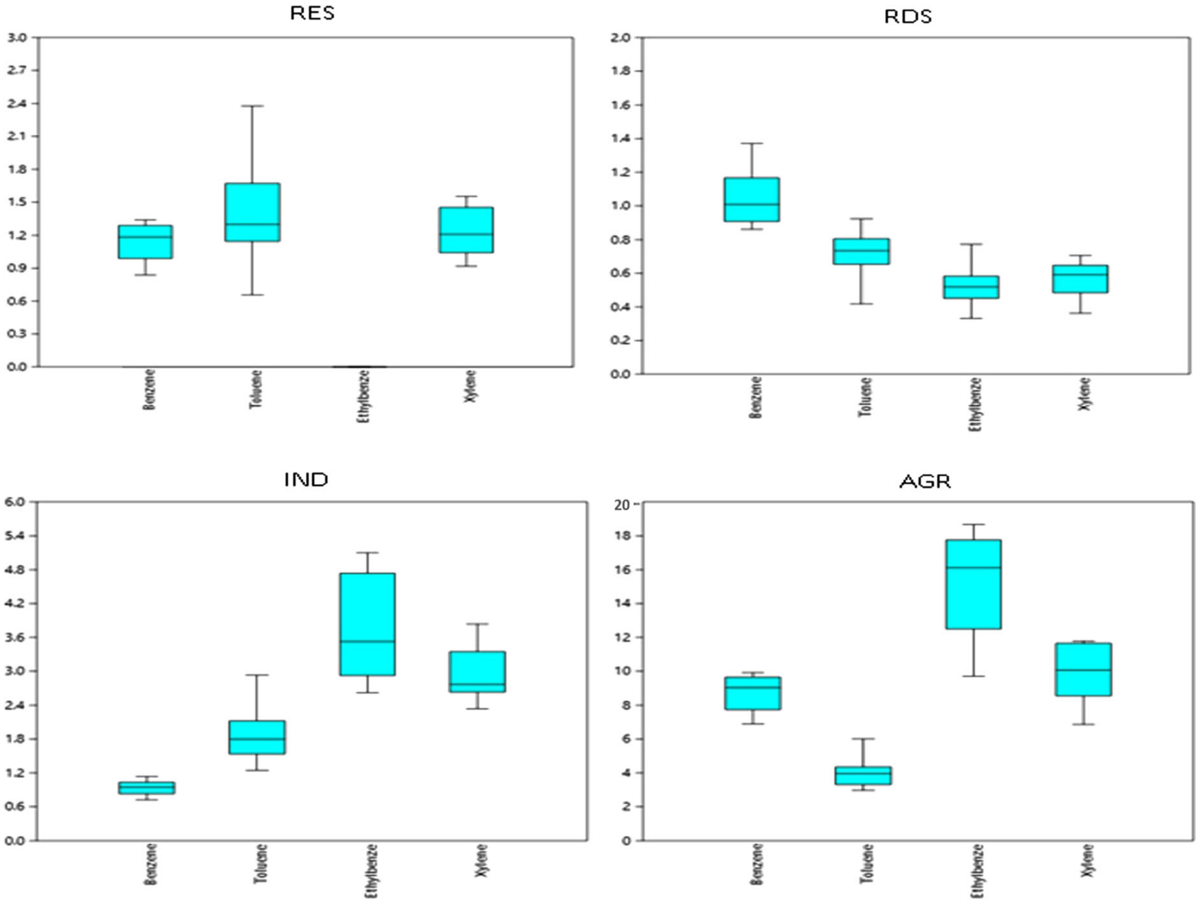

Fig. 3 Boxplot for indoor–outdoor ratios at different sites of Gorakhpur

ratio in a specific range from 0.26 to 0.50 (Bruno et al. 2006; Caselli et al. 2010; de Gennaro et al. 2015). In the present study, B/T ratio at the roadside site was 0.48 which is probably due to traffic related emissions as the sampling site was adjacent to a busy road. Similarly, at the residential site, B/T ratio of 0.42 may be attributed to the frequent use of diesel gen-sets due to erratic supply of electricity. However, B/T ratios at the industrial (0.91) and agricultural sites (0.75) were quite high suggesting emissions from distant sources. At the industrial site, higher B/T ratio may be attributed to emissions from different industries located within the range 1–5 kms from the sampling site. Similarly, at the agricultural site, higher B/T ratio may probably be due to incineration as well as diesel gen-sets used for irrigation. The ratios of xylene to ethylbenzene (X/E) and xylene to benzene (X/B) have also been used to examine the photochemical aging of air mass (Tunsaringkarn et al. 2014). The X/E ratios at roadside, industrial and agricultural sites were 1.26, 1.01 and 1.03, respectively, while the X/B ratios were 0.18, 0.31, 0.10 and 0.11 at residential, roadside, industrial and agricultural site, respectively. Lower values of X/E and X/B at industrial and agricultural sites also indicate emissions originating from a distant source.

Factor analysis

The principal sources effecting BTEX levels were determined by means of a varimax rotated factor analysis. Factor analysis includes a mathematical process that converts a number of (possibly) correlated variables into a (smaller) number of uncorrelated variables. The set of multiple intercorrelated variables was interchanged by small number of independent factors using orthogonal rotations. This is achieved by estimating the correlation matrix, i.e., by calculating the eigenvalues and eigenvectors. Correlation between variables and the factors are obtained by the component loadings attained after varimax rotation. KMO (Kaiser–Meyer–Olkin) value for each variable was also calculated, which gives sampling appropriateness, and data only having eigenvalues bigger than one were included in the matrix. The primary matrix was transmuted into an easier matrix, using varimax technique for rotation of the factor matrix. In the present study, the statistical package for social scientists (SPSS version 20.0) computer software was used to execute factor analysis. Table 6 depicts the results attained by varimax rotated factor analysis. Loadings>0.7 were considered to be statistically significant. As evident from Table 6, at entire sites only one factor is extracted for indoors. Residential, industrial, roadside and agricultural sites exhibited 83.8%, 85.7%, 72.5% and 86.4% variance of data set, respectively. At outdoor residential and industrial sites, only one factor is extracted having 72.1% and 73.9% variance of data set, respectively, whereas roadside and agricultural sites are having two factors each with 49.3%, 50.7% and 56.6%, 43.4% variance of data set, respectively.

Indoor probable sources

As evident from Table 6, the common indoor combustion activities such as cooking, smoking, use of incense and mosquito repellant coils may be regarded as a major source of BTEX in indoor air at each site. Besides the use of LPG, a variety of other cooking fuels such as kerosene, coal, wood and cow dung cakes were also found in use at different sites, which probably enhanced the indoor levels of BTEX.

Outdoor probable sources

Table 6 also explains that incineration may be the dominant source of BTEX at agricultural site due to

Table 5 Average interspecies ratios for ambient BTEX observed at different sites

	RES	RDS	IND	AGR
B/T	0.42	0.48	0.91	0.75
X/E	NA*	1.26	1.01	1.03
X/B	0.18	0.31	0.10	0.11

*Concentration of ethylbenzene at this site was below detection level

Table 6 Results of factor analysis with varimax rotation on BTEX at Gorakhpur

BTEX	Residential		Industrial		Roadside			Agricultural		
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor		Indoor	Outdoor	
	Factor 1	Factor 1	Factor 1	Factor 1	Factor 1	Factor 1	Factor 2	Factor 1	Factor 1	Factor 2
Benzene	0.76	0.75	0.93	0.39	0.75	0.99	0.41	0.92	0.23	0.96
Toluene	0.93	0.27	0.94	0.98	0.73	0.97	0.37	0.97	0.71	0.39
Ethylbenzene	0.97	–	0.88	0.94	0.97	0.23	0.74	0.86	0.96	–
Xylene	0.98	0.74	0.96	0.98	0.93	0.02	0.79	0.97	0.89	0.30
Eigenvalue	3.35	3.01	3.47	2.96	2.90	1.97	2.08	3.46	2.26	1.77
% of Variance	83.8	72.1	85.7	73.9	72.5	49.3	50.7	86.4	56.6	43.4
Cumulative %	83.8	72.1	85.7	73.9	72.5	49.3	100	86.4	56.6	100
Predicted Sources	Combustion activities (cooking and smoking)	Diesel and kerosene gen-sets	Combustion activities (cooking and smoking)	Coal and oil	combustion activities				Combustion activities (cooking and smoking)	Vehicular activities
Asphalt road surfaces	Wood/cow dung cakes/coal		Incineration/						stubble/moorland heather/brushood/ straw burning/	Diesel and kerosene gen-sets

Loading greater than 0.7 is significant

the practice of dumping entire wastes at outskirts of the city nearby the agricultural area, which is incinerated at a regular interval of time. In addition, for irrigation purpose, diesel-powered pumping sets were also used. In the same manner, source of BTEX at the residential site might be emissions by diesel gen-sets which are used as an alternate source of electricity due to its erratic supply. At the roadside, vehicular activities may be the main source of BTEX as large number of diesel- and petrol-fuelled vehicles (2, 3 and 4 wheelers) are commonly used for transportation. On other hand, at the industrial site, oil burning and coal combustion may be the dominant source of BTEX because they are used in different industries/factories for furnace heating purpose. Thus, factor analysis illustrates a combined influence of combustion and vehicular activities.

Analysis of variance (one-way ANOVA)

The results obtained through factor analysis were examined with the help of one-way analysis of variance (ANOVA). At each site, certain indoor and outdoor activities affecting the air quality were pointed out with the help of questionnaire. Then each activity was analyzed with ANOVA in order to determine its significance. Table 7, 8 show the variation in indoor and outdoor BTEX levels, respectively, due to different indoor/outdoor activities which are describes as follows.

Use of different cooking fuels

The indoor air quality basically depends on the type of fuel used for cooking up to an extent, which in turn depends upon the economic status of the family. Thus, the use of cow dung cakes and mixed fuel (wood/coal/kerosene) is quite common among the lower income families especially at industrial and agricultural sites, while LPG which is comparatively expensive is usually used by the middle higher income families. Table 7 indicates that indoor levels of BTEX were highest in homes using cow dung followed by mixed fuel (wood/coal/kerosene), while lowest levels were obtained in homes using LPG as a cooking fuel. At all other sites, except agricultural site, LPG was the preferred fuel. Even in economically superior residential and roadside areas, apart from LPG, few of the families also acknowledged the use of mixed fuel for cooking especially during winters. The results of one-way ANOVA show that, at residential site, homes using mixed fuel had significantly higher levels of benzene ($F=6.039$, $p=0.020$), toluene ($F=4.814$, $p=0.036$), ethylbenzene ($F=6.518$, $p=0.016$) and xylene ($F=4.210$, $p=0.049$) than those using LPG. Similarly at roadside site, benzene ($F=7.147$, $p=0.012$), toluene ($F=8.075$, $p=0.008$), ethylbenzene ($F=5.182$, $p=0.030$) and xylene ($F=5.835$, $p=0.022$) had significantly higher concentration in homes using mixed fuel. At industrial and agricultural site homes, three categories of cooking fuel, i.e., solid biomass fuel, mixed fuel and LPG were found in use. One-way ANOVA showed that at industrial site, homes using these three categories of cooking fuel had significantly different levels of benzene ($F=4.782$, $p=0.016$), toluene ($F=4.308$, $p=0.023$), ethylbenzene ($F=5.419$, $p=0.010$) and xylene ($F=3.352$, $p=0.049$). In the same way at agricultural site, the difference was significant for benzene ($F=12.249$, $p<0.001$), toluene ($F=10.757$, $p<0.001$), ethylbenzene ($F=5.916$, $p=0.007$) and xylene ($F=10.277$, $p<0.001$).

Smoking

Many previous studies show that the indoor levels of benzene are higher in homes where smoking is prevalent (Godish 2001; Duarte-Davidson et al. 2001). In the present study, it was observed that at each site, the indoor concentrations of not only benzene but also of toluene, ethylbenzene and xylene were higher in homes where smoking was prevalent than homes where no smoking was held (Table 7). But, the results of one-way ANOVA showed that only benzene levels were significantly different ($p<0.05$) in homes with different smoking status at residential site ($F=4.711$, $p=0.038$) and industrial site ($F=6.257$, $p=0.018$). However, at agricultural site the indoor concentrations of benzene ($F=6.038$, $p=0.020$) as well as ethylbenzene ($F=4.376$, $p=0.045$) were significantly higher in homes where smoking was prevalent. Also tobacco smoke is regarded as a major source of ethylbenzene in indoor air (Wallace et al. 1987; Parra et al. 2008). Surprisingly at roadside site, smoking did not contribute significantly to indoor levels of BTEX.

Table 7 Variation of indoor BTEX levels

Site	Activities	Categories	*N*	Benzene		Toluene		Ethylbenzene		Xylene	
				Mean (μg/m^3)	*p*-value	Mean (μg/m^3)	*p*-value	Mean (μg/m^3)	*p*-value	Mean (μg/m^3)	*p*-value
RES	Cooking fuel	Mixed fuel	9	18.3	0.020*	36.5	0.036*	0.5	0.016*	2.4	0.049*
		LPG	23	13.4		27.7		0.3		1.7	
	Smoking	Yes	6	16.3	0.038*	34.2	0.054	0.5	0.280	2.4	0.075
		No	26	14.4		29.3		0.3		1.7	
	Incense/mosquito repellants	Yes	27	15.0	0.107	30.5	0.115	0.4	0.268	1.9	0.101
		No	5	13.6		28.5		0.3		1.6	
RDS	Cooking fuel	Mixed fuel	6	39.3	0.012*	45.8	0.008*	4.3	0.030*	4.4	0.022*
		LPG	26	23.2		34.3		3.3		4.0	
	Smoking	Yes	8	38.6	0.081	42.5	0.139	4.1	0.065	4.2	0.120
		No	24	22.0		34.5		3.2		4.0	
	Incense/mosquito repellants	Yes	25	28.5	0.426	38.4	0.722	3.6	0.315	4.1	0.366
		No	7	17.9		29.8		2.9		3.8	
IND	Cooking fuel	Cow dung	9	36.3	0.016*	76.5	0.023*	15.1	0.010*	12.0	0.049*
		Mixed fuel	13	28.9		67.2		14.0		10.3	
		LPG	10	25.4		53.0		12.6		10.3	
	Smoking	Yes	11	34.0	0.038*	67.4	0.096	15.0	0.248	11.6	0.163
		No	21	27.7		64.3		13.3		10.4	
	Incense/mosquito repellants	Yes	22	32.1	0.381	66.4	0.160	14.8	0.105	10.9	0.311
		No	10	25.1		63.2		11.9		10.5	
AGR	Cooking fuel	Cow dung	18	173.2	<0.001*	108.1	<0.001*	25.9	0.007*	18.3	<0.001*
		Mixed fuel	10	121.8		80.4		19.6		13.5	
		LPG	4	115.9		74.7		14.3		9.9	
	Smoking	Yes	15	160.4	0.020*	99.4	0.160	23.9	0.045*	17.2	0.394
		No	17	140.8		91.7		21.2		14.4	
	Incense/mosquito repellants	Yes	18	158.3	0.274	98.7	0.198	22.8	0.662	15.9	0.300
		No	14	139.3		90.9		22.1		15.6	

**p*-value<0.05

Mixed fuel: wood/coal/kerosene, LPG: liquefied petroleum gas

Table 8 Variation of outdoor BTEX levels

Site	Activities	Categories	*n*	Benzene		Toluene		Ethylbenzene		Xylene	
				Mean (µg/m^3)	*p*-value	Mean (µg/m^3)	*p*-value	Mean (µg/m^3)	*p*-value	Mean (µg/m^3)	*p*-value
RES	Diesel/kerosene gen-sets	Yes	6	15.3	0.047*	24.7	0.251	BDL		1.7	0.018*
		No	26	12.3		21.6				1.4	
RDS	Vehicular activities	High	21	29.1	0.028*	54.6	0.015*	8.2	0.054	7.6	0.505
		Moderate	11	17.7		38.6		3.8		6.2	
IND	Distance from industry	Near (0–2 km)	19	33.2	0.408	39.1	0.005*	4.0	0.040*	3.9	0.023*
		Distant(> 2 km)	13	30.4		29.3		3.5		3.4	
AGR	Incineration/stubble/moorland heather/brushwood/straw burning	Yes	20	18.2	0.246	26.1	0.022*	1.6	0.016*	1.7	0.039*
		No	12	15.2		20.2		1.3		1.4	
	Diesel/kerosene gen-sets	Yes	7	21.1	0.034*	24.3	0.090	1.6	0.640	1.6	0.372
		No	25	16.0		23.8		1.5		1.6	

*p-value<0.05

Use of incense/mosquito repellants and diesel/kerosene generators

Higher concentrations of benzene and toluene have also been associated with use of mosquito repellant coils, incense burning (Lee and Wang 2006) and also by the use of diesel generators due to erratic supply of electricity (Jobson et al. 2005; Ferreira et al. 2008). In the present study, at each locality, homes using incense/mosquito repellants and diesel/kerosene-powered generators were found higher indoor–outdoor levels of BTEX as compared to homes not using them. It was observed that at indoors, the differences in concentrations were not significant (p>0.05) at any site. However, in ambient air, at the residential site, benzene (F=4.290, p=0.047) and xylene (F=6.259, p=0.018), while at the agricultural only benzene (F=4.932, p=0.034) had significantly higher levels at homes using diesel-powered generators.

Incineration

A few incineration activities were found at the agricultural site. It was observed that homes near the incineration point had higher ambient levels of BTEX. Results of one-way ANOVA show that the difference was significant for toluene (F=5.833, p=0.022), ethylbenzene (F=6.510, p=0.016) and xylene (F=4.658, p=0.039).

Vehicular activity

In order to study the influence of vehicular emissions on ambient levels of BTEX, at roadside site, homes were categorized according to the observed traffic density, i.e., moderate and high traffic. Results of one-way ANOVA show that at roadside site, the homes along high traffic road had significantly higher ambient levels of benzene (F=5.328, p=0.028) and toluene (F=6.655, p=0.015) than homes along moderate traffic road. The ambient levels of ethylbenzene and xylene were also found to be higher at homes along high traffic road; however, the difference in concentrations were not significant (p>0.05).

Industrial emissions

BTEX compounds are commonly used as industrial solvents especially for various industrial processes like degreasing and coating. BTEX are also used as initial components for the synthesis of various drugs, paints and polymers (APA 2001; Nikodinovic et al. 2008). As a result of extensive usage of BTEX compounds in industries, the emission of BTEX at industrial areas is quite natural (ATSDR 2004; Nikodinovic et al. 2008). In order to study the effect of industrial emissions on ambient levels of BTEX, homes in the industrial area were divided into two categories on the basis of distance from nearest industry. It was observed that the homes which were fairly close (0–2 kms) to industries had higher ambient levels of BTEX than homes which were quite distant (>2 kms). Results of one-way ANOVA show that the difference was significant for toluene (F=9.176, p=0.005), ethylbenzene (F=4.609, p= 0.040) and xylene (F=5.737, p=0.023).

Health risks of BTEX

Table 9 illustrates integrated lifetime cancer (ILTCR) and non-cancer (HQ) risks (for 15 years residing time for an individual). At all the sites, the estimated cancer risk for benzene, i.e., at residential area (9.3E-06 at indoor and 8.2E-06 at outdoor), roadside area (1.7E-05 at indoor and 1.6E-05 at outdoor), industrial area (1.9E-05 at indoor and 2.0E-05 at outdoor) and at agricultural area (9.5E-05 at indoor and 1.1E-05 at outdoor) exceeded the threshold value of 1E-06 indicating more cancer risk from benzene due its high carcinogenicity. But the hazard of melanoma from ethylbenzene is more at indoors of industrial and agricultural area having 1.2E-06 and 2.0E-06, respectively.

Figure 4 shows a graphical representation comparing the ILTCR values obtained for indoor and outdoor concentrations of benzene and ethylbenzene at each site. It is evident from the figure that at most of the sites, in general, the ILTCR values for indoor and outdoor ethylbenzene levels were either very low or close to the recommended value of 1×10^{-6}. However, the ILTCR values for indoor as well as outdoor concentrations of benzene were higher than 1×10^{-6}, suggesting ‘possible risk’ (1×10^{-4}>ILTCR 1×10^{-5}) to ‘probable risk’ (1×10^{-5}>ILTCR 1×10^{-6}) for the occurrence of cancer. The table also specifies that at all the sites (indoor and outdoor), benzene gives the uppermost non-cancer risk (HQ) trailed by toluene, ethylbenzene and xylene, respectively. However, the individual HQs for BTEX were much lower than unity at all the sites except for benzene at agricultural site (indoor) and industrial site (outdoor) signifying no significant threat of chronic non-cancer health effects in pollutant specific target organs for the city population (ASTDR 2010; Majumdar et al. 2011). The higher value of HQ (5.0) at agricultural indoors may be due to indoor burning of wood, cow dung cakes and coal for heating as well as cooking purpose. Similarly, at outdoor industrial locations, higher HQ (1.1) for benzene may be attributed to the industrial emissions.

Comparison of indoor–outdoor BTEX levels with other studies of the world

Table 10 shows a comparison of indoor/outdoor BTEX levels with other studies from different parts of the world. In the present study, the observed benzene levels at indoor/outdoor (27.8/20.3 µg m^{-3}) were quite higher than those reported in other studies at Hong Kong (4.9/1.9 µg m^{-3}), Mexico (7.4/7.2 µg m^{-3}), Belgium (2.2/1.1 µg m^{-3}), Egypt (7.1/7.8 µg m^{-3}), Turkey (2.3/1.2 µg m^{-3}), Japan (1.3/1.0 µg m^{-3}) and Iran (15.8/8.6 µg m^{-3}). However, the concentrations of toluene at indoor/outdoor (49.3/28.0 µg m^{-3}) were lower than those found in Hong Kong (59.1/36.6 µg m^{-3}), Mexico (76.2/41.8 µg m^{-3}) and Iran (69.7/40.6 µg m^{-3}). The indoor ethylbenzene levels (6.6 µg m^{-3}) were lower than those reported from Iran (12.1 µg m^{-3}) and xylene concentrations (6.6 µg m^{-3}) were lesser than found in Hong Kong (9.2 µg m^{-3}), Mexico (25.8 µg m^{-3}), Egypt (15.6 µg m^{-3}), Japan (8.4 µg m^{-3}) and Iran (48.1 µg m^{-3}). On the other hand, the outdoor levels of ethylbenzene/xylene (2.3/2.5 µg m^{-3}) were higher than those reported in Belgium (0.6/1.1 µg m^{-3}), Turkey (0.3/0.4 µg m^{-3}) and Japan (1.5/2.3 µg m$^{-3)}$.

Conclusion

During winter season, indoor and outdoor concentrations of BTEX were investigated at four different sites of Gorakhpur during winter season (November 2015 to February 2016). The indoor levels of BTEX were highest at the agricultural site, followed by industrial, roadside and residential sites, whereas for outdoor air, the highest BTEX levels were observed

Table 9 Estimate of non-cancer and cancer risk at different sites

Residential				
	Indoor		Outdoor	
	HQ	ILTCR	HQ	ILTCR
Benzene	4.8E-01	9.3E-06	4.3E-01	8.2E-06
Toluene	6.0E-03		4.4E-03	
Ethylbenzene	3.5E-04	3.2E-08	BDL	BDL
Xylene	1.5E-02		1.6E-02	
Roadside				
	Indoor		Outdoor	
	HQ	ILTCR	HQ	ILTCR
Benzene	8.7E-01	1.7E-05	8.4E-01	1.6E-05
Toluene	7.3E-03		9.8E-03	
Ethylbenzene	3.5E-03	3.1E-07	6.5E-03	5.8E-07
Xylene	4.1E-02		7.0E-02	
Industrial				
	Indoor		Outdoor	
	HQ	ILTCR	HQ	ILTCR
Benzene	9.9E-01	1.9E-05	1.1	2.0E-05
Toluene	1.3E-02		7.0E-03	
Ethylbenzene	1.4E-02	1.2E-06	3.8E-03	3.4E-07
Xylene	1.1E-01		3.6E-02	
Agricultural				
	Indoor		Outdoor	
	HQ	ILTCR	HQ	ILTCR
Benzene	5.0	9.5E-05	5.7E-01	1.1E-05
Toluene	1.9E-02		4.7E-03	
Ethylbenzene	2.2E-02	2.0E-06	1.5E-03	1.3E-07
Xylene	1.6E-01		1.6E-02	

HQ hazard quotient, *ILTCR* integrated life time cancer risk

BDL: below detection limit

at the roadside site, followed by industrial, agricultural and residential sites. The indoor concentrations of BTEX were higher than outdoors, at all sites except the roadside site. At each site, poor correlation was observed between indoor and outdoor levels suggesting dominance of indoor sources. Factor analysis was utilized for source characterization of indoor and outdoor BTEX. The achieved factors were then related to different indoor/outdoor sources with the help of one-way ANOVA. It was noticed that, at each site, the indoor BTEX levels were influenced by the type of cooking fuel and smoking status of the occupants, while the outdoor levels were affected by incineration, use of diesel/kerosene generators, vehicular and industrial emissions at different sites. The cancer risk assessment (ILTCR) revealed ‘probable’ to ‘possible’ risk of cancer due to prevailing levels of benzene. The HQ values for individual BTEX compounds were lower than 1 at all the sites except for benzene at the agricultural site indoors and the industrial site outdoors.

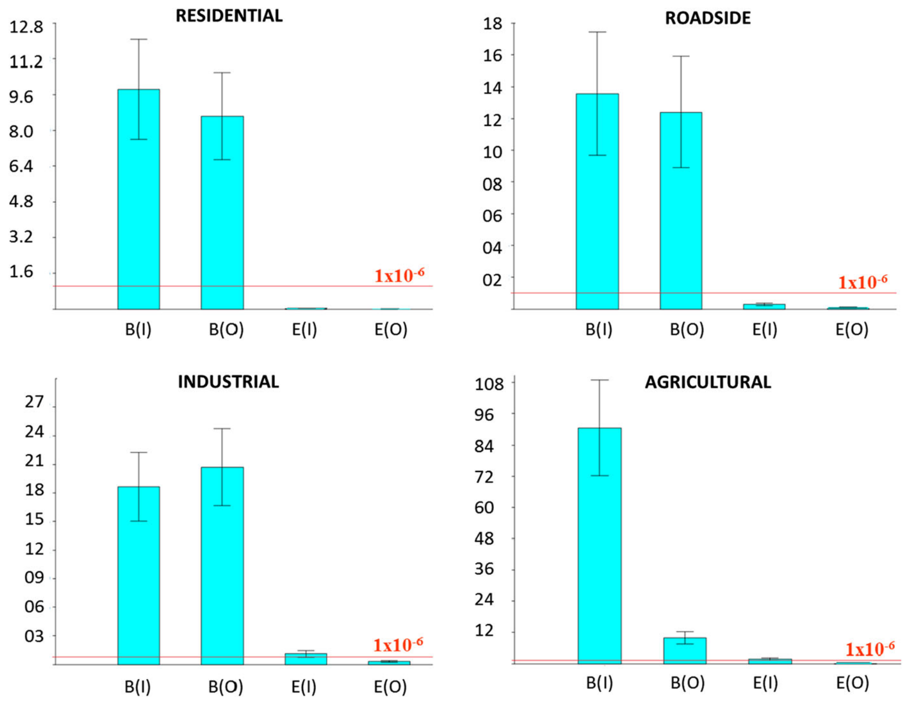

Fig. 4 ILTCR values for indoor and outdoor concentrations of benzene and ethylbenzene investigated at each site. *Y* axis= ILTCR ($\times 10^{-6}$ mg^{2}kg^{-2} day^{-2}) B(I)=benzene@indoors B(O)= benzene@outdoors E(I)=ethylbenzene@indoors E(O)= ethylbenzene@outdoors

Table 10 Worldwide comparison of indoor–outdoor BTEX levels (μg m^{-3})

Country	Benzene		Toluene		Ethylbenzene		Xylene		Reference
	In	Out	In	Out	In	Out	In	Out	
Hong Kong	4.99	1.94	59.13	36.56	2.72	5.40	9.16	8.82	Guo et al. (2003)
Mexico	7.4	7.2	76.2	41.8	5.9	5.9	25.8	24.8	Serrano et al. (2004)
Belgium	2.2	1.05	4.25	2.27	0.62	0.57	2.04	1.07	Stranger et al. (2007)
Egypt	7.08	7.81	23.4	22.84	3.18	3.07	15.56	15.45	Silke et al. (2010)
Turkey	2.29	1.23	26.55	6.11	0.73	0.26	0.98	0.43	Demirel et al. (2014)
Japan	1.3	1.0	12.0	7.0	4.4	1.5	8.4	2.3	Uchiyama et al. (2015)
Iran	15.18	8.65	69.7	40.56	12.07	4.92	48.08	7.44	Hazrati et al. (2016)
India	27.8	20.3	49.3	28.0	6.6	2.3	6.6	2.5	Present Study

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Compliance with ethical standards

Conflict of interest The authors have no conflicts of interest to declare that are relevant to the content of this article.

Consent to publish Copy Attached.

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