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Investigation of outdoor BTEX: Concentration, variations, sources, spatial distribution, and risk assessment

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HIGHLIGHTS

- Seasonal variation, spatial mapping, photochemical aging, and risk assessment of BTEX in the ambient air of Tehran were investigated. .
- The maximum and minimum concentrations of BTEX were observed for toluene and ethylbenzene, respectively.
- Spatial distribution of BTEX pollution showed that the highest concentrations were found along the major roads because of heavy traffic.
- The cancer risk of benzene and noncarcinogenic risk of BTEX were in the acceptable range.

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ABSTRACT

The aim of this study was to measure BTEX (benzene, toluene, ethylbenzene, and xylenes) concentrations in the ambient air of Tehran, the capital of Iran, and investigate their seasonal variations, probable sources, spatial mapping, and risk assessment. The concentrations of BTEX were measured using a continuous monitoring device installed in seven stations around the city. Spatial mapping procedure was conducted using the inverse distance weighting (IDW) method. Monte Carlo simulation was used to assess the carcinogenic and noncarcinogenic risks imposed by BTEX. The highest and lowest annual mean concentrations of toluene and ethylbenzene were recorded as 16.25 and 3.63 μg m^{-3}, respectively. The maximum (6.434) and minimum (3.209) toluene/benzene (T/B) ratio was observed in summer and winter, respectively. The spatial distribution of BTEX pollution indicated that the highest concentrations were found along the major roads because of heavy traffic. Spearman's rank correlation coefficients and concentration ratios showed that BTEX were produced by the multiemission sources. The mean of inhalation lifetime cancer risk (LTCR) for benzene was 3.93×10^{-7}, which is lower than the limits recommended by the United States Environmental Protection Agency (US EPA) and the World Health Organization (WHO). The hazard quotient (HQ), noncarcinogenic risk index, for all BTEX compounds was <1. The obtained results showed no threat of BTEX concentrations to human health. However, as the concentrations of BTEX will increase due to the rapid growth of vehicles and industrial activities, much effort is required to control and manage the levels of these compounds in the future.

1. Introduction

Traffic and industrial activities together with natural emissions are the major sources of air pollution in urban areas. A large

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number of pollutants such as sulfur oxides (SOx), nitrogen oxides (NOx), particulate matter (PM), carbon oxides (COs), ozone (O_3), radioactive pollutants, and volatile organic compounds (VOCs) are released by these sources into the ambient air. In recent years, there has been an increasing interest in research on VOCs not only for concerns about their carcinogenic and noncarcinogenic effects on human population but also for their photochemical reactions, which can produce secondary pollutants such as O_3 and proxy acetyl nitrate (PAN) (Ghozikali et al., 2016; Guimarães et al., 2014; Lin et al., 2016; Pekey and Yilmaz, 2011).

Over the past two decades, a considerable number of studies have been conducted on benzene, toluene, ethylbenzene, and ortho (o), para (p), and meta (m)-xylene (BTEX) as the most important VOCs because of their high potential to exert carcinogenic effects and relatively high abundance in ambient air. The most abundant and hazardous BTEX compound is benzene, which has been categorized by the International Agency for Research on Cancer (IARC) as a known carcinogenic to humans (Group 1) (Demirel et al., 2014).

Two important characteristics of BTEX are their presence in the ambient air and the correlations with their atmospheric concentrations (Bruno et al., 2008; Hoque et al., 2008; Khoder, 2007; Miller et al., 2010). The latter, in the BTEX studies, is used to determine the sources and photochemical ages of these compounds.

Toluene-to-benzene (T/B) ratio has been widely used by many researchers worldwide as an emission source index of BTEX (Buczynska et al., 2009; Hoque et al., 2008; Miller et al., 2011). The T/B ratio in the range of 1.5–4.0 indicates that transportation is the original source of BTEX in the region concerned. In addition, the ratio of (m + p)-xylene to ethylbenzene (m + p-X/E) has been known as a photochemical age index (Zhang et al., 2008). Although emission rate of (m + p)-xylene is 3.6 times more than ethylbenzene (Monod et al., 2001), the (m + p)-xylene is removed by chemical reactions about 3 times faster than ethylbenzene. Therefore, the (m + p)-X/E ratio is reduced by photochemical aging (Nelson and Quigley, 1983). Consequently, this ratio can be used to determine the period of time in which BTEX entered the ambient air.

Although extensive studies have been carried out on BTEX, they have been mostly restricted in terms of intermittent measurement frequency and low number of sampling sites.

Most studies have been carried out only in maximum 12 sites, for 1 or 2 weeks and in one season (Brocco et al., 1997; Bruno et al., 2008; Ho et al., 2004, 2009; Khoder, 2007; Moriarty, 1988; Smith et al., 2007). Such approaches have failed to address the seasonal trend of BTEX concentrations and their exposure assessments as the atmospheric reactions of these compounds are dependent on time (Miller et al., 2012). In the present study, however, the BTEX concentrations were obtained through continuous measurement device, which can record the BTEX concentrations throughout the year. To the best of our knowledge, this study is unique in this field and can fill this research gap. Therefore, it was designed to investigate the seasonal variations, spatial mapping, photochemical aging, and risk assessment of BTEX in the ambient air of Tehran, the capital of Iran.

2. Material and methods

2.1. Study area

Tehran has a population of approximately 9 million, according to the last census report in 2011. It is the largest city and urban area of Iran, the second largest city in western Asia, and the third largest city in the Middle East. Its longitude and longitude are 51°17′E–51°33′E and 35°35′N–35°48′N, respectively, and it is located at about 1040–2500 m above the sea level. Tehran has a total area of approximately 680 km^2. It is categorized as one of the most polluted cities at the global level. The most important environmental problem in this city is extreme air pollution, which mainly originates from the transportation (80%) and industrial activities (20%) (Amini et al., 2014). Fig. 1 presents the map of Tehran and sampling locations. These locations were selected according to factors such as proximity to highways, traffic situation, and the status of residential and industrial areas.

2.2. Data collection and analysis

BTEX concentrations were measured using continuous VOC monitoring device model VOC71M-PID (France), which is based on the gas chromatography method combined with a photoionization detector (PID) or flame ionization detector (FID). Sampling was conducted every day continuously from March 2012 to March 2013. The sampling device is a fully automated instrument that can monitor low level of specific VOCs such as BTEX. It is particularly well adapted for application in air quality monitoring (urban and industrial sites) programs. The sampling is conducted in cyclic mode with two tubes filled with selective sorbents. When one tube collects a sample, the other one desorbs it. This allows the instrument to achieve approximately 100% sampling time coverage. The desorbed sample is then injected into a fused silica capillary column for separation. The controlled temperature programs permit the fast and accurate separation of VOCs. Compounds are identified by their elution times through the capillary column.

Data management and analysis were performed using SPSS software (version 22). The interrelationship between BTEX compounds was assessed using Spearman's rank correlation coefficient. T/B, (m + p)-X/B, and (m + p)-X/EB ratios were calculated for each measurement station to evaluate photochemical aging.

2.3. Spatial distributions

To conduct spatial analysis, ArcGIS 10.1 software, developed by ESRI Company, was used. The inverse distance weighting (IDW) interpolation technique was used to develop independent raster layer for annual mean concentration of each pollutant and determine the distribution of BTEX compounds. The raster calculator function was then used to overlay each layer and produce yearly average maps of BTEX. Many researchers have used IDW method for mapping air pollutants such as sulfur dioxide in Turkey (Mulholland et al., 1998; Tolbert et al., 2000), ozone in northern Georgia (Bell, 2006), and PMs in the USA and China (Whitworth et al., 2011). IDW is, in fact, a nonstatistical method, which is usually applied in environmental studies to predict the concentration of pollutants at unmeasured locations through the optimal spatial prediction technique. It is very useful when the distribution of the estimated parameters is abnormal. IDW model assumes that the predictions are in a linear function of available data. The IDW model follows Eq. (1) (Xie et al., 2011):

$$\lambda i = \frac{Di - \alpha}{\sum_{i=1}^{n} Di - \alpha}, \quad (1)$$

where λ_i is the weight of the station i, D_i is the distance between the station i and unknown point, α is the weighting power, and n is the total number of known points used in the interpolation. The higher weighting powers are assigned to the values nearer to the interpolated points. A decrease in weight will be observed with increasing distance.

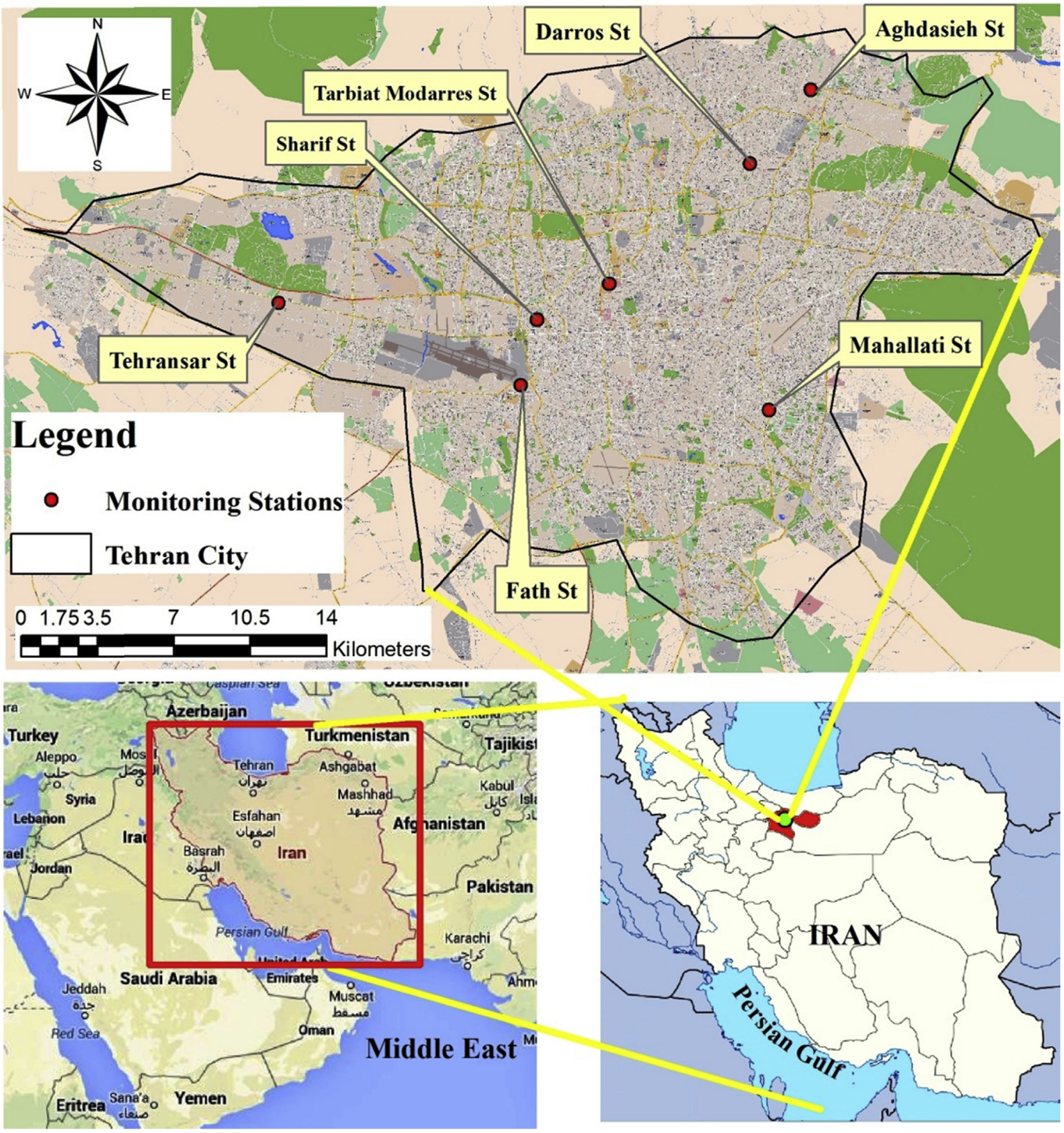

Fig. 1. Location of study area and their monitoring stations.

2.4. Health risk assessment

The noncarcinogenic hazard of BTEX and the lifetime cancer risk (LTCR) of benzene through the inhalation pathway were calculated to estimate the human health impacts of BTEX compounds in the ambient air of Tehran. The LTCR was calculated by multiplying chronic daily intake (CDI) of benzene by cancer slope factor (CSF), which is determined by Integrated Risk Information System (IRIS). LTCR is calculated as

$$LTCR = CDI \times CSF \tag{2}$$

CDI was calculated as follows:

$$CDI = \frac{(C \times CF \times IR \times ED \times EF)}{(BW \times AT)}, \tag{3}$$

where C is the contaminant concentration ($\mu g\ m^{-3}$), CF is the conversion factor ($mg\ \mu g^{-1}$), IR is the inhalation rate ($m^3\ day^{-1}$), ED

is the exposure duration (year), EF is the exposure frequency (days year^{-1}), BW is the body weight (kg), and AT is the average lifetime (days).

Because of the uncertainty in the values of some variables imported to cancer and noncarcinogenic risk equations, inaccurate or biased risk estimates are unavoidable. To overcome this problem, sensitivity analysis is used. In other words, the sensitivity analysis technique is used to determine how different values of input variables will affect an estimated risk under a given set of assumptions. In this study, sensitivity analysis was conducted by the Monte Carlo simulation technique using Oracle® Crystal Ball software version 11.1.2.3.

The noncarcinogenic hazard of BTEX was measured in terms of HQ, which can be calculated as follows:

$$HQ = \frac{LEC}{RfC}, \quad (4)$$

where LEC and RfC are the annual average of daily received concentration (mg m^{-3}) and noncarcinogenic reference concentration of the pollutant (mg m^{-3}), respectively.

The literature review was used to exert the values of some parameters to determine the CDI, LTCR, and HQ. The values of IR, ED, EF, BW, and AT found in previous studies were 0.65–0.83 m^3 h^{-1} (Hoseini et al., 2016; Hou et al., 2012), 70 years (Demirel et al., 2014), 350 days year^{-1} (Hou et al., 2012), 71 ± 13.6 kg (Hou et al., 2012), and 25,500 days (Massolo et al., 2010), respectively. For calculating the CDI, the annual average of BTEX concentrations was used. The CSF for benzene was obtained from the IRIS as 0.029 mg kg^{-1} day^{-1} (USEPA, 2009). It should be noted that there is no CSF for other BTEX compounds (TEX). Therefore, cancer risk for TEX compounds cannot be estimated. In addition, the RfC of BTEX was obtained from the IRIS as 0.03, 5, 0.01, and 1 mg m^{-3} for benzene, toluene, xylenes, and ethylbenzene, respectively (USEPA, 2009).

3. Results and discussion

3.1. BTEX concentrations

A statistical summary of benzene, ethylbenzene, toluene, xylenes, and total BTEX concentrations is shown in Fig. 2. The maximum (4.291 ppb or 16.25 μg m^{-3}) and minimum (0.837 ppb or 3.63 μg m^{-3}) annual mean concentrations were observed for toluene and ethylbenzene, respectively. The annual mean concentrations of benzene, (m + p)-xylene, and o-xylene were 1.056, 2.929, and 1.044 ppb (or 3.444, 12.734, and 4.53 μg m^{-3}), respectively. These results are in agreement with those obtained by other studies, in which toluene was reported to have the highest concentration of BTEX in the urban areas. For example, the values of toluene reported by Hazrati et al. (2016), Pekey and Yilmaz (2011), Caselli et al. (2010), and Borgie et al. (2014) were, 45.56, 35.51, 6.21, and 11520 μg m^{-3}. However, the total BTEX concentration found in this study was considerably higher than that obtained by other studies conducted in the UK (Derwent et al., 2000), Germany (Schneider et al., 2001), and USA (Smith et al., 2007). This difference can be explained by the variation in traffic, the quality of fuels and vehicle, and the industrial emissions. These results, however, seem to be relatively consistent in terms of concentration with those obtained by Pekey in Turkey (Pekey and Yilmaz, 2011), which reported the annual mean concentration of toluene to be about 18.75 μg m^{-3}. Moreover, the rank order observed in this study confirms those observed in earlier studies (Guo et al., 2007; Miller et al., 2011; Mulholland et al., 1998; Parra et al., 2006), in which the highest and lowest concentrations were reported for toluene and ethylbenzene, respectively.

3.2. Annual and seasonal trends of BTEX

The descriptive statistic of BTEX data indicated that the maximum seasonal average concentrations of benzene, (m + p)-xylene, o-xylene, toluene, and total BTEX were observed in spring, while for ethylbenzene, it was observed in winter. The minimum

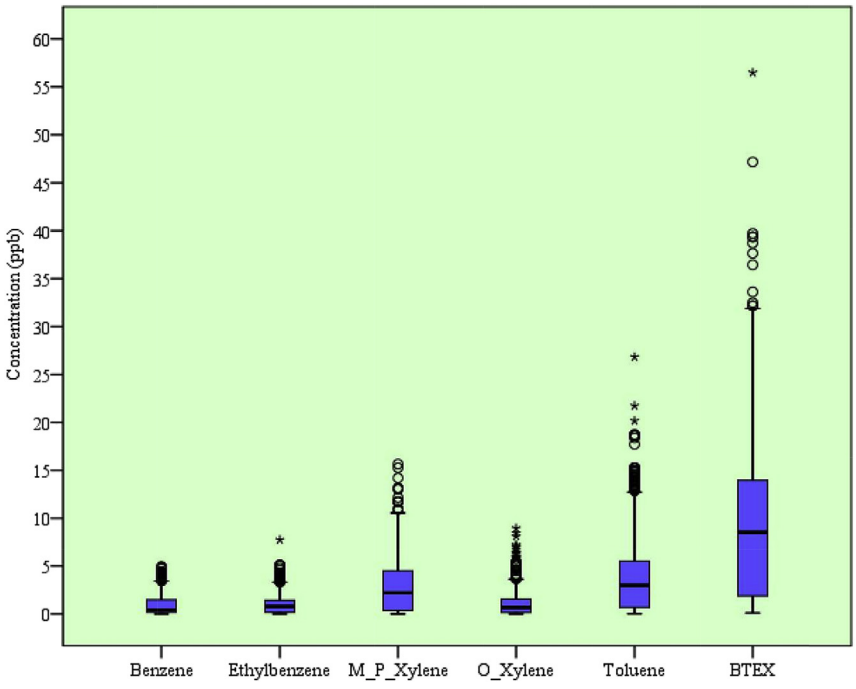

Fig. 2. Descriptive statistics of BTEX concentrations (ppb).

concentration of total BTEX was observed in winter and then in autumn (8.682 and 8.882 ppb, respectively). The most possible reason for the decrease of total BTEX concentration in winter and autumn is rainy condition, which is often frequent in these seasons. Rainfall rapidly transfers the atmospheric BTEX into other environmental media such as soil, plants, and roadbed. As a result, the monitoring device can measure the released BTEX (Mullaugh et al., 2015). The minimum benzene concentration (0.589 ppb) was observed in summer. These findings do not support the results of previous studies (Batterman et al., 2002; Hansen and Palmgren, 1996; Kourtidis et al., 2002), in which the lowest concentration of BTEX was reported for the warmer seasons. In another study conducted in the United States (Pankow et al., 2003), higher toluene and benzene concentrations were recorded in winter; this result was inconsistent with our study result.These differences can be explained in part by the difference in sampling condition (sampling frequency, sampling period of time, and type of sampler). In most of previous studies, measurements were made using noncontinuous devices for only 2 weeks in a month or in one season, while in this study, the samples were obtained using continuous device throughout the year. Although these results are in contrast to those of some studies, they are consistent with those obtained by Ho et al. (2004), who reported that the highest xylene concentration (3.99 μg m^{-3}) and the lowest benzene concentration (0.32 μg m^{-3}) in Hong Kong were observed in summer, and no seasonal variation was found for toluene and ethylbenzene (Ho et al., 2004). Furthermore, Miller et al. (2012) observed the highest BTEX concentrations in spring (Miller et al., 2012).

3.3. Correlations and ratios between BTEX compounds

Nonparametric Spearman's rank correlation was used to determine the correlations between the BTEX compounds according to the annual and seasonal mean concentrations (Table 1). As shown in Table 1, there are significant positive correlations between BTEX concentrations. The observed correlations between BTEX compounds can indicate that the emission sources of these compounds are probably similar. These results are supported by other researchers who reported that there are strong correlations between BTEX compounds (Hoque et al., 2008; Miller et al., 2010; Rad et al., 2014). The correlation coefficients were stronger in cooler seasons than warmer seasons. The lowest correlation coefficient ($r < 0.55$) was seen between benzene and total BTEX. A possible explanation for this result may be related to the different sources of BTEX compounds. The major emission sources of benzene are vehicles, while the other constituents of BTEX are emitted by both vehicles and industrial activities. These findings are comparable to those obtained by previous studies (Miller et al., 2012; Parra et al., 2006; Smith et al., 2007; Su et al., 2010). The concentration ratio is a well-established parameter for determining the emission sources of BTEX compounds (Kerbachi et al., 2006; Khoder, 2007). Table 2 presents the T/B, (m + p)-X/B, and (m + p)-X/EB ratios according to annual and seasonal mean concentrations.

The T/B ratio in the range of 1.5–4.3 indicates that the traffic and vehicular emissions are the main sources of benzene and toluene in the ambient air. The T/B ratio higher than this range shows that these compounds mainly originated from the stationary point sources such as industries (Buczynska et al., 2009; Hoque et al., 2008).

The observed ranges of the (m + p)-X/EB and (m + p)-X/B ratios were 0.63–36.196 and 0.887–7.649, respectively. Following increase in daylight time, concentrations of ethylbenzene and xylenes were decreased. These results are likely related to higher reactivity of xylenes and ethylbenzene than benzene and toluene (Prinn et al., 1987). The mean values of the (m + p)-X/EB and (m + p)-X/B ratios observed in this study were 7.906 and 3.537, which are higher than those observed in other urban areas. These differences might be attributed to the presence of compounds with young photochemical age and fresh emission sources in the study area.

Table 2
Ratio of BTEX species at different monitoring stations.

Monitoring stations	T/B	(m + p)-X/EB	(m + p)-X/B
Aghdasieh	2.307	2.633	2.109
Darrous	2.448	0.63	0.887
Fath	3.775	4.770	3.948
Mahallati	7.393	3.794	5.899
Tarbiat Modares	3.938	36.196	1.896
Tehransar	3.180	2.31	2.375
Sharif	8.922	4.976	7.649
Mean	4.566	7.906	3.537
Max	8.922	36.196	7.649
Min	2.307	0.63	0.887
Spring	5.696	3.208	3.28
Summer	6.434	2.897	5.13
Autumn	3.575	4.805	3.762
Winter	3.209	2.011	2.5

Table 1
Spearman's correlation coefficients between annual and seasonal mean concentration of BTEX compounds (**correlation is significant at the 0.01 level).

	Pollutant	Benzene	Ethylbenzene	(m + p)-Xylene	o-Xylene	Toluene	BTEX
Annual	Benzene	1.000					
	Ethylbenzene	0.628**	1.000				
	(m + p)-Xylene	0.497**	0.531**	1.000			
	o-Xylene	0.566**	0.534**	0.937**	1.000		
	Toluene	0.700**	0.691**	0.867**	0.871**	1.000	
	BTEX	0.72	0.717	0.908	0.910	0.985	1.000
Warmer season	Benzene	1.000					
	Ethylbenzene	0.613	1.000				
	(m + p)-Xylene	0.406	0.443	1.000			
	o-Xylene	0.421	0.462	0.955	1.000		
	Toluene	0.633	0.608	0.856	0.876	1.000	
	BTEX	0.666	0.639	0.901	0.913	0.986	1.000
Cooler season	Benzene	1.000					
	Ethylbenzene	0.704	1.000				
	(m + p)-Xylene	0.648	0.640	1.000			
	o-Xylene	0.0.747	0.608	0.901	1.000		
	Toluene	0.79	0.701	0.911	0.881	1.000	
	BTEX	0.800	0.716	0.934	0.914	0.98	1.000

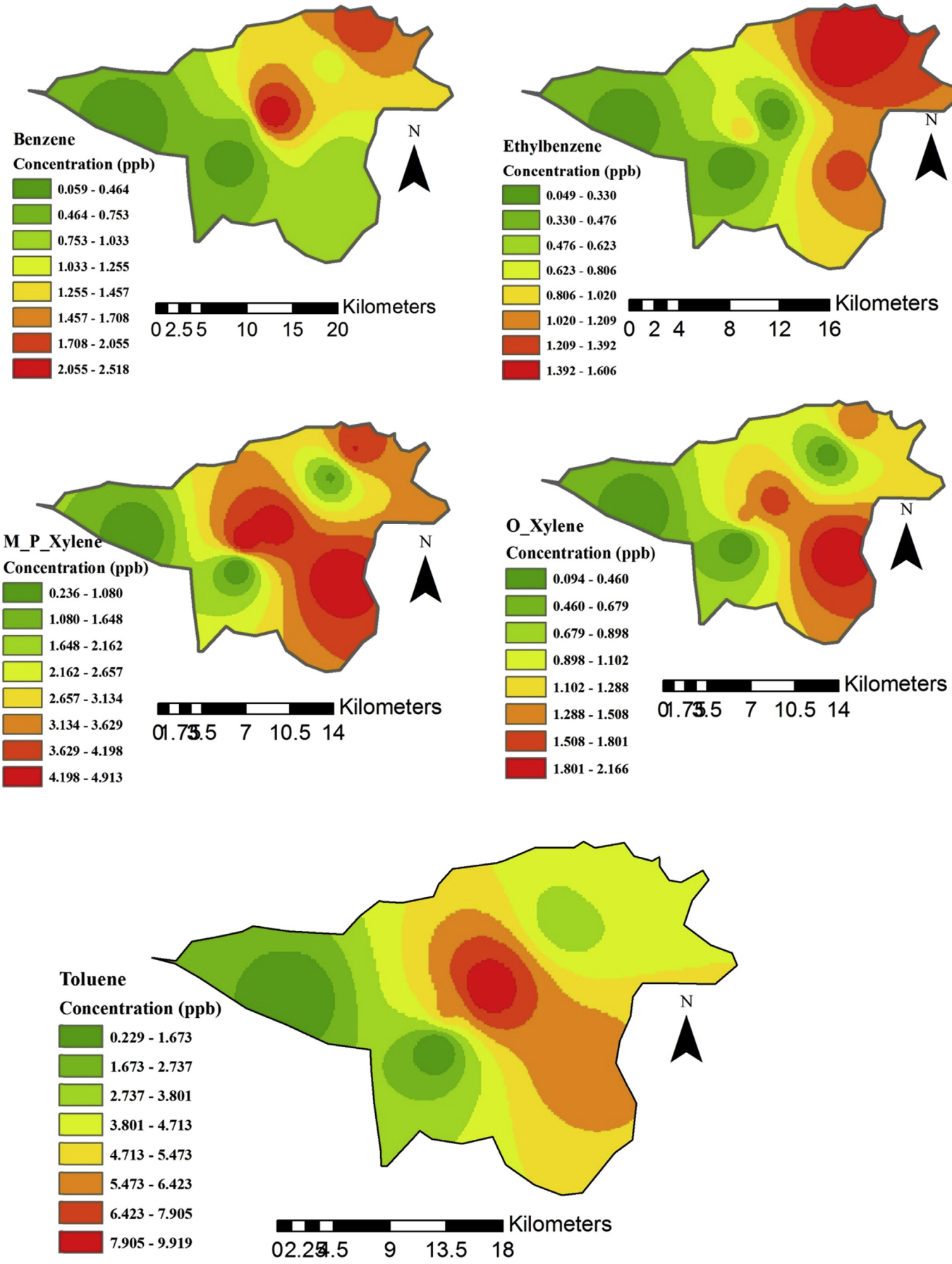

Fig. 3. Spatial mapping of annual mean concentrations (ppb) of BTEX species.

3.4. Spatial analysis of BTEX

The spatial mappings of the annual mean concentration of each BTEX compound and total BTEX are shown in Figs. 3 and 4. These maps show that the maximum concentrations of BTEX were observed in Tarbiat Modares and Mahallati stations. These results might be attributed to the heavy traffic and buildings with high density in these areas, which can trap the pollutants. Another reason is the wind direction, which is mainly from the west to the east and can consequently lead the pollutants to move from the west areas to center and then to the east areas. Individual BTEX compounds showed the same pattern as the total BTEX so that the highest concentration of each compound was observed in areas near highways and heavy traffic conditions. The concentrations of benzene in heavy traffic stations (Tarbiat Modares and Mahallati) were about 3.85–11.44 times higher than that in residential areas (Tehransar and Darros stations). In a consistent study, Thorsson and Eliasson (2006) showed that increase in distance from emission sources (heavy traffic) led to rapid decrease in benzene concentrations. The results for the other BTEX compounds were the same as that of benzene (Fig. 3).

3.5. Health risk assessment

The LTCR of benzene determined by WHO ranges from 1×10^{-6} to 1×10^{-5}, while that recommended by US EPA is $< 10^{-6}$. As shown in Fig. 5, the mean LTCR of benzene for outdoor exposure and for adults is 3.937×10^{-7}, which is lower than the limits recommended by WHO and US EPA.

The result of sensitivity analysis in the form of contribution to the variance is presented in Fig. 6. The percent value related to each parameter determines the contribution of that parameter to the obtained value of LTCR. As shown in the figure, body weight has the highest negative contribution (−51.6%) to the mean LTCR.

The results of HQ are shown in Table 3. If the hazard quotient (HQ) is 1, it can be stated that the concentrations are higher than the threshold limit (RfC) and it is expected to produce harmful effects to the exposed population. Hence, there is need to assess the risk and identify the people being exposed to emissions. If HQ ≤ 1, no health effects are expected, as the exposure is lower than the benchmark limit. According to these data, the HQ of all BTEX compounds is < 1. Therefore, the results obtained in this study show that the noncarcinogenic risk of BTEX compounds is

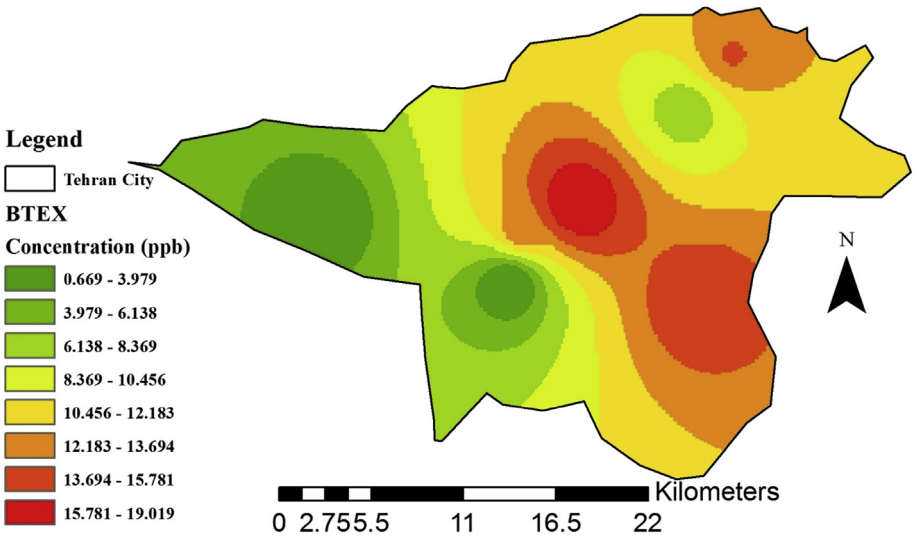

Fig. 4. Spatial mapping of annual mean concentrations (ppb) of total BTEX.

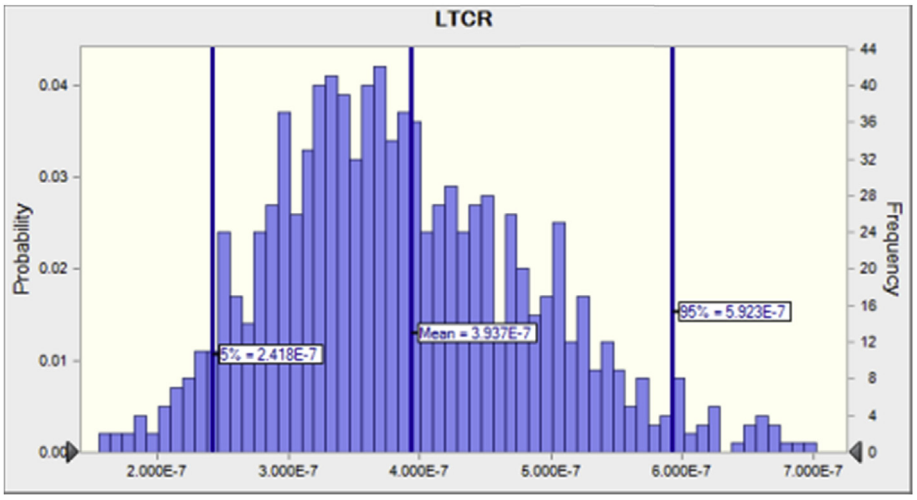

Fig. 5. Predicted probability of lifetime cancer risk (LTCR) for benzene.

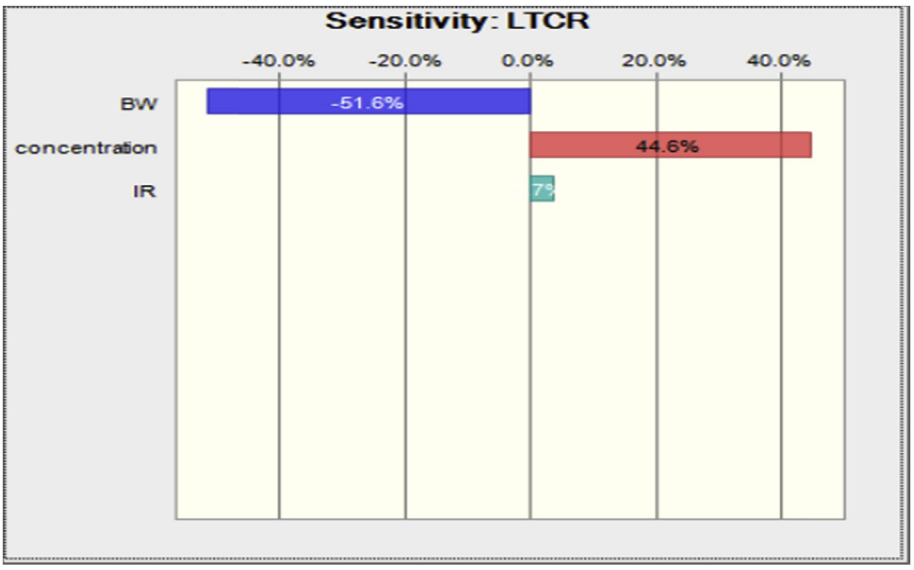

Fig. 6. Sensitivity analysis of lifetime cancer risk (LTCR) model for benzene, (BW: body weight, IR: inhalation rate).

Table 3
Noncarcinogen risk of BTEX in terms of hazard quotient (HQ).

Compound	RfC (mg/m^3)	Concentration (μg/m^3)		Hazard quotient (HQ)	
		Mean	SD	Mean	SD
Benzene	3 (10^{-2})	1.311	0.221	4.376 (10^{-2})	7.329 (10^{-3})
Toluene	5	5.397	1.824	1.065 (10^{-3})	3.700 (10^{-4})
Ethylbenzene	1	1.73	0.83	1.750 (10^{-3})	8.068 (10^{-4})
o-Xylene	1 (10^{-1})	1.676	0.493	1.651 (10^{-2})	5.012 (10^{-3})
(m + p)-Xylene	1 (10^{-1})	4.473	1.17	4.459 (10^{-2})	1.172E (10^{-2})

negligible.

4. Conclusion

This study aimed to measure the concentrations of BTEX compounds in the ambient air of Tehran metropolis and investigate their spatial analysis, seasonal variations, and risk assessment. The most obvious findings of this study are as follows:

1) The annual mean concentrations of benzene, (m + p)-xylene, o-xylene, toluene, and ethylbenzene were 1.056, 2.929, 1.044, 4.291, and 0.837 ppb (or 3.444, 12.734, 4.53, 3.63, and 16.25 μg m^{-3}), respectively.
2) The maximum average concentrations of benzene, (m + p)-xylene, o-xylene, toluene, and total BTEX were observed in spring, while that for ethylbenzene was recorded in winter.
3) The strong positive correlation between BTEX compounds concentrations indicates that the emission sources of all compounds are almost similar.
4) According to the annual mean ratios of (m + p)-X/EB, fresh emission sources in the study area and young age photochemical compounds in the ambient air are responsible for the higher ratio.
5) Spatial analysis indicated that heavy traffic areas had the highest BTEX concentrations, which were reduced with the increase in distances from highways and heavy traffic conditions.
6) The cancer risk of benzene and noncarcinogenic risk of BTEX are in the acceptable range. However, these findings cannot guarantee the health of the exposed population.

In general, the current data highlight the importance of BTEX control in large cities with heavy traffic and industrial activities. There is an urgent need to conduct more studies to identify and control the emission sources of BTEX to reduce their concentrations in the future. In this regard, the most important things that can be implemented include fuel and vehicle standardization, developing public transport systems, and industrial emission control.

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