Spatial distribution and health risk of exposure to BTEX in urban area: a comparison study of different land-use types and traffic volumes

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Abstract Many previous studies have investigated BTEX concentrations in urban areas; however, the available evidence on the association of different land-use types and BTEX concentrations is still scarce. In this study, the BTEX concentrations were measured and compared in different land-use types and traffic volumes of Mashhad metropolis, Iran. Sampling was conducted in summer and winter of 2018 based on NIOSH 1501 method in six land-use types, including *Residential*, *Commercial/official*, *Industrial*, *Greenspace*, *Transportation,* and *Tourism*. The spatial autocorrelation model was used to investigate the emission pattern. The Monte Carlo simulation technique and sensitivity analysis were used to assess the health risk of exposure to BTEX compounds. The median [interquartile range (IQR)] of benzene, toluene, ethylbenzene m-xylene, o-xylene and total BTEX concentrations based on overall mean were 4 (2.23), 8.37 (4.48), 1.2 (1.46), 0.89 (2.59), 0.8 (1.73) and 17.7 (8.19) $\mu g/m^3$, respectively. Benzene and toluene had clustered emission patterns (z-score > 1.96). Exposure to benzene in the study area had a carcinogenic risk for inhabitants. The concentration of BTEX compounds was significantly different based on land-use type. The maximum and minimum concentrations of BTEX were observed in *Transportation* and *Greenspace* land uses, respectively. The BTEX concentrations in summer were significantly higher than in winter, and traffic had a significant effect on BTEX concentrations. Overall, our results supported a significant relationship between land-use type and BTEX concentrations in the urban area. Moreover, ambient

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benzene concentration had a carcinogenic risk potential for inhabitants of study area.

Introduction

In the recent decade, air pollution has been introduced as one of the major problems in the urban areas, especially in metropolitan cities (Gulia et al. 2015; Nabizadeh et al. 2018; Mokhtari et al. 2019). Growing urbanization, industrial activities and consumerism have led to higher pollutant emissions and decrease air quality in urban areas (Miri et al. 2019; Ghaffari et al. 2017). The concentration and type of ambient pollutants are largely dependent on their emission source. Volatile organic compounds (VOCs) are released into the atmosphere through natural and anthropogenic sources (Miri et al. 2016a, b, c). Traffic and industrial activities have been recognized as the main anthropogenic sources of VOCs in the urban area (Zhang et al. 2012). After discharge of these pollutants to the air, they can participate in photochemical reactions and produce secondary pollutants like tropospheric ozone, peroxyacetyl nitrates (PAN) (Hajizadeh et al. 2018).

Benzene, toluene, ethylbenzene, xylenes (BTEX) have been identified as carcinogenic, mutagenic and toxic at detectable concentrations in the ambient air (Khoder 2007). Benzene is listed as a carcinogenic compound for humans (EPA 1989). Several adverse health effects, including chronic and acute nonlymphocytic and myeloid leukemia, anemia and immunological effects, have been associated with exposure to benzene (Bird et al. 2010; Pyatt and Hays 2010; Belson et al. 2007). Moreover, exposure to xylene can affect the nervous system, irritation in the eyes, nose and throat and damage the lungs (Kandyala et al. 2010; Niaz et al. 2015; Garg and Gupta 2019).

The available evidence on BTEX compounds in Asian cities reported a high level of these pollutants in the heavy traffic areas, e.g., near to traffic lanes and transportation areas (e.g., bus terminals) (Truc and Kim Oanh 2007; Chan et al. 2003; Phuc and Kim Oanh 2018) and other local activities such as industrial and commercial activities of inside cities, loading and refueling activities at gasoline stations, residential cooking and open burning of solid waste and agricultural residues (Dehghani et al. 2017; Zalel et al. 2008; Mohammadi et al. 2020; Mehdi Mokhtari et al. 2016). Moreover, it has been reported that BTEX was one-third lower inside parks compared to built-up areas (Milazzo et al. 2019). However, to our knowledge, there is no study on comparing the ambient BTEX concentrations in different land-use types as well as different traffic density, simultaneously. Therefore, in the present study, BTEX concentrations were measured and compared in six different land-use types and traffic volumes of Mashhad metropolis, Iran. The spatial distribution and emission pattern of each compound were investigated, and finally, Monte Carlo simulation technique was used to assess the probabilistic health risk of exposure to BTEX.

Material and methods

Study area

This study was conducted in Mashhad metropolis, Iran. Mashhad is the capital of Razavi Khorasan Province and is located in northeastern Iran (longitude of 59° 35′ E to 59° 74′ E and latitude of 36° 14′ N to 36° 48′ N). This city is the second-largest metropolitan city of Iran. The area of Mashhad is about 300 km². According to the last census in 2016, the city's population is about 2,800,000 people (Miri et al. 2016a; Iran 2016). Mashhad has an arid climate with annual average rainfall and relative humidity of 251 mm and 53%, respectively. The annual mean, minimum and maximum temperatures in the city are 14, −28 (in January) and 44 °C (in August), respectively. Sampling locations, highways, main streets and different land-use types of Mashhad are presented in Fig. 1.

Sampling and BTEX analysis

The samples were collected during the summer and winter of 2018 in the morning between 8:00 and 10:00 AM local time. The land-use map of the study area prepared by the municipality of Mashhad (2017) was used to select the sampling location. Six important land-use types, including *Residential*, *Commercial/ official*, *Industrial*, *Greenspace*, *Transportation,* and
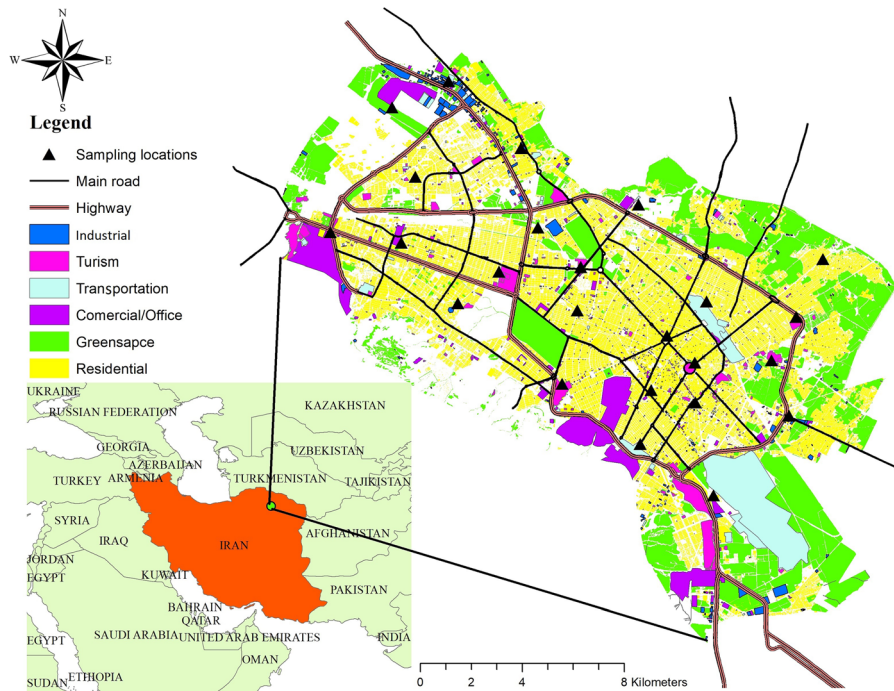
Fig. 1 Study area with different land-use types, major roads, and sampling locations

Tourism, were determined in ArcGIS v10.5 software. Four sampling locations of each land-use type were selected (two samples in high-traffic areas and two samples in low-traffic areas), in such a way that all areas of the city were covered (overall, 24 sampling locations). Due to the limitation in the budget, it was not possible to select more sampling locations. High and low traffic density areas were determined based on the traffic volume layer prepared by Transportation and Traffic Organization of Mashhad (2017). According to the traffic density of Mashhad, we selected 3000 vehicles per day as a cutoff between high and low traffic density areas. The days with unsuitable weather conditions, e.g., rainy days, were excluded from our sampling. The National Institute for Occupational Safety and Health (NIOSH)-1501 method (i.e., using charcoal sorbent tubes (SKC Inc., England, 226–01) and SKC pump with a flow rate of 0.2 L/min) was applied for sampling BTEX compounds in Mashhad. A soap bubble flow meter was used to calibrate the pumps before sampling, manually. The sorbent tubes were stored at 4 °C after sampling and transported to the laboratory for analysis.

The BTEX compounds were desorbed by 1 mL carbon disulfide (CS_2). A gas chromatograph with a flame ionization detector (GC-FID, Agilent Technology, 7890) was used to measure the BTEX compounds immediately after the desorption of the samples. Table S1 of Supplemental Materials represents the details of the GC-FID condition during sampling analysis. Moreover, validation of BTEX measurements, including details of the calibration curve, the limit of detection (LOD), method quantitation limit (MQL), and blank method, was fully described on page 8 of Supplemental Materials.

Spatial analysis

Global Moran's Index (Moran's I) was used to assess the spatial distribution pattern (which could be dispersed, random, or clustered) of BTEX compounds in summer and winter in Mashhad. In this model, spatial autocorrelation of each pollutant can be measured based on its value and location. In Moran's I, the null hypothesis is that the distribution of values is random across the study area (Baller et al. 2001; Anselin 1993). In the spatial autocorrelation model, in addition to Moran's I, z score and P value can be measured to evaluate the significance of mentioned

index (Miri et al. 2018a, b). Details of Moran's I were fully described in page 9 of Supplemental Material.

Ordinary kriging (OK) was used to produce a spatial mapping of BTEX compounds in summer and winter. The optimum transportation type (Log or Box-Cox) and the optimum semivariogram type based on minimum root-mean-square error (RMSE) were applied to reduce estimate errors. RMSE and %RMSE were applied for validation of model performance. RMSE% was also used as another validation indicator to tackle the sensitivity of the RMSE to the outlier. The RMSE% less than 40% indicated good model performance; values ranged between 40 and 70% were considered an acceptable range and higher than 70% indicated that the estimated values not better than average values and model are rejected (Hengl et al. 2004). Details of OK models and model performance are provided in pages 10 and 11 of Supplemental Materials.

Health risk assessment

Health risks of exposure to BTEX compounds (i.e., carcinogenic and noncarcinogenic risks) were evaluated using a method developed by USEPA (EPA 1989). We used Monte Carlo simulations to calculate the lifetime cancer risk (LTCR) of inhalation exposure to benzene and noncarcinogenic risk [i.e., hazard quotient (HQ)] of inhalation exposure to BTEX. The risk assessment was performed with 10,000 iterations using Crystal Ball software (version 11.1.2.4, Oracle, Inc., USA). The effect of imported variables in assessing estimated LTCR was achieved using sensitivity analysis. Details of risk assessment and sensitivity analysis were provided in pages 11 and 12 of Supplemental Materials.

Statistical analysis

The Shapiro–Wilk test was used to assess the distribution of BTEX data. The seasonal differences (summer vs. winter) and differences in BTEX concentrations based on traffic (low vs. high traffic volume) were assessed using T test and Mann–Whitney test. The difference in BTEX concentrations based on different land-use types was evaluated using one-way ANOVA and the Kruskal–Wallis. Moreover, post hoc test was performed to compare the concentration of each BTEX species in different land-use

types. Stata software version 15 (Stata Corp LP, College Station, Texas) was performed for all analyses.

Results

BTEX concentration

Statistical summary of measured BTEX compounds measured in two seasons (summer and winter) and based on the overall mean in Mashhad's ambient air is shown in Fig. 2. The median [interquartile range (IQR)] of benzene, toluene, ethylbenzene m-xylene, o-xylene and total BTEX concentration in summer were 4.72 (2.34), 9.77 (4.92), 1.36 (2), 0.8 (2.91), 0.8 (2.50) and 21.08 (9.86) μg/m^3, respectively. The median (IQR) of these compounds in winter was 3.08 (2.04), 6.46 (4.07), 0.76 (0.91), 0.8 (2.08), 0.8 (1) and 13.63 (7.42) μg/m^3, respectively. The median (IQR) of benzene, toluene, ethylbenzene m-xylene, o-xylene and total BTEX concentration based on overall mean were 4 (2.23), 8.37 (4.48), 1.2 (1.46), 0.89 (2.59), 0.8 (1.73) and 17.7 (8.19) μg/m^3, respectively. Toluene had the maximum concentration between BTEX compounds in both seasons. Moreover, xylene and ethylbenzene had minimum concentrations in summer and winter, respectively. The mean temperature, humidity, wind direction and wind speed measured in the summer were 24.5 °C, 25%, 50 and 8 m/s, respectively; however, for winter, they were 6.6 °C, 54%, 300 and 11 m /s, respectively.

Correlation between BTEX compounds

Results of Spearman rank correlations among BTEX species based on overall and seasonal average are shown in Fig. 3. The benzene concentration was significantly correlated with toluene, m-xylene, ethylbenzene and total BTEX in summer ($r = 0.44$ to 0.99, P-value < 0.05). Furthermore, toluene concentration was significantly correlated with other compounds ($r = 0.46$ to 0.99, P-value < 0.05) except o-xylene ($r = 0.20$) in summer (Fig. 3). Based on winter measurements, a strong significant correlation was observed between benzene with toluene, m-xylene and total BTEX ($r = 0.41$ to 0.99, P-value < 0.05). A significant correlation was observed between toluene and m-xylene ($r = 0.41$, P value < 0.05). However,
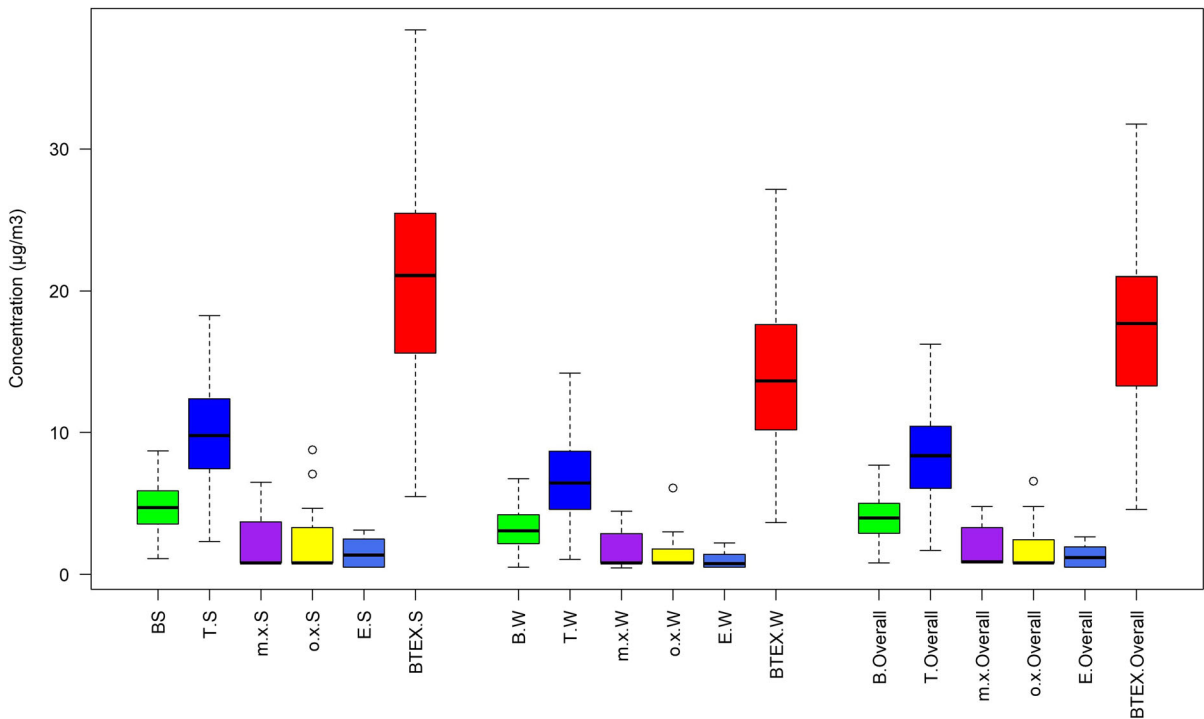
Fig. 2 Descriptive statistics of BTEX concentrations based on seasonal and the overall mean

there was a week correlation between o-xylene and other BTEX species based on summer and winter and overall mean (Fig. 3).

The averages (ranges) of toluene/benzene ratio for summer, winter and overall mean were 2.08 (1.73 to 2.10), 2.09 (1.97 to 2.21) and 2.09 (1.88 to 2.14), respectively.

Spatial distribution

The OK models for BTEX species in summer and winter are presented in Figs. S1 and S2 of Supplemental Materials, respectively. Based on RMSE and % RMSE, OK models for benzene, toluene and total BTEX had a good performance to estimate pollutant concentration in summer and winter (%RMSE < %40). The OK models for ethylbenzene had acceptable performance (%RMSE between 40 and %70). However, the %RMSE for m-xylene and o-xylene was higher than the acceptable range (%RMSE > %70) (Table S2 of Supplemental Material). The maximum concentration of benzene and toluene in summer was observed in the center of the study area (near main roads with heavy traffic, *Transportation,* and *Tourism*

land-use type). However, the highest ethylbenzene and o-xylene concentrations were observed in the west and northwest areas of Mashhad (near highways and *Industrial* land-use type). The maximum concentration of m-xylene was estimated near transportation land use and highways in the southwest of the study area in summer. It should be mentioned that the distribution of BTEX compounds in winter was almost the same as in summer (Figs. S1 and S2 of Supplemental Materials).

The results of the spatial emission pattern (Moran's I) are presented in Table 1. In this model, the z score value lower than −1.65 indicates a dispersed emission pattern, z score between −1.65 and 1.65 represents random emission and z score higher than 1.65 indicates clustered emission pattern (Miri et al. 2018a, b; Dai et al. 2014). The spatial distribution patterns of benzene and toluene in summer, winter and overall mean were clustered. Given the z-score of 2.74 for benzene and 2.76 for toluene in summer, there was less than 1% likelihood that these clustered patterns could result from random chance. For benzene and toluene in winter with z score of 2.38 and 2.29, there was less than 5% likelihood that these clustered
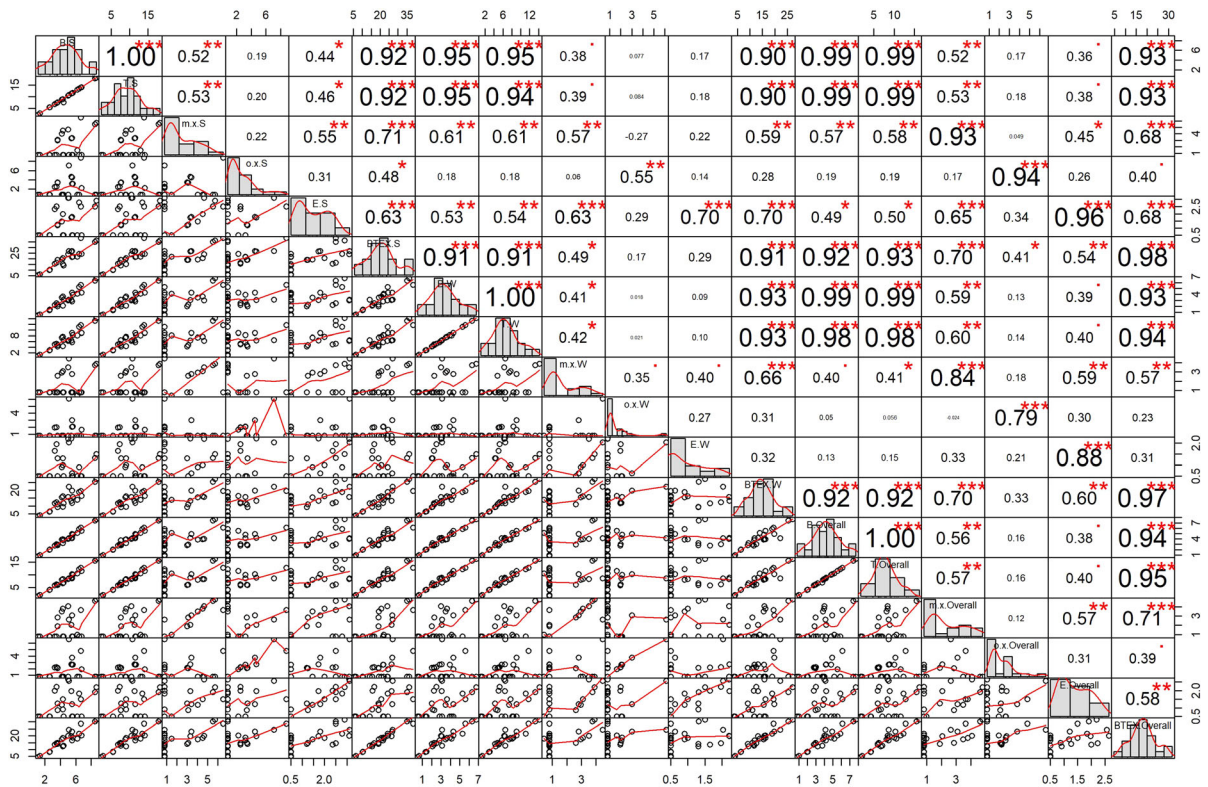
Fig. 3 Correlation matrix, scatter plot and histogram of BTEX concentrations (*significant level of 0.05, **significant level of 0.01 and ***significant level of 0.001)

	Pollutant		Moran's Index	z-score	p value	Distribution pattern
Table 1 Emission patterns of BTEX compounds in Mashhad	Summer	Benzene	0.41	2.74	0.01	Clustered
		Toluene	0.41	2.76	0.01	Clustered
		Ethylbenzene	− 0.10	− 0.34	0.73	Random
		m-xylene	0.01	0.27	0.79	Random
		o-xylene	-0.18	− 0.86	0.39	Random
		Total BTEX	0.09	0.78	0.43	Random
	Winter	Benzene	0.35	2.38	0.02	Clustered
		Toluene	0.33	2.29	0.02	Clustered
		Ethylbenzene	− 0.20	− 0.91	0.36	Random
		m-xylene	− 0.09	− 0.27	0.79	Random
		o-xylene	− 0.18	− 1.07	0.29	Random
		Total BTEX	0.18	1.38	0.17	Random
	Overall mean	Benzene	0.40	2.67	0.007	Clustered
		Toluene	0.39	2.65	0.008	Clustered
		Ethylbenzene	− 0.15	− 0.62	0.54	Random
		m-xylene	− 0.05	− 0.03	0.97	Random
		o-xylene	− 0.24	− 1.25	0.21	Random
		Total BTEX	0.13	1.03	0.30	Random

Table 2 Hazard quotient (HQ) and chorionic daily intake (CDI) of exposure to BTEX compound in Mashhad

Pollutant			Percentile 5th	Median	Mean	SD	Percentile 95th
Summer	Benzene	CDI	5.41×10^{-4}	1.10×10^{-3}	1.21×10^{-3}	5.54×10^{-4}	2.26×10^{-3}
		HQ	7.82×10^{-2}	1.47×10^{-1}	1.58×10^{1}	2.29×10^{-2}	2.76×10^{-1}
	Toluene	CDI	1.12×10^{-3}	2.30×10^{-3}	2.53×10^{-3}	1.16×10^{-3}	4.72×10^{-3}
		HQ	9.71×10^{-4}	1.84×10^{-4}	1.98×10^{-3}	7.96×10^{-4}	3.47×10^{-3}
	Ethylbenzene	CDI	1.14×10^{-4}	3.18×10^{-4}	3.87×10^{-4}	2.69×10^{-4}	8.93×10^{-4}
		HQ	4.79×10^{-4}	1.27×10^{-3}	1.51×10^{-3}	9.80×10^{-4}	3.37×10^{-3}
	Xylene	CDI	3.24×10^{-4}	966×10^{-4}	1.19×10^{-3}	8.59×10^{-4}	2.80×10^{-3}
		HQ	1.41×10^{-2}	3.86×10^{-2}	4.66×10^{-2}	3.15×10^{-2}	1.06×10^{-1}
winter	Benzene	CDI	3.14×10^{-4}	7.27×10^{-4}	8.32×10^{-4}	4.57×10^{-4}	1.70×10^{-3}
		HQ	4.49×10^{-2}	9.70×10^{-2}	1.08×10^{-1}	5.39×10^{-2}	2.10×10^{-1}
	Toluene	CDI	6.55×10^{-4}	1.52×10^{-3}	1.73×10^{-3}	9.45×10^{-4}	3.51×10^{-3}
		HQ	5.65×10^{-4}	$1,22 \times 10^{-3}$	1.36×10^{-3}	6.69×10^{-4}	2.61×10^{-3}
	Ethylbenzene	CDI	8.32×10^{-5}	2.18×10^{-4}	2.60×10^{-4}	1.68×10^{-4}	5.77×10^{-4}
		HQ	3.54×10^{-4}	8.76×10^{-4}	1.02×10^{-3}	6.06×10^{-4}	2.17×10^{-3}
	Xylene	CDI	2.17×10^{-4}	6.22×10^{-4}	7.66×10^{-2}	5.45×10^{-4}	1.79×10^{-3}
		HQ	9.19×10^{-3}	2.48×10^{-2}	3.0×10^{-2}	2.0×10^{-2}	6.77×10^{-2}
Overall mean	Benzene	CDI	4.29×10^{-4}	9.16×10^{-4}	1.02×10^{-3}	4.99×10^{-4}	1.96×10^{-3}
		HQ	6.20×10^{-2}	1.22×10^{-1}	1.33×10^{-1}	5.76×10^{-2}	2.41×10^{-1}
	Toluene	CDI	9.02×10^{-4}	1.92×10^{-3}	2.14×10^{-3}	1.05×10^{-3}	4.11×10^{-3}
		HQ	7.77×10^{-4}	1.54×10^{-3}	1.67×10^{-3}	7.24×10^{-4}	3.03×10^{-3}
	Ethylbenzene	CDI	1.02×10^{-4}	2.69×10^{-4}	3.21×10^{-4}	2.08×10^{-4}	7.17×10^{-4}
		HQ	4.33×10^{-4}	1.08×10^{-3}	1.26×10^{-3}	7.57×10^{-4}	2.70×10^{-3}
	Xylene	CDI	1.66×10^{-4}	4.17×10^{-4}	4.88×10^{-4}	2.99×10^{-4}	1.05×10^{-3}
		HQ	7.06×10^{-3}	1.66×10^{-2}	1.91×10^{-2}	1.08×10^{-2}	3.94×10^{-2}

patterns could result from random chance. Gives z score of 2.67 for benzene and 2.65 for toluene in overall mean, there are less than 1% likelihoods, in which the observed clustered patterns could be due to random chance (Table 1). For other BTEX species based on seasonal and overall mean, the emission pattern did not appear to be significantly different than random (z score ranged between −1.25 and 1.38) (Table1).

Health risk assessment

The hazard quotient (HQ) results and chronic daily intake (CDI) of exposure to BTEX based on seasonal and overall mean concentrations are presented in Table 2. The minimum mean (percentile 95th) and maximum mean (percentile 95th) of HQ were estimated for ethylbenzene in winter [1.02×10^{-3} (2.17×10^{-3})] and benzene in summer [1.58×10^{-1} (2.76×10^{-1})]. The BTEX compounds in summer and winter had not noncarcinogenic risk for inhabitants of Mashhad (HQ < 1) (Table 2).

Figure 4 shows the results of LTCR through exposure to benzene based on summer, winter and the overall mean. The mean and 95th percentile of LTCR for summer were 2.67×10^{-6} and 4.79×10^{-6}, for winter 1.83×10^{-6} and 3.73×10^{-6} and for overall mean concentration 2.25×10^{-6} and 4.32×10^{-6}. The LTCR values indicated that benzene concentration in Mashhad had a carcinogenic risk for inhibitions (USEPA thresholds level 1×10^{-6}) (M. Miri et al. 2016a, b, c).

Sensitivity analyses of LTCR for annual and seasonal mean showed that the most influential variable in increasing the carcinogenic risk was pollutant concentration (C) (correlation rank ranged
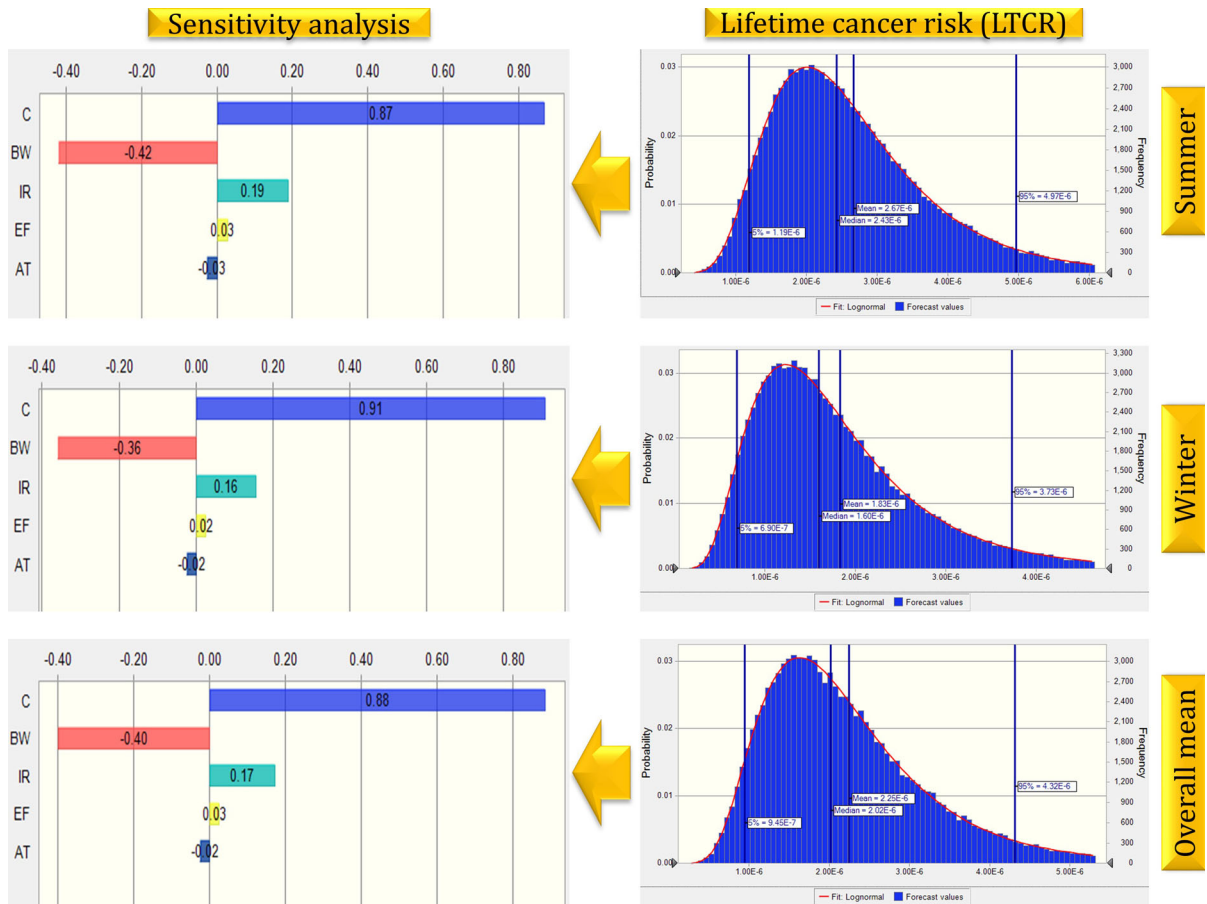
Fig. 4 Carcinogenic risk and sensitivity analysis of exposure to benzene

0.87–0.91). In contrast, body weight (BW) had an inverse correlation with LTCR (correlation rank ranged from −0.36 to −0.42) (Fig. 4).

Association with land-use type and traffic

Benzene

A significant difference was observed between the mean concentrations of benzene in summer and winter (mean difference = 1.51, 95% confidence interval (CI): 1.25, 1.78, *P* value < 0.001) (Table S3 of Supplemental Materials). Moreover, benzene concentration in low-traffic areas was significantly lower than high-traffic areas (*P* value = 0.03, 0.02 and 0.03, for summer, winter and overall mean, respectively) (Table S4 of Supplemental Materials).

A significant difference was observed between benzene concentrations in different land-use types

(*P* value = 0.002 for all models). The results of post hoc analyses showed that *Transportation* land use had the highest and *Greenspace* land use had the lowest benzene concentration in summer, winter and overall mean (Table 3). The overall order for benzene based on land-use type was *Transportation* > *Tourism* > *Commercial/official* > *Industrial* > *Residential* > *Greenspace* (Table 3).

Toluene

A significant difference was observed between toluene concentrations in summer and winter (mean difference = 3.13, 95% CI: 2.53, 3.74, *P* value < 0.001) (Table S3 of Supplemental Materials). Furthermore, this pollutant concentration in low-traffic areas was significantly lower than high-traffic areas (*P* value = 0.03, 0.02 and 0.03, respectively) (Table S4 of Supplemental Materials).

Table 3 Concentration of BTEX compound in different land-use types based on summer, winter and the overall mean

Pollutant		Residential	Commercial/official	Industrial	Greenspace	Transportation	Tourism
Summer	Benzene	4.23 (0.98)*	5.44 (1.27)	4.55 (1.05)	1.92 (0.81)	6.42 (2.51)	6.01 (0.58)
	Toluene	8.86 (2.05)	11.02 (3.2)	9.54 (2.2)	4.04 (1.7)	13.49 (5.27)	12.63 (1.22)
	Ethylbenzene	2.77 (0.40)	1.57 (1.24)	1.5 (0.31)	0.5 (0)	2.22 (0.62)	0.5 (0)
	m-xylene	1.76 (1.93)	2.23 (2.85)	3.2 (0.18)	0.8 (0)	4.93 (0.8)	0.98 (0.35)
	o-xylene	3.23 (2.69)	2.80 (3.99)	4.02 (0.66)	0.8 (0)	0.8 (0)	2.36 (1.13)
	Total BTEX	20.85 (5.48)	23.05 (11.19)	22.8 (4.38)	8.06 (2.51)	27.85 (9.14)	22.47 (1.35)
Winter	Benzene	2.71 (0.40)	4.02 (1.7)	2.91 (1.02)	0.98 (0.48)	5.06 (1.72)	3.81 (0.66)
	Toluene	5.75 (0.76)	8.44 (3.57)	6.12 (2.15)	2.06 (1)	10.62 (3.62)	7.79 (1.45)
	Ethylbenzene	1.96 (0.23)	0.8 (0.39)	1.16 (0.79)	0.5 (0)	1.18 (0.28)	0.5 (0)
	m-xylene	2.39 (1.88)	0.8 (0)	1.75 (1.1)	0.8 (0)	3.43 (0.47)	0.71 (0.18)
	o-xylene	2.71 (2.33)	0.8 (0)	1.35 (1.1)	0.8 (0)	0.8 (0)	1.66 (0.61)
	Total BTEX	15.52 (4.80)	14.87 (5.08)	13.29 (4.62)	5.15 (1.47)	21.08 (6.08)	14.47 (1.96)
Overall mean	Benzene	3.47 (0.69)	4.73 (1.48)	3.73 (3)	1.45 (0.64)	5.74 (2.11)	4.91 (0.58)
	Toluene	7.31 (1.4)	9.73 (3.3)	7.83 (2.1)	3.05 (1.35)	12.05 (2.44)	10.21 (1.24)
	Ethylbenzene	2.36 (0.3)	1.19 (0.6)	1.33 (0.46)	0.5 (0)	1.70 (0.45)	0.5 (0)
	m-xylene	2.08 (1.59)	1.51 (1.43)	2.48 (0.59)	0.8 (0)	4.18 (0.64)	0.84 (0.09)
	o-xylene	2.97 (2.5)	1.8 (2)	2.68 (0.77)	0.8 (0)	0.8 (0)	2.01 (0.87)
	Total BTEX	18.19 (5.05)	18.96 (7.87)	18.05 (4.29)	6.06 (2)	24.47 (7.61)	18.47 (1.53)

*Concentration of all compounds as mean and standard deviations. Post hoc analysis was used to compare BTEX concentration between different land-use types

A significant difference was observed between the toluene concentrations based on different land-use types (P value = 0.004, 0.002, and 0.002 for summer, winter and overall mean, respectively). The maximum and minimum toluene concentrations were observed in *Transportation* and *Greenspace* land use (Table 3). The overall order was *Transportation > Tourism > Commercial/official > Industrial > Residential > Greenspace* for toluene concentration (Table 3).

Ethylbenzene

The average concentration of ethylbenzene in summer was significantly higher than winter (mean difference = 0.49, 95% CI: 0.19, 0.80, P-value = 0.003) (Table S3 of Supplemental Materials). The summer mean concentration of ethylbenzene in high-traffic area was significantly higher than low-traffic areas (P-value = 0.049); however, there was not a significant difference for the mean concentration of ethylbenzene in winter as well as the overall mean between high-

and low-traffic areas (P-value = 0.539 and 0.115, respectively) (Table S4 of Supplemental Materials).

A significant difference was observed between ethylbenzene concentration and different land-use types in summer, winter and overall mean (P value = 0.005, 0.005 and 0.002, respectively). The highest concentration of ethylbenzene was observed in *Residential* land use, and the lowest concentration was in *Greenspace* and *Tourism* land use (Table 3).

Xylene

The concentration of o-xylene was significantly higher in summer compared to winter (P value = 0.01). However, the difference in m-xylene concentration in summer and winter (P value = 0.052) was not statistically significant (Table S3 of Supplemental Materials). The concentration of m-xylene in low-traffic areas was significantly lower than in high-traffic areas. However, there was no significant difference between the concentrations of o-xylene in high- and low-traffic

areas for summer, winter and overall mean (Table S4 of Supplemental Materials).

A significant difference was observed between xylene concentration and different land-use types in summer, winter and overall mean (P value < 0.05). The highest concentration of m-xylene was observed in *Residential* land use, and the lowest concentration was in *Greenspace* land use (Table 3).

Total BTEX

The concentration of total BTEX in summer was significantly higher than in winter (mean difference = 6.78, 95% CI: 5.15, 8.42, P value < 0.001) (Table S3 of Supplemental Materials). The concentration of total BTEX in low-traffic areas was significantly lower than high-traffic areas (P value < 0.05) (Table S4 of Supplemental Materials).

A significant difference was observed between total BTEX concentration and different land-use types (P value < 0.05). The highest and lowest BTEX concentration were observed in *Transportation* and *Greenspace* land-use types (Table 3). The order of overall mean of total BTEX concentration based on land-use type was *Transportation* > *Commercial/official* > *Tourism* > *Residential* > *Industrial* > *Greenspace* (Table 3).

Discussion

To the best of our knowledge, this is one of the first studies on comparing ambient BTEX concentrations in six different land-use types as well as high- and low-traffic areas. We found a significant difference between the concentrations of BTEX compounds in different land-use types. The highest concentration of these compounds was observed in *Transportation*, and the lowest concentration was observed in *Greenspace* land-use type. Moreover, BTEX concentration in summer was significantly higher than winter, and traffic significantly affected BTEX concentrations. Benzene and toluene in summer, winter and overall mean had clustered emission patterns, and exposure to benzene had carcinogenic risk for Mashhad's inhabitants (higher than USEPA thresholds level, i.e., 1×10^{-6}).

BTEX concentration

In our study, the highest and lowest BTEX concentrations were observed for toluene and ethylbenzene. Other studies in Spain, China, Canada (Li et al. 2014; Miller et al. 2012; Wang et al. 2010; Parra et al. 2009; Rad et al. 2014) and Iran (Miri et al. 2016a, b, c; Hazrati et al. 2016) showed the same results. Based on the overall mean, the range of total BTEX was from 4.58 to 31.75 μg/m³, which was lower than previous studies in India (150–400 μg/m³) (Hoque et al. 2008) and Turkey (3.7–335.5 μg/m³) (Pekey and Yılmaz 2011). In a study by Carlsen et al. (2017), the concentration of BTEX compounds in Almaty, Kazakhstan, was investigated and compared with 19 major cities around the world (Carlsen et al. 2018). Comparison of our results with these cities indicated that Mashhad was the 16th polluted city among these major cities worldwide.

A strong correlation was observed between benzene and other compounds, especially toluene, in summer and winter, indicating that these pollutants could have the same emission sources (Golkhorshidi et al. 2019; Hajizadeh et al. 2018). Previous studies in other urban areas also reported similar results (Phuc and Kim Oanh 2018; Liu et al. 2018; Hajizadeh et al. 2018). Moreover, the mean T/B ratios for summer, winter and overall mean were 2.08, 2.09 and 2.09, respectively. The concentration ratio is a well-established parameter for identifying the BTEX compounds in ambient air (Khoder 2007). The T/B ratio between 1.5 and 4.3 indicated that the main emission sources of toluene and benzene are vehicular emission and traffic (Miri et al. 2016a, b, c). Therefore, the main source of toluene and benzene in the ambient air of Mashhad could be traffic.

Spatial analysis

The spatial autocorrelation model (Moran's I) indicated that benzene and toluene have a clustered emission pattern in summer and winter, while other compounds have random emission patterns. Our finding indicated that benzene and toluene have neighboring pollutant emissions (Z score higher than 1.96), and maybe some specific sources, including heavy traffic points (e.g., intersections and squares), bus satiation and terminal and other point emission sources, had a high effect on the concentration of these

pollutants in Mashhad (Dong and Liang 2014). The emission pattern of pollutants in the urban area can affect the distribution pattern of related diseases. Overall, the people who live near pollutants' emission sources are at a higher risk (Requia et al. 2017; Zhou et al. 2018). Although we could not find any study on the distribution pattern of BTEX compounds in urban area, other studies which investigated the distribution pattern of other air pollutants indicated that there was an overlap and clustering relationship between hot spot areas of air pollutants (e.g., particulate matters) and lung cancer, mortality rate, asthma incidence and hospital admission (McEntee and Ogneva-Himmelberger 2008; Zhang and Tripathi 2018; Han et al. 2017).

Risk assessment

The risk assessment results indicated that Mashhad's inhabitants were at the cancer risk related to exposure to benzene (LTCR $> 10^{-6}$). Previous studies in other countries, e.g., India (LTCR $= 4.6 \times 10^{-6}$) (Masih et al. 2016), China (LTCR $= 1.34 \times 10^{-5}$) (Li et al. 2014) and Thailand (LTCR $= 1.75 \times 10^{-4}$) (Tunsaringkarn et al. 2012) and Iran (LTCR for morning $= 1.96 \times 10^{-4}$ and evening $= 2.49 \times 10^{-4}$) (Dehghani et al. 2018) reported consistent results with our findings. However, the LTCR of exposure to benzene in Tehran, Iran and Nanjing, China, was at a safe level (Miri et al. 2016b; Hu et al. 2010).

In our study, the risk estimation uncertainties of exposure to BTEX were assessed using Monte Carlo simulation technique. The most important variables that could affect LTCR were identified through sensitivity analysis. Nevertheless, the risk assessment process could be affected by other uncertainties. In this study, the health risk of exposure to BTEX was only investigated based on the inhalation pathway. However, BTEX could affect human health through dermal exposure (Malik et al. 2019), water and foods (i.e., ingestion pathway) (Arambarri et al. 2004; Kelley et al. 1997) that were not investigated in our study. Furthermore, occupational exposure (Omidi et al. 2019) and indoor exposure (Hazrati et al. 2015; Guo et al. 2003) to the BTEX compound were not evaluated. Besides, the spring and autumn were lost from our measurements, while the concentrations of the BTEX compounds were measured using an appropriate method and the uncertainties in

measurements were reduced using duplicate experiments. Finally, in this study, the health risk of exposure to ambient BTEX in Mashhad could be underestimated because the occupational and indoor exposure to BTEX was not evaluated; moreover, dermal and ingestion exposure pathways were not assessed, which could increase the carcinogenic and noncarcinogenic risks. Thus, additional data of different sources and different exposure pathways should be considered in future studies to accurate and precise estimate risks of exposure to BTEX compounds.

Comparison between seasons, traffic and different land-use types

In this study, a significant difference was observed between the concentrations of BTEX compounds in summer and winter (summer > winter). Hajizadeh et al. 2018 reported a significant difference between BTEX compounds in summer and winter (Hajizadeh et al. 2018). Moreover, other studies reported that the correlation between the BTEX compounds in summer and winter was stronger than in other seasons (Amini et al. 2017; Miri et al. 2016a, b, c). Although high correlation indicated the same origin and emission source of these pollutants, the higher temperature in summer can increase the evaporation of BTEX compounds from fuel storage tanks, fuel pumps and vehicles; therefore, the concentration of these pollutants could significantly increase in summer (Hajizadeh et al. 2018; Perez-Rial et al. 2009). In contrast, the lower concentration of BTEX compounds in winter can be due to more atmospheric precipitation and washout of these pollutants to surface area (Mullaugh et al. 2015).

The concentration of BTEX compounds in high-traffic areas was significantly higher than low-traffic areas in summer, winter and the overall mean. Traffic is one of the main sources of air pollutants (e.g., BTEX) in the urban area (Phuc and Kim Oanh 2018; Buczynska et al. 2009; Rad et al. 2014). In addition to vehicle exhaust emissions, BTEX can release to the atmosphere from buck, crankshaft, cartel, injection fuel system, etc., (Kato et al. 2008). Lower technology of vehicles and fuel quality in developing countries (e.g., Iran) can exacerbate the problem (Truc and Kim Oanh 2007; Phuc and Kim Oanh 2018). A study by Phuc et al. 2018 in Hanoi, Thailand, reported that there was a strong relationship between the concentration of

BTEX compounds and traffic flows, especially in rush hours (Phuc and Kim Oanh 2018). Moreover, they reported that BTEX concentrations in more heavily traveled roads were higher than the small residential road (Phuc and Kim Oanh 2018). These results are in agreement with our findings.

The mean concentrations of BTEX compounds in summer, winter and overall mean were significantly different in six land-use types. The maximum BTEX concentration was observed in *Transportation* land use, and *Greenspace* land use had minimum concentration. Each land-use type can act as a point source of air pollution in urban areas. Usually, *Transportation* land use has heavy traffic. Moreover, negative acceleration and on-road mode work of vehicles in the stations and terminals (bus or/and taxi) could result in higher pollutant emissions (Golkhorshidi et al. 2019; Dehghani et al. 2018). A study by Golkhorshidi et al. 2019 in four intercity bus terminals in Tehran, Iran, reported that the BTEX concentrations in working days were significantly higher than nonworking days and moreover reported diesel bus emissions and fuel evaporation were introduced as the main source of BTEX emissions in bus terminals (Golkhorshidi et al. 2019). In another study by Dehghani et al., 2018, BTEX concentrations in an intercity bus terminal and around it in Shiraz, Iran, were investigated and reported BTEX concentrations in the terminal and street with heavy traffic around it were higher than other areas (Dehghani et al. 2018).

Green space can act as a biofilter against for ambient air pollutants (Miri et al. 2018a, b; Dadvand et al. 2015; Keymeulen et al. 2001). In a study by Milazzo et al. 2019, the potential of city parks to reduce exposure to BTEX compounds in Alabama, the USA, was investigated, and they reported that except for benzene, the concentration of TEX in parks was significantly lower than residential areas (Milazzo et al. 2019). Although the available evidence on the association between greenness and BTEX concentrations in ambient air is still scarce, previous studies on the other air pollutants indicated a negative association between greenspace and air pollution concentration (Miri et al. 2018a, b; Dadvand et al. 2015). These results are consistent with our study.

In our study, total BTEX concentration in *Industrial* land use was lower than *commercial/official*, *Residential* and *Truism* land use. However, a study by Liu et al. 2018 reported that *Industrial* land use had a higher total BTEX concentration than *Residential* and *commercial/official* land use (Liu et al. 2018). Moreover, a study by Dehdari Rad et al. 2014 reported that *Industrial* land use had higher total BTEX concentration than *Residential* land use (Rad et al. 2014). These studies are inconsistent with our study. These different results can be due to different industrial activities in different cities. Some industries used VOCs in their process, which can release BTEX compounds into the atmosphere. Moreover, Mashhad is a megacity with high population density; therefore, the residential areas can act as a point source of BTEX pollutants. Furthermore, most of *Truism* land uses are in the center of Mashhad, which has high traffic density and has high population density; thus, the concentration of BTEX compounds could increase in this land-use type.

Limitations

This study had limitations that should be considered in future studies. The number of monitoring satiations was low, considering the study area. Moreover, BTEX concentrations were just measured in winter and summer, and we lost spring and autumn, which can affect our results. There are more than ten land-use types, but we select six more important of them. The limitation in the number of sampling locations and loss of two seasons were due to the insufficient budget of our study; therefore, a longer sampling period in all land-use types is recommended in future studies.

Conclusion

We found that the concentrations of BTEX compounds between different land-use types were significantly different. The minimum and maximum concentrations of BTEX were observed in *Greenspace* and *Transportation* land uses, respectively. Moreover, we found a significant difference in BTEX concentration between summer and winter as well as between high- and low-traffic areas. Toluene and benzene have clustered emission patterns, and exposure to benzene had a carcinogenic risk for Mashhad's inhabitants. Due to rapid urbanization and increased vehicles in the cities, reducing pollutant emissions and their health effects are the most important issue. The finding of this study coupled with future studies can help urban

planners and decision-makers to improve urban air quality and community health. Future studies on BTEX concentrations in different cities, other land-use types and longer sampling time are recommended.

Compliance with ethical standards

Conflict of interest All authors declare that they have no conflicting interests.

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