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A case study of BTEX characteristics and health effects by major point sources of pollution during winter in Iran☆

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ABSTRACT

This study characterized spatio-temporal variations in the concentration of benzene, toluene, ethylbenzene, and xylene (BTEX) compounds in the vicinity of gas and compressed natural gas (CNG) stations in Tehran, Iran. Health risk assessment (HRA) was computed using Monte Carlo simulations (MCS) for evaluating inhalation lifetime cancer risk (LTCR), the hazard quotient (HQ), and sensitivity analysis (SA) for BTEX exposure in different age groups (birth to <81) and as a function of distance (0–250 m) from the center of the stations. For all monitoring stations, the average values of benzene, toluene, ethylbenzene, and xylene in winter were 466.09 ± 132.25, 873.13 ± 233.51, 493.05 ± 141.22, and 910.57 ± 145.40 μg m^{-3}, respectively. The mean wintertime ratios of T/B for the 12 stations ranged from 1.69 to 2.04. Furthermore, there was no significant relationship between the concentration of BTEX with either the specific month or distance from the center of stations ($p > 0.05$). Factors promoting BTEX formation in the study region were fuel evaporation and gas/CNG station emissions. The LTCRs for the target compounds in the winter for different age groups and distances from the center of stations was limited to 2.11×10^{-4} to 1.82×10^{-3} and 2.30×10^{-4} to 2.01×10^{-3}, respectively, which exceeded proposed values by U.S. EPA. Moreover, the HQs for BTEX for three age groups and distances were limited to between 2.89×10^{-5} and 9.33×10^{-2}, which were lower than the acceptable limit (HQs < 1). The results of this work are applicable to similar areas that are heavily populated with vehicular traffic. This study motivates a closer look at mitigation strategies to limit the health effects of carcinogenic emissions such as benzene and ethylbenzene from gas/CNG stations.

1. Introduction

Gas and compressed natural gas (CNG) stations are recognized as the main sources of volatile organic compounds (VOCs) in urban areas worldwide (Bauri et al., 2016; Correa et al., 2012a; Cruz et al., 2017; Hazrati et al., 2016b; Huang et al., 2018; Xiong et al., 2016). These point sources can emit significant levels of benzene, toluene,

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ethylbenzene, and xylenes (BTEX) into the ambient air (Correa et al., 2012a; Fazlzadeh Davil M, 2011; Moolla et al., 2015a; b; Tunsaringkarn et al., 2012). Air pollution in many developing countries has been aggravated with time because of increasing population via urbanization, which resulted in a rapid increase in the number of gas/CNG stations leading to more emissions posing major health risks for public health (Correa et al., 2012a; Cruz et al., 2017; Esmaelnejad et al., 2015; Esteve-Turrillas et al., 2007; Goyal, 2003; Hazrati et al., 2016a; Hazrati et al., 2016b; Huang et al., 2018; Okonkwo et al., 2014; Ravindra et al., 2006; Suthawaree et al., 2012; Tunsaringkarn et al., 2012; Yimrungruang et al., 2008). BTEX species have been the subject of many toxicological and health effects studies (Chauhan et al., 2014; Dehghani et al., 2018a; Esteve-Turrillas et al., 2007; Moolla et al., 2015b; Partovi et al., 2018; Zabiegała et al., 2010). Exposure to BTEX concentrations in the ambient air pose harmful health effects such as cancer, teratogenic impacts, and neurological disorders (Chauhan et al., 2014; Esteve-Turrillas et al., 2007; Moolla et al., 2015b; Xiong et al., 2016; Zabiegała et al., 2010). Benzene (B), ethylbenzene (EB), xylenes (Xs), and toluene (T), based on the International Agency for Research on Cancer (IARC), were classified as group 1 (carcinogenic to humans), group 2B (possibly carcinogenic to humans), group 3 (not classifiable as to its carcinogenicity to humans), and group 3 (not classifiable as to its carcinogenicity to humans), respectively (IARC, 2014; Lim et al., 2014). Based on previous evidence, benzene is one of the main carcinogenic species harmful for humans and can cause disorders such as hematological disorders, polycythemia, aplastic anemia, chronic myeloid leukaemia, myocardial infarction, nasopharyngeal cancer, respiratory diseases, and myeloproliferative disease (Correa et al., 2012a; Duarte-Davidson et al., 2001; Durmusoglu et al., 2010; Glass et al., 2014; Koh et al., 2015; Moolla et al., 2015a; Neghab et al., 2017; Ran et al., 2018; Robinson et al., 1997; Smith, 2010; Suh et al., 2011; Xiong et al., 2016; Ye et al., 2017). According to previous studies, toluene exposure can lead to premature delivery, congenital malformations, retardation, and neurobehavioral and reproductive problems (ATSDR-DTHHS, 2015; Donald et al., 1991; Foo et al., 1990; Greenberg, 1997; Soni et al., 2018; Tunsaringkarn et al., 2012).

The toluene ($\mu g\ m^{-3}$) to benzene ($\mu g\ m^{-3}$) (T/B) ratio has been commonly used as an indicator for emission sources of pollution (traffic versus non-traffic sources) (Parra et al., 2006). In addition, Tunsaringkarn et al. (2012), Dehghani et al. (2018), Chan et al. (1996), Tiwari et al. (2010), and Khoder (2007) suggested that T/B ratios exceeding one indicate high influence from non-traffic emissions (Chan et al., 1996; Dehghani et al., 2018c; Tunsaringkarn et al., 2012) (Khoder, 2007; Tiwari et al., 2010). For instance, Tunsaringkarn et al. (2012) showed that the ratio of toluene ($\mu g\ m^{-3}$) to benzene ($\mu g\ m^{-3}$) (T/B) in the vicinity of gasoline stations was 2.08 and thus $T/B > 1$ is an indicator of high influence of non-traffic emissions (Tunsaringkarn et al., 2012). The same study indicates that measured toluene concentration is high and the main source of toluene was linked to gas and CNG stations (Tunsaringkarn et al., 2012). In addition to the T/B ratio, the ratios of Xs/B and EB/B ($\mu g\ m^{-3}/\mu g\ m^{-3}$) are useful as they quantify the degree of photochemical reactivity and photochemical aging in the ambient air of stations (Bauri et al., 2016; Bretón et al., 2017; Ho et al., 2004; Hoque et al., 2008; Hsu and Huang, 2009; Kerbachi et al., 2006; Kerchich and Kerbachi, 2012; Khoder, 2007; Tiwari et al., 2010). For example, Kerchich and Kerbachi (2012) reported that the ratios of X/B and EB/B exceeding one (X/B and $EB/B > 1$) suggest that sampled air masses were not photochemically aged because insufficient amounts of hydroxyl radicals ($^{\bullet}OH$) in the ambient air were available to react with target components such as xylenes and ethylbenzene (Kerchich and Kerbachi, 2012).

In order to determine the cancer and non-cancer risk of BTEX components, Monte Carlo simulations (MCS) have been applied to calculate inhalation lifetime cancer risk (LTCR) of benzene and ethylbenzene and the hazard quotient (HQ) of BTEX for urban pollution and indoor air of beauty salons (Baghani et al., 2018; Dehghani et al., 2018b; Delikhoon et al., 2018a). Also, LTCR and HQ values were 0.002–0.6 and 1.75×10^{-4} to 9.55×10^{-7}, respectively, for a gas station in Bangkok, Thailand (Tunsaringkarn et al., 2012). In contrast, the LTCR and HQ values for a petrol station in Bangkok, Thailand were 2.50×10^{-4} to 7.81×10^{-6} and 0.004 to 0.351, respectively (Kitwattanavong et al., 2013). In addition, the results of the Monte Carlo simulation technique and sensitivity analysis (SA) for evaluating carcinogenic and non-carcinogenic risk of BTEX compounds showed that the concentration of BTEX had the highest positive impact on the average lifetime cancer risk (Dehghani et al., 2018b).

Tehran, Iran represents a major urban center with many gas/CNG stations and thus is a hotspot for BTEX emissions. The aim of the present study is to report on concentrations of BTEX and associated health effects for twelve gas/CNG stations in Tehran during winter. Discussion is devoted to a spatiotemporal characterization of concentrations and health effects of BTEX as a function of distances from the center of the stations up to 250 m away. Lastly, attention is given to the possibility of photochemical aging in the ambient air of twelve gas/CNG stations. This is the first such study investigating BTEX in this understudied, yet largely populated, region in the Middle East with attention geared towards spatial and diurnal trends, health effects of BTEX, and amount of BTEX emissions in gas stations versus CNG stations.

2. Material and methods

2.1. Study area

The capital of Iran is Tehran (35.6892° N, 51.3890° E) (Fig. 1) with around 13.31 million inhabitants according to a census report in 2016 (Statistical Centre of Iran (SCI), 2016). This megacity located in northern Iran is classified as one of the most polluted around the world (Amini et al., 2017; Crosbie et al., 2014; De Koning et al., 1986; Mage et al., 1996). Fig. 1 shows a map of Tehran and the four regions (categorized as 6, 7, 11, 12) within which measurements were conducted. Measurements were specifically conducted at two CNG stations and 10 gas stations, which were chosen using criteria related to proximity to residential areas (and thus less industrial influence), proximity to the center of Tehran, population characteristics, existence of a CNG station close to a gas station for at least one of the four regions, and traffic conditions.

2.2. Sampling and analysis

Sampling was performed according to the NIOSH 1501 method (NIOSH, 2003; Rezazadeh Azari et al., 2012; Wang et al., 2005) and carried out over 2 h (9:00–11:00 AM local time) from 22 December 2017 to 28 February 2018 (in winter: December, January and February) by active sampling (Low Flow Sample Pump 222 Series, SKC Inc.) with charcoal sorbent tubes (SKC Inc.) at a flow rate of $0.2\ L\ min^{-1}$ (Dehghani et al., 2018c; Samarghandi et al., 2014). Samples were collected every-third-day at all 12 stations. Sampling was done at the center of all gas and CNG stations (Mirzaye Shirazi and Shoush stations) and also around the periphery of the centers of stations at different distances from the center points (e.g., 50, 150, and 250 m away) at a height of 2 m. We collected 380 samples (300 for 12 gas and CNG stations from December to February and 80 samples from 0 to 250 m for Mirzaye Shirazi and Shoush stations for January).

Before analysis, the two parts of the sampling tubes (the front

and back) were placed into two separate vials and the BTEX pollutants were extracted using 1 mL carbon disulfide from charcoal tubes (ASTM, 1995; NIOSH, 2003). The vials including carbon disulfide and charcoal were slowly shaken for 30 min. The solvent was placed in GC vials and target compounds were measured by a GC (Chrompack-Netherlands (NL)-CP9001) equipped with an FID detector using a capillary column (CP-Sil 8 CB-Chrompack, 30 m × 0.32 mm, 0.20 μm in film thickness). The injector was operated in split mode with a ratio of 1–5 at 250 °C. Nitrogen (99.999%) served as the carrier gas at a flow rate of 2.5 mL min^{-1}. One μL aliquots were derived from GC vials and injected in a capillary column. The oven temperature was programmed at 40 °C for 10 min and then 10 °C min^{-1} to 200 °C.

A portable weather station (Preservation Equipment Ltd, UK and Campbell Scientific, Inc., USA) was used for measurement of parameters such as wind speed (m s^{-1}), temperature (°C) and relative humidity (%) at the study sites.

2.3. Quality assurance/quality control (QA/QC)

The two sections of the sampling tubes were separately analysed for the possibility of breakthrough and there was no evidence of target pollutants in the second section of charcoal tube. Thirty eight samples were collected during the winter as blank samples and the target pollutant concentrations (μg m^{-3}) in these tubes were limited to 0.00–0.028 for benzene, 0.00–0.039 for toluene, 0.00–0.204 for ethylbenzene, and 0.00–0.321 for xylenes. The mean recovery was 97% (95–113%) for the target components and standard deviations (SDs) of BTEX components were lower than 5%. Calibration curves were developed using six points based on standard solutions ranging from 1 to 60 ppm. The results showed that the coefficients of determination (R^2) for BTEX contaminants were as follows using the calibration curves: 0.998 for benzene, 0.997 for toluene, 0.999 for ethylbenzene, and 0.998 for xylenes.

2.4. Statistical analysis

All analyses were carried out by the statistical program R (version 3.0.1). The relationships between BTEX concentrations and meteorological conditions such as wind speed (m s^{-1}), temperature (°C), and relative humidity (%) were quantified using Spearman's rank correlation coefficient or Spearman's rho. Toluene to benzene (T to B), xylenes/benzene (Xs/B), and ethylbenzene to benzene (EB to B) ratios were calculated in order to infer insight about photochemical aging and to determine emission sources of target components. In this study, the Fligner-Killeen test was applied to assess for homogeneity of variance. If the p-value obtained from the Fligner-Killeen test exceeded 0.05, the ANOVA test was performed for further analysis. But, if the p-value was less than 0.05, the Kruskal-Wallis test was applied for further analysis. Finally, Tukey's multiple comparison test (Tukey's HSD) was applied in the comparative analysis between the mean concentrations of BTEX at different sampling locations. In the end, health risk assessment (HRA) was computed using Monte Carlo simulations (MCS) for evaluating the hazard quotient (HQ) and sensitivity analysis (SA) for BTEX concentrations. Probability distributions of cancer risks are included in supplementary information (SI). Figures were depicted by GraphPad Prism 7 and R Statistical Software version 3.0.1.

2.5. Spatial distributions

The Arc GIS 10.4.1 software package was utilized for spatial analysis. The inverse distance weighting (IDW) interpolation method was used to make raster layers for the mean concentration of target components and to show the distribution of BTEX contaminants for 12 stations. The IDW method is centered around the following equation (Dehghani et al., 2018c; Delikhoon et al., 2018b; Hazrati et al., 2016b; Hazrati et al., 2015; Jalali et al., 2014; Li et al., 2014; Li et al., 2008):

$$\lambda_i = \frac{D_i - \alpha}{\sum_{i=1}^{n} D_i - \alpha} \quad (1)$$

Where α = the weighting power, λ_i = the weight of the x sample station, and D_i = the distance between the station i and an uncertain point that was varied in the present study.

The maximum values were dedicated to values close to the interpolated point; on this matter, Dehghani et al. (2018c), Xie et al. (2011), and Marshall et al. (2008) reported that when the distance increases, the weight decreases (Bauri et al., 2016; Dehghani et al., 2018c; Xie et al., 2011). The number of stations utilized in the interpolation (n) was 12 in this work.

2.6. Health risk assessment (HSA) for target components

The United States Environmental Protection Agency (U. S. EPA) has reported that the acceptable limit for carcinogenic compounds is equivalent to a risk level of 1×10^{-6} (Dehghani et al., 2018c; EPA, 2003; Tam and Neumann, 2004). In addition, "an acceptable risk for humans" in terms of the lifetime cancer risk (LTCR) is reported to be lower than 1×10^{-6} (Dehghani et al., 2018c; EPA, 2003; Hazrati et al., 2016b; Ho et al., 2004; Tam and Neumann, 2004; Tunsaringkarn et al., 2012).

In order to determine the cancer and non-cancer risk of humans to BTEX components, inhalation lifetime cancer risk (LTCR) of benzene and ethylbenzene (Eq. (2)) and the hazard quotient (HQ) of BTEX (Eq. (3)) were computed with the data acquired in this work. Concentrations of BTEX were applied to estimate inhalation lifetime cancer risk (LTCR) (Eq. (2)), Hazard Quotient (HQ) (Eq. (3)), and the lifetime average daily dose (LADD (mg kg^{-1} day^{-1})) (Eq. (4)). The LTCR was estimated according to following equation (Dehghani et al., 2018c; Delikhoon et al., 2018b; Hazrati et al., 2016a; Hazrati et al., 2016b; Hazrati et al., 2015; Jalali et al., 2014; Li et al., 2014; Li et al., 2008):

$$LTCR = LADD \times CSF \quad (2)$$

Where LADD and CSF are lifetime average daily dose (mg kg^{-1} day^{-1}) and the cancer slope factor (mg kg^{-1} day^{-1}), respectively.

In addition, the hazard quotient was calculated according to following equation:

$$HQ = \frac{LADD}{RfD} \quad \text{where (Unsafe) } 1 < HQ \leq 1 \text{ (Safe)} \quad (3)$$

where HQ and RfD are hazard quotient (mg kg^{-1} day^{-1}) and reference dose (mg kg^{-1} day^{-1}), respectively.

$$LADD = (C \times EF \times ED \times IR) / (BW \times AT) \quad (4)$$

Where C represents ambient concentration of pollutants (μg m^{-3}), EF and ED are exposure frequency (days $year^{-1}$) and exposure duration (year), respectively, and IR, BW, and AT are human inhalation rate (m^3 day^{-1}), body weight (kg), and average lifetime (yr), respectively (Table S1).

Hazard quotients exceeding one (HQ > 1) indicate unsafe risk, whereas HQ ≤ 1 represents safe risk (Dehghani et al., 2018c; Delikhoon et al., 2018b; Demirel et al., 2014; Durmusoglu et al., 2010; Hazrati et al., 2016a; Hazrati et al., 2016b; Hazrati et al.,

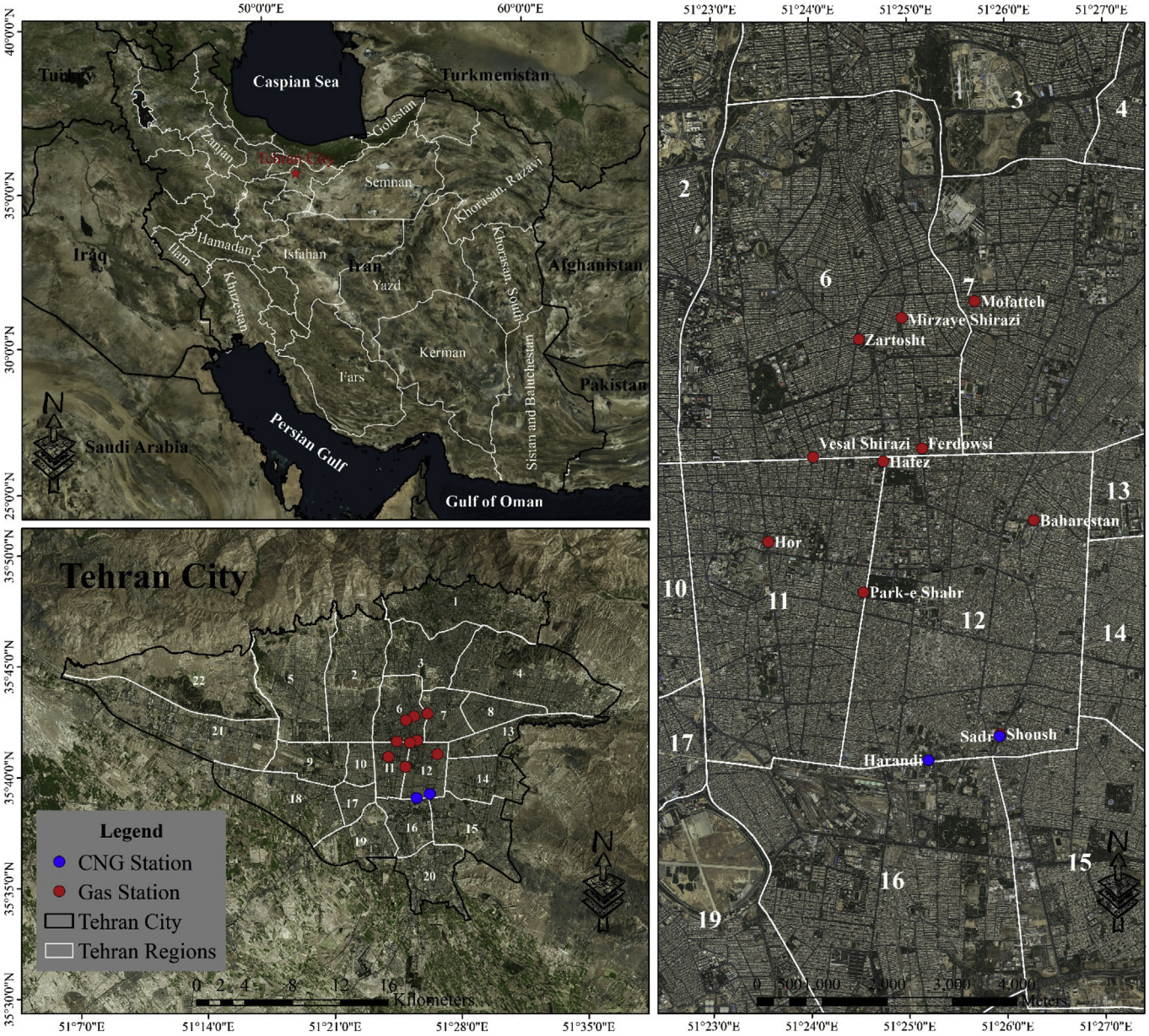

Fig. 1. Map of Tehran (Iran) and sampling sites in four regions of Tehran (6, 7, 11, and 12), including two CNG stations and 10 gas stations.

2015; Jalali et al., 2014; Li et al., 2014; Li et al., 2008; Moolla et al., 2015b; Tunsaringkarn et al., 2012).

3. Results and discussion

3.1. BTEX concentrations in compressed natural gas (CNG) and gas stations

The mean ± standard deviation (SD) values of BTEX (μg m^{-3}), temperature (°C), relative humidity (%) and wind speed (m s^{-1}) in compressed natural gas (CNG) and gas stations during the winter are shown in Table S2. The total mean wind speed, temperature, and relative humidity during sampling for 12 sampling sites were, respectively: 4.17 ± 0.58 m s^{-1}, 12.38 ± 0.96 °C and 71.31 ± 5.88%. A comparison of BTEX concentrations (μg m^{-3}), temperature (°C), relative humidity (%), and wind speed (m s^{-1}) for CNG and gas stations between this and other studies is summarized in Table S3.

For all 12 monitoring stations, the total mean and SD concentrations (μg m^{-3}) of benzene, toluene, ethylbenzene, and xylene in winter were 466.09 ± 132.25, 873.13 ± 233.51, 493.05 ± 141.22, and 910.57 ± 145.40, respectively. Mean BTEX concentrations at gas stations exceeded those at CNG stations. The results of this work showed that xylenes were the most abundant of the four species for 11 monitoring stations (because of insufficient hydroxyl radicals ($^{\bullet}$OH) for reacting with xylene (Kerchich and Kerbachi, 2012)), except for the Mirzaye Shirazi gas station where toluene was the most abundant.

The finding in this work that xylenes were the most abundant is consistent with past reports of data recorded at gas stations in Rio de Janeiro (Brazil) (61.2 μg m^{-3}) (Correa et al., 2012b), petrol stations in Tehran (Iran) (299.11 ppm) (Eisaei et al., 2015), bus diesel-refueling bays in Johannesburg (South Africa) (850.97 ppb) (Moolla et al., 2015a), urban areas in Rio de Janeiro (Brazil) (14.4 μg m^{-3}) (Martins et al., 2007), Tehran (Iran) (9–138 μg m^{-3}) (Hajizadeh

et al., 2017), Yazd (Iran) (340 μg m^{-3}) (Mehrjerdi et al., 2014), cabin air inside taxis in Tehran (Iran) (6.47 ppb) (Bakhtiari et al., 2018), public gasoline stations in Dammam and al-Khobar (Saudi Arabia) (0.27 ppm) (Alyami, 2017), gas stations in Athens (Greek) (423 μg m^{-3}) (Soldatos et al., 2003), oil and gas stations in the Junggar Basin (northwest China) (0.92 ppb) (Huang et al., 2018), a parking area in Rio de Janeiro (Brazil) (118.93 μg m^{-3}) (de Castro et al., 2015), and the Tijuca District (near to diesel, CNG and oxygenated gasoline vehicular fleet) in Rio de Janeiro (Brazil) (24.3 ppb) (Martins et al., 2010). Reasons for xylenes being highest in concentration could be related to differences in the content of target compounds in fuel, insufficient hydroxyl radicals (•OH) for reacting with xylene, fuel evaporation, and gas/CNG station emissions. Le Ha et al. (2017) and Martins et al. (2010) reported for outdoor air Hanoi (Vietnam) and in a diesel, compressed natural gas (CNG) and oxygenated gasoline vehicular fleet in Rio de Janeiro (Brazil), respectively, the following order of abundance: xylenes > toluene > ethylbenzene > benzene (Le Ha, 2017; Martins et al., 2010). In addition, Goyal (2003) and Martins et al. (2007) stated that concentrations of benzene and toluene were depressed when natural gas was applied as fuel, consistent with findings of this study (Goyal, 2003; Martins et al., 2007).

Fig. S1 displays a box plot of BTEX species concentrations at all sampling locations during winter. The minimum concentrations (μg m^{-3}) of BTEX species were measured at the Harandi CNG station: 130.32 for benzene, 230.47 for toluene, 176.76 for ethylbenzene, 554.23 for xylenes. The maximum BTEX species concentrations (μg m^{-3}) were detected at the Shoush gas station: 598.34 for benzene, 1076.29 for toluene, 627.93 for ethylbenzene, and 1054.32 for xylenes. The reason fort why the concentrations of BTEX species at the Shoush gas station exceeded those of other stations may have been the short distance between the Sadr CNG station and Shoush gas station in the study area (region 12) (Fig. 1).

The only recorded guideline value among BTEX species for the study region is for benzene, which is reported by Iran's Environmental Protection Organization (IEPO) as being 5 μg m^{-3} (IEPO, 2012). Hence, in our study, the concentration of benzene in winter was higher compared to the recommended value by IEPO (5 μg m^{-3}) (IEPO, 2012). State and local agencies have no monitoring programs, which is worthwhile to initiate based on results of this study.

3.2. Spatial distribution of BTEX levels

Several studies have performed the IDW method for the spatial distribution of pollutants such as for BTEX concentrations in the ambient air of in Shiraz (Iran) (Dehghani et al., 2018c), NO/NO_2/CO/O_3 in the ambient air of Vancouver (Canada) (Marshall et al., 2008), $PM_{2.5}$/NO_2/O_3 in California (Jerrett et al., 2013), O_3/NO_2/SO_2 in Abidjan, west Africa (Bahino et al., 2018), polycyclic aromatic hydrocarbons (PAHs) in urban stream sediments in Pennsylvania (USA) (Witter et al., 2014), volatile organic compounds (VOCs) in Greater Cairo (Egypt) (Matysik et al., 2010), and formaldehyde and acetaldehyde in the ambient air of in Shiraz (Iran) (Delikhoon et al., 2018b). Recently, the method of IDW was used to estimate levels of contaminants in the ambient air all over the world (Delikhoon et al., 2018b; Wang et al., 2014; Yanosky et al., 2008).

Hence, the IDW method was used for the spatial distribution of BTEX pollutants in the ambient air of gas/CNG stations in Tehran (Iran). The relative amount of BTEX concentrations (μg m^{-3}/μg m^{-3}) at each gas station compared with either of the CNG stations (Harandi or Sadr) was calculated by dividing the mean concentrations of BTEX components at each gas station by the same pollutant at either the Harandi or Sadr CNG station. According to Table S4, each gas station yielded 2.88–4.14 (benzene), 3.45–4.31 (toluene), 2.60–3.60 (ethylbenzene), and 1.54–1.87 (μg m^{-3}/μg m^{-3}) (xylenes) times higher concentration compared to the CNG stations, which is likely attributed to the different fuel types produced from various refineries in Iran. Increasing numbers of CNG stations have been linked to reductions in VOC emissions, especially for benzene and toluene (Dehghani et al., 2018c; Goyal, 2003; Hazrati et al., 2016b; Martins et al., 2007; Martins et al., 2010).

3.3. Interrelationships between BTEX concentrations and meteorological conditions

Table S5 describes Spearman's rank correlations between BTEX species, based on mean concentrations, and meteorological parameters for the winter season. The correlation coefficients (r) between BTEX species exceeded 0.890. These findings revealed that BTEX species had the similar sources and the CNG and gas stations were the main sources of BTEX in the four studied regions of Tehran. The maximum correlation coefficient ($r = 0.978$, $p < 0.01$) was observed to be between benzene and ethylbenzene. Previous studies reported similarly high correlations between BTEX species such as for a hot spot for urban pollution in Shiraz (Iran) (Dehghani et al., 2018c), in the petrochemical industrial area in Yokohama (Japan) (Tiwari et al., 2010), in urban/suburban in Martorell (Spain) (Baldasano et al., 1998), in ambient air in Daegu (Korea) by (Choi et al., 2009), in indoor microenvironments in Sakaka (Saudi Arabia) (El-Hashemy and Ali, 2018), in the semi-urban in Orleans, (France) (Jiang et al., 2017), and in nine gas stations in Chonburi (Thailand) (Yimrungruang et al., 2008).

In this study, a significant correlation was observed between BTEX species and temperature ($p < 0.01$). Concentrations of BTEX species increased as a function of temperature, indicative of evaporative processes as a key source of BTEX species in the ambient air of CNG and gas stations. Furthermore, this work and other studies have similarly calculated a negatively correlation between BTEX species with wind speed in areas such as a petrol station in Bangkok (Thailand), an oil and gas station in Junggar Basin (China), and at a gasoline pump in Finland (Huang et al., 2018; Rattanajongjitrakorn and Prueksasit, 2014; Vainiotalo et al., 2006). Hence, higher BTEX concentrations were estimated in this work to occur at higher temperatures and lower wind speeds. No significant correlation was observed between BTEX species and humidity ($p > 0.01$).

3.4. Temporal variations of BTEX

Temporal variations of benzene, toluene, ethylbenzene, and xylenes (BTEX) (μg m^{-3}) from December to February were examined based on sampling location (Figs. 2 and 3). The relative concentrations of each of the four BTEX species remained the same for each month (December, January, February), indicating that these contaminants exhibited a similar distribution spatial pattern regardless of monthly-driven factors, consistent with former studies (Bauri et al., 2016; Rad et al., 2014; Tiwari et al., 2010; Zalel and Broday, 2008). In contrast, different temporal variations for the target pollutants were reported by Huang et al. (2018), Alghamdi et al. (2014), Menchaca-Torre et al. (2015), and Ho et al. (2004), owing to differences in photochemical activity, source emissions, and meteorological conditions (Alghamdi et al., 2014; Ho et al., 2004; Huang et al., 2018; Menchaca-Torre et al., 2015). The highest mean concentration of BTEX compounds were observed in January for benzene and toluene and December for ethylbenzene and xylenes, while the lowest concentration of target contaminants was observed in February for ethylbenzene and December for benzene, toluene, and xylenes.

3.5. Spatial and temporal statistical analysis of BTEX compounds

3.5.1. Statistical analysis of temporal variability

Fig. 4 shows a box plot of BTEX concentrations for different months (December, January, February) in winter. The output of the Fligner-Killeen test demonstrated that p-values for benzene, toluene, ethylbenzene, and xylenes in different months (December, January and February) were 0.675, 0.961, 0.916, and 0.738, respectively. The p-values are more than the level of 0.05 ($p > 0.05$), indicating that there were insignificant differences as a function of month. Hence, parametric ANOVA test was used for further analysis.

The results of the ANOVA test illustrated that the p-values for benzene, toluene, ethylbenzene, and xylenes were, respectively, 0.964, 0.956, 0.987, and 0.930, again suggesting that the difference in BTEX species in different months were insignificant.

3.5.2. Statistical analysis of spatial variability

The output of the Fligner-Killeen test showed that p-values for benzene, toluene, ethylbenzene, and xylenes across all sampling stations were 0.716, 0.363, 0.479, and 0.330, respectively. The results of the ANOVA test illustrated that the p-values for benzene, toluene, ethylbenzene, and xylenes were lower than 0.05 ($p < 0.05$), suggesting that the difference in BTEX species in different locations were significant. Consequently, the results of the Tukey multiple comparisons test showed that the mean concentrations of BTEX species between two sampling locations of Ferdowsi-Baharestan, Mirzaye Shirazi-Baharestan, Mofatteh-Baharestan, Mirzaye Shirazi-Ferdowsi, Mofatteh-Ferdowsi, Zartosht-Ferdowsi, Hor-Hafez, Park-e Shahr-Hafez, Shoush-Hafez, Sadr-Harandi, Park-e Shahr-Hor, Shoush-Hor, Mofatteh-Mirzaye Shirazi, Zartosht-Mirzaye Shirazi did not show significant differences ($p > 0.05$). However, there were significant differences between the mean concentrations of BTEX species in other sampling locations ($p < 0.05$).

Also, the findings of Tukey's test indicated that the most significant difference among all sampling locations for benzene, toluene, ethylbenzene, and xylenes occurred between Shoush and Harandi ($p < 0.05$).

3.5.3. Statistical analysis in CNG and gas station

The box plot of BTEX species at CNG versus gas stations is shown in Fig. S2. The results of the Fligner-Killeen test revealed that p-values for benzene, toluene, ethylbenzene, and xylenes compared between CNG and gas stations were 0.019, 0.007, 0.006, and 0.004, respectively. This demonstrates that the differences of BTEX concentrations between CNG and gas stations were significant ($p < 0.05$). The results of Kruskal-Wallis test, which compares normally distributed variables for more than two groups for BTEX concentrations showed that the chi-square test statistic was 15.71, regardless of species, which was significant at a level of 0.05 ($p < 0.05$). In other words, there was certainly a significant difference between the concentrations of BTEX at CNG versus gas stations ($p < 0.05$).

3.5.4. Statistical analysis for different distances

Fig. S3 displays box plot of BTEX concentrations as a function of distance from the center of stations up to 250 m away. As can be seen from Fig. S3 and the results of the Fligner-Killeen test, p-values for benzene, toluene, ethylbenzene, and xylenes at different distances were 0.816, 0.828, 0.818, and 0.681, respectively ($p > 0.05$). Therefore, the difference between the variances in BTEX levels at different distances from stations was not significant ($p > 0.05$). Results of the ANOVA test showed that the p-values for benzene, toluene, ethylbenzene, and xylenes were, respectively, 0.373, 0.268, 0.375, and 0.467, again indicating that the difference in BTEX species at different distances was not significant.

3.6. The ratios between target components

3.6.1. T/B ratios

The ratio of toluene to benzene (T ($\mu g\ m^{-3}$) to B ($\mu g\ m^{-3}$)) for the 12 sampling locations is summarized in Fig. 5, with the significance of the values being that this ratio is commonly used as an indicator for source emissions of target pollutants (traffic versus non-traffic sources) (Bretón et al., 2017; Chan et al., 1996; Dehghani et al., 2018c; Parra et al., 2006; Tunsaringkarn et al., 2012).

Tunsaringkarn et al. (2012), Dehghani et al. (2018), Chan et al. (1996), Tiwari et al. (2010), and Khoder (2007) suggested that T/B ratios exceeding one indicate high influence from non-traffic emissions (Chan et al., 1996; Dehghani et al., 2018c; Tunsaringkarn et al., 2012) (Khoder, 2007; Tiwari et al., 2010). In this study, the average ratios of T/B for the 12 stations ranged from 1.69 to 2.04, indicative of the importance of non-traffic emissions

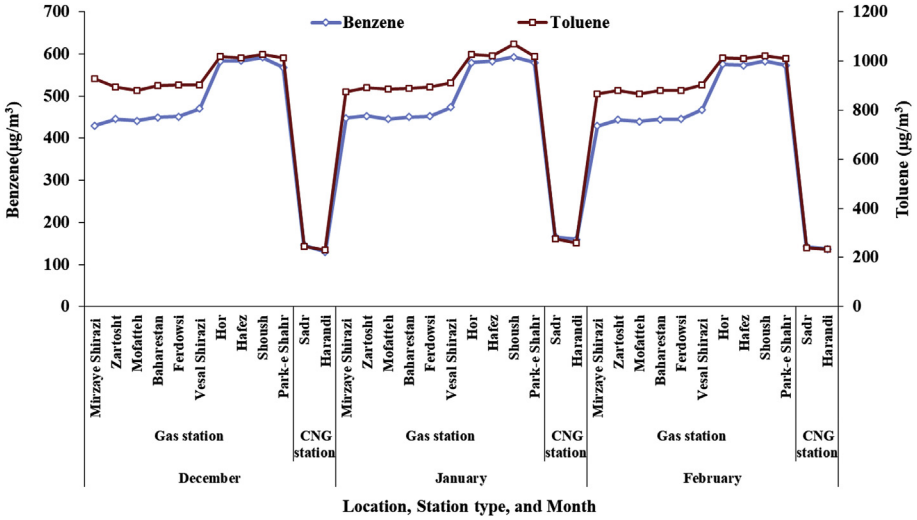

Fig. 2. Temporal variations (December to February) of benzene and toluene ($\mu g\ m^{-3}$) at different sampling locations during winter.

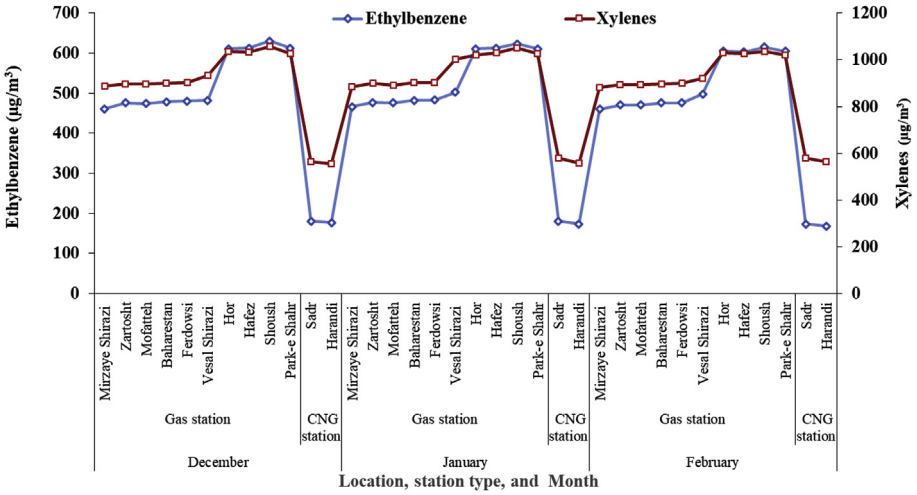

Fig. 3. Temporal variations (December to February) of ethylbenzene and xylenes (µg m^{-3}) at different sampling locations during winter.

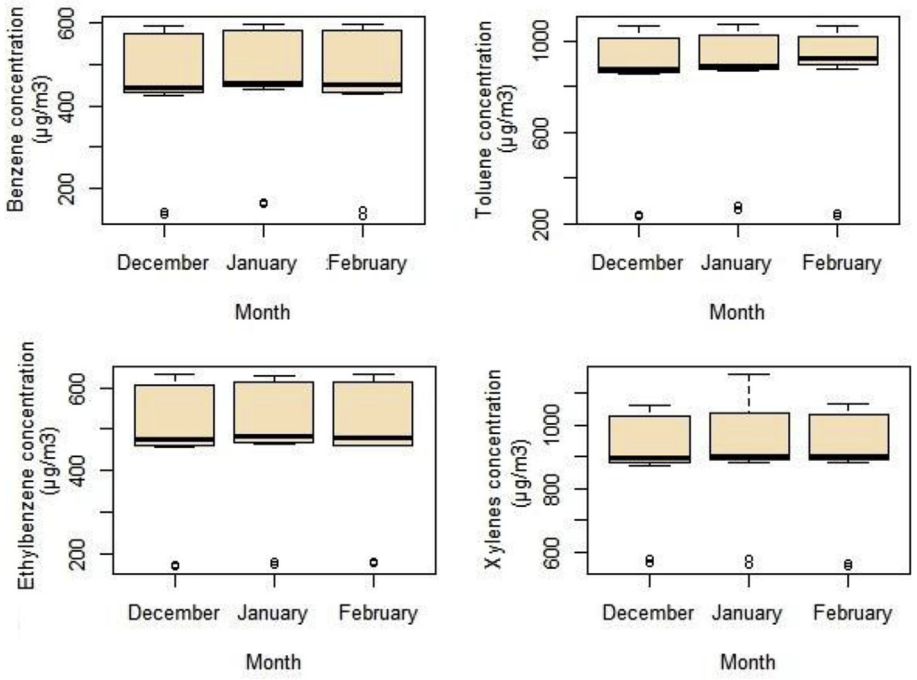

Fig. 4. Box plot of BTEX concentrations in different winter months (December, January, February).

(e.g., gas and CNG stations) as the major emission source of target components in the study area. The ratio values of T/B in this study were comparable to those at a gas station in Bangkok, Thailand (2.08) (Tunsaringkarn et al., 2012), in ambient air of Ahvaz, Iran (3.25) (Rad et al., 2014), in urban areas in Greater Cairo–Ramsis, Egypt (2.45) (Khoder, 2007), in urban areas in Greater Cairo-Haram, Egypt (2.42) (Khoder, 2007), in a rural area in Menofiya-Kafr El-Akram, Egypt (1.29) (Khoder, 2007), in ambient urban air in Carmen-Campeche, Mexico (2.56–2.70) (Cerón et al., 2013), at industrial locations in Yokohama, Japan (>4.5) (Tiwari et al., 2010), and at a gas station in Yazd, Iran (2.94) (Mehrjerdi et al., 2014). Furthermore, T/B ratios in this study were higher than those detected in previous studies including ambient air of the Cholul site (for four seasons: 0.09–0.30) and in the center of the municipality of Merida (SEDUMA site) (for four seasons: 0.05–0.13) in the Yucatan Peninsula (Mexico) (Bretón et al., 2017), in the semi-urban air in Orleans (France) (0.31) (Jiang et al., 2017), and in ambient air of Mumbai, India (0.64–0.56) (Srivastava et al., 2006). Variations in the T/B ratio could be explained by differences in the areas studied (e.g., gas stations, CNG stations, semi-urban, urban, rural, industrial) and types of vehicles (Alghamdi et al., 2014; Amini et al., 2017; Bretón et al., 2017; Jiang et al., 2017; Miller et al., 2011; Tiwari et al., 2010).

3.6.2. *Xs/B and EB/B ratios*

The ratios of Xs/B (µg m^{-3}/µg m^{-3}) and EB/B (µg m^{-3}/µg m^{-3}) were used to gain insight into the extent of photochemical reactivity and photochemical aging in the ambient air of the studied stations (Bauri et al., 2016; Hoque et al., 2008; Kerchich and Kerbachi, 2012; Khoder, 2007). Ratios of X/B and EB/B exceeding

one (X/B and EB/B > 1) have been proposed to be reflective of sampled air masses that were not photochemically aged because of insufficient hydroxyl radicals (•OH) for reacting with target components such as xylenes and ethylbenzene (Kerchich and Kerbachi, 2012). Fig. 5 represents the ratios of xylenes to benzene (Xs (μg m^{-3}) to B (μg m^{-3})) and ethylbenzene to benzene (EB (μg m^{-3}) to B (μg m^{-3})) for the 12 sampling locations. Accordingly, in the study area, the mean values of the EB/B and X/B ratios were limited to 1.05–1.21 and 1.77–3.91, respectively. Hence, Xs and EB were relatively stable in this the study area (gas and CNG stations) compared with a previous study that reported that these pollutants cannot remain for a long time in the ambient air of Point Matatula (American Samoa), Cape Grim (Tasmania), and Aldrigole (Ireland) (Prinn et al., 1987). Moreover, the Xs/B ratio values (1.77–3.91) in this study exceeded one, in contrast to other areas like in the ambient air of Bangkok, Pathumwan District, Thailand (0.70) (Tunsaringkarn et al., 2015), in a hot spot of urban pollution in Shiraz, Iran (0.49–0.89) (Dehghani et al., 2018c), in the ambient air of Ahvaz, Iran (0.8) (Rad et al., 2014), and in the suburban Ankara atmosphere, Turkey (0.18–1) (Yurdakul et al., 2013).

The EB/B ratio (1.77–3.91) in this work is also more than that in the suburban Ankara atmosphere, Turkey (0.38) (Yurdakul et al., 2013), in urban air of Gdansk, Gdynia and Sopot, Poland (0.20) (Marć et al., 2014), in urban air of Shiraz, Iran (0.19–0.28) (Dehghani et al., 2018c), in ambient air of Manila, Philippines (0.6) (Gee and Sollars, 1998), and in ambient air of Bangkok, Thailand (0.6) (Gee and Sollars, 1998). Tiwari et al. (2010) and Gaur et al. (2016) reported that the ratio of EB/B in an industrial area of Yokohama, Japan (Tiwari et al., 2010) and in a hot spot (Mathura road) in New Delhi, India (Gaur et al., 2016) were limited from 1.3 to 3.8 and 1.09, respectively. In addition, Gee and Sollars (1998) reported that EB/B ratios in urban air of Quito (Ecuador), Santiago (Chile), São Paulo (Brazil), and Caracas (Venezuela), respectively, were 2.3, 2.3, 2.6, and 2.8 (Gee and Sollars, 1998).

3.7. HRA for BTEX compounds at different distances from stations

Chosen parameter values utilized for HRA and sensitivity analysis (SA), including values to compute LADD, LTCR, and HQ, are summarized in Table S1. Table S6 reports results of LTCR, HQ, and SA for model simulations of BTEX species in the ambient air of the 12 stations and at different distances from station centers. The mean ± (SD) LTCRs calculated for benzene and ethylbenzene in different age groups (birth to <81) were higher than the proposed limits by the U.S. EPA. On the other hand, the LTCRs for target compounds for different age groups and distances were limited to 2.11×10^{-4} to 1.82×10^{-3} and 2.30×10^{-4} to 2.01×10^{-3}, respectively, which exceeded recommended values by the U.S. EPA.

These LTCR values were compared with previous studies in different regions around the world: intercity Karandish bus

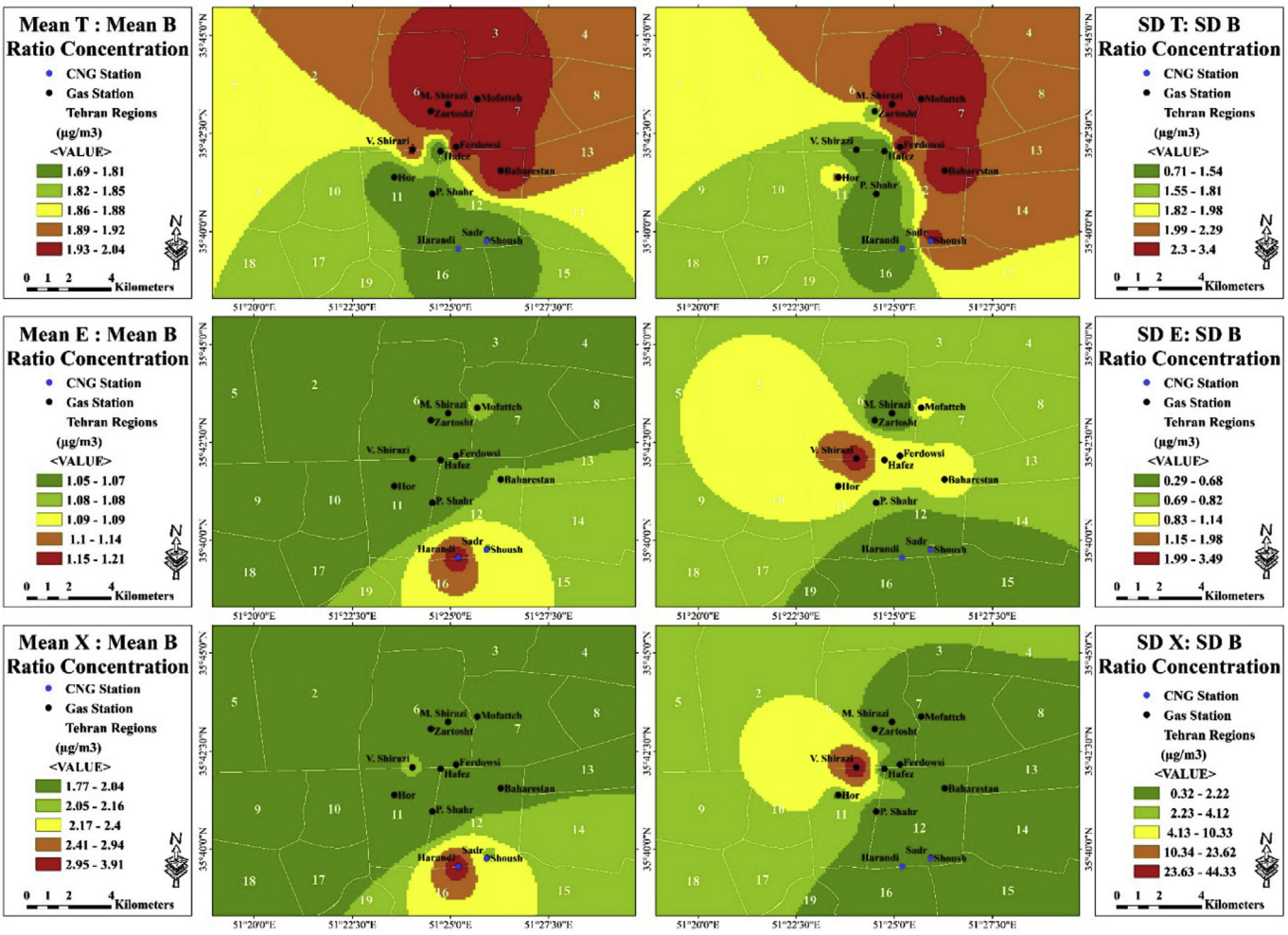

Fig. 5. The ratios of toluene to benzene (T (μg m^{-3}) to B (μg m^{-3})), xylenes to benzene (Xs (μg m^{-3}) to B (μg m^{-3})), and ethylbenzene to benzene (EB (μg m^{-3}) to B (μg m^{-3})) for 12 sampling locations during the winter.

terminal in Shiraz, Iran (1.96×10^{-4} to 2.49×10^{-4}) (Dehghani et al., 2018c); Oregon (Beaverton, Downtown, Forest Heights, Lafayette, and Roselawn), Portland (from 1.08×10^{-5} to 1.94×10^{-5}) (Tam and Neumann, 2004); nine gas stations in Chonburi, Thailand (199.6×10^{-6}) (Yimrungruang et al., 2008); service stations and petroleum refineries in Canada, United Kingdom and Australia (340×10^{-6} to 1800×10^{-6}) (Edokpolo, 2014); gasoline and CNG stations in Ardabil, Iran (1884×10^{-6}) (Hazrati et al., 2016b); parking lot in Bangkok, Thailand (4.37×10^{-6} to 1.47×10^{-6}) (Loonsamrong et al., 2015); service station environments in different countries such as France, Spain and Mexico (2×10^{-6} to 340×10^{-6}) (Edokpolo et al., 2014); petrol station in Bangkok, Thailand (1.82×10^{-4} to 2.50×10^{-4} for benzene and 7.81×10^{-6} to 1.04×10^{-5} for ethylbenzene) (Kitwattanavong et al., 2013); 10 gas stations in the Salvador and Feira de Santana, Brazil (1.82×10^{-4}) (Cruz et al., 2017); diesel station at a bus depot in Johannesburg, South Africa (3.78×10^{-4}) (Moolla et al., 2015a); and a gasoline station in Bangkok, Thailand (1.75×10^{-4} to 9.55×10^{-7}) (Tunsaringkarn et al., 2012).

The findings of this study showed that the HQs of the BTEX target compounds for different age groups and distances (birth to <81) were lower than the acceptable limit (HQs < 1). In contrast, values from other regions are as follows: 18 sampling locations in the ambient air of Shiraz, Iran (from 2.59×10^{-3} to 8.71×10^{-1} for benzene, toluene and ethylbenzene compounds) (Dehghani et al., 2018c); indoor air of beauty salons in Ardabil, Iran (from 0.108×10^{-1} to 6.23×10^{-2} for benzene, toluene and ethylbenzene compounds) (Baghani et al., 2018); gas station in Bangkok, Thailand (0.002–0.6) (Tunsaringkarn et al., 2012); petrol Station Workers in Bangkok, Thailand (0.004–0.351) (Kitwattanavong et al., 2013); parking lot in Bangkok, Thailand (0.006–0.360) (Loonsamrong et al., 2015); and nine gas stations in Chonburi (Thailand) (0.03–0.4) (Yimrungruang et al., 2008).

In contrast, HQs of target compounds ranged from 0.01 to 1.70, 0.097 to 17.73, 0.012 to 15.78, 5.76×10^{-3} to 1.84, and 0.0012 to 77, respectively, for the following cases: Heibati et al. (2017) in petroleum product distributors in Sari, Mazandaran, Iran (Heibati et al., 2017), Hazrati et al. (2016b) in atmospheric air of gasoline and CNG stations in Ardabil, Iran (Hazrati et al., 2016b), Moolla et al. (2015a) in diesel station at a bus depot in Johannesburg, South Africa (Moolla et al., 2015a), Cruz et al. (2017) in the ambient air of gas stations in the Salvador and Feira de Santana, Brazil (Cruz et al., 2017), and Edokpolo et al. (2015) in service station and petroleum refinery environments in Australia, Canada and United Kingdom (Edokpolo et al., 2015).

As can be seen from Table S1, the variables applied for sensitivity analysis (SA) of the model Monte Carlo simulations for ethylbenzene and benzene contained exposure duration (ED), body weight (BW), exposure frequency (EF), averaging time (AT), and inhalation rate (IR). As can be observed from Table S6, the most positive effects on the average LTCR for various age groups (birth to <6, 6 to <21 and 21 to <81) stemmed from body weight (BW) and concentrations of benzene and ethylbenzene (C). Similarly, Baghani et al. (2018) and Dehghani et al. (2018) have stated that concentrations of target pollutants (C) and BW had the most positive impact on the mean LTCR for the air in 50 beauty salons in Ardabil (Iran) and in the ambient air of Shiraz (Iran), respectively (Baghani et al., 2018; Dehghani et al., 2018c).

4. Conclusions

This study concentrated on characterizing the health effects and concentrations of BTEX compounds in the atmospheric ambient air of gas and CNG stations in four regions of Tehran, Iran. The mean concentrations of BTEX species measured at the center of the gas stations (0 m) were 0.93–1 times less than those at a distance of 250 m. Spatial analysis showed that concentrations of BTEX species reduced with increasing distance from the stations. No significant relationship was observed between either the specific month or distance from the center of the station with concentration of BTEX ($p > 0.05$). The mean LTCRs for benzene and ethylbenzene in winter for different age groups and distances from stations were higher than the proposed value by the U.S. EPA. The HQ of BTEX components in three age groups and different distances were less than recommended limits. The results of this work are applicable to similar areas with high populations and many gas/CNG stations with the goal of motivating strategies to curb the negative effects of emissions from these point sources. Our findings indicate that controlling emissions of BTEX compounds and monitoring employees' exposure and protecting public health near these areas are important. It is important to decrease exposure of people to high BTEX concentrations in hot spots such as gas/CNG stations and to move such facilities outside urban centers. Additionally, regulations to reduce pollution from gas/CNG stations are recommended and populated buildings would benefit from being far from such pollutant hot spots.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.01.070.

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