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Benzene, toluene, ethylbenzene and xylene concentrations in atmospheric ambient air of gasoline and CNG refueling stations

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Abstract This study aimed to assess workers' exposure to benzene, toluene, ethylbenzene, and xylene (BTEX) compounds in refueling stations of Ardabil city (Iran). Twenty-four refueling stations including 15 petrol and 9 compressed natural gas (CNG) stations from different regions were selected and monitored for ambient BTEX concentrations. Air samples were taken based on NIOSH Manual of Analytical Method no 1501. Target compounds were extracted using CS_2 and analyzed by GC equipped with FID. Average concentrations of benzene, toluene, ethylbenzene, and xylene were obtained 2.01, 1.80, 2.72, and 1.65 mg/m^3, respectively. Benzene concentrations exceeded the occupational exposure limit set by the Iran Ministry of Health and Medical Education. Its concentrations were significantly higher in commercial areas (2.72 mg/m^3) compared to suburban areas (1.89 mg/m^3). BTEX concentrations in gasoline stations were slightly, but not significantly, higher than those in CNG stations. Long-term exposure cancer risk of $1884\times10^{-6}\pm390\times10^{-6}$ and hazard index of 22.83±3.66 were estimated for benzene and BTEX compounds, respectively. The results declare the necessity for controlling BTEX emission (mainly benzene) and monitoring employee's exposure in refueling stations.

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Introduction

Development of industries in recent decades has led to serious environmental problems including emission of volatile organic compounds (VOCs) into the urban atmosphere. Benzene, toluene, ethylbenzene, and xylene, known as BTEX, are environmentally important VOCs and emitted into the atmosphere from both artificial and natural sources (Caselli et al. 2010; Davil et al. 2013; Fazlzadeh Davil et al. 2012; Liu et al. 2009; Sturaro et al. 2010; Tiwari et al. 2010). These compounds are used as raw materials or solvents in various industries and released into the environment through evaporation, leaking from underground fuel tanks as a result of poor maintenance, vehicle exhaust emissions, combustion of fossil fuels, and evaporative emissions from refueling vehicles, and therefore, traffic-related sources are considered as the most important sources for indoor and outdoor ambient air pollution (Bailey and Eggleston 1993; Bauri et al. 2015; Caselli et al. 2010; Esteve-Turrillas et al. 2007; Rad et al. 2014; Singh et al. 1992; Truc and Kim Oanh 2007). Sources for BTEX in indoor environments include infiltration of outdoor air pollution, smoking, paints, adhesives, and other VOC-emitting materials utilized in building interiors (Hazrati et al. 2015; Singh et al. 1992; WHO 2000). BTEX compounds are known as a significant cause of cancer in humans and may develop neurological disorders and symptoms such as weakness, loss of appetite, fatigue, confusion, and the nausea (Hoskins 2011).

Benzene is the most toxic compound within the BTEX, and long-term exposure to its low concentrations may increase the incidence of leukemia and aplastic anemia in humans (Baker et al. 1985; Mehlman 1990; Niri et al. 2009; Wong 1995). The

World Health Organization has reported that prolonged exposure to 1.7 μg/m^{3}of benzene causes leukemia in 10 people per million (WHO 2000). The International Agency for Research on Cancer (IARC) has classified benzene as an intense carcinogenic compound and ethylbenzene as a suspected carcinogenic compound (IARC 1999). On the other hand, the presence of VOCs, such as BTEX, increases levels of photochemical oxidants especially ozone in the atmosphere that is potentially dangerous to human health and the environment (Atkinson 2000; Khoder 2007).

Several studies have monitored air quality of petrol stations in terms of BTEX concentration (Backer et al. 1997; Cheng et al. 1990; Egeghy et al. 2000; Esteve-Turrillas et al. 2007; Lagorio et al. 1998; Tironi et al. 1986; Vainiotalo et al. 1999). Most of these studies considered the petrol-fueled vehicles as the emission sources for these compounds. Gasoline is a mixture of over 200 petroleum-derived chemicals, mainly low-molecular-mass compounds (e.g., paraffinic, naphtenic, olefinic, and aromatic) with a range of boiling points from 38 to 150–205 °C and a carbon number distribution of C4–C12. Its composition varies depending on the crude oil origin and the refining process. Among other chemical hazards, the group of aromatic components of gasoline constituted by benzene, toluene, ethylbenzene, and xylenes (BTEXs) are considered to be the most hazardous components of most gasoline fuels (El-Naggar and Majthoub 2013; Periago and Prado 2005).

It is believed that approximately 60–85 % of atmospheric benzene is derived from mobile sources, and therefore, higher concentrations are often found inside motor vehicles, inside petrol stations, and adjacent to major roadways (Carey 1987; Winebrake and Deaton 1999; Winebrake et al. 2001). Petroleum derivatives and consequently petrol stations, as the important sources, may contribute to BTEX compounds in urban atmosphere. Therefore, it is important to monitor BTEX levels in petrol stations' ambient air to pursue proper planning and management policies. Since there are no data published on VOC concentrations in Ardabil city, concentrations of BTEX compounds were monitored in ambient air of refueling stations.

Methods

In a cross-sectional study, 15 petrol stations and 9 compressed natural gas (CNG) stations from different regions of Ardabil city were selected and their ambient air was monitored for BTEX compounds. Among these, 7 stations were located in residential areas, 3 in commercial, and 14 in the suburban areas (Fig. 1). Air samples were taken from standing breathing zone of employees, ~150 cm above the ground level.

Air sampling process

Air samples were taken based on NIOSH Manual of Analytical Method no 1501. SKC personal sampling pumps equipped with adjustable low-flow holders were used for air sampling. Flow rates were calibrated using a soap bubble flow meter. Air sampling was performed at the flow rate of 0.2 L/min and continued for 50 min to collect a total air volume of 10 L. Coconut charcoal sorbent tubes (SKC, Anasorb CSC, 6×70-mm size, 2 sections, 50/100-mg sorbent, and 20/40 mesh) were used for this purpose. The tubes were attached to collar of workers, and after completion of the sampling period, the tubes were detached and transported to laboratory according to the manufacturer guideline. They were stored at −20 °C and analyzed within 72 h.

Sample preparation and analysis

BTEX compounds were extracted from charcoal tubes by 2 mL of GC-grade carbon disulfide (CS_2). The vials containing CS_2 and charcoal were gently shaken for 20 min. The solvent was transferred into GC vials and BTEX compounds were quantified by a GC (Agilent 7890A) equipped with an FID detector using a capillary column (TRB-1 ms, 30 m×

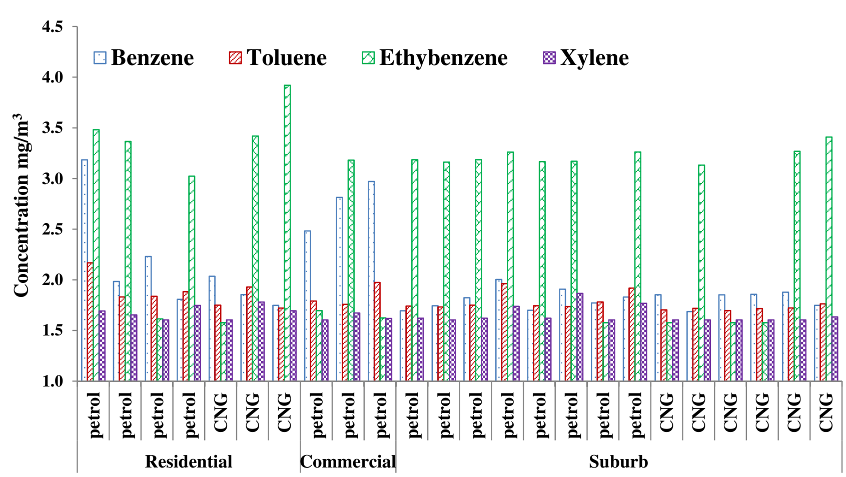

Fig. 1 Concentration of BTEX compounds at the different sampling sites

0.32 mm, 0.25 μm in film thickness). The injector was maintained at the split mode with the ratio of 1:5 and temperature of 300 °C. Nitrogen (99.999 %) was used as the carrier gas at a flow rate of 2.6 mL/min. Aliquots of 1 μL were taken from the vial and injected into a capillary column. Injector temperature was set at 250 °C. Oven temperature was programmed at 40 °C for 10 min and then 10 °C/min to 230 °C (Hazrati et al. 2015; NIOSH 2003; Rezazadeh Azari et al. 2011).

QC and QA measures

In order to control breakthrough of the sorbent tubes, front and back sections of the tubes were analyzed separately, and none of the target compounds was detected in the back section for all samples. Recovery of analytical method was tested by injecting 10 μg of standard solution, and average recovery of 92 % (87–102 %) was found. Method and field blanks were taken (one for each batch of five) and subjected to the same preparation and analytical procedures. Concentration of target compounds were found to be less than 5 % of the values quantified for the samples, and therefore, no correction was made for blank values.

The GC detector was calibrated by running a five-point calibration standard solution of BTEX ranging from 0.01 to 50 ppm, and R^2 values of 0.992, 0.981, 0.980, and 0.983 were obtained for benzene, ethylbenzene, toluene, and xylene, respectively.

Statistical analysis and the risk assessment

Other variables including the region that stations were located (i.e., residential, commercial, and suburban areas), type of the fuel served (i.e., gasoline or CNG), and station information were collected using a self-designed questionnaire. Levels of BTEX in indoor air were compared with national occupational limit levels. Data obtained were analyzed by *t* test and non-parametric tests using SPSS version 16, and the confidence level was set at 95 %.

The risk assessment analysis was comprised based on the recommended reference doses and work condition of the refueling stations' employees (Table 1), and the calculations were carried out as reported by Majumdar et al. (2011) for the BTEX compounds.

Table 1 Details of the risk assessment parameters

	Value	Unit
Inhalation rate, adult (IRa)	0. 83	m^3/h
Exposure duration, adult (EDa)	8	h/day
Body weight, adult (Bwa)	70	kg
Days per week exposure (D)	6	day
Weeks of exposure (WK)	48	week
Years of exposure (YE)	30	year
Years in lifetime (YL)	70	year
Slope factor or carcinogenic potency slope (SF)	Benzene=0.029 (Guo et al. 2004)	mg/kg day
Reference dose (RfD)	Benzene=0.00855 Toluene=1.4 Ethylbenzene=0.286 Xylene=0.029	mg/kg day

RfD=RfC (inhalation reference concentration mg/m^3)×20 (assumed adult inhalation rate m^3/day)×1/Bwa (kg); based on RfCs for USEPA, IRIS (benzene=0.03 mg/m^3, toluene=5 mg/m^3, ethylbenzene=1 mg/m^3, and xylenes=0.1 mg/m^3) (USEPA 2007).

Table 2 Minimum, maximum, mean, and standard deviation of the BTEX compounds in fuel stations

Contaminant	Minimum (mg/m^3)	Maximum (mg/m^3)	Average (SD) (mg/m^3)	OEL (mg/m^3)
Benzene	1.69	3.18	2.01(0.41)	1.6
Toluene	1.70	2.17	1.80 (0.11)	75
Ethylbenzene	1.58	3.92	2.72 (0.82)	87
Xylene	1.61	1.87	1.65 (0.07)	434

Results

The ambient BTEX concentrations in petrol and CNG stations are presented in Fig. 1 and Table 2. The average concentration of benzene (2.01±0.41 mg/m^3) was higher than (1.6 mg/m^3) the occupational exposure limit (OEL) recommended by Iran Ministry of Health and Medical Education (Table 2). The average concentrations of 2.01±0.41, 1.80±0.11, 2.72±0.82, and 1.65±0.07 mg/m^3were determined for benzene, toluene, ethylbenzene, and xylene for all the monitored refueling stations, respectively. Within the BTEX compounds, only benzene concentrations significantly differed between the areas monitored (i.e., residential, commercial, and suburban) (*p* value=0.02) with the commercial area having the highest average benzene concentration of 2.72 mg/m^3 (Table 3). BTEX concentrations were

Table 3 Average concentrations of BTEX compounds based on types of areas and fuel

Area/Fuel type	Benzene (mg/m^3)	Toluene (mg/m^3)	Xylene (mg/m^3)	Ethylbenzene (mg/m^3)
Residential	2.06	1.85	1.67	2.94
Commercial	2.72	1.88	1.61	1.66
Suburban	1.89	1.76	1.65	2.75
p value ANOVA	0.020	0.108	0.545	0.143
Gasoline	2.13	1.84	1.66	2.79
CNG	1.83	1.74	1.63	2.6
p value *t* test	0.094	0.049	0.310	0.597

Table 4 The risk assessment results for workers exposure to BTEX

	Risk			HQ		
	Min	Max	Mean	Min	Max	Mean
Benzene	1574×10^{-6}	2971×10^{-6}	$1884\times10^{-6}\pm390\times10^{-6}$	14.81	27.95	17.73±3.67
Toluene	–	–	–	0.091	0.116	0.097±0.0061
Ethylbenzene	–	–	–	0.41	1.03	0.72±0.23
Xylene	–	–	–	4.15	4.83	4.29±0.19

higher in petrol serving stations compared to the CNG refueling stations (Table 3). However, only toluene concentrations were statistically different between the stations serving different fuel types (p value=0.049).

The risk assessment for long-term exposure of employees to BTEX compounds showed notably high cancer risk values for benzene exposure in the refueling stations (Table 4). The average cancer risk of $1884\times10^{-6}\pm390\times10^{-6}$ was significantly higher than the unit cancer risk value (i.e., 1×10^{-6}). Moreover, the average non-cancer risk values, expressed as hazard quotient (HQ), for exposure to benzene (17.73±3.67) and xylene (4.29±0.19) were higher than the unit value of 1. The average value of hazard index (HI) for long-term exposure to BTEX compounds was 22.83±3.66, which is appreciably higher than the unit value of 1.

Discussion

BTEX concentrations in the ambient air of refueling stations

The fuelling process was done by the car owners, and the petrol pumps were not equipped with a vapor recovery system to collect the vapor coming from the car tank during the filling process in our study. According to the results, workers of the refueling stations are exposed to high concentrations of benzene in such public environments. Although ethylbenzene, toluene, and xylene average concentrations were higher than the typical outdoor air levels, they did not exceed national OEL time-weighted average (TWA) values.

Benzene concentrations in ambient air of petrol stations have been monitored in some countries. The levels reported for Sudan, Saudi Arabia, Thailand, Brazil, and Italy (Caselli et al. 2010; Correa et al. 2012; El-Naggar and Majthoub 2013; Salih and Younis 2013; Tunsaringkarn et al. 2012) are well lower than the concentrations found in the present study. This might be due to differences in VOC contents of the petrol, fuel vapor control systems applied in refueling stations, and the volume of petrol sold per day.

However, other studies conducted in Tehran (Iran) and the USA (Egeghy et al. 2000; Fazlzadeh Davil et al. 2012; Mosaddegh et al. 2014) found the concentrations much higher than the values reported in our work. The exact reason for these differences are not clear; however, the high concentration might reflect less safe refueling methods, high number of cars served, and consequently high volume of petrol sold by Tehran stations. In the USA case, exposure to benzene was measured during self-service refueling process and that petrol vapor releasing from the car tank while refueling might be responsible for the quantified high concentrations. The average benzene levels before and after refueling were about 8.6 and 160 μg/m^3, respectively.

Table 5 BTEX levels (μg/m^3) reported for fuel station ambient air

Country	City	Benzene	Toluene	Ethylbenzene	Xylene	Reference
Sudan	Khartoum	281	–	–	–	Salih and Younis (2013)
Thailand	Bangkok	295	736	27	50	Tunsaringkarn et al. (2012)
Spain	Murcia	163	753	–	316	Periago and Prado (2005)
Iran	Tehran	5200	10254	1996	15320	Rezazadeh Azari et al. (2011)
Brazil	Rio de Janeiro	29.7	47.7	23.3	61.2	Correa et al. (2012)
USA		2900	–	–	–	Egeghy et al. (2000)
Iran	Hamadan	–	–	–	4567.5	Bahrami et al. (2008)
Iran	Yazd	1932	667	148	340	Mosaddegh et al. (2014)
Italy	Rome	181.8	452.1	–	–	Lagorio et al. (1998)
Iran	Ardabil	1690	1700	1580	1610	Present study

Unlike benzene, the toluene, ethylbenzene, and xylene concentrations found in the present study are well below the OEL-TWA values and lower than the results reported for ambient air of most petrol stations studied worldwide (Table 5). However, air pollution due to toluene and ethylbenzene and xylene (TEX) in Tehran refueling stations was appreciably higher than those of found in our work (Rezazadeh Azari et al. 2011).

The average concentrations of BTEX compounds in this study are much higher than the average values reported for urban atmospheric ambient air (Buczynska et al. 2009; Caselli et al. 2010; Kerchich and Kerbachi 2012; Sturaro et al. 2010). This may imply the presence of pollutant sources and a lower rate of photochemical reactions responsible for atmospheric degradation of BTEX due to covered roof and lack of intense solar radiation in refueling stations (Sturaro et al. 2010).

Gasoline is a complex mixture of mainly low-molecular-mass paraffinic, naphtenic, olefinic, and aromatic compounds. Its composition varies depending on the crude oil origin and the refining process applied as well as the national regulations limiting specific constituents of the petrol. Based on regulations adopted by European Parliament, the aromatic content of the gasoline must be <35 % *v*/*v*. This figure for benzene is <1 % *v*/*v* (Commission 2009). The concentrations of aromatic compounds especially BTEX detected in ambient air of different gasoline stations should normally vary based on the standards applied for manufacturing of the fuel served. Since different refineries in the Islamic Republic of Iran produce petrol with different quality in terms of aromatic content, therefore, air pollution to BTEX compound may vary in gasoline stations based on the fuel-providing refinery.

Unlike benzene concentrations that were significantly higher in commercial (2.72 mg/m^3) comparing to suburban areas (1.89 mg/m^3), TEX concentrations were not significantly differed between the stations located in different areas, namely, residential, commercial, and suburbs ($p>0.05$). Since industrial settlements, commercial centers, and urban traffic vary in different areas, relatively similar levels of TEX found for the stations located at different areas imply that air quality at petrol stations are not influenced by surrounding atmospheric air quality.

Average BTEX concentrations in ambient air of gasoline pump stations (Table 3) were higher than those of CNG stations, though the differences were not statistically significant. The maximum benzene concentrations were observed in the gasoline station No. 1, 8, and 12 and the minimum concentrations were recorded for CNG station No.9, 15, and 24. This might be due to more strict safety regulations applied for CNG refueling stations as well as the fact that BTEX compounds were present at very low concentrations (*v*/*v*) in CNG fuel (Bakar 2008).

Exposure risks of BTEX in the refueling stations

The hazard index of 22.83±3.66 shows a great concern in terms of workers' exposure to BTEX compounds at refueling stations. The main causes of the concern are benzene and xylene, which have HQs considerably higher than the unit value. Many non-cancer effects were reported for exposure to the BTEX compound such as disturbances of memory, mood, equilibrium, and sleep that occurred simultaneously with headache and indigestion (Kilburn et al. 1985). Chronic human inhalation exposure to xylenes can cause kidney, liver, and nerve damage. It can also affect the central nervous system with symptoms such as headache, fatigue, dizziness, and memory loss (Gunathilaka 2003). A significant relationship has been reported between benzene and toluene exposure with fatigue among the refueling station workers (Tunsaringkarn et al. 2012). They also reported a high cancer risk value of 175×10^{-6} for long-term exposure of workers in refueling stations. This high cancer risk agrees well with the results of the present study.

Conclusions

BTEX concentrations present in refueling stations are generally higher than typical outdoor air levels suggesting petrol and CNG stations as sources for atmospheric air pollution. In terms of BTEX levels, air quality of gasoline fuel stations is poorer than that of the CNG stations. This highlights the necessity for improvement in quality of gasoline supplied (i.e., meeting standards of VOC and benzene content *v*/*v* of the fuel) as well as the enhancement of safety measures applied while refueling at petrol stations. Among the BTEX compounds, ethylbenzene has the highest average concentration followed by benzene. However, only benzene levels were higher than the occupational exposure limits set by the Iran Ministry of Health. Also, there is a big concern about non-cancer effects for xylene and especially cancer risk for benzene exposure for the workers of refueling stations. Therefore, assessment of worker exposure through monitoring benzene and its derivatives in biological samples and comparing with biological exposure indices are highly recommended.

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