Original Article

Monitoring of benzene, toluene, ethyl benzene, and xylene isomers emission from Shahreza gas stations in 2013

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ABSTRACT

Aims: The aims of this study were to monitor the concentration of benzene, toluene, ethyl benzene and xylenes (BTEX) in the ambient air of the city of Shahreza gas stations and to identify the spreading distance of the pollutants from the fueling stations.
Materials and Methods: Sampling was carried out from the air of 10 existing fuel stations, (2 compressed natural gas and 8 gasoline and diesel stations) and points of 50, 150 and 250 m away from the stations during cold and warm seasons in 2013. Air samples were taken via active sampling process using activated carbon tubes, extracted by carbon disulfide and analyzed by a gas chromatograph coupled to a flame ionization detector.
Results: The averages of all achieved BTEX concentrations were under/around the permitted guideline levels for occupational exposure. According to the ambient air guidelines, the benzene level was much higher than the suggested levels in all the stations. However, the average concentrations of toluene, ethyl benzene, and xylene were not exceeded from the standards. The seasonal variation had no influence on the concentrations of BTEX. There was no significant difference between the pollutants concentrations at points 50, 150 and 250 m away from the stations.
Conclusions: Fuel stations could be the main sources of volatile organic compounds emission in the city of Shahreza. The number and volume of refueling in the gas stations influence the emission rates. Therefore, it is suggested to take preventive actions such as repairing of pumps and tanks leak and installing vapor return systems at the time of fuel transferring.

Key words: Air pollution, benzene, toluene, ethyl benzene and xylenes, gas station

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INTRODUCTION

Volatile organic compounds (VOCs) are consisted of about 1,000 components including hydrocarbons, aromatic compounds, oxygenated and chlorinated compounds. They are emitted to the atmosphere from anthropogenic and biogenic sources and also be formed in the atmosphere by photochemical and photolysis reactions.[1] Benzene, toluene, ethyl benzene, and xylene (BTEX) isomers are

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among these aromatic compounds which are monocyclic, more volatile and similar to each other in other physical and chemical features. Because of their solubility and volatility, these compounds have the features of diffusion and emission in the environment.[2,3] Their main sources of emission include refueling of motor vehicles, combustion of gasoline, petrochemical industries and various combustion processes.[4] BTEX mostly release to the atmosphere through vehicle exhaust, motor carburetor, refueling evaporation in fuel and automobile service stations.[2,3,5] Its emission from the transportation sector is more than the half of total emissions, and gas stations are the second major source of emission.[6] BTEX also plays an important role in the atmospheric chemistry. It has been recognized as an important photochemical precursor for tropospheric ozone and second organic aerosols.[7]

Although the BTEX can enter to the human body via congestion, skin absorption and breathing, the latter is the most important way of exposure. Human exposure to BTEX can have serious health consequences such as neurological diseases or variety of cancers.[4] BTEX compounds are in the list of hazardous pollutants.[5,7] Gasoline is a complex combination, which is formed from 50 different hydrocarbons that some of them such as benzene are dangerous.[8] According to the International Agency for Research on Cancer, benzene has been classified as group 1 carcinogenesis.[5,9] Therefore, monitoring of these harmful compounds is becoming an important issue.

Large number of studies have proved a strong association between occupational exposure to benzene by inhalation and an increased incidence of certain types of leukemia.[5] Benzene affects bone marrow, which is the tissue that produces blood cells.[10] Other adverse health effects of benzene include acute myelogenous leukemia, blood diseases, plastic anemia, injury of the immune system, menstruation disorders.[4] The main effect of toluene on human is on the central nervous system. Its exposure can cause tiredness, dizziness, faintness, memory loss, nausea, appetence decrease, hearing loss, and unconsciousness. Acute exposure to elevated levels of ethylbenzene may result in neurological effects such as light-headiness, dizziness and eyes irritation.[10] Ethyl benzene has the features of spouting and can cause blood cancer. Chronic exposure to xylene may also affect the central nervous system.[11] Exposure to xylene may cause skin irritation, skin stimulation, dryness, skin rapture, blister and skin dermatitis.[2] Very high level of xylene in atmosphere can cause unconsciousness and the death of individuals.[10]

The allowed level of these compounds in the atmosphere for BTEX are 0.5, 50, 100, and 100 ppm respectively.[2] Also, the National Institute for Occupational Safety and Health (NIOSH) issued guidelines for BTEX are 0.1, 100, 100, and 100 ppm, respectively.[12] The European Union and United State Environmental Protection Agency have announced that the mean annual standards of ambient air for benzene is 5 and 10 μg/m^3 respectively. These guidelines for toluene, ethyl benzene and xylene are 0.92, 0.106 and 0.22 ppm, respectively.[8,13]

Several researches have been conducted in association with the monitoring of BTEX compounds in the atmosphere of different regions.[13-19] Some of them have reported about exceeded amounts of BTEX emission in the air of fuel stations.[6] For instance, monitoring of BTEX emission in Rio de Janeiro gas station, Sergio *et al.*, reported an average value of 29.7 μg/m^3 for benzene, 47.7 μg/m^3 for toluene, 23.3 μg/m^3 for ethyl benzene, 46.9 μg/m^3 for m+p-xylene and 14.3 μg/m^3 for o-xylene.[6] Among them, only the amount of benzene was upper than the European standards. Ambient level of different VOCs at 5 busy refueling stations in Kolkata, India was monitored by Dutta *et al*. The average exposure levels for benzene and toluene were 3.9 and 5.5 fold higher than those standards in ambient air.[20] Mehrjerdi *et al*. measured the BTEX emission in Yazd, Iran gas stations area using solid phase micro-extraction technique in winter 2010. They reported mean concentrations of 1932 ± 807 μg/m^3 for benzene, 148 ± 89 μg/m^3 for toluene, 667 ± 405 μg/m^3 for ethyl benzene and 340 ± 216 μg/m^3 for xylene.[2] Esteve-Turrillas *et al*., conducted a research on monitoring BTEX inside vehicles and at filling stations by semi-permeable devices. They measured BTEX levels of 0.03-79 mg/m^3 for filling times from 2 to 40 min, especially during refueling of vehicles with gasoline.[4] Zhang *et al*., determined the atmospheric level of BTEX during the 2008 Olympic games in the urban area of Beijing. During the games, the mean daytime concentrations of benzene, toluene, ethyl benzene, m+p-xylene and o-xylene were 2.37, 3.97, 1.92, 3.51 and 1.90 μg/m^3, respectively, which were 47-64% lower than those after the games.[21]

Based on similar studies conducted all over the world and particularly in Iran, and due to having similar construction of gas stations in Iran, it is suggested that the levels of BTEX emission in the gas stations of urban area can be possibly high. The main aims of this research were to determine the concentration of BTEX in the ambient air of the city of Shahreza gas stations area and to identify the spreading distance of the pollutants from the emission sources. The effects of meteorological parameters and seasonal variation on the pollutants concentrations were also investigated.

MATERIALS AND METHODS

Study area

This study was carried out in Shahreza, one of the Isfahan province cities, located in the center of Iran with a population of more than 130,000 and area of 2,820 km.[2,22] Its geographical longitude and latitude are 51°, 52′ east and 32°, 1′ south, with an elevation of 1,825 m from sea level. Shahreza is not an industrialized city; however, there are some industrial region and service areas such as vehicle mechanics and other workshops inside and around the city. There is also another big industrial region located 20 km far away in which its pollutants do not influence the city. The number of vehicles in the city is more than 60,000 with an annual increase of 10%. Fuels used in this city include gasoline, diesel, compressed natural gas (CNG) and liquid petroleum gases. Entirely there are 8 fuel stations of gasoline and diesel and 3 CNG stations in the city.

Sampling points and periods

Sampling was conducted at cold (February and March) and warm (July and September) seasons of year 2013. The sampling points was 16 and from each point, each season 2-4 samples were collected (sample size = 78). Sampling was done from 8 fuel stations, 2 CNG stations and 6 points of 50, 150 and 250 m away from the 2 busy fuel stations for evaluating the spreading of pollutions from the refueling points. Figure 1 shows the sampling points on the city map.[6,8] Samples were taken from 1.7 m above the ground and in the fuel stations 2 m far from the filling pumps among the routine daily activities.[6,14] In a time of sampling, meteorological parameters such as temperature, wind speed, and relative humidity of the air were also measured. Sampling was not taken in much unstable atmosphere with speedy winds and in rainy days.

Sampling methods and analyses

Air samples were taken via active sampling process using a low flow rate sampling pump (SKC Inc., England, model 222-3). An air flow of 100 ml/min was passed through a dual packed activated carbon tube (SKC Inc., England, 226-01) for 3 h to swapped with adsorb the organic pollutions.[14] Sampling and BTEX analysis were carried out based on the NIOSH

Figure 1: Sampling points (fuel stations) on the Shahreza city map for benzene, toluene, ethyl benzene and xylenes monitoring

and SKC instructions guidelines.[20] In this way, the activated carbon inside the tube was separated to two parts of 50 and 100 mg by polyurethane foam and settled in glass tube with 7 cm long and 4 mm inner diameter.[14] Prior to sampling, a gas meter device calibrated the sampling pump. All the chemicals used in this research such as carbon disulfide (CS_2), standards of BTEX isomers were HPLC grade and supplied by Merck Company.[2]

Samples were transferred to the laboratory in the same day under cold condition inside an ice box. Activated carbon materials of the tube were separately transferred to 1.8 ml vials, and 1 ml CS_2 was added to the vial as solvent. The caps of vials were closed and then settled in vibrator for 30-60 min for proper desorption of BTEX from activated carbon into the solvent.[14,20] BTEX contents of the extracts were immediately analyzed using a gas chromatograph coupled to a flame ionization detector (Agilent Technology, 7890, GC-FID). A fused silica column, HP-5 (5% phenyl, 95% dimethyl polysiloxane; 30 m × 0.25 mm I.D, 0.25 μm), was employed for separation with helium (purity 99.995%) as a carrier gas at flow rate of 1 ml/min. The amount of injection was 1 μl with a split ratio of 1-5 and the temperature of injection line was 200°C. The temperature program of the column oven was 40°C for 1 min, 40°C-150°C with a ramp of 10°C/min and staying at this temperature for 3 min. Detector temperature was set at 250°C, hydrogen flow rate was 30 ml/min, air flow was adjusted at 300 ml/min and nitrogen gas flow was programmed at 30 ml/min used as a purge gas. This method gives a detection limit of nearly 1.0 μg/L of injection solution.

For the calibration of the instrument, known concentrations of BTEX prepared from the dilution of pure standards in CS_2 were introduced to the GC. To test repeatability and accuracy of the system, 10% of the samples were analyzed in triplicate and correlations of about 99% were achieved. In order to correct the sampling and preparation errors, blank samples, prepared from the extraction of the raw adsorbent without passing the air through it, were analyzed. To control possible penetration of the pollutants during sampling, the adsorbents of both parts of the tube were analyzed.

RESULTS

The concentrations of BTEX in ambient air of fuel stations in the cold and warm seasons of the year and the average of them were represented in Table 1. Figure 2 compares the average amounts of evaluated parameters in both cold and warm seasons. Table 2 presents the concentration of BTEX in ambient air of CNG stations and the average of them at different seasons. The results of BTEX concentration in the ambient air at 50, 150, and 250 m far from two busy fuel stations (A and B) in the cold and warm seasons are shown in Table 3. Table 4 shows the relations between concentrations of pollutants and the volume and number of refueling in the some busy fuel stations. Table 5 presents the amounts and the average of evaluated meteorological parameters during sampling of all places in both cold and warm seasons of the year. Figure 3 illustrated the correlation of the volume of fuel transferred and the pollutants emission levels.

Table 1: BTEX concentrations in the ambient air of Shahreza fuel stations in winter and summer 2013

Season	Sample code*	BTEX concentrations (μg/m³)			
		Benzene	Toluene	Ethyl benzene	Xylenes
Winter (February to March)	A1-4	522.3	1045.4	222.5	1024.9
	B1-4	201.2	397.9	129.8	353.4
	C1-2	73.7	77.8	15.9	49.7
	D1-2	289.7	268.8	92.8	159.6
	E1-4	345.0	607.2	123.5	473.9
	F1-3	482.6	673.8	143.1	469.8
	G1-2	106.5	193.8	31.7	132.7
	H1-4	127.1	273.3	46.0	217.8
Average		268.5	442.2	100.6	360.2
SD		171.2	316.2	68.7	310.7
Summer (August to September)	A5-8	699.4	1290.2	282.4	1320.4
	B5-8	579.0	948.0	152.4	864.6
	C3-4	100.8	165.0	13.2	87.8
	D3-4	224.3	232.3	22.7	125.7
	E5-7	300.8	678.1	156.2	734.2
	F4-5	232.6	323.3	54.9	250.3
	G3-4	153.5	285.2	61.1	178.0
	H5-6	194.3	343.8	61.7	382.3
Average		310.6	533.2	100.6	492.9
SD		213.5	402.5	90.8	439.2

*A to H denote the gasoline and diesel filling stations. SD: Standard deviation, BTEX: Benzene, toluene, ethyl benzene and xylenes

Table 2: BTEX concentrations in ambient air of CNG fuel stations at different seasons

Sample code	BTEX concentrations (μg/m³)			
Winter	**Benzene**	**Toluene**	**Ethel benzene**	**Xylene**
CNG 1	56.61	105.42	ND	29.47
	42.50	67.15	12.78	37.13
CNG 2	44.97	31.09	16.94	35.18
	ND	26.10	12.78	28.14
Average	48.03	57.44	14.17	32.48
Summer				
CNG 1	21.39	27.38	ND	ND
	36.49	107.35	11.59	32.21
CNG 2	31.57	46.16	17.02	33.61
	36.33	59.76	11.98	40.65
Average	31.45	60.16	13.53	35.49

BTEX: Benzene, toluene, ethyl benzene and xylenes, CNG: Compressed natural gas

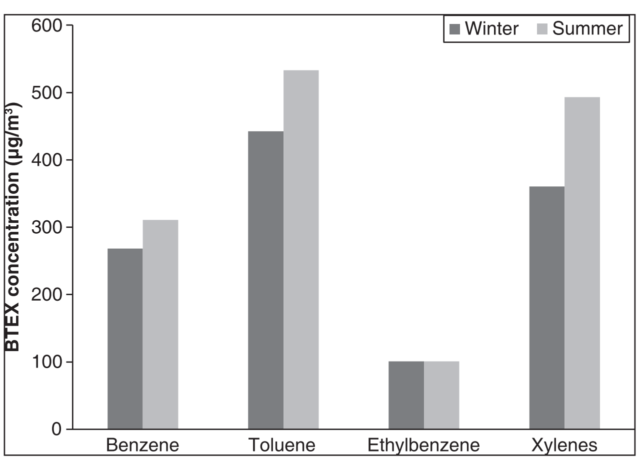

Figure 2: Comparison of the average benzene, toluene, ethyl benzene and xylenes concentration measured in the ambient air of gas stations area in cold and warm seasons

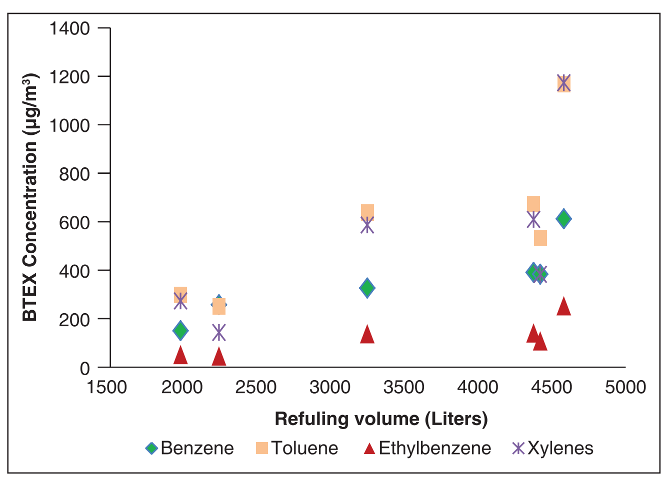

Figure 3: Benzene, toluene, ethyl benzene and xylenes concentrations in the ambient air of gas stations area in relation to the volume of fuel transferred within 3 h

Table 3: BTEX concentration in the ambient air at 50 m, 150 m and 250 m far from the fuel stations in winter and summer 2013

Season	Sample code*	BTEX concentrations (μg/m³)			
		Benzene	Toluene	Ethyl benzene	Xylenes
Winter (February to March)	A-50	27.1	44.06	10.4	20.1
		40.0	31.5	17.2	29.4
	A-150	36.7	52.6	ND	21.4
		26.2	31.4	11.6	36.4
	A-250	ND	34.8	8.0	28.1
		36.3	60.0	21.2	42.6
	B-50	31.0	53.1	18.5	38.1
		38.3	40.6	ND	26.3
	B-150	ND	27.4	23.6	25.9
		33.3	68.8	12.0	36.0
	B-250	25.2	32.5	19.7	29.8
		21.4	59.8	13.2	30.9
Average		32.0	43.9	15.8	31.2
Summer (August to September)	A-50	33.1	64.2	18.4	41.2
		50.3	47.5	12.1	37.1
	A-150	32.8	13.4	ND	7.3
		ND	22.4	5.2	10.6
	A-250	21.4	11.2	4.0	5.5
		12.6	21.8	7.0	10.9
	B-50	42.5	60.1	17.2	39.1
		33.7	36.1	11.3	24.4
	B-150	28.3	62.5	10.0	25.0
		24.8	11.1	4.0	6.8
	B-250	13.3	25.2	5.9	3.9
		16.1	8.4	4.6	5.5
Average		24.9	26.0	7.1	13.7

*These codes represents distance of sampling points from fuel station A and B. BTEX: Benzene, toluene, ethyl benzene and xylenes

Table 4: BTEX concentrations in relation to the rate of refuelling and fuel volume transfer

Fuel station	Number of refueling	Refueling volume (L)	BTEX concentrations (μg/m³)			
			Benzene	Toluene	Ethylbenzene	Xylenes
A	292	4,582	610.83	1167.82	252.45	1172.65
B	286	4,377	390.08	672.99	141.10	608.99
D	113	2,249	256.98	250.52	46.06	142.66
E	209	3,252	326.02	637.56	137.51	585.47
F	243	4,422	382.58	533.56	107.78	381.98
H	97	1,989	149.53	296.82	51.23	272.62

BTEX: Benzene, toluene, ethyl benzene and xylenes

DISCUSSION

Benzene, toluene, ethyl benzene, and xylene concentrations and the guidelines

The average concentrations of BTEX isomers in the air of gas stations in winter were 268.5 ± 171.2, 442.3 ± 316, 100.7 ± 68.6, and 360.2 ± 310.6 $\mu g\ m^{-3}$ and those of in summer were, 310.6 ± 213.5, 533.2 ± 402.5, 100.6 ± 90.8, and 492.9 ± 439.2 $\mu g\ m^{-3}$, respectively [Table 1]. Based on occupational standards for personnel exposure issued by NIOSH[23] and the standards suggested by Iranian technical committee of occupational health[2] for BTEX, the average concentrations of subjected pollutants were under/around the permitted standards level. However, these standards are applicable provided that all the personnel only work 8 h a day and use personal protection devices. Unfortunately, in some of the gas stations the 8 h work shift does not apply and their shift may last 12-24 h. In spite of this, in some of the fuel stations such as A, B and E even the lowest concentration was more than the average of all stations, and in 20 cases of sampling from the fuel stations (38% of the samples) the concentration of benzene was upper than NIOSH occupational standard.

According to the standards of these compounds in ambient air, concentrations of benzene in all the stations were 100–200 μg/m³ above the suggested standards. However, the average concentrations of toluene, ethyl benzene, and xylene were not exceeded from the standards.[8,13] BTEX levels in the CNG stations and 50, 150, and 250 m distances of gas stations were acceptable and under the ranges of ambient air standards except benzene that was a little higher. The results of the present study are similar with the results of work conducted by Mehrjerdi *et al.*, in Yazd; however, they reported higher amount for benzene that could be attributed to the sampling method.[2] The achieved concentrations of BTEX in the research of Keshavarzi *et al.*, carried out in Tehran gas stations were also much higher than our

Table 5: Meteorological parameters measured in the sampling point at different season

Winter				Summer			
Code*	Temperature (°C)	Humidity (%)	Wind speed (m/s)	Code	Temperature (°C)	Humidity (%)	Wind speed (m/s)
A1-5	10.00	31.25	1.25	A5-7	29.75	30.00	0.63
B1-4	9.13	34.38	1.30	B5-7	33.25	27.50	1.00
C1-2	12.00	38.75	1.25	C3-4	31.00	20.00	0.63
D1-2	10.75	30.00	1.13	D3-4	29.50	20.00	1.00
E1-4	11.63	42.50	1.06	E5-7	30.66	20.83	0.83
F1-3	13.16	33.33	1.58	F4-5	35.75	20.00	0.75
G1-2	13.00	33.75	1.25	G3-4	34.50	25.00	1.50
H1-4	13.50	30.00	1.00	H5-6	28.00	18.75	0.88
CNG 1 (2)	11.25	45.00	0.75	CNG 1	30.00	22.50	1.00
CNG 2 (2)	10.00	40.00	1.50	CNG 2	30.75	25.00	1.13
Average	11.44	35.90	1.21	Average	31.32	22.96	0.93

*A to H denote the gasoline and diesel filling stations

results. They suggested that busy and crowded gas stations and mostly refueling of NISSAN Van by SAIPA Company could be the main reasons of these higher concentrations.[8] However, the low concentrations of BTEX in our research could be attributed to the increased price of gasoline, the low amount of refueling in any time, the promotion of public culture about refueling technique and paying attention to the reformation of consumption pattern and meteorological condition. Sergio *et al*. monitored BTEX in the gas station area in Brazil and reported lower concentrations than our work. Probably that fuel stations are equipped to leak control systems, vapor recycling, and returning systems.[6]

Evaluation of dispersion distance from gas stations

As Table 3 shows, the differences in the BTEX average concentrations measured in the distances of 50, 150, and 250 m from stations A and B were not considerable. Dispersion distance of fuel station as a point source of emission depends on the concentrations of pollutants on the air of station area, temperature and the wind speed and its direction. Higher pollutants emission from the stations and higher air temperature would result in the large distance of the emission.[8] Regarding the measured concentrations of the pollutants far from the stations it could be concluded that the gas stations cannot be considered as the only sources of these pollutants. Thus, other sources of emission such as the movement of vehicles and some small local industries could be effective on the air pollution. Some research has reported the radial effect of 30 m for the gas station.[8] In our study, there were not meaningful differences between the concentrations at the mentioned distances, and they were not decreasing by getting far from the stations. So that, the effects of the gas stations cannot go so far and may be limited down to 50 m.

The effect of weather condition on Benzene, toluene, ethyl benzene, and xylene concentrations

As Figure 2 shows, the concentrations of BTEX were slightly high in the warm season as compared with the cold season but the difference is not as significant as expected. It could be partially because of rapid evaporation of gasoline at high temperature; however, in winter refineries may increase the vapor pressure of gasoline by adding more volatile compounds which causes the evaporation of BTEX does not hinder at cold season.[8] This can compensate the effect of temperature reduction on the evaporation rate in winter. Nevertheless, the effects of other parameters such as background and upwind concentrations cannot be ignored which needs extensive research at least throughout the year with large sampling points. As Table 5 shows, the pollutants concentrations at unstable weather with high wind speed are lower than those at stable atmosphere. This is in accordance with the fact that at unstable atmosphere the dispersion and dilution rate of the pollutants is high.

The effects of refueling volume and numbers on benzene, toluene, ethyl benzene, and xylene concentrations

Because of installing electronic facilities on the pumps of gas stations, it was possible to get information such as refueling number and the volume of fuel transferred at any period. So that, these data was collected from 6 gas stations for the 3 h air sampling times. According to Table 4 and Figure 3 the gas station with a large number of refueling and more volume of refueling has higher pollutant emission compared to the others. In spite of this, in the more crowded stations on the time of fuel transfer into the vehicle fuel tanks some fuel overflow and spills were observed during the air sampling times. Furthermore, in some stations such as A and F higher emissions of BTEX could be attributed to the close distance of filling pumps to each other.

CONCLUSION

Fuel stations are one of the main sources of VOCs emission in Shahreza city. Within the BTEX compounds only the concentration of benzene, the most hazardous one, is higher than the maximum allowed level in the air of gas stations that may put at risk the public health and the occupational health of personals. The concentrations of BTEX are slightly high in the warm season compared to the cold season; however, the difference is not considerable as expected. The number and volume of refueling in the gas stations influence the emission rates of BTEX. Therefore, due to having carcinogenic and neurological effects of these pollutants, purposive and scientific plans for abatement of VOCs emission from the fuel stations must be organized. It is also suggested to do preventive actions such as repairing of pumps and tanks leak using vapor return systems at the time of transferring fuel, and setting the pumps on desired distances.

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