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Measurement of BTEX (benzene, toluene, ethylbenzene and xylene) concentration at gas stations

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Abstract

Background: Fuel stations are one of the major sources of air pollution with volatile organic compounds, especially the four main petrol compounds benzene, toluene, ethylbenzene, and xylene (BTEX). BTEX in gasoline enters the air of gas stations due to high evaporation of gasoline. Therefore, determining the concentration of these compounds in gas stations in crowded and busy cities is one of the important priorities of environmental health, which is doubly important in terms of its negative effects on health.

Methods: In this descriptive cross-sectional study, a total of 39 samples were collected from 13 gas stations. Sampling was performed in autumn 2018 in three working shifts (morning, noon, and night). The method NIOSH-1501 (i.e., using charcoal sorbent tubes and SKC pump with a flow rate of 0.2 L/min) was used for sampling the BTEX compounds. The mean difference and correlation of BTEX compounds based on meteorological parameters and the number of nozzles in gas stations were assessed using one-way ANOVA and correlation tests.

Results: The mean and standard deviation of benzene, toluene, ethylbenzene, and xylene concentrations in the air of fuel stations were 2.784 ± 1.461, 3.495 ± 1.390, 2.091 ± 0.811, and 1.140 ± 0.419 mg/m3, respectively. The relationship between BTEX compounds and meteorological parameters such as humidity and exposure time is very important. There is a strong correlation between the concentrations of BTEX compounds. The highest correlation was observed between benzene and toluene and the lowest one was observed between benzene and xylene. In this study, no significant relationship was observed between air temperature and concentration of BTEX compounds, but there was a relationship between relative humidity and the concentration of BTEX compounds.

Conclusion: The average benzene concentration in the air at the fuel stations was about 5.5 times the standard limit. Authorities should improve fuel quality and reduce its evaporation through engineering measures to overcome the issue.

Keywords: BTEX, Gas stations, Iran

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Introduction

Traffic, industrial activities, and natural emissions are the major sources of air pollution in cities. A large number of pollutants, including volatile organic compounds (VOCs), enter the ambient air through these sources (1). Oil, gas, and petrochemical activities have a special role in the release of VOCs into the environment (2). These compounds, which are produced through various processes, in the form of volatile liquids or solids, have organic carbon and high evaporation rate (3). After suspended particles, the highest frequency and diversity of emissions are related to VOCs (4). Benzene, toluene, ethylbenzene, and xylene isomers (commonly called BTEX) are the most common VOCs (5). BTEX compounds are the most abundant VOCs in the atmosphere and surface waters. Plastics, paints, resins, rubber, adhesives, lubricants, detergents, etc. are known as the sources of BTEX pollutants (6). BTEX compounds in industrial products easily enter the environment and are widely found in the general environment today (7).

Toxicokinetic studies on humans and animals show that BTEX compounds are well absorbed, due to their lipophilic nature, they are distributed in adipose-rich tissues such as the brain, bone marrow, and body fat, and are rapidly eliminated from the body (8). The World Health Organization (WHO) has identified BTEX compounds as potent carcinogens (7). The most dangerous of these compounds, which was classified as a human carcinogen in 1982 by the International Agency for Research on

Cancer (IARC), is benzene. The US Environmental Protection Agency (USEPA) prioritizes BTEX compounds (9). In studies investigating the toxic effects of BTEX on human health, these compounds have been found in blood and urine (10) and have shown adverse effects on the human respiratory system and lungs (11). Aplastic anemia, pancytopenia, and neurological disorders such as blurred vision are other chronic complications of exposure to these compounds (12). Apart from toxicity, BTEX compounds also act as precursors to contaminants such as peroxyacetyl nitrate, ozone, free radicals, and nitrogen oxides (13).

The use of motor vehicles has become an integral part of human life for many years for intercity, intercity communication, and doing various things. Over time, more and more gasoline- and gas-powered vehicles have been seen in the roads, which has subsequently led to the expansion of stations and increased fuel supply (14). According to a study by Watson et al, gasoline vehicles emit four times more VOCs than diesel vehicles (15). BTEX compounds are the main constituents of gasoline and due to the high evaporation rate of these compounds, they can enter the ambient air from the exhaust, vehicle carburetor engine, and petroleum product distribution stations (3).

Since air pollutants alter BTEX effect quality, it seems necessary to evaluate and monitor the concentration of these compounds in ambient air. On the other hand, determining the concentration of these compounds in the air of gas stations as places that cause cross-sectional exposure to BTEX compounds for customers and permanently for station staff is more important. However, the available evidence on the measuring BTEX compounds in gas stations in low- and middle-income countries (e.g., Iran) with lower fuel quality is scarce yet, and to the best of our knowledge, there is no study on the association of BTEX concentration at gas stations with methodological parameters and the number of nozzles in these countries. Therefore, the present study was performed to determine the concentration of these compounds in the air of gas stations in the Mashhad metropolis as a case study and representative of major cities in Iran.

Materials and Methods

In this descriptive cross-sectional study, the emissions of benzene, toluene, ethylbenzene, and xylene in the air of 13 gas stations in Mashhad as a case study were investigated. Sampling was conducted in 13 gas stations of the city, the sampling location of refueling stations is shown in Figure 1.

To select the sampling sites, the city was divided into five areas: North, South, East, West, and Central, and the sites were selected to cover all areas and represent the entire city. Therefore, the air of the station with code (A), (B), (C), (D), (E), (F), (G), (H), (I), (J), (K), (L), and (M) were sampled. Sampling was performed in three shifts in the morning, at noon and night from each location according to NIOSH 1501 method (16) in autumn 2018. Overall, 39 samples were collected from the gas stations. At the time of sampling, atmospheric conditions were measured and recorded in terms of temperature, humidity, and wind speed by WBGT meter and anemometer.
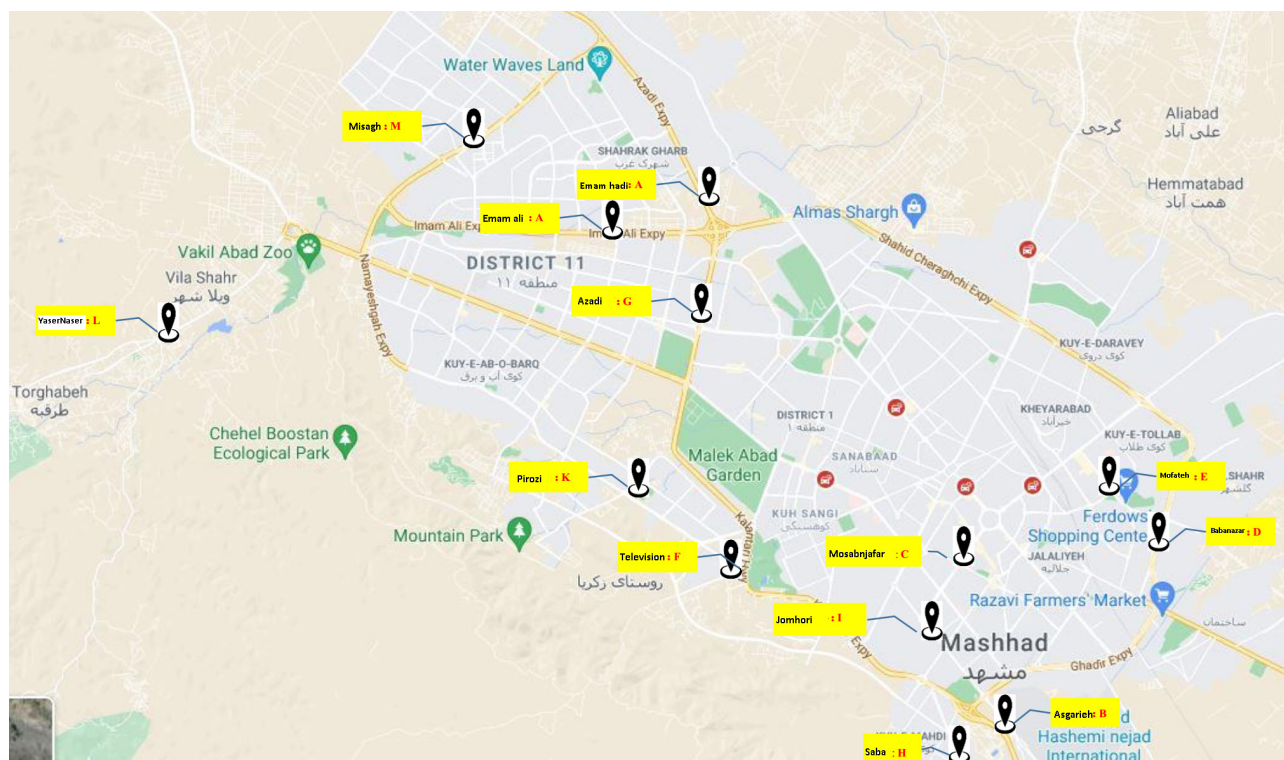

Figure 1. Sampling location of refueling stations in Mashhad city, northeastern of Iran, as a case study.

Preparation of air samples

A personal pump (SKC Ltd, DT11 8ST, UK) with an adjustable flow rate below 200 mL/min was used for sampling. The air at the sampling site was pumped through activated carbon adsorbent tubes to absorb the desired compounds. Activated carbon were contained in closed glass tubes made by SKC, its length was 7 cm and its inner diameter was 4 mm, and its content consisted of two separate parts of activated carbon 50 and 100 mg. The two sections are separated by a 2 mm layer of polyurethane foam. To perform the sampling operation, first, the individual sampling pump was calibrated by a soap bubble device by placing a flowmeter and a double-ended adsorbent tube in the circuit. The pump flow rate was set at 0.2 L/min during the sampling steps and the sampling time for each sample was about 1 hour and 15 minutes. All sorbent tubes were placed at the height of about 2 m above the ground level. At each sampling, the two ends of the absorber tube were broken and the absorber tube was connected to the pump inlet according to the arrow sign indicating the direction of airflow. Out of every 10 adsorbents, 2 were considered as control samples. To determine the background concentration and test error, carbon adsorbents were extracted without passing sampling air. After sampling, the tubes were transferred to Department of Analytical Chemistry Laboratory, Sabzevar University of Medical Sciences, at a temperature of 4°C. The samples were stored in the refrigerator until analysis. It should be noted that the shelf life of the samples from the day of sampling to the day of analysis at the mentioned temperature is 30 days. In this study, the interval between sampling and analysis was 7 days.

Chemical analysis of samples

To extract the contaminants, the front part of the adsorbent was broken and its contents were poured into 2-ml vials (washed with CS2), and then, 1 ml CS2 with 99% purity was added to the vials. The vials were closed with a septum cap, and the samples were placed in a shaker for 30 minutes. After that, one μl of the solution inside the vial was removed by a 10 μL syringe (Hamilton Company) and injected at the injection port of the gas chromatograph of the mass spectrometer (model CP3800, Inc., Variance company). The extracted samples were injected into a gas chromatography device with a flame ionization detector (GC-FID, Agilent Technology, 7890) to measure the concentration of BTEX compounds. The calibration curves were made using a standard mixture of BTEX (2000 μg/mL in methanol) to reach a response signal (peak graph founding in GC-FID) as standard concentrations in 10 points. The recognized calibration curves for the BTEX compounds were obtained to have R-squared values ≥ 0.999. Finally, the measured BTEX compounds were reported in mg/m³. The shutdown time of the device was 1.9-3.5 minutes, the injection

site temperature was 180°C, the initial temperature of the column was 30°C, and the storage time at the initial temperature was 12 min. The temperature increased by 20°C per minute until it reached 180°C, and then, stopped at this temperature. The total analysis time was 2 minutes, and column specifications were (SGE Code: 25 mm × 0.22 mm, Capillary serial number: 4792B09, P/N: 054253, CO.N: SGG).

The precision of Method Quantitation Limit (MQL) values was evaluated by relative standard deviations (RSD). The results confirm that the RSD of each calibration BTEX level (3 replicates) was not higher than 5% for BTEX. The method detection limit (MDL) for each BTEX compound were found to be in the range of 0.12 to 0.16 μg/m³. The value of BTEX in blank samples were ranged less than the limit of detection (LOD).

Data analysis

Data were analyzed using SPSS version 19. In this regard, the Kolmogorov-Smirnov test was used to evaluate the distribution of pollutants and meteorological data. Parametric tests such as Pearson correlation and one-way analysis of variance (ANOVA) were used to examine normal variables, and Spearman and Kruskal-Wallis correlation tests were used to analyze abnormal variables. Statistical significant level was considered at $P = 0.05$.

Results

The results of assessment of the normal status of BTEX compounds and meteorological factors indicated that all of them were normal except for xylene isomers. Examination of BTEX normality in three times of the morning, noon, and night using the Shapiro-Wilk test showed that the data related to benzene and ethylbenzene concentration were normal. Data on the concentration of BTEX compounds (mg/m³) by sites examined are presented in Table 1. Accordingly, the highest concentration of all BTEX compounds was related to the site with code C and the lowest one was related to the site with code L. The mean and standard deviation of humidity, air temperature, and wind speed reported by the stations as well as the number of nozzles in each station are shown in Table 2. The relationship between BTEX and meteorological factors and the number of nozzles at fuel stations was investigated using Pearson and Spearman correlation tests (Table 3). Pearson correlation test was used for normal variables and Spearman correlation test was used for abnormal variables. Considering that P value less than 0.05 indicates a significant correlation between the two variables, the relationship between humidity and BTEX was significant ($P < 0.05$). On the other hand, the closer the correlation (r) to 1 or -1, the stronger the correlation, so there was the highest correlation between toluene concentration and humidity. Since the positive correlation shows a direct relationship and the negative correlation shows an inverse

Table 1. Mean and standard deviation of BTEX by position

Station	Benzene (mg/m3)	Toluene (mg/m3)	Ethyle Benzene (mg/m3)	Xylene (mg/m3)
A	4.486±1.200	5.129±1.017	2.788±0.661	1.291±0.586
B	2.691±0.718	3.421±1.203	2.107±0.127	0.916±0.104
C	5.635±1.875	6.324±1.921	3.482±0.667	1.610±0.642
D	2.652±1.004	3.235±1.027	2.504±0.552	1.291±0.619
E	3.119±1.223	4.061±1.316	2.745±0.657	1.501±0.535
F	4.051±1.579	4.170±1.474	2.532±0.646	1.270±0.292
G	2.041±0.864	2.687±0.580	1.635±0,476	0.987±0.253
H	2.038±0.096	2.925±0.642	1.727±0.224	1.160±0.381
I	1.881±0.0543	2.734±0.851	1.453±0.110	7.939±0.079
J	2.304±0.494	3.156±0.704	2.066±0.111	10.47±0.058
K	1.589±0.248	2.443±0.505	1.212±0.165	0.872±0.196
L	1.024±0.149	1.909±0.300	0.942±0.175	0.658±0.079
M	3.822±1.646	4.357±0.994	2.628±1.371	1.540±0.629
Total	2.785±1.461	3.495±1.390	2.091±0.811	1.140±0.419

Table 2. Mean and standard deviation of humidity, air temperature, and wind speed reported by the stations and number of nozzles in each station

Station	Humidity (%)	Air temperature (°C)	Wind speed (m/s)	Number of Nozzles
A	20.3±8.0	28.3±5.2	0.5±0.4	21
B	17.3±3.5	31.2±2.6	0.6±0.2	16
C	25.2±9.5	23.9±9.9	0.9±0.2	16
D	29.1±5.1	28.4±3.6	0.7±0.3	18
E	28.3±8.4	30.3±2.5	1.1±0.8	18
F	19.5±5.9	31.8±4.5	0.2±0.2	30
G	25.9±11.0	30.1±3.4	0.3±0.2	20
H	26.3±10.2	30.0±2.8	0.3±0.2	15
I	27.8±3.4	32.3±3.3	0.3±0.2	38
J	31.8±4.5	29.4±2.1	1.0±0.7	20
K	34.7±3.7	24.2±3.6	0.7±0.5	22
L	35.2±2.6	26.1±2.4	0.6±0.1	22
M	28.1±2.2	30.3±1.3	0.4±0.3	24

relationship between the two variables, the relationship between BTEX and humidity and the number of nozzles in the stations was inverse.

The relationship between pollutant concentration and measurement time was investigated using regression model. Also, variables that had a significant correlation with the concentration of pollutants were included in the model. Based on this result, the relationship between benzene concentration and measurement time as well as air humidity was investigated. The benzene concentration in the morning was 1.5669 mg/m³ less than that at night ($P = 0.002$). Also, benzene concentration at noon was 1.7413 mg/m³ lower than that at night ($P = 0.001$). In addition, there was a significant relationship between humidity content and benzene concentration ($P = 0.002$) so that for one unit increase in humidity, benzene concentration decreased by 0.0918 mg/m³. Toluene concentration in the morning was 1.6354 mg/m³ lower than that at night ($P = 0.001$). At noon, the concentration of toluene was 1.2995 mg/m³ less than that at night ($P = 0.009$). The results showed that there was a significant relationship between toluene concentration and air humidity ($P = 0.003$) so that for one unit increase in air humidity, toluene concentration decreased by 0.0842 mg/m³. The concentration of ethylbenzene in the morning was 0.6694 mg/m³ lower than that at night ($P = 0.03$), while at noon, it was 0.7425 mg/m³ lower than that at night ($P = 0.02$). There was also a significant relationship between humidity and ethylbenzene concentration ($P = 0.01$) and for one unit increase in humidity, ethylbenzene concentration decreased by 0.0457 mg/m³. The results showed a significant relationship between measurement time and xylene concentration ($P = 0.001$), so that its concentration was 0.5259 mg/m³ lower in the morning than that at night. The correlation of BTEX pollutants with each other is shown in Table 4.

Table 3. Correlation of BTEX with meteorological factors and number of nozzles

Pollutants	B	T	E	X
Humidity	$r_p = -0.36$ $P = 0.03$	$r_p = -0.41$ $P = 0.01$	$r_p = -0.34$ $P = 0.04$	$r_s = -0.33$ $P = 0.05$
Air temperature	$r_p = 0.09$ $P = 0.60$	$r_p = 0.14$ $P = 0.41$	$r_p = 0.12$ $P = 0.50$	$r_s = 0.32$ $P = 0.05$
Wind speed	$r_p = -0.03$ $P = 0.86$	$r_p = 0.03$ $P = 0.84$	$r_p = 0.09$ $P = 0.59$	$r_s = -0.02$ $P = 0.91$
Number of nozzles	$r_s = -0.18$ $P = 0.30$	$r_s = -0.17$ $P = 0.32$	$r_s = -0.29$ $P = 0.09$	$r_s = -0.14$ $P = 0.41$

R_p: Pearson correlation.
R_s: Spearman correlation.

Table 4. Correlation of BTEX pollutants with each other

Pollutants	B	T	E
B	-	-	-
T	$r_p = 0.96$ $P>0.0001$	-	-
E	$r_p = 0.92$ $P>0.0001$	$r_p = 0.88$ $P>0.0001$	-
X	$r_s = 0.78$ $P>0.0001$	$r_s = 0.79$ $P>0.0001$	$r_s = 0.81$ $P>0.0001$

R_p: Pearson correlation.
R_s: Spearman correlation.

Discussion

According to Iran Occupational Health Committee, the permissible limits of benzene, toluene, ethylbenzene, and xylene are 0.5, 50, 100, and 100 ppm (3.2, 205.5, 473, and 473 mg/m³), respectively (17). Accordingly, the concentration of benzene in all stations was higher than the permissible limit and the concentration of toluene, ethylbenzene, and xylene isomers in all stations was less than the permissible limit. In this study, the highest concentration of BTEX compounds was observed in one site. The study of the station with code C, considering that it has the maximum value of all four parameters, showed that the traffic location of this station in the city center and near the holy shrine is the cause of high concentrations of BTEX compared to other stations(Figure 2). In addition, it seems that the small structure and limited space of the

site C have caused the concentration of VOCs in this site to be higher than that in other sites. In this study, benzene concentrations ranged from about 1 to more than 5 ppm (3.2-16 mg/m³) at different sites. These values are higher than the values of benzene measured in the petrochemical industry in the study of Rene et al, which was reported to be in the range of 0.450-2.000 ppm (1.44-6.4 mg/m³) (18). The average concentration of benzene in the air of fuel stations in the present study was 2.784±1.462 ppm (8.9±4.68 mg/m³), while the average concentration of benzene in the respiratory air of workers working in Tehran gas stations in the study of Rastkari et al was reported to be 5.900±1.930 ppm (18.8±6.17 mg/m³), which is two times more than this amount (19). Due to the use of petroleum products in industries such as dyeing, the concentration of BTEX compounds in the environments using these products has been investigated, and similar concentrations of these compounds have been reported with the concentrations in the air of refueling stations. In the study of Salmani et al on the ambient air of car painting workshops in Yazd, the average concentrations of BTEX compounds were reported to be 2.14, 4.09, 1.08, and 3.05 ppm (6.48, 16.81, 5.11, and 14.43 mg/m³), respectively (20), which are close to the values obtained in the present study. In a study conducted in Firuzabad, India, the concentration of BTEX compounds in gas stations was reported to be 0.675-0.784 ppm (2.16-3.7 mg/m³), which is much lower than that reported in the present study (21).

The comparative evaluation of the results of the present study with those of some related studies performed at different parts of the world is shown in Table 5.

According to the table, the concentration of pollutants in the air of fuel stations is close to each other and higher than that reported in other countries, which can depend on several factors. Differences in refineries and refining processes, and consequently, the chemical composition of gasoline and its aromatic contents, as well as preventing or reducing the entry of gasoline vapors into the atmosphere due to the presence of gasoline vapor recovery systems and advanced gasoline waste control systems in other

Table 5. Comparison of the results of the present study with those of some related studies performed in different parts of the world

Benzene (mg/m³)	Toluene (mg/m³)	ethylbenzene (mg/m³)	Xylene (mg/m³)	Sites	References
2.785	3.495	2.092	1.14	Mashhad, Iran	The present study
0.23	0.3923	0.0933	0.3802	Khorasgan, Iran	(22)
0.286	0.495	0.112	0.414	Shahreza, Iran	(23)
1.932	0.667	0.148	0.34	Yazd, Iran	(3)
0.15	2.2	0.049	0.192	Valensia, Spain	(24)
0.0018	0.0049	0.0015	0.0044	Belgium	(25)
0.599	1.694	0.096	0.533	Thailand	(26)
0.0297	0.0477	0.0233	0.0306	Brazil, 2000	(27)
1.787	9.145	0.973	0.476	Shahindej, Iran	(14)
2.01	1.8	2.72	1.65	Ardabil, Iran	(28)
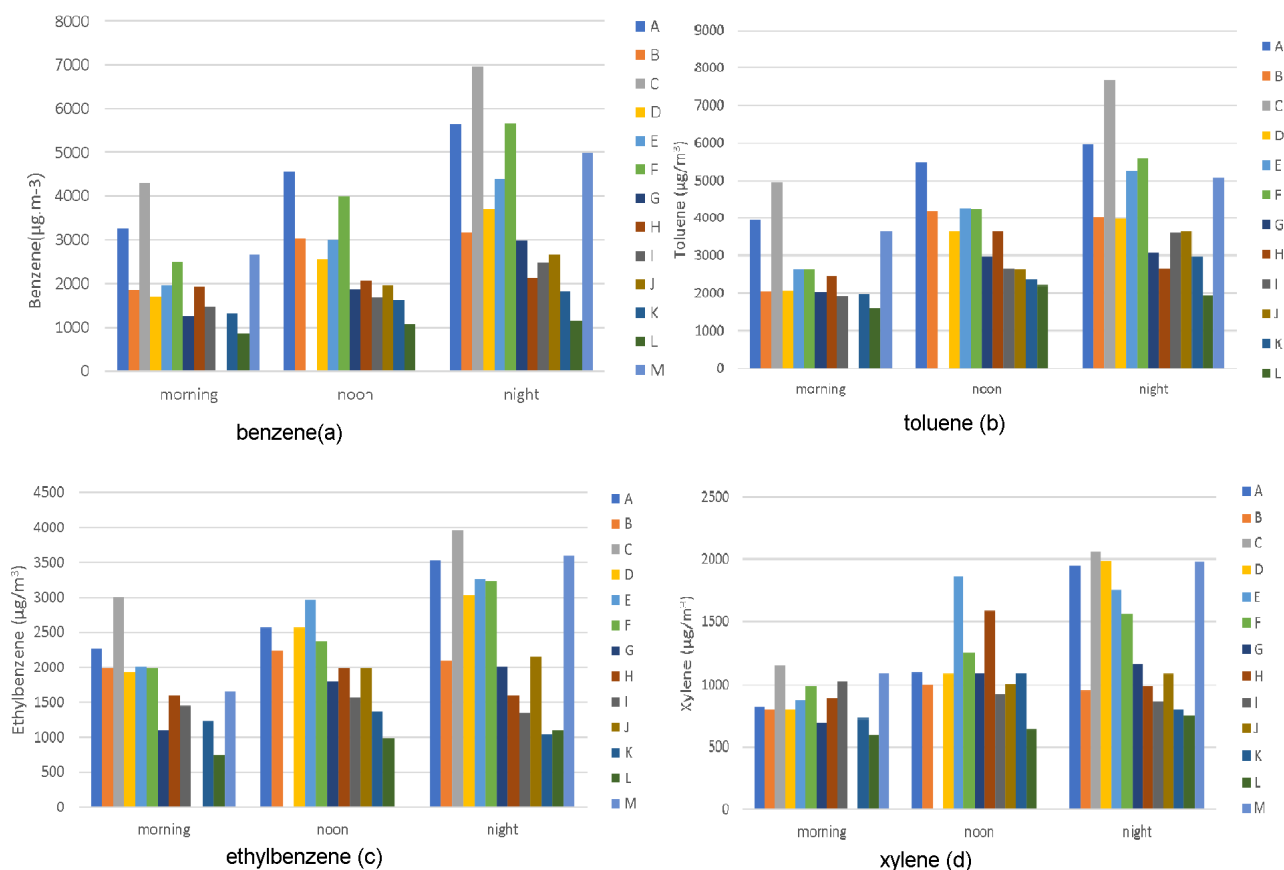
Figure 2. Comparison of the concentrations of toluene (a), benzene (b), ethylbenzene (c), and xylene (d) (µg/m³), in the air of different refueling stations by sampling time.

countries can be among these reasons. The number of service stations per day and population density, along with economic, social, and cultural factors, affect the amount, type, and correctness of gasoline use.

Soltanpour et al conducted a systematic review and probabilistic health risk assessment on the concentration of benzene, toluene, ethylbenzene, and xylene in ambient air of the gas stations in Iran, and suggested that the health risk assessments of exposure to BTEX could be useful in choosing suitable control measures. This health risk assessment is suggested to be developed based on data measurement of different sites in a variety of the centers that may be exposed to BTEX (29).

However, numerous studies, environmental monitoring of residents around these stations as well as workers who are in contact with these pollutants (both in the distribution sector and in the industry of production of these materials) emphasized the need to assess the risks of emissions in urban areas (30-34).

Effects of inhaled combined of BTEX and health symptoms associated with occupational exposure of gasoline station workers to BTEX compounds have been studied in recent studies (35,36).

In addition to concerns about BTEX pollution in urban areas, some natural treatment systems such as phytoremediation of BTEX from indoor air or reduction of BTEX in automotive paint sludge by vermicomposting

process in recent studies are promising news for pollution control (37,38).

Heibati et al evaluated the average exposure of workers in different stages of loading as well as tanks and transport drivers and firefighters and office workers and found that the highest concentration was related to workers loading in tankers, and the concentration of benzene was reported 5.390 mg/m³ (39). The concentration of BTEX in station C in the present study is very close to that reported in this study.

Conclusion

This study showed that although the concentrations of toluene, ethylbenzene, and xylene compounds were within the standard permissible limits, but the concentration of benzene can increase several times the permissible limit in crowded cities, as in this case study in Mashhad, benzene concentration was reported up to 5.5 times the permissible limit, which requires the planning of control managers on the environmental aspects of gas stations. It requires the attention of the authorities to take interventions such as improving the quality of fuel and reducing its evaporation through engineering measures.

This study showed that by decreasing the temperature in different places in the morning, the concentration of VOCs in BTEX increased. In the morning, to make it easier to start the car, the gasoline vapor pressure is increased by

increasing the percentage of volatile gasoline components, which leads to faster evaporation of gasoline and increases the concentration of compounds in the station area in the morning.

This study has shown the extent of the pollution so that the authorities in the distribution of these fuels can take a specific action in a national program to equip the stations with warning systems above the concentration limit. The suggestion is that all operating stations and fuel station workers should be trained in hazardous exposure to BTEX compounds and adherence to occupational safety protocols and occupational health and safety guidelines before official employment and professional startup at the stations or starting work, and should be under the control and supervision of an occupational health organization with a continuous and scientific training program. In terms of physical equipment, fuel stations or station managers should control the installation of equipment on fuel injection nozzles to control evaporation of these materials, and install warning signs and information posters to inform customers about the dangers of exposure to these compounds.

In future research, it is suggested to develop the range of BTEX concentration measurements to various large cities, in addition to gas stations, to the industrial sites of production of these products, to the workers of transportation of these products, and to storage tanks of these products and transfer stations. Also, the risk of exposure to these compounds should be evaluated.

Ethical issues
In this article, the authors considered all the ethical points in collecting data and confidently declare that this manuscript is the original work of the authors, and all data collected during the study are as presented in the manuscript, and no data from the study has been or will be published elsewhere separately. This article is a research project and simultaneous thesis of Ms. Zohreh Payendeh Sharif, which was approved by the Vice-Chancellor for Research and Technology of Mazandaran University of Medical Sciences and the Faculty of Health of Sabzevar University of Medical Sciences (Approval code: 3027), and the Ethics Committee of Mazandaran University of Medical Science Ethical code: IR.mazums.rec.1396.3027).

Competing interests
The authors declare that there is no conflict of interests.

Authors' contributions
All authors have contributed to various sections of this research from guidance and advice to collection of samples, writing and editing the article, and all items related to the article.

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