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TRAFFIC - RELATED CONCENTRATIONS OF BTEX, FORMALDEHYDE AND ACETALDEHYDE IN TEHRAN; CONCENTRATIONS AND SPATIAL VARIABILITY

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ABSTRACT:

Introduction: Traffic and mobile sources are possibly responsible to the most of the ambient volatile organic compounds (VOCs) in urban areas. This study aimed to measure and determine the traffic - related concentrations of benzene, toluene, ethylbenzene, xylenes, formaldehyde, and acetaldehyde at the main streets of Tehran, Iran.
Materials and methods: The samples were taken from highly populated streets or main roads with heavy traffic in Central, Northern, Eastern, Southern, and Western areas of Tehran. In total, 33 points for BTEX and 23 points for formaldehyde and acetaldehyde were selected for sampling. The sampling and analysis were performed according to NIOSH methods 1501 and 2016.
Results: The averages (± SD) of benzene, toluene, ethylbenzene, xylenes, formaldehyde and acetaldehyde concentrations in Tehran were 15.04 (± 9.18), 23.42 (± 8.73), 4.97 (± 2.55), 11.81 (± 4.46), 107.11 (± 30.58) and 57.10 (± 18.28) ppbv, respectively. Benzene concentrations were 3.30 to 26.00 times higher than air quality standard of European Union (5 μg / m^3). High concentrations of BTEX and formaldehyde / acetaldehyde were found in central and eastern areas, respectively. High correlation coefficients were found between BTEX species (r = 0.77-0.95) and also formaldehyde and acetaldehyde (r = 0.98). The highest coefficient of variation (CoV) as a measure of spatial variability was observed for benzene (54.5 %).
Conclusions: The high outdoor concentrations observed in this study needs to be decreased immediately, especially in case of benzene.

INTRODUCTION

Ambient air may contain volatile organic compounds (VOCs) as one of the main issues of air quality in the cities. VOCs' importance is mainly because of their toxicity for human health and the fact that they are precursors of photochemical

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products including ozone [1, 2]. Aromatic volatile organic compounds and carbonyl compounds such as benzene, toluene, ethylbenzene, p-xylene, m-xylene, o-xylene (BTEX), formaldehyde, and acetaldehyde have been the subjects of many toxicological studies. Multiple carcinogenic and non - carcinogenic health effects have been found to be induced by BTEX, formaldehyde, and acetaldehyde. Therefore, high concentrations of these compounds have been reported in urban areas [3, 4]. These compounds may originate from the large fleet of old gasoline fueled vehicles and anti - knock additives in unleaded - gasoline [1, 5]. Low - quality engine and fuel and lack of improper catalyst converters exacerbate the emissions of BTEX [6]. In addition to the emissions from mobile sector, other sources may release BTEX, formaldehyde, and acetaldehyde in urban sites. These include domestic cooking, open burning of solid wastes, evaporative emissions from gasoline stations, and industrial activities near the cities [7, 8].

Tehran as the capital of Iran have a population about 9 million people, and is one of the most populated cities in the Middle East [9]. Air pollution is a major problem for this city [10, 11], and cause many deaths annually [12 - 15]. There are about 3 million personal vehicles in Tehran, of which 25 % are more than 10 years old and 75 % have emissions with Euro -2 standard and less [16]. These large fleet of vehicles release substantial amount of air pollutants. It is estimated that about 1.5 million tons of pollutants especially particulate matter are produced in Tehran annually [17, 18]. Mobile sources are responsible for about 86 % of VOCs in Tehran. Other sources of VOCs include stationary sources including gas stations (10.5 %), energy sector (1.6 %), households and commercials (1.4 %), industries (0.3 %), and outbound terminals (0.15 %). 86 % of volatile organic compounds (VOCs) [19]. The rest of 14 % of VOCs in 2013 emitted from stationary sources including gas stations (10.5 %), energy conversion (1.6 %), households and commercials (1.4 %), industries (0.3 %), and terminals (0.15 %) [20, 21].

Some studies have been conducted to characterize and determine the types and concentrations of VOCs in Tehran. In a study, the authors investigated the in - vehicle concentrations of BTEX, formaldehyde, and acetaldehyde in Iranian taxis. They reported high concentrations of these compounds, and concluded that the age and model of vehicle, and type of fuel affects the VOCs' levels in cabin of vehicle [8]. In another study, the concentrations of BTEX and criteria air pollutants were investigated within the Mehrabad international airport. The concentrations of benzene, toluene, and ethylbenzene were different in runway and passengers waiting hall [22]. Using the data on the continuous monitoring of BTEX at 7 sites in Tehran showed that the highest BTEX levels were found along the major roads [23]. However, in another study, the authors measured the BTEX concentrations in 179 sites, and investigated their spatiotemporal status. They found that the concentrations of BTEX near the gas stations are higher than those in other sites [24]. These two latter studies showed inconsistent results.

Since mobile sources and traffic is possibly responsible to the most of the ambient VOCs in Tehran, it is necessary to conduct another study and measure VOCs' levels exclusively emitted from traffic. This helps our knowledge in terms of the air quality within the main roads, emissions from vehicles, and also the human health. In addition, since the size and composition of fleet, and the chemical composition of fuels may change over time, it is required to monitor the ambient air quality in case of BTEX. To our knowledge, no comprehensive studies have been conducted about the outdoor concentrations of formaldehyde and acetaldehyde in Tehran. Therefore, this study was aimed to measure and determine the traffic - related concentrations of benzene, toluene, ethylbenzene, xylenes, formaldehyde, and acetaldehyde at the main streets of Tehran, Iran.

MATERIALS AND METHODS

Study design

This study was carried out in Tehran during the summer of 2016. The samples were taken from highly populated streets or main roads with heavy traffic in Central, Northern, Eastern, Southern, and Western areas of Tehran. These streets are surrounded by residential areas, shop stores, and cultural centers. Many people are exposed to the air pollutants emitted from the traffic jam in these streets.

Four to seven points were selected in each areas. One sample was collected from each point. In total, 33 samples for BTEX and 23 samples for formaldehyde and acetaldehyde were collected. The sampling was performed at the height of 1.5 m.

Sampling and analysis

The samplings were performed in accordance to NIOSH 1501 procedure [25], which is an active sampling method. The pump (Universal 224-44 MTX, SKC, USA) was calibrated before each day using a standard gas flow meter (Model 4140, TSI Inc., USA). Coconut shell charcoal tubes (100 mg / 50 mg) were used to sample BTEX. Before sampling, nitrogen gas was passed from the tubes to eliminate any previous contamination. For sampling, the tubes were attached to the pump, and the flow was set on 200 mL / min for 30 min. Air temperature and pressure and relative humidity were recorded during each sampling using a digital portable device (LUTRON- PHB-318). After finishing the sampling, the tubes were sealed and placed in cold box at a temperature below 4 °C.

Extraction and analysis of the samples were also implemented according to NIOSH 1501 method [25]. The BTEX adsorbed on the charcoal was desorbed after being exposed by 1 mL of CS_2 (76.13 g / mol, Merck, Germany) alongside with sonication for 30 min. Gas chromatography / flame ionization detector (GC-FID: Agilent 7890B, Agilent Technologies, Waldbronn, Germany) was used to measure the concentrations of BTEX. Helium a flow rate of 2.6 mL / min was employed as the carrier gas. The samples were injected to the column with a 5:1 split ratio. The temperatures of injector and detector were set at 250 °C and 300 °C, respectively. Column temperature was raised from 40 °C by 10 °C / min to 230 °C.

Formaldehyde and acetaldehyde were sampled using a cartridge containing silica gel coated with 2,4 - dinitrophenylhydrazine and a flow rate of 500 mL / min. The samples were extracted using 10 mL of carbonyl-free acetonitrile and 0.5 h sonication. Then, the samples were analyzed by high - performance liquid chromatography (Agilent 1100 system, Agilent Technologies, Waldbronn, Germany) The details of this methodology are presented in NIOSH method 2016 [25].

Statistical analysis

The values obtained from analytical measurements were inserted to a Microsoft Excel spreadsheet. SPSS 18 was used to calculate the descriptive statistics of concentrations. The figures were drawn using the Microsoft Excel 2013. To investigate the spatial variability, the coefficient of variation (CoV) was calculated for the compounds, using the following equation:

$$CoV = (\sigma/\mu) * 100 \quad (1)$$

Where σ and μ are the standard deviation and average of each compounds' concentration.

RESULTS AND DISCUSSION

Description of concentrations

Air pollution is known as a major problem in Megacity of Tehran [26 - 28]. Fig. 1 shows the concentrations of benzene, toluene, ethylbenzene, and xylenes in each of the samples. The concentrations have been categorized based on the area of the sampling. Toluene showed to have higher concentration than benzene, ethylbenzene, and xylenes. In addition, formaldehyde and acetaldehyde concentrations have been illustrated in Fig. 2. The concentrations of formaldehyde were higher than those for acetaldehyde. The maximum concentrations of benzene, toluene, ethylbenzene, and xylenes were 40.68, 51.50, 10.60, and 23.26 ppbv, respectively. Additionally, the minimum concentrations of benzene, toluene, ethylbenzene, and xylenes were 5.16, 10.44, 1.52, and 5.76 ppbv, respectively. Maximum and minimum concentrations of formaldehyde were 154.13 and 45.67 ppbv, respectively. Maximum and minimum concentrations of acetaldehyde were observed to be 85.07 and 23.35 ppbv, respectively.

Benzene concentrations were 3.30 to 26.00 times higher than air quality standard of European Union (5 μg / m^3) [29]. These extremely high levels of benzene is due to the proximity of sampling points to main streets. It is reported that the highest concentrations of BTEX were found along the major roads because of heavy traffic [23]. In the study, the ranges of observed concentrations for benzene, toluene, ethylbenzene, and xylenes in Tehran were 2.1-25.8, 6.1-88.9, 1.4-9.8, and 6.4-69.3 μg / m^3, respectively [24]. In the other study it was reported that the maximum (4.291 ppb) and minimum (0.837 ppb) annual average concentrations of BTEX were observed for toluene and ethylbenzene, respectively [23]. In a Chinese study, the maximum observed hour-average benzene concentration was 254 μg / m^3 [30].

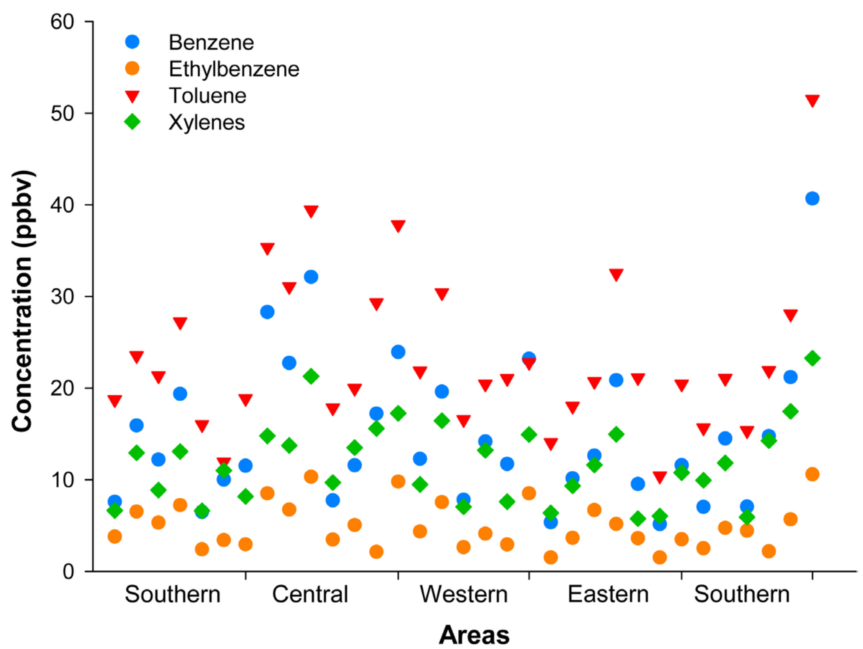

Fig. 1. Concentrations of BTEX samples in selected areas

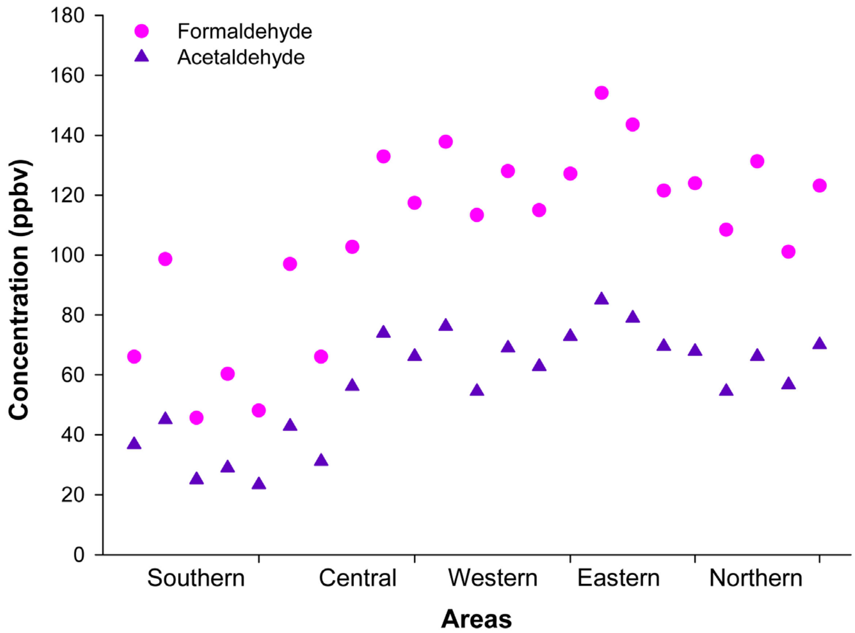

Fig. 2. Concentrations of formaldehyde and acetaldehyde samples in selected areas

Spatial variability

The concentrations of BTEX compounds in each area were averaged. The results are illustrated in Fig. 3. Although, the concentrations of each compound in Central and Northern areas were higher than those in other areas, no statistically significant differences were found ($P_{value} > 0.05$). The maximum average (±SD) concentrations of benzene, toluene, ethylbenzene, and xylene were 20.52 (± 8.81), 30.13 (± 8.45), 6.58 (± 3.15), 15.12 (± 3.58) ppbv, respectively. The minimum average (±SD) concentrations of benzene, toluene, ethylbenzene, and xylene were found in Eastern areas, and observed to be 10.76 (± 5.30), 19.62 (± 6.93), 3.67 (± 1.86), and 9.27 (± 3.45) ppbv, respectively.

The average concentrations of formaldehyde and acetaldehyde in each area are presented in Fig. 4. Formaldehyde concentrations in Southern areas were significantly lower than those in other areas ($P_{value} < 0.05$). Acetaldehyde concentrations in Southern areas was significantly lower than only the concentrations in Eastern areas ($P_{value} < 0.05$). No significant differences were found for other areas ($P_{value} > 0.05$). The average (± SD) concentrations of formaldehyde in Southern, Central, Western, Eastern, and Northern areas of Tehran were 63.77 (± 21.26), 103.24 (± 25.02), 124.28 (± 10.15), 135.78 (± 15.71), 116.01 (± 13.70) ppbv, respectively. The average (± SD) concentrations of acetaldehyde in Southern, Central, Western, Eastern, and Northern areas were 31.81 (9.01), 54.04 (17.31), 67.06 (8.60), 75.34 (8.13), 61.86 (7.45) ppbv, respectively.

The coefficient of variation (CoV) as a measure of spatial distribution was calculated for the compounds. The CoV values showed that the highest spatial variability was observed for benzene (54.5 %), followed by ethylbenzene (51.4%), xylenes (37.8%), toluene (37.3%), acetaldehyde (32.0 %), and formaldehyde (28.6 %). In a previous study, CoVs of BTEX compounds were investigated in Tehran, and reported that the highest spatial variability of annual mean estimates in all sites was observed for toluene (43.7%) followed by m - xylene (42.0 %) > o - xylene (40.3

%) > benzene (39.7 %) > ethylbenzene (38.1 %) > p - xylene (36.4 %) [24].

In another study, the maximum concentrations of BTEX were observed in areas heavy traffic and high density of buildings, which can trap the pollutants. In their study, the concentrations of benzene in heavy traffic areas were 3.85-11.44 times higher than that in residential areas [23]. In our study, the higher concentrations of BTEX and formaldehyde/acetaldehyde were found in Central and Eastern areas, respectively. This can be due the fact that the direction of wind in Tehran is mainly from the west to the east, and can consequently lead the pollutants to move from the west areas to center and then to the east areas [22, 23].

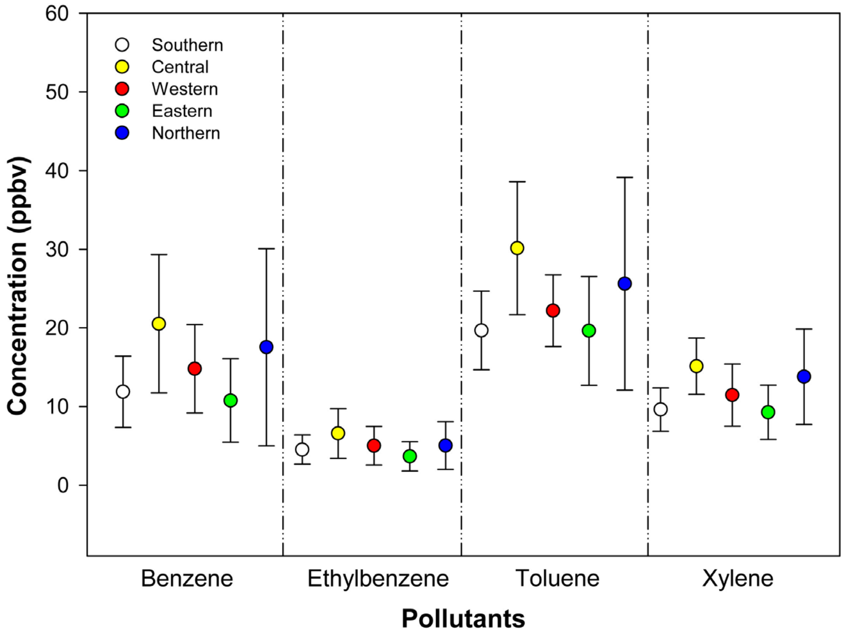

Fig. 3. Average (± SD) concentrations of BTEX compounds in selected areas

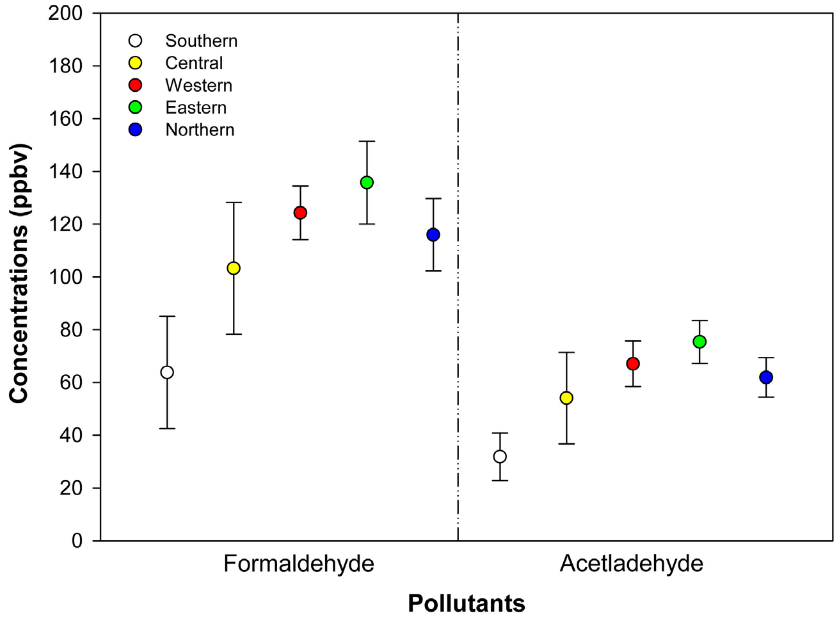

Fig. 4. Average (± SD) concentrations of formaldehyde and acetaldehyde in selected areas

Overall averages

Figs. 5 and 6 show the box plots of concentrations in all of the samplings. The minimum, quartile 25th, median, quartile 75th, maximum, and outlier values are presented in these figures. In case of BTEX, toluene and ethylbenzene had highest and lowest concentrations rather than other compounds, respectively. The averages (± SD) of benzene, toluene, ethylbenzene, and xylenes concentrations in Tehran were 15.04 (± 9.18), 23.42 (± 8.73), 4.97 (± 2.55), and 11.81 (± 4.46) ppbv, respectively. In addition, the averages (± SD) of formaldehyde and acetaldehyde concentrations were 107.11 (± 30.58) and 57.10 (± 18.28) ppbv, respectively. In a previous study, the authors reported that the annual median of benzene, toluene, ethylbenzene, and xylenes in all of the measurements were 7.8, 23.2, 5.7, 22.1 µg / m^3, respectively [24]. In another study, the annual average concentrations of benzene, (m , p) -xylene, and o-xylene in Tehran were 1.056, 2.929, and 1.044 ppb, respectively [23]. In a study in China, daily concentrations of benzene, toluene, ethylbenzene, p,m-xylenes and o-xylene were 56, .[121, 21, 64 and 23 µg / m^3, respectively [30

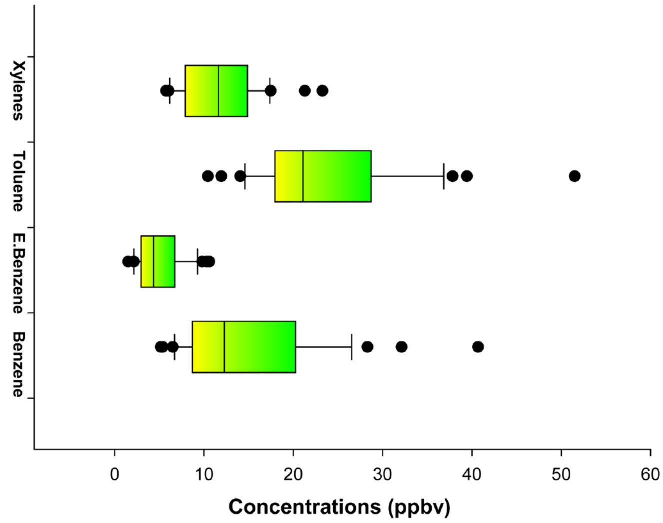

Fig. 5. Statistics of BTEX compounds in all of the samplings
(minimum, quartile 25th, median, quartile 75th, maximum, and outlier values)

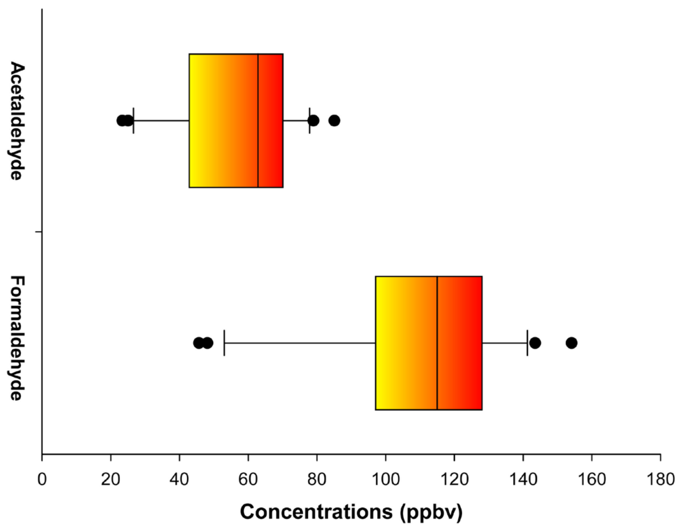

Fig. 6. Statistics of formaldehyde and acetaldehyde in all of the samplings
(minimum, quartile 25th, median, quartile 75th, maximum, and outlier values)

Correlation analysis

The correlation between the concentrations of the interest compounds were determined using the Pearson's correlation coefficient, and the results are presented in Table 1. Statistically significant coefficients are shown in bold numbers. High correlations (r = 0.77 - 0.95) were found between the benzene, toluene, ethylbenzene, and xylenes. This means that the variations of each compound follow the variations of other compounds. Additionally, and extremely high correlation coefficient (r = 0.98) was found for the formaldehyde and acetaldehyde. However, the correlation between BTEX and formaldehyde/acetaldehyde was weak and statistically insignificant. The high correlation coefficients can be mainly due to the common sources of these compounds released to the ambient air of Tehran.

In a study, the estimated annual mean concentrations were spatially correlated for all pollutants (range = 0.81-0.98). The correlations between annual mean benzene and other compounds ranged from 0.81 to 0.96, with a minimum for o- xylene and a maximum for ethylbenzene [24]. In another study, there were significant positive correlations between BTEX concentrations. The observed correlations between BTEX compounds indicated that the emission sources of these compounds are probably similar [23]. In a study in China, high correlations among BTEX species were .[found [30

CONCLUSIONS

The BTEX, formaldehyde, and acetaldehyde samples were collected from 33 and 23 points in Tehran respectively. The higher concentrations of BTEX and formaldehyde / acetaldehyde were found in central and eastern areas, respectively. Large amount of population live in these areas. High concentrations of the studied toxic compounds were found, especially in terms of benzene. Benzene concentrations in all samples were several times higher than air quality standards. As benzene is introduced as definite human carcinogen, these high outdoor concentrations needs to be decreased immediately. This would decrease the concentrations of other pollutants too, since the correlation analysis showed they have common sources. The same pattern was found for formaldehyde and acetaldehyde. Same control strategies and actions could be employed for the pollutants with common sources.

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COMPETING INTERESTS

The authors declare that there are no competing interests.

Table 1. Pearson's correlation coefficients of BTEX, formaldehyde and acetaldehyde compounds

	Benzene	Toluene	Ethylbenzene	Xylenes	Formaldehyde	Acetaldehyde
Benzene		0.95	0.86	0.91	0.16	0.18
Toluene			0.81	0.86	0.19	0.25
Ethylbenzene				0.77	0.06	0.09
Xylenes					0.31	0.30
Formaldehyde						0.98
Acetaldehyde						

*significant coefficients are bold.

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ETHICAL CONSIDERATIONS

"Ethical issues (Including plagiarism, Informed Consent, misconduct, data fabrication and / or falsification, double publication and/or submission, redundancy, etc) have been completely observed by the authors."

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