

Characteristics and Health Risk of BTEX at Selected Different Microenvironments in an Industrial-Urban Area, Iran

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ABSTRACT: The present study monitors BTEX concentration in outdoor and indoor air of eight different microenvironments during summer 2017 and winter 2018 at Asaloyeh city, Iran's energy capital. It samples BTEX compounds by charcoal tubes, analyzing the samples by means of a gas chromatograph with a flame ionization detector. According to the obtained results, outdoor concentrations of BTEX have been higher than the indoor ones, for both seasons, with the highest outdoor and indoor BTEX being 21.70 and 18.59 $\mu\text{g}/\text{m}^3$, respectively. Toluene has been the most abundant substance, among the investigated BTEX in all sampling points. Based on the MIR scale, m, p-xylene is the most dominant contributor to ozone formation potential among BTEX species. Indoor to outdoor (I/O) ratios of BTEX compounds range from 0.53 to 0.88 and 0.41 to 0.77 in winter and summer, respectively. The cumulative hazard index (HI) is within an acceptable range. The LTCR value of benzene concentration, obtained, exceeds the value of $1.0\text{E}-06$, recommended by USEPA. Sensitivity analysis shows that benzene concentration, exposure duration, and inhalation rate have a greater impact on health risk assessment.

Keywords: Air quality, BTEX, Temporal variation, LTCR, Spatial distribution.

INTRODUCTION

It is well recognized that motor vehicle exhausts, gasoline evaporation, and leakage from both compressed natural gas and liquefied petroleum gas station play an important role in quality of urban atmosphere (Kalenge et al., 2013; Yurdakul et al., 2017). On the other hand, the development of industries, especially chemical industries, petroleum or gas refineries, and petrochemical plants, in the vicinity of urban areas exacerbates the effect of pollutants (Baltrenas et al., 2011; Zhang et al., 2017). Volatile organic compounds (VOCs) like benzene, toluene, ethylbenzene, and xylene (BTEX) are among the important groups of

the hazardous air pollutants (HAPs), emitted from mobile and stationary sources such as vehicle exhausts and industrial processes (Chen et al., 2016). VOCs undergo monitoring as they are a key contributor to the increase in global greenhouse effect, photochemical reactions, and threats to public health (Zhao et al., 2004; Demirel et al., 2014). The photochemical reactions can generate ground-level ozone (GLO), peroxyacetyl nitrate (PAN), and secondary organic aerosol (SOA). The former (GLO) is created by chemical reactions between VOCs and nitrogen oxides in the presence of sunlight (Bauri et al., 2016; Wang et al., 2016). The main health concerns, associated with exposure to high concentrations of

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GLO, include shortness of breath, cough, asthma attacks, respiratory disease, cardiopulmonary disease, and death. Living near roads with heavy traffic has been associated with increased rates of asthmatic and allergic symptoms (Buonocore et al., 2009; Han & Naehar, 2006). Generally, the effect level depends on the duration of exposure, the gender, and age of the person, not to mention the kind and concentration of chemicals and particles, exposed (Axelsson et al., 2010).

According to USEPA (2013), indoor concentrations of some pollutants are typically two to five times greater than outdoor ones. According to previous studies, BTEX concentrations in indoor spaces of buildings can be attributed to several factors, such as building material (insulation, paint, plywood, and adhesives), furnishing material (furniture, wall coverings, etc.), combustion appliances (vehicle emission, tobacco smoking, etc.), consumer products (cleaning and personal care products), indoor activities (cooking, tobacco smoking, use of solvents), and poor ventilation (Fenech et al., 2010; Missia et al., 2010; Masih et al., 2017).

Indoor air pollution can have harmful health effects, as most people spend over 80-90% of their time indoors, especially at home and workplaces. Some microenvironment like classrooms needs to be assessed since children and teenagers spend about 25 hours per week at school. The biggest health risks, associated with indoor air pollutants, are VOCs (Azuma et al., 2016; Dai et al., 2017). Previous studies have shown that exposure to BTEX can result in short-term effects such as irritation of the mucous membranes, eyes, and throat, as well as long-term health effects including mutagenic, carcinogenic, and teratogenic problems (Ramirez et al., 2012; Ren et al., 2017). Based on the International Agency for Research on Cancer classification (IARC) and the World Health Organization (WHO), among the BTEX, while toluene and xylene are classified as group D carcinogen or non-carcinogen,

benzene belongs to group one carcinogens and ethylbenzene is categorized as a potential carcinogen, (Demirel et al., 2014). According to WHO estimates, the concentrations of benzene in the air that can lead to an excess lifetime risk of leukemia of 10^{-4} , 10^{-5} , and 10^{-6} are 17, 1.7, and 0.17 $\mu\text{g}/\text{m}^3$, respectively (WHO, 2000). The current study aims at investigating the outdoor-indoor concentration levels of BTEX, their spatial and temporal characteristics, and ozone formation potentials. To do so, it uses inter-species ratios of BTEX to characterize major sources and photochemical age, and potential public health risk of exposure to these pollutant in eight microenvironments at Asaloyeh county, Iran.

MATERIALS AND METHODS

Asaloyeh is located in southeastern Bushehr province in Iran ($27^{\circ} 49' \text{ N}$, $52^{\circ} 62' \text{ E}$), covering an area of 389 km^2 . Based on the last census report in 2016, it had a population of 73,958, in 13,100 families (Statistical Center of Iran, 2016). The main industries include gas refinery, petrochemical, and power plants. South Pars gas field is one of the largest gas reservoirs in the world (Atabi et al., 2016).

For outdoor and indoor air measures of BTEX, 192 (96 outdoor and 96 indoor) samples were collected from eight stations, including a residential worker camp (RWC), an elementary school (ELS), a high school (HIS), Nakhle-taghi Park (NTP), a hospital (HOS), a fire station (FIS), a shopping center (SHC), and a sport complex (SPC) in two periods of time from July to September 2017 and from January to March 2018. Sampling points for BTEX pollutants were selected according to the following criteria: the sensitive urban microenvironment, densely populated areas, the area affected by the industrial zone, traffic, and distance from the road and gasoline station. Fig. 1 shows the map of geographical location of Asaloyeh city and the sampling points of the study area.

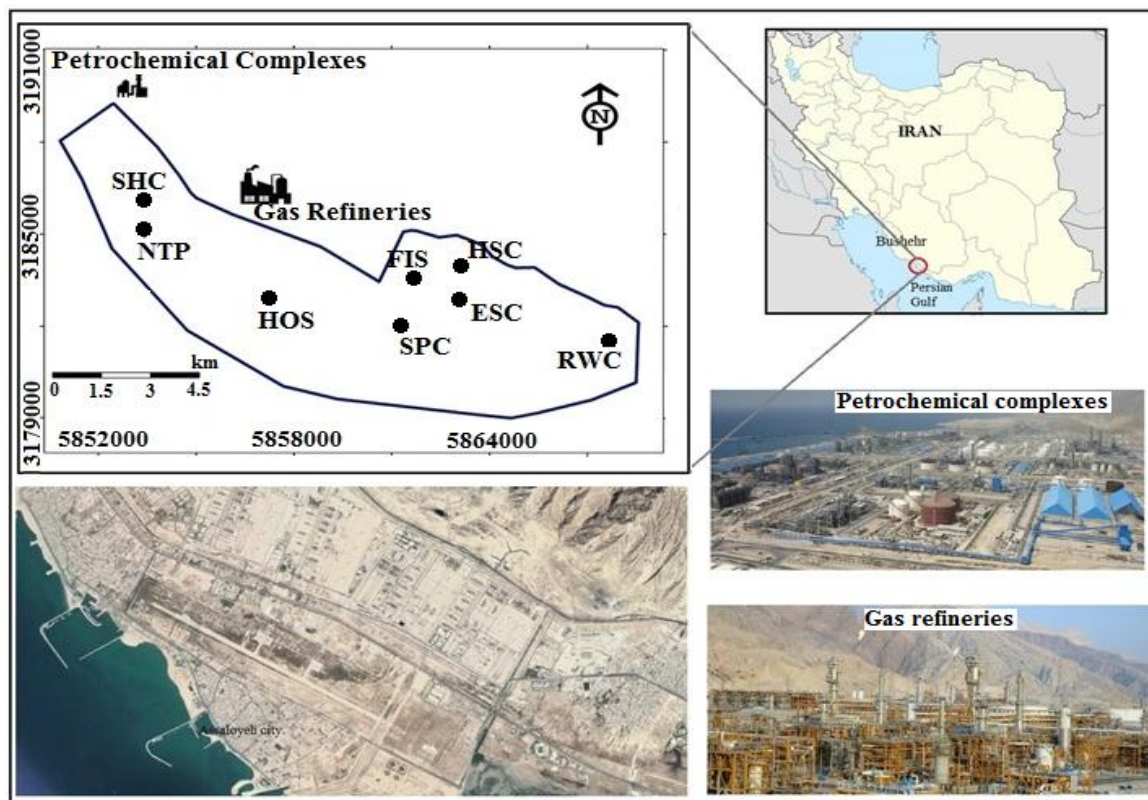


Fig. 1. Map of the study area and the sampling points

BTEX samples were taken based on the National Institute for Occupational Safety and Health (NIOSH) manual of analytical method 1501. Sampling was carried out by a low-flow air sampling pump (SKC Inc.) at a constant flow rate of 0.20 l/min for 2 hours, using charcoal sorbent tubes (SKC Inc.). All the samples were stored at -4°C and transported to the laboratory. The charcoal beds containing BTEX sample were transferred into 2.0 ml vials, there to get extracted by adding 1.0 ml of carbon disulphide (CS_2) with an agitation of 30 min in the ultrasonic bath. After extraction and separation, it was analyzed, using a gas chromatography/flame ionization detector (Clarus 600 model). The injector and detector temperatures were kept at 250 and 300°C , respectively. The initial oven temperature was set to 40°C for 10 min, then to rise to 230°C at a constant rate of 10°C per minute. The carrier gas used was pure helium gas at a flow rate of 2.0 ml/min, a volume of 1.0 μl , and a 5:1 split.

The recovery average for BTEX compounds was between 85% and 115% (Hazrati et al., 2016; Masih et al., 2016; Dehghani et al., 2017).

To evaluate the health risk of people, living in the investigated regions, risk assessment tool was used to estimate the potential carcinogen and non-carcinogen adverse health effects. The general equation of the potential dose and the toxicological risk to BTEX can be seen below:

$$I = C_A \times \frac{IR \times ET \times EF \times ED}{BW} \times \frac{1}{AT} \quad (1)$$

Non-carcinogenic hazards were determined, using a term called hazard quotient (HQ), expressed as the probability of an individual suffering an adverse effect. Reference exposure levels (RELs) is the dose at which adverse health impact will occur in exposed individuals in comparison with unexposed ones. Hazard index (HI) is the sum of the HQs of various pollutants (Demirel et al., 2014; Miri et al., 2016;

Zhang et al., 2017). HQ and HI was calculated using Equations (2) and (3), respectively:

$$HQ = \frac{I}{REL} \quad (2)$$

$$HI = \sum_{i=1}^n (HQ_i) \quad (3)$$

The carcinogenic risks, associated with benzene and ethylbenzene exposure, are expressed in terms of increased probability

of contracting cancer during a person's lifetime. Toluene and Xylene do not have cancer unit risk factors yet. Lifetime cancer risk (LTCR) was calculated using equation (4) (OEHHA, 2003; Tam & Neumann, 2004) below. Table 1 shows the description of the variables, used in the calculation of acute and chronic exposure rate.

$$\text{Lifetime cancer risk} = I(\mu\text{g} / \text{m}^3) \times \text{cancer unit risk factors}(\mu\text{g} / \text{m}^3) \quad (4)$$

Table 1. The variables, used in the calculation of exposure rate and risk assessment factors

Variable	Description	Value	Unit
I	Pollutant intake dosage	--	µg/kg/day
C	Contaminant concentration	--	µg/m ³
IR	Inhalation rate	0.83	m ³ /h
BW	Body weight	70	kg
ED	Duration of exposure	58	year
ET	Exposure time	12	h/day
EF	Exposure frequency	350 - 365	day/year
AT	Averaging time	25550	days
REL	Reference exposure level		
	Benzene	6.0E+01	µg/m ³
	Toluene	3.0E+02	µg/m ³
	Ethylbenzene	2.0E+03	µg/m ³
	Xylene	7.0E+02	µg/m ³
	Cancer unit risk factors for		
	Benzene	2.9E-05	µg/m ³
	Ethylbenzene	2.5E-06	µg/m ³

Statistical analysis took place in SPSS packaging (Version 22), with Spearman's correlation used to identify the coefficient among BTEX species. Also, ArcGIS software (Version 10.3) was used to contact spatial analysis. In order to quantify the uncertainty in health risk assessment, Monte Carlo simulation and sensitivity analysis were implemented, using Crystal Ball risk analysis software (Version 11.1.2.4).

RESULTS AND DISCUSSION

In outdoor air, the concentration of benzene ranged between 1.61 µg/m³ and 4.83 µg/m³, with an average value of 3.19 µg/m³. Toluene ranged from 1.11 µg/m³ to 14.27 µg/m³, with an average value of 6.08 µg/m³. Concentration of ethylbenzene was

between 0.42 µg/m³ and 1.98 µg/m³, with its average value being 1.54 µg/m³. As for m,p-xylene, its concentration was limited to the ranged of 0.72-4.74 µg/m³, having an average value of 3.55 µg/m³, whereas o-xylene ranged from 0.08 µg/m³ to 1.92 µg/m³ and its average value was 1.12 µg/m³. Its results showed that the average annual concentration of benzene was below the threshold value of 5 µg/m³ (European Environment Agency, 2017) in all sampling points. There is no European legislation air quality standard for toluene, ethylbenzene, and xylene levels, but WHO has recommended a guideline value of 260 µg/m³ (weekly average concentration of toluene) to protecting against developmental neurotoxicity (Alexopoulos et al., 2006). In this study, the average

concentration of toluene was below the limit value of 260 $\mu\text{g}/\text{m}^3$, proposed by WHO. The average concentration of BTEX species in the present study turned out to surpass those, reported in Lithuania (in the vicinity of oil refinery at the Baltic Sea region), Turkey (in the vicinity of mega industrials such as the petroleum refinery and petrochemicals complex), Spain (in urban atmospheres and around the chemical and petrochemical industries), and China (in urban areas with heavy traffic and petroleum-chemical industrials).

However, these results were significantly lower than the ones, measured in Iran (in urban atmospheres), Kuwait (in urban residential and commercial area), Canada (in urban atmospheres), and India (at urban and industrial area). Table 2 compares the BTEX concentrations, reported in the present study, to those, reported in other literature. Due to two major sources (traffic density and fuel quality), BTEX concentrations in the urban areas turned out to be higher than the industrial ones.

Table 2. Comparison of average outdoor concentration of BTEX in other worldwide researches

Country	Location	Concentration ($\mu\text{g}/\text{m}^3$)				Reference
		B	T	E	X	
Kuwait	Al-Jahra	1.1	7.9	0.7	4.3	(Al-Khulaif et al., 2014)
China	Jinan	0.24	0.08	0.82	1.94	(Zhang et al., 2017)
Canada	Ontario	0.76	2.75	0.45	1.83	(Miller et al., 2012)
Iran	Tehran	7.8	23.2	5.7	17.4	(Amini et al., 2017)
Iran	Yazd	21	38	14	41	(Hajizadeh et al., 2018)
Iran	Ahvaz	1.78	5.19	0.51	1.13	(Rad et al., 2014)
Italy	Bari	0.8	0.9	0.2	1.1	(Caselli et al., 2010)
Canada	Edmonton	7.8	23.2	5.7	11.7	(Bari & Kindziarski, 2017)
Lithuania	Mazeikiai	1.2-2.4	1.3-3.5	0.3-0.7	1.0-2.4	(Marciulaitiene et al., 2017)
Spain	Southern Catalonia	1.18	3.90	0.85	1.32	(Ras et al., 2009)
Turkey	Aliaga (Industry)	4.7	3.9	0.99	2.76	(Civan et al., 2015)
	Aliaga (Urban)	0.68	1.6	0.25	0.47	
India	Dehradun	29.7	83.5	9.9	42.8	(Bauri et al., 2016)
Iran	Asaloyeh	3.19	6.08	1.54	4.67	Present study

According to average concentration of BTEX for each sampling locations (Table 3), the highest level of BTEX were registered in FIS point, close to the industrial zone, gasoline station, and the road with moderate daily traffic, along with SHC point, near the industrial zone and the road with moderate daily traffic. In this study, BTEX concentration in FIS and SHC points were ~1.7 times higher than HIS, ELS, SPC, HOS, and NTP, and ~2.5 times higher than RWC point. BTEX concentrations in this study decreased as the distance from the source increased. At all sampling points, the concentration of BTEX were the highest in winter season, during which time, the average (\pm standard deviation) concentrations of benzene, toluene, ethylbenzene, m,p-xylene, and o-

xylene were the highest at FIS point with 3.72 ± 0.28 , 11.25 ± 0.85 , 1.86 ± 0.08 , 3.98 ± 0.66 , and 1.31 ± 0.28 $\mu\text{g}/\text{m}^3$, followed by SHC point with 3.19 ± 0.35 , 9.92 ± 1.17 , 1.65 ± 0.10 , 3.92 ± 0.19 , and 0.59 ± 0.30 $\mu\text{g}/\text{m}^3$, respectively. The lowest average concentration for benzene, toluene, and m,p-xylene was observed at RWC point with 2.08 ± 0.21 , 2.16 ± 0.63 , and 1.28 ± 0.27 $\mu\text{g}/\text{m}^3$, respectively. This point was situated close to the villages, far away from the industrial zone and daily traffic. Whereas ethylbenzene and o-xylene concentrations were the lowest at HOS and SPC points with 1.11 ± 0.14 and 0.21 ± 0.12 $\mu\text{g}/\text{m}^3$, respectively. Generally, BTEX concentrations are lower in summer than winter, due to metrological factors. In winter, due to calm conditions and high

stability of the atmosphere, there is less dispersion, leading to pollutant accumulation, while in summer, dispersion is fast due to high temperature turbulence and unstable atmosphere. Also, removal of BTEX by OH radicals in summer leads to dilution of these compounds (Rad et al., 2014; Singh et al., 2016). The temperature (T) and solar radiation (SR) in Asaloyeh exhibited the highest averages in summer (35.5°C - 30.2 W/m²) and the lowest in winter (12.7°C - 18.6 W/m²), respectively. As T and SR increased, VOCs' vapor, emitted from condensate storage tanks, and evaporation from industrial wastewater basin are expected to be greater. However, high SR and T are suitable conditions for formation of OFP by consuming VOCs through photochemical reactions. In the present study, the OH radicals and unstable atmosphere play an important role in

atmospheric degradation process and dilution of the VOCs, respectively. These results are similar to previous studies (Hoque et al., 2008; Mullaugh et al., 2015; Rad et al., 2014; Singh et al., 2016).

According to the results, average concentration of toluene (~40.3%) were mostly abundant among investigated BTEX species in the summer and winter seasons at all locations. The second dominant species were benzene (~25.4%), followed by m,p-xylene (~18.3%) > ethylbenzene (~12.3%) > o-xylene (~3.7%). Previous studies have reported toluene as having the highest concentrations in urban and industrial atmospheres (Amini et al., 2017; Civan et al., 2015; Dehghani et al., 2018). The results showed that the toluene concentration increased in the following order: RWC < SPC < NTP < HOS < ELS < HIS < SHC < FIS.

Table 3. The average (±SD) concentration of BTEX (µg/m³) in indoor and outdoor air at different microenvironment

	RWC	ELS	HIS	NTP	HOS	FIS	SHC	SPC
<i>Outdoor-summer</i>								
Benzene	1.93±0.26	2.78±0.22	2.98±0.27	1.99±0.29	2.08±0.42	3.02±0.24	3.13±0.52	2.0±0.23
Toluene	2.00±0.32	3.07±0.72	4.73±0.45	3.60±0.32	3.43±0.69	8.75±0.41	7.42±0.81	3.17±0.40
Ethylbenzene	1.29±0.09	0.67±0.07	1.36±0.10	1.31±0.16	1.11±0.14	1.12±0.08	1.44±0.07	1.29±0.08
m,p-Xylene	1.74±0.21	1.18±0.50	2.83±0.18	1.05±0.22	1.32±0.35	2.59±0.55	3.42±0.06	2.05±0.24
o-Xylene	0.36±0.11	0.21±0.23	0.23±0.31	0.43±0.12	0.14±0.28	0.81±0.30	0.41±0.15	0.11±0.19
<i>Outdoor-winter</i>								
Benzene	2.08±0.21	2.96±0.26	3.13±0.45	2.57±0.13	2.35±0.44	3.72±0.28	3.19±0.35	2.29±0.27
Toluene	2.16±0.63	5.48±0.76	5.89±0.27	4.01±0.42	4.54±1.09	11.25±0.85	9.92±1.17	3.97±0.46
Ethylbenzene	1.56±0.09	1.56±0.07	1.60±0.07	1.41±0.71	1.35±0.10	1.86±0.08	1.65±0.10	1.60±0.08
m,p-Xylene	0.25±0.07	0.77±0.16	2.40±0.18	0.57±0.21	0.90±0.13	3.98±0.41	3.91±0.09	2.05±0.20
o-Xylene	0.40±0.12	0.55±0.25	0.44±0.30	0.55±0.05	0.24±0.20	1.31±0.28	0.59±0.30	0.21±0.12
<i>Indoor-summer</i>								
Benzene	0.79±0.13	1.69±0.42	1.91±0.19	-	1.00±0.15	2.17±0.31	2.04±0.12	1.23±0.20
Toluene	0.91±0.09	1.98±0.51	3.64±0.28	-	2.34±0.11	7.05±0.61	6.00±0.31	2.22±0.31
Ethylbenzene	0.38±0.05	0.12±0.02	0.50±0.09	-	0.22±0.05	0.25±0.03	0.49±0.11	0.41±0.15
m,p-Xylene	1.74±0.21	1.18±0.50	2.83±0.18	-	1.32±0.35	2.59±0.55	3.42±0.06	2.05±0.24
o-Xylene	0.13±0.02	0.18±0.07	0.16±0.03	-	0.11±0.02	0.53±0.17	0.33±0.09	0.08±0.01
<i>Indoor-winter</i>								
Benzene	0.95±0.17	1.87±0.10	2.00±0.12	-	1.35±0.14	2.65±0.42	2.15±0.16	1.58±0.22
Toluene	1.06±0.19	4.42±0.62	4.08±0.42	-	3.43±0.87	10.30±0.25	8.83±0.70	2.88±0.51
Ethylbenzene	0.65±0.09	0.67±0.03	0.78±0.15	-	0.43±0.16	0.95±0.18	0.71±0.14	0.68±0.12
m,p-Xylene	0.89±0.17	2.39±0.24	3.58±0.33	-	1.78±0.28	3.62±0.16	3.43±0.11	2.31±0.19
o-Xylene	0.18±0.02	0.43±0.09	0.28±0.13	-	0.12±0.06	1.01±0.18	0.39±0.14	0.28±0.02

Correlation coefficients could reveal the possible sources of BTEX (Tiwari et al., 2010). As such, the study showed significant positive correlation between ethylbenzene and xylene, especially at FIS point ($r > 0.95$). Moreover, a strong correlation between ethylbenzene and xylene suggested that they could be emitted from the gasoline stations (Khoder, 2007). This was consistent with previous studies, showing high correlations between ethylbenzene and xylene in Yokohama, Japan (Tiwari et al., 2010), Yazd, Iran (Hajizadeh et al., 2018), Ontario, Canada (Miller et al., 2012), and Delhi, India (Hoque et al., 2008). Benzene displayed low values of correlation coefficients ($r < 0.60$) with TEX compounds at HIS, FIS, and SHC points, indicating various emission sources of BTEX species (Hoque et al., 2008; Tong et al., 2013). Two main sources, namely vehicular exhaust or fugitive emissions of VOCs leaking from industrial equipment on one hand and evaporation from condensate storage tanks and gas flaring on the other, simultaneously contributed to BTEX emission in these three sampling points. Furthermore, RWC, ELS, SPC, NTP, and HOS points showed good correlation coefficients ($r > 0.80$) between benzene and TEX compounds, suggesting that BTEX were emitted from a common source, possibly vehicular emission. In order to identify this source, inter-species ratios of BTEX, especially the T/B ratio, was used as a diagnostic indicator (Caselli et al., 2010; Khoder, 2007).

The ratio of toluene to benzene (T/B) could reveal the emission sources of target compounds (Niu et al., 2012). T/B ratio within the range of 1 indicates the traffic-originated emission sources, while a higher ratio proposes that there are additional sources, beside vehicle exhaust (Hoque et al., 2008; Tiwari et al., 2010). Values above than 3 indicate volatile organic compounds, released into the atmosphere by local resources and industrial activities (Ceron-Breton et al., 2015). Here, the average of

T/B ratios in both winter and summer ranged from 1.03 to 3.08 and from 1.04 to 3.35, respectively, with its average value, observed to be maximum in SHC (3.03-3.35), FIS (3.08-3.21), and HIS (3.06-3.18) in winter and summer, respectively. Other locations displayed low values, indicating that toluene and benzene's emission sources could be the same, similar to those in Helsinki, Finland (2.64), Izmir, Turkey (1.87-2), Delhi, India (1.80-2.54), Algiers, Algeria (roadside, 1.45, and urban, 1.58), La Plata, Argentina (industrial area, 2.7, urban area, 3.21, and control area, 2.44), and Yazd, Iran (1.4-1.8) (Edwards et al., 2001; Muezzinoglu et al., 2001; Hoque et al., 2008; Kerchich and Kerbachi, 2012; Lerner et al., 2014; Hajizadeh et al., 2018). On the other hand, these results were lower than the ones, observed in Hong Kong (9.24), Fuji, Japan (8.69), and Bangkok, Thailand (5.7), attributing the emissions to industrial sources and toluene-rich fuels (Kume et al., 2008; Laowagul et al., 2009; Lee et al., 2002a).

Benzene has lower reactivity than ethylbenzene and xylene. The tropospheric lifetime of benzene, ethylbenzene, m-xylene, o-xylene, and p-xylene are estimated to be 9.4 days, 1.6 days, 11.8 h, 20.3 h and 19.4 h, respectively (Monod et al., 2001). The ratio of xylene to benzene (X/B) indicates the photochemical age of air parcel or a predominant emission source. Higher and lower values of X/B ratios show fresh air and old air mass (due to the transport), respectively (Khoder, 2007; Niu et al., 2012). The low ratios (0.47-1.42) of X/B in this study indicated which imply aging of the air mass and the transport were the main emission sources at all locations. X/B ratios found in the study were similar to those in Izmir, Turkey (1.87-2), Xiamen, China (0.70-3.40), La Plata, Argentina (1.60 - 2.33), Orleans, France (0.28 - 1.09), Shiraz, Iran (0.49 - 0.89) and Yazd, Iran (0.8-3.6) (Muezzinoglu et al., 2001; Niu et al., 2012; Lerner et al., 2014; Jiang et al.,

2017; Dehghani et al., 2018; Hajizadeh et al., 2018). The degradation rate of xylenes in the ambient air are faster than of ethylbenzene. Based on studies, xylenes are three times more reactive with the OH radical compared to ethylbenzene. The ratio of xylene to ethylbenzene (X/E) is used to evaluate VOC transfer from emission source. The decrease in the X/E ratio is dependent on the distance from the emission source (Lan and Minh, 2013; Monod et al., 2001). In the present study, the results showed that X/E ratio increased in the following order: RWC (1.10-0.85) < NTP (1.13-1.32) < HOS (1.32-1.76) < SPC (1.67-2.02) < ELS (2.07-2.21) < HIS (2.25-2.59) < SHC (2.66-2.73) < FIS (3.04-2.84) in the summer and winter, respectively. These findings confirm sampling locations distance from emission source. The X/E ratio of 0.14-2.64 in El Paso, USA (Raysoni et al., 2017), 2.4-5.3 in Algiers, Algeria (Kerchich & Kerbachi, 2012), 2.9 in Bari, Italy (Caselli et al., 2010) are similar to the ratios reported in this study.

VOCs play key roles in ground level ozone and photochemical smog formation. Generally, maximum incremental reactivity (MIR) is widely used to assess ozone formation potential (OFPs) from VOCs. The amount (g) of ozone, formed per gram of VOCs added to the initial VOCs-NO_x mixture, MIR shows how much the compound can contribute to the formation of ozone in the air. Its coefficients are intended to be employed in relatively high NO_x conditions, which can be used as an important tool in ozone control programs. Here, the MIR coefficients were adopted from previous studies (Carter, 1994; Li et al., 2015; Tong et al., 2013; Vo et al., 2018).

BTEX species plays a major role in the formation of ozone (Niu et al., 2012; Zhang et al., 2017). In the current study, m,p-xylene is the most dominant contributor to ozone formation, among BTEX species. The high reactivity of m,p-xylene (with an MIR of 8.2) were the main

reason for this. In addition, having an MIR rate of 2.70, toluene was the second largest contributor to ozone formation and benzene (MIR=0.42) had the lowest OFP value in the area. The reason was that concentration of benzene turned out to fall lower than toluene. As such, results of this study were similar to those, reported by Tiwari et al. (2010) in Yokohama, Japan; Hoque et al. (2008) in Delhi, India; Bauri et al. (2016) in Dehradun, India; and Na et al. (2005) in Seoul, South Korea. The average OFP value rose in the following order: RWC (24.34) < NTP (30.45) < HOS (36.39) < SPC (41.80) < ELS (47.44) < HIS (56.48) < SHC (68.56) < FIS (78.11) and RWC (18.10) < ELS (22.31) < HOS (25.21) < NTP (25.91) < SPC (30.52) < HIS (42.40) < FIS (54.42) < SHC (55.95) in winter and summer, respectively.

Table 3 shows indoor average concentration of BTEX species, found in the microenvironment during summer and winter. The average concentrations of BTEX were higher in winter than summer, ranging from 2.47 to 12.03 µg/m³ in summer and from 3.78 to 18.59 µg/m³ in winter. Indoor average concentration was in the following order: toluene > m,p-xylene > benzene > ethylbenzene > o-xylene for winter, and toluene > benzene > m,p-xylene > ethylbenzene > o-xylene for summer. Similar to outdoor concentrations of BTEX, indoor toluene was the most dominant compound in all the sampling locations, which was in good agreement with other previous studies (e.g., Du et al., 2014; Hazrati et al., 2016; Masih et al., 2017; Uchiyama et al., 2015). The highest and lowest average indoor BTEX concentration belonged to FIS and RWC, with 30.59 and 6.25 µg/m³, respectively, indicating that average concentration of benzene, toluene, ethylbenzene, m,p-xylene, and o-xylene displayed their highest records in FIS and SHC microenvironments, followed by HIS, ELS, HOS, SPC, and RWC. Highest concentration of BTEX in FIS and SHC may

be explained by the building materials, poor ventilation, and outdoor sources like vehicles and industrial activities. The average concentration of benzene was lower than the mean annual concentration standards of the European Union ($5\mu\text{g}/\text{m}^3$) in all samples (European Environment Agency, 2017).

Average concentration of benzene, toluene, ethylbenzene and xylene in all different locations (being 0.87-2.30, 0.99-8.6, 0.32-0.63, and 0.59-3.26 $\mu\text{g}/\text{m}^3$, respectively) was found to fall lower than those, reported in Bangkok, Thailand (8.08, 110, 12.1, and 10.9 $\mu\text{g}/\text{m}^3$); Los Angeles, USA (3.54, 15.26, 2.55, and 4.49 $\mu\text{g}/\text{m}^3$); Rio-de-Janeiro, Brazil (54.14, 209.24, 45, and 59.46 $\mu\text{g}/\text{m}^3$); Ardabil, Iran (15.8, 69.70, 12.07, and 48.08 $\mu\text{g}/\text{m}^3$); and Gorakhpur, India (12.68, 28.93, 4.11, and 3.11 $\mu\text{g}/\text{m}^3$) (de Castro et al., 2015; Hazrati et al., 2016; Masih et al., 2017; Ongwande et al., 2011; Su et al., 2013). However, average BTEX concentrations reported in Antwerp, Belgium (2.2, 4.25, 0.62, and 2.04 $\mu\text{g}/\text{m}^3$) were lower than the results, found in this study (Stranger et al., 2007).

The BTEX statistical analysis was conducted to determine the potential sources for emission. For this purpose, the ratio of indoor-outdoor (I/O) was put into good use. In order to calculate these ratios, dividing the average indoor concentrations were divided by the average outdoor concentrations for BTEX compounds. The average I/O concentration ratio was reported for BTEX > 1 in Asan and Seoul, South Korea (Son et al., 2003); Kalkata, India (Majumdar et al., 2012); Valencia, Spain (Esplugues et al., 2010); Aveiro, Portugal (Vicente et al., 2017); and Ardabil, Iran (Hazrati et al., 2016), suggesting the presence of indoor sources for these compounds.

The average I/O concentration ratio in the present study varied between BTEX species. It was found to be below 1. I/O ratio of ≤ 1.0 , indicating influence of

outdoor sources and lack of significant indoor sources (Edwards et al., 2001; Hadei et al., 2018). This is similar with previous studies in the Los Angeles, Hong Kong, and Mexico City (Hun et al., 2011; Lee et al., 2002b; Serrano-Trespacios et al., 2004). The I/O concentration ratio depends on the sampling region (the status of the country), the sampling location (road or park for outdoor), traffic density, variety and number of industries, product use inside the homes, period and points of sampling, and atmospheric conditions (Do et al., 2013). Lower ratio of I/O in the present study than the US EPA report on the status of indoor air quality (USEPA, 2013) may be due to the use of natural gas and electric heater for cooking and heating in the area, the use of metal cabinets in the kitchen instead of medium-density fiberboard cabinets, lack of rooms coloring, restrictions on smoking inside the room, and good ventilation system. It should be noted that the natural gas contains low levels of BTEX (Colborn et al., 2013).

Results showed that the average I/O concentration ratio for BTEX ranged from 0.53 to 0.88 and 0.41 to 0.77 in winter and summer, respectively. The average I/O ratio of benzene, toluene and xylene were > 0.70 in FIS, compared to other points and the I/O ratio of ethylbenzene was the lowest in all sampling locations. Fig. 2 demonstrates the relations between indoor and outdoor concentrations of BTEX species. High correlation between indoor and outdoor toluene ($R^2=0.98$) and xylene ($R^2=0.99$) showed that their levels were affected by the outdoor environment.

In this study, despite the fact that average BTEX concentration was found more in winter than summer, the difference was insignificant. No significant seasonal variations of BTEX were observed, probably due to continuous emissions from major petrochemical and gas industrial facilities (Kim et al., 2019).

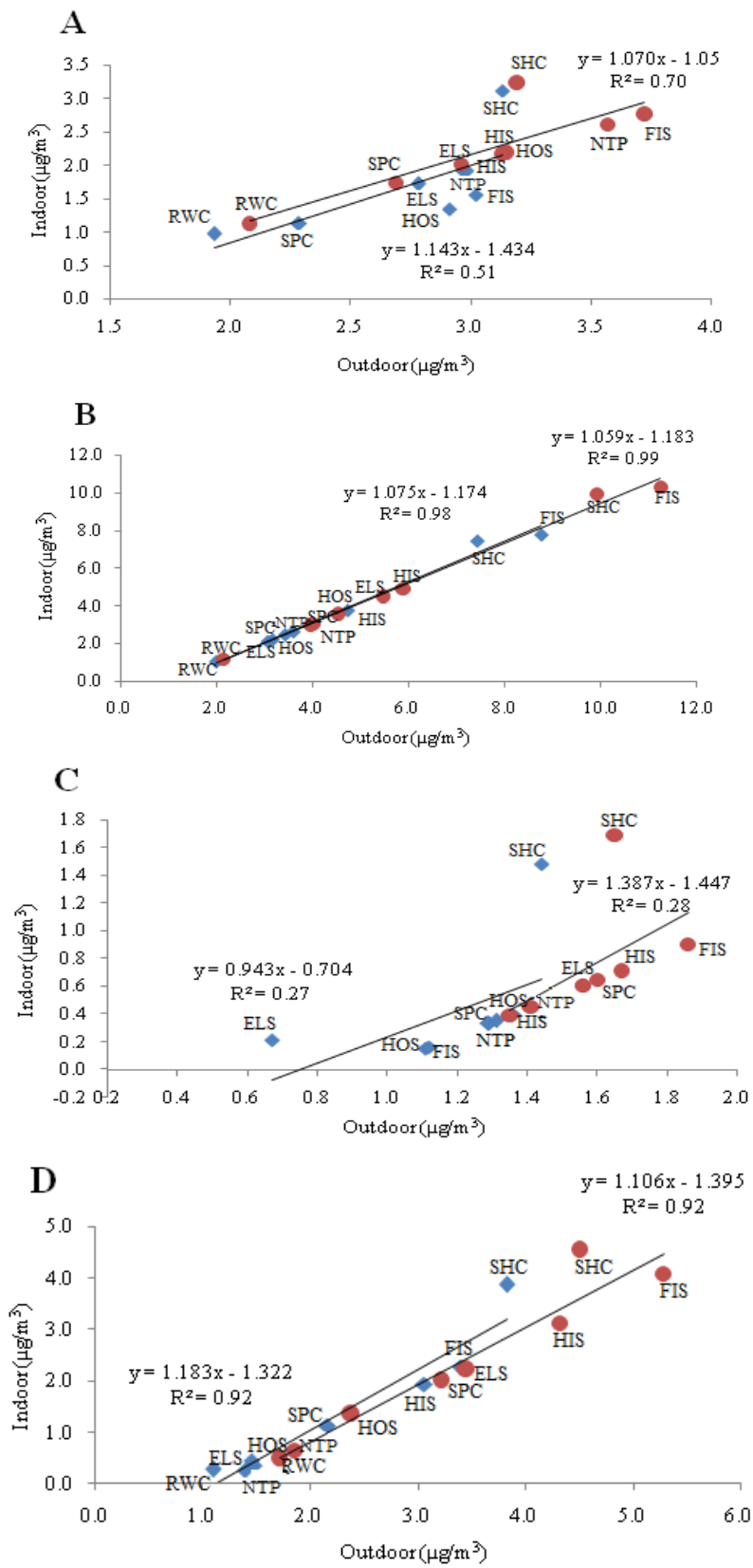


Fig. 2. Correlation of I/O concentration for benzene (A), toluene (B), ethylbenzene (C), and xylene (D) in summer (◆) and winter (●) at different microenvironments

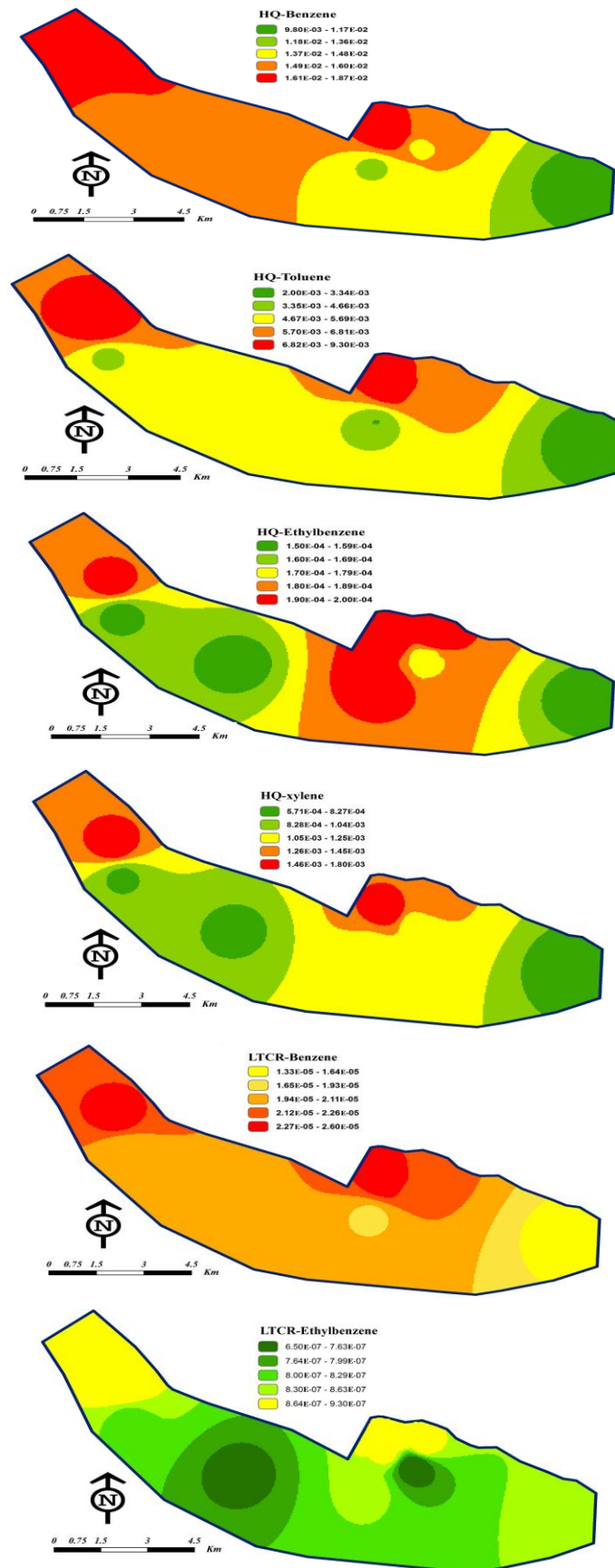


Fig. 3. Spatial distribution of HQ for BTEX species and LTCR for benzene and ethylbenzene

Fig. 3 illustrates the spatial distribution of HQ values outdoors, in accordance with modeled BTEX data. The $HQ \leq 1$ shows that adverse health effects are not likely to occur, and thus can be considered as having negligible hazard, even to a sensitive individual. On the contrary, $HQ > 1$ means that there may be some risks to sensitive individuals as a result of exposure. In the current study, benzene gave the highest HQ followed by toluene, xylene, and ethylbenzene, both indoors and outdoors. The HQ for all pollutants was below the above-mentioned value in all sampling locations, indicating no serious risk of non-cancer health effects. Cumulative non-cancer hazard index (HI) values for BTEX species both outdoors and indoors turned out to be the highest in FIS (3.0E-02, 2.1E-02) followed by SHC (2.7E-02, 1.8E-02), HIS (2.3E-02, 1.5E-02), ELS (2.1E-02, 1.2E-02), HOS (2.0E-02, 9.1E-03), NTP (2.2E-02), SPC (1.7E-02, 9.8E-03), and RWC (1.2E-02, 5.5E-03).

The LTCR of benzene, determined by USEPA, was $< 1.0E-06$, while the WHO has suggested an LTCR rate between $1.0E-05$ and $1.0E-06$ as acceptable (Dehghani et al., 2018; Lerner et al., 2014). The

predicted LTCR value for benzene concentration in indoor microenvironment was higher than the threshold value, being $1.0E-06$ in FIS, followed by SHC, HIS, and ELS with $1.7E-05$, $1.4E-05$, $1.3E-05$, and $1.2E-05$, respectively, while the lowest LTCR value belonged to RWC with $6.1E-06$. The average LTCR of benzene and ethylbenzene air was $1.9E-05$ and $7.3E-07$ outdoors, respectively, showing the LTCR of benzene above the limits, recommended by USEPA. Fig. 3 gives the spatial distribution of LTCR for benzene and ethylbenzene in all sampling locations. Cumulative risk values was found in FIS point ($2.7E-05$), followed by SHC ($2.5E-05$), HIS ($2.3E-05$), ELS ($2.2E-05$), HOS ($2.1E-05$), NTP ($1.9E-05$), SPC ($1.8E-05$), and RWC ($1.4E-05$). Considering LTCR value of benzene in the present study, it is noteworthy that similar findings have been reported by Ramirez et al. (2012), Zhang et al. (2012), Chen et al. (2016), and Dehghani et al. (2018), with mean LTCR values $3.4E-04$, $4.1E-05$, $6.4E-05$, and $1.9E-04$, respectively. However, different results have been obtained by Masih et al. (2016) and Miri et al. (2016).

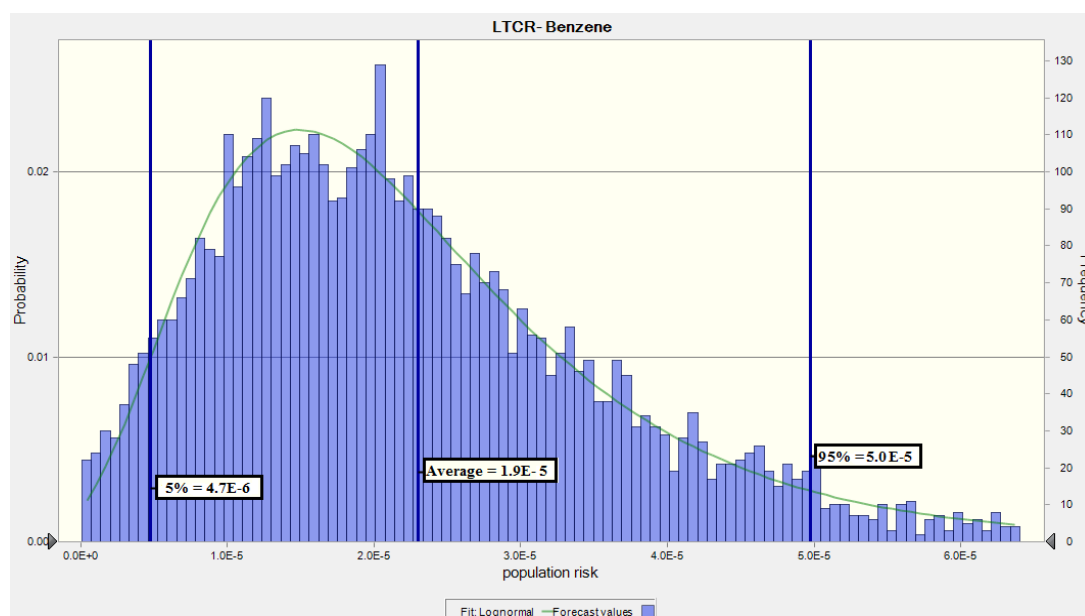


Fig. 4. LTCR for personal exposure to benzene in the study area (based on an acceptable risk level of $1.0E-6$)

When only a point value is used to assess the risk of exposure to pollutants for a population, it is associated with great uncertainty. Therefore, this study implemented Monte Carlo simulation and sensitivity analysis so as to quantify the uncertainty. To obtain a realistic picture of risk distribution, there were 5000 iterations performed and a risk histogram was created. Fig. 4 displays the probability distribution (average of 5%, and 95%) of the estimated LTCR for personal exposure to outdoor concentration of benzene. Results of sensitivity analysis show that benzene concentrations (ranging from 52.1% to 59.3%), exposure duration (ranging from 14.8% to 18.5%), and inhalation rate (ranging from 8.7% to 12.2%) contributed to the total risk variance.

CONCLUSION

The present study determined the characteristics and health risk assessment of BTEX in the outdoor and indoor air of eight different microenvironment in an industrial-urban area. Both indoor and outdoor average concentration of BTEX was higher in winter than summer. Also, BTEX concentrations were greater outdoors than indoors. Indoor to outdoor ratios of BTEX were <1 in all sampling microenvironments. Spatiotemporal distribution revealed that BTEX concentration declined by increasing the distance from emission source, including industrial zone, gas station, and traffic road. In this study, toluene concentration turned to be dominant in all sampling points. Health risk assessment, based on the estimated data, proved that the levels of benzene were higher than the acceptable level of $1.0E-06$. However, HQ risk for BTEX compounds in all areas was lower than 1, showing no concern of increased health risk. To reduce and eliminate the effects of BTEX compounds, it is recommended to change fuel quality and keep maximum distance between the road and sensitive urban location. On the

other hand, so far as the public health is concerned, to avoid any likewise adverse impact of the industrial area, it is essential to create the buffer area between industrial and residential zones.

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