Levels of BTEX in ambient air in two urban sites located in the center zone of Orizaba Veracruz, Mexico during autumn 2014 and assessment of the carcinogenic risk levels of benzene

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Abstract**—**Atmospheric levels of BTEX (benzene, toluene, ethyl benzene and p-xylene) were measured in two urban sites located in Orizaba, Veracruz, Mexico using an active sampling during autumn 2014. 1.5 h-samples were collected using carbon packed cartridges at 07:00 h, 14:00 h and 17:30 h, and then analysed using gas chromatography with flame ionization detector (GC-FID). The relative abundance of BTEX followed the order for both sites: benzene> toluene >p-xylene>ethyl benzene. Mean concentrations for benzene were 74.50 $\mu g\ m^{-3}$ and 91.16 $\mu g\ m^{-3}$, for toluene were 5.32 $\mu g\ m^{-3}$ and 19.74 $\mu g\ m^{-3}$, for ethylbenzene were 3.34 $\mu g\ m^{-3}$ and 2.52 $\mu g\ m^{-3}$ and for p-xylene were 2.25 $\mu g\ m^{-3}$ and 2.72 $\mu g\ m^{-3}$, for the first and the second site, respectively. For the first site, benzene and ethyl benzene did not show a specifically behaviour and the highest mean levels for toluene were obtained during the afternoon sampling period. p-xylene showed the highest levels during midday. For the second site, the highest concentrations of benzene, toluene and ethyl benzene were found during the midday sampling period. During the whole sampling period the influence of the vehicular sources was evident due to the highway Veracruz-Mexico which is located in these directions (SE, S, SSE) for both sampling sites. We can infer that the high benzene concentrations found in this study were influenced by vehicular traffic. The life time cancer risk due to exposure to ambient benzene in both study sites exceeded the acceptable values reported by USEPA and health concerns of benzene exposure should be considered in this region of Veracruz, Mexico.

Keywords**—**BTEX, Orizaba, VOCs, air pollution, autumn, Mexico

I. INTRODUCTION

AMBIENT monitoring of Volatile Organic Compounds (VOCs) is a field of increasing interest due to their effects on human health and their contribution to the formation of tropospheric ozone [1] and additionally, due to their possibilities of causing adverse health effects [2-4]. Alkanes, alkenes, alkynes, aldehydes, alcohols, ketones, chlorofluorocarbons, chlorinated organic compounds and aromatics are included within the VOC group. Aromatic Hydrocarbons includes a sub-group named as “BTEX” which includes to benzene, toluene, ethyl-benzene, 1,2,4-trimethyl-benzene, o-xylene, m-xylene and p-xylene, all of them considered as air toxic pollutants and some of them as carcinogenic species [5]. Some VOCs such as benzene, 1,3-butadiene and formaldehyde have also mutagenic and carcinogenic effects after long-term exposure [6-7].

On the other hand, these compounds have multiple and different sources: they can be can emitted from biogenic sources [8]; from mobile sources [9-10] from evaporative emissions in fuel and gas storage [11-12], from fuel

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combustion [13], from biomass burning [14]; and from industrial processes and solvents [13]. Determination of VOCs levels in ambient air in urban areas is crucial to understand their influences on human health and urban air quality, and to identify their probable emissions sources of these pollutants. In spite of the well-known toxic effects of BTEX, data available on BTEX levels in Mexico are very limited. Few studies have reported BTEX levels in Mexico mega-cities [15-18] and there are not studies on other important cities in Mexico.

Veracruz is one of the most important states in Mexico due to its economy which is based on agriculture and petroleum. The state has about one-fourth of Mexico's petroleum reserves and ranks third in petroleum production [19-20]. In the central area of this state, important cities are located including its capital, Xalapa city. About one third of the state's population lives in urban centers, especially in the central region. One of the most important cities is Orizaba, which is located in the mountainous central region, at 266 km from Mexico City, adjacent to Rio Blanco, on Federal highways 180 and 190. The municipality population is of more than 120,000 inhabitants and it has an area of 27.97 km^2 [19]. Its economy has grown significantly in the last years. The industry is mainly focused in the following areas: breweries, paper, cement, pharmaceutical, iron and steel factories, soft drinks and food, leather and shoemaker companies among others. For all these reason, Orizaba Valley has not only grown in population if not also in the number of vehicles, industries and services. As a result of this increased economic development, air quality in this region has suffered a gradual decline in recent years. In this study, we monitored BTEX levels at two urban sites located in Orizaba city within the mountainous central region of Veracruz, Mexico during autumn 2014. The purposes of this study are: (1) to investigate the exposure levels of BTEX in ambient air in two urban sites in Orizaba, Veracruz, Mexico; (2) to explore their probable emission sources using meteorological analysis (wind roses and backward trajectory analysis using HYSPLIT from NOAA) and chemometric analysis (principal component analysis: PCA); (3) to explore their diurnal patterns; and (4) to estimate the potential health risks of benzene in these sites.

II. STUDY AREA

A. Observation Site

The observation site is located near the urban center of Orizaba, Veracruz, in the west area (18° 49' 54.5" N; 97° 06' 48.6" W), at 1251 masl. Its climate is pleasant, though often cloudy and rainy and the soil of the valley is extraordinary fertile. Overlooking the valley from the north is the Pico de Orizaba, a volcano with 5636 m high which is the highest mountain in Mexico and the third highest in North America.

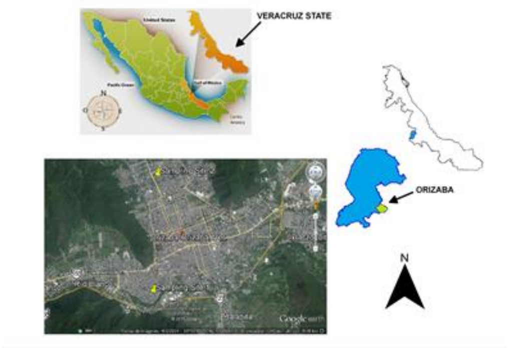


Fig. 1 Sampling Site Location.

The mean annual rainfall is 1800 mm. January is the coldest month (7-10 ºC). The average annual high and low temperatures are of 27 ºC and 13 ºC, respectively. Mean annual temperature is of 21 ºC. Relative humidity from November to February is high almost all the time (almost 100%) and thermal inversions are common in winter. A detailed map of the sampling sites is presented in Figure 1. The specific observation sites are located within urban, residential, educational and commercial areas where there also are several avenues with high vehicular traffic volume.

III. METHODOLOGY

A. Sampling Method

A total of 72 samples were collected from November 7 to November 29, 2014, during the autumn season. Benzene (B), ethyl benzene (Ebz), toluene (T) and p-xylene (X) were determined in all ambient air samples. Samples were collected using glass tubes containing 226-01 Anasorb CSC (SKC) with following features: length 70 mm; inner diameter 4.0 mm; outer diameter 6 mm packed in two sections with 100 mg and 500 mg of active carbon, separated by a glass wool section (Method INSHT MTA/MA-030/A92)[21, 22]. The downstream end of the glass tube was connected to a calibrated flow meter. Ambient air was passed through the glass tubes at a flow rate of 200 ml min^{-1} at 1.5-hour intervals (morning, midday and afternoon). Samples with a sampling volume lower than 5 liters were discarded. Sampling was carried out using a Universal XR pump model PCXR4 (SKC), at three sampling periods (local time): B1 (from 07:00 to 08:30 h), B2 (from 14:00 to 15:30 h) and B3 (from 17:30 to 19:00 h). During sampling, the Swagelok® fitting was removed from the marked end, and a diffusion cap was fitted to the end of the sampling tube. Prior to the main study, several pilot experiments were conducted to evaluate the suitability of the sampling procedure described here. These pilot experiments included the determination of appropriate sampling times. In addition, desorption efficiency (DE) was calculated for each glass tubes lot and for each analyte, and tubes with ED lower than 75% were discarded. Sampling tubes were protected from adverse weather conditions by aluminum shelters. After the exposure time, the adsorption tubes were labeled and capped tightly with PTFE caps and transferred to the laboratory in cold boxes. This procedure was applied to both clean and sample tubes for storage prior to use or analysis. Field blanks were transported to the field along with samplers and stored in the laboratory during the sampling period. Samples were analyzed within three weeks after sample collection at the Environmental Sciences Laboratory of the Autonomous University of Carmen City (UNACAR).

B. Analytical Method

Samples were extracted with 1 ml of CS_2 for each section of the sample tubes, and shaken for 30 s to assure maximum desorption. Extracted samples were analyzed using a TRACE GC Ultra gas chromatograph (Thermoscientific) and one flame ionization detector (FID; Thermoscientific Technologies, Inc) (Method INSHT MTA/MA-030/A92) [22]. The analytical column used was a capillary column (57 m, 0.32 mm i.d., 0.25 µm film thickness). Operation of the instrument was controlled using a Trace Chemstation data system. The oven temperature program was initially set to 40 °C for 4 min, then increased at a rate of 5 °C/min up to 100 °C, and finally maintained for 10 min at 100 °C. The FID temperature was set to 250 °C using a hydrogen/air flame with constant flows of 35 ml min^{-1} and 350 ml min^{-1} for ultra-pure hydrogen and extra-dried air, respectively. The ultra-pure nitrogen carrier (99.999%) gas flow rate was 1 ml min^{-1} (INSHT 1992). Four BTEX were investigated: benzene, p-xylene, ethylbenzene, and toluene. Five-point calibration was performed using 99.98% Sigma-Aldrich analytical reagents at a concentration of 2 ppm for each BTEX.

The established calibration curves for the four investigated BTEX were found to have R-square values of 0.999. Method detection limits (MDL) for each compound were calculated by multiplying the standard deviation obtained from seven replicate measurements of the first level of calibration by 3.14 (Student's t-value). The analytical results showed that the MDLs for the four VOC compounds of benzene, ethyl benzene, p-xylene, and toluene, were 0.0517, 0.0566, 0.0600, and 0.025 µg m^{-3}, respectively. The amount of BTEX in blank samples was below the limit of detection for all compounds studied.

C. Monitoring of Meteorological Parameters

Wind conditions (speed and direction), relative humidity, temperature and barometric pressure were monitored during summer and autumn, 2013 (from July 3 to November 14) using a Davis Vantage Pro II model portable meteorological station. Wind frequency statistics were determined using WRPLOT software [23]. This station was located specifically in the sampling site.

D. Correlation and Principal Component Analysis (PCA)

Pearson correlation analysis was applied to all data collected at the sampling site. To assess the relationships between BTEX concentrations, meteorological parameters and criteria air pollutants; a factor analysis (Principal Component Analysis) was applied using XLSTAT software (Statistics Package for Microsoft Excell) [24].

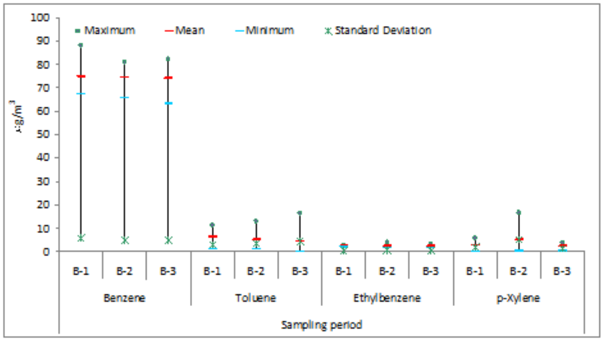
E. Health Risk Assessment

The carcinogenic potential of benzene is broadly known [25]. European Union recommends an annual limit of 5 µg m^{-3} for benzene in ambient air and the inhalation Minimal Risk Level (MRL, at a cancer risk of 1 in 10,000) of 4.0 ppbv has been established by USEPA for this pollutant [26]. We use the methodology proposed by Zhang et al. 2015 in order to determine daily exposure (E), the lifetime cancer risk (LTCR) and the non-cancer risk (HQ).

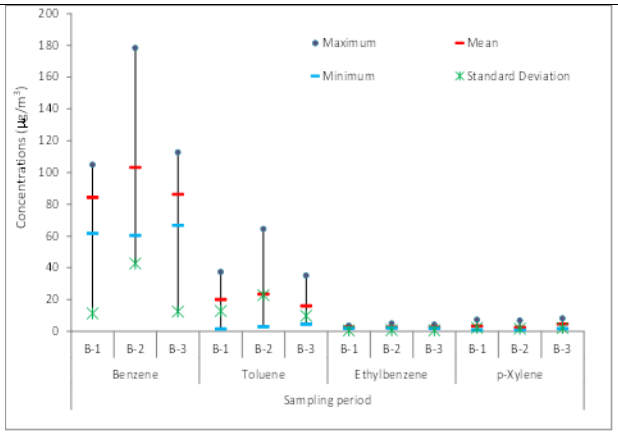
IV. RESULTS

A. Diurnal Variation

Diurnal variation and descriptive statistics for autumn sampling period can be observed in Fig 2a and 2 b for the sampling sites 1 and 2, respectively. For the Sampling Site 1 (SS1), during this period, Benzene (B), Toluene (T), Ethylbenzene (Ebz), and p-xylene (X) showed different diurnal patterns. B and Ebz did not show significant differences among the different sampling periods (B1: morning sampling, B2: midday sampling, and B3: afternoon sampling). T presented higher levels during the mornings (B1), decreasing during the midday (B2) and presenting the lowest values during the afternoons (B3). X had the highest concentrations during midday (B2) and the lowest levels during the afternoon sampling period (B3). For the Sampling Site 2 (SS2), B, T and Ebz showed their highest concentrations during the midday sampling period whereas X had its maximum level during the afternoon sampling period. The relative abundance of BTEX exhibited the following order for both sampling sites: B > T> X > Ebz. Mean concentration levels were: 74.51 µg m^{-3} and 91.16 µg m^{-3} for B., 5.33 µg m^{-3} and 19.74 µg m^{-3} for T., 2.26 µg m^{-3} and 2.52 µg m^{-3} for Ebz., and 3.35 µg m^{-3} and 2.72 µg m^{-3} for X; for SS1 and SS2, respectively.



a)



b)

Fig. 2 Diurnal variation and descriptive statistics for measured BTEX during the autumn sampling period, 2014. B1 (07:00-08:30 h), B2 (14:00-15:30 h) and B3 (17:30-19:00 h). a) Sampling Site 1: SS1. b) Sampling Site 2: SS2.

BTEX levels in different cities are influenced by several factors including sampling season, sampling duration, sampling location, meteorological conditions, site topography and sampling methods. Bearing these factors in mind, BTEX levels in the study site could be compared in a cautious way with those levels reported in other cities around the world (Table 1). It can be observed that, during autumn sampling period, for SS1, B levels found in this study were similar to those reported for Greater Cairo and Algeriers, whereas, T levels were lower than those found in Seoul, Korea and higher than those reported by Pankow [27] in New Jersey. Ebz levels were similar to those found in Seoul and X levels were higher than those reported for New Jersey. For SS2, B levels were higher than those reported for Haram, Greater Cairo; T and X levels were within the range of concentrations reported for Algeriers, Africa. Ebz concentrations were lower than those registered in Seoul, Korea but higher than those reported for New Jersey.

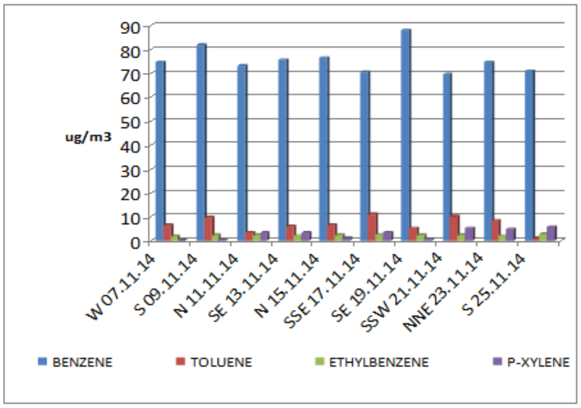
B. Meteorological influence

Winds during the autumn sampling period were quite unstable but always showing a south component coming from the highways 180 and 190 located in this direction for two sampling sites. Figures 3a, 3b and 3c and 4a, 4b and 4c show the daily meteorological influences for each BTEX for B1, B2 and B3 sampling periods, for SS1 and SS2, respectively. During the mornings (B1), BTEX showed the highest values when winds blew from SSE and S for SS1. During B1 period, B and T showed the highest concentrations when winds came from NNE for SS2. During the midday sampling period (B2) for SS1, the highest values of concentration were found for B when winds blew from SW, for T when air masses came from SE, for Ebz when winds came from SSW and for X when winds blew from SSE. BTEX showed higher levels when winds came from S and SSW during midday sampling period for SS2. During the afternoon sampling period (B3) for SS1, winds had a more stable behavior: B presented the highest levels when winds came from SSE, whereas, T, Ebz and X showed the highest concentrations when winds blew from SE.

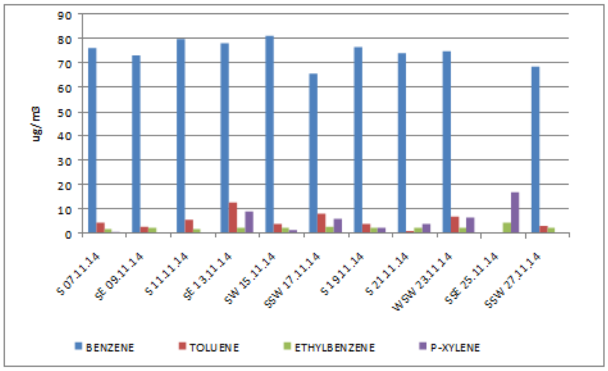
Table 1 Comparison of atmospheric concentrations of BTEX found in this study with data of other studies around the world.

	B		**T**		**Ebz**		**X**	
	µg m^{-3}	ppbv	µg m^{-3}	ppbv	µg m^{-3}	ppbv	µgm^{-3}	ppbv
Rowan College, NJ [27]	-	0.36	-	0.5	-	0.09	-	0.3
Haram, Greater Cairo [28]	33.3-58.5	-	78.4-138.6	-	16.6-32.4	-	51.51-99.64	-
Seoul, Korea [29]	-	1.00	-	6.4	-	0.7	-	2.3
Algeriers, Africa [30]	1.1-26.8		3.5-63.3	-	2-12	-	4.9 - 46.8	-
Present Study								
SS1	74.5	24.2	5.3	2.0	2.2	0.5	3.3	0.7
SS2	91.1	29.6	19.7	7.7	2.5	0.5	2.7	0.6

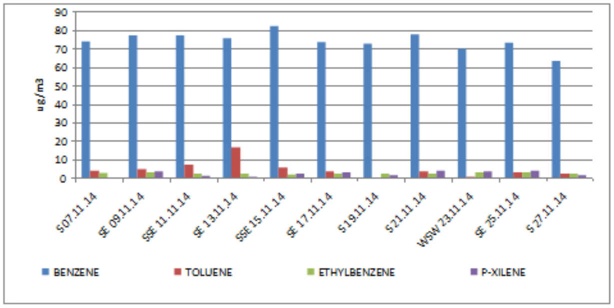
For SS2, during the afternoon sampling period (B3), B and T levels were higher when winds came from S and SW. Whereas, Ebz showed its maximum levels when winds blew from S and for X when air masses came from SSE and SW. Figures 5a, 5b and 5 c and Figures 6a, 6b and 6c show representative wind roses for the autumn period which were constructed by WRPLOT from Lakes Environmental Inc. [23], for SS1 and SS2, respectively. In addition 24 h backward air masses trajectories were calculated in order to infer the origin of air masses at three different levels (50, 100 and 500 magl) using the hybrid Lagrangian model HYSPLIT from NOAA (Figures 7a, 7b and 7c for SS1 and Figures 8a, 8b and 8c for SS2). The wind conditions are used to identify the probable sources of the measured BTEX compounds. During the whole sampling period the influence of the vehicular sources was evident due to the highway Veracruz-Mexico which is located in these directions (SE, S, SSE and SW). We can infer that the high benzene concentrations found in this study were influenced by vehicular traffic.



a) B1

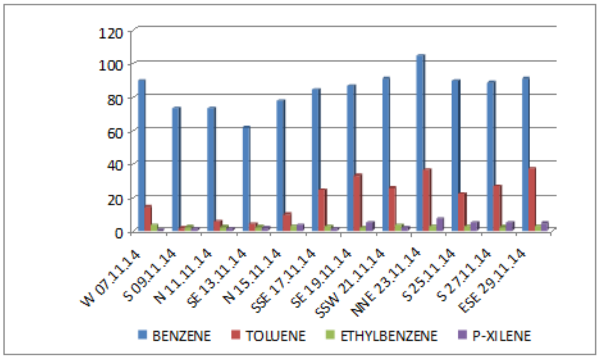


b)B2

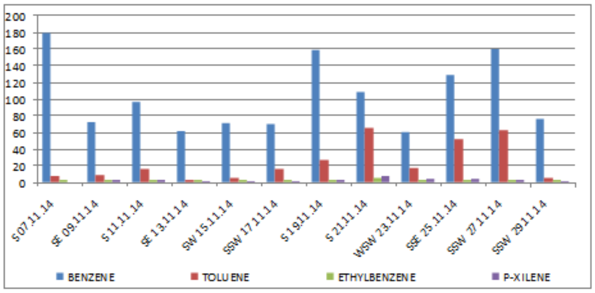


c) B3

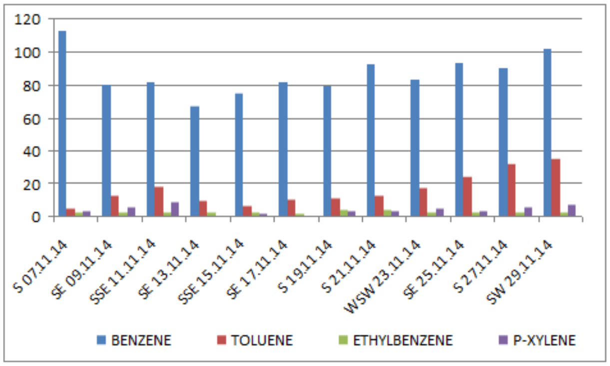
Fig. 3 Meteorological influence on BTEX levels for sampling site 1 (SS1): a (B1: 07:00- 080:30 h); b (B2: 14:00-15:30 h); and c (B3: 17:30-19:00 h).



a) B1



b) B2



c) B3

Fig. 4 Meteorological influence on BTEX levels for sampling site 2 (SS2): a (B1: 07:00- 080:30 h); b (B2: 14:00-15:30 h); and c (B3: 17:30-19:00 h).

a)

b)

c)

Fig. 5 Representative wind roses for a) November 19, 2014, b) November 17, 2014 and c) November 25, 2014.

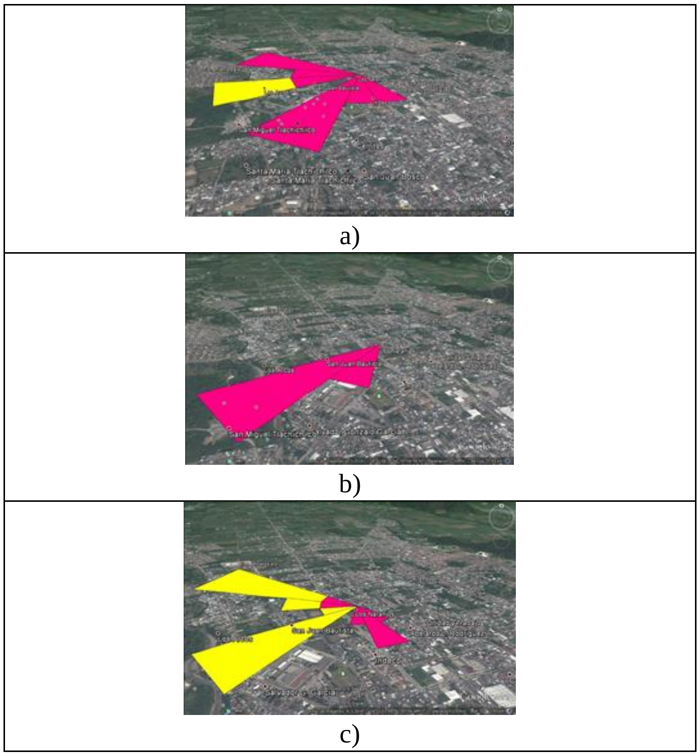


Fig. 6 Representative wind roses for a) November 19, 2014, b) November 17, 2014 and c) November 25, 2014.

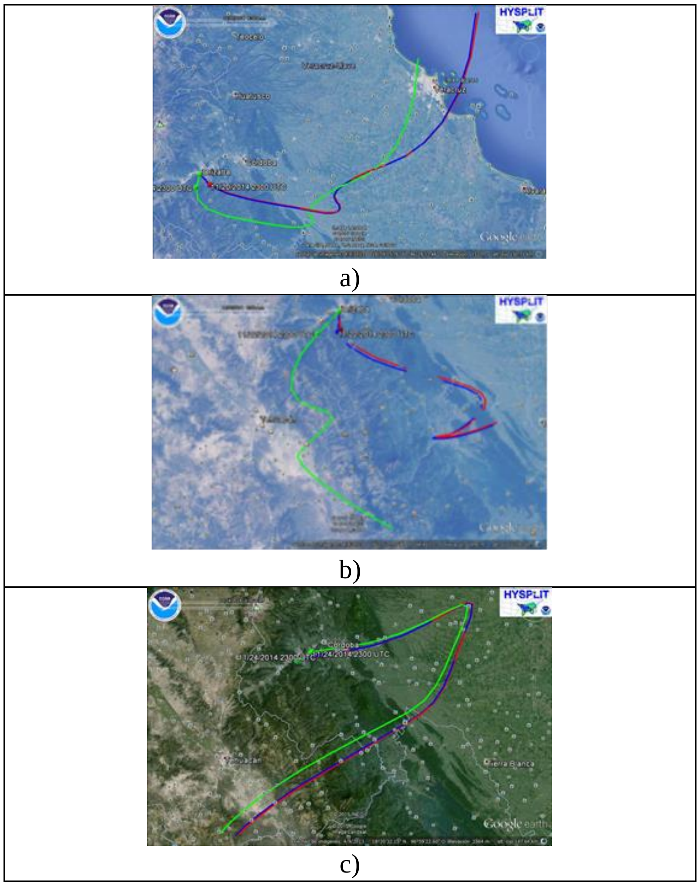


Fig. 7 Representative 24-h backward air masses trajectories for summer period: a) November 21, 2014, for b) autumn period: November 23, 2014 and for c) November 25, 2014.

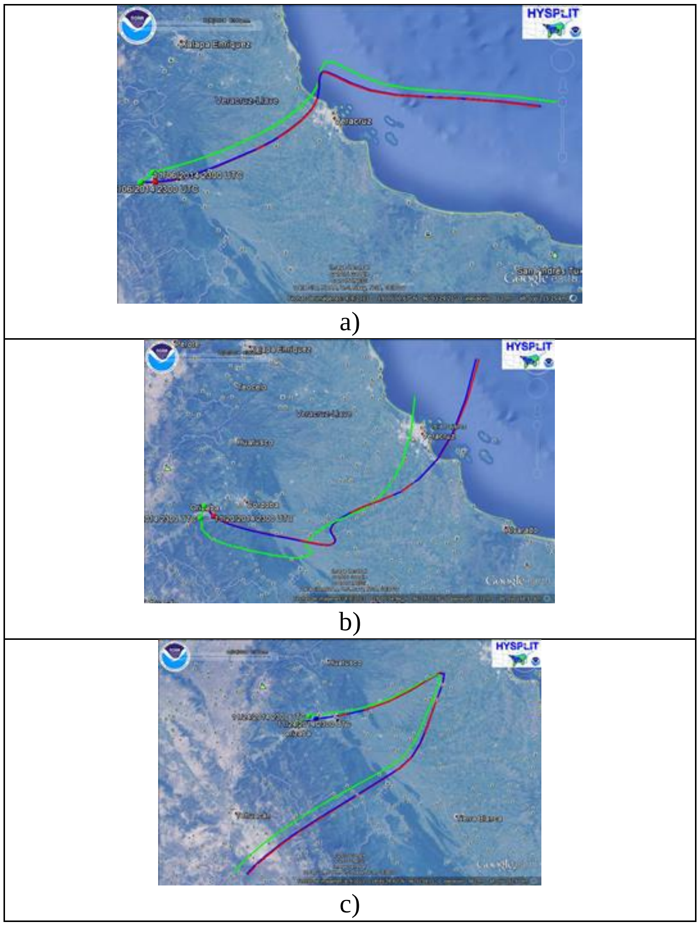


Fig. 8 Representative 24-h backward air masses trajectories for summer period: a) November 7, 2014, for b) autumn period: November 21, 2014 and for c) November 25, 2014.

C. Toluene to Benzene ratio (T/B ratio)

The T/Bz ratio has been commonly used as an indicator of traffic emissions. Bz and T are constituents of gasoline and are emitted into the atmosphere by motor vehicle exhaust. The toluene content of gasoline and motor vehicle exhaust is 3-4 times higher than Bz content [31]. T/Bz values lower than 2-3 are characteristic of vehicular emissions in many urban areas worldwide [32, 16-17]; whereas values higher than 3 may indicate that BTEX levels could be associated with industrial facilities and area sources (evaporative emissions, painting, cooking processes, among others). This ratio was of 0.0864 for B1, of 0.0689 for B2 and 0.0583 for B3, for SS1 being higher during the morning sampling period. For SS2, this ratio showed mean values of 0.2247 for B1, of 0.2128 for B2 and 0.1824 for B3. These values are in agreement with typical values of vehicular emissions reported for other urban areas, suggesting that this site was under the influence of mobile sources.

D. p-Xylene to Ethylbenzene ratio (X/Ebz ratio)

The p-Xylene to Ethylbenzene (X/Ebz) ratio is commonly used as an indicator of the photochemical age of the air masses. A ratio of 3.6: 1 of (X/Ebz) has been established as a typical emission relation for these species[43-44]. This ratio is related to the atmospheric residence time of these pollutants: high values of indicate aged air masses (old emissions), and low values indicate fresh air masses (recent emissions). Kuntasal et al. [33] used a value of 3.8 for this ratio. Fresh gasoline emissions provide values between 3.8 and 4.4 for this ratio. In this study, the whole period registered low values for this ratio, indicating that most of the air masses correspond to "fresh emissions". A mean value of 1.2798 for this ratio was obtained for SSI (B1: 1.1104; B2: 1.8025; B3: 0.9263). For SS2, mean X/Ebz ratios were of 1.3814 for B1, 0.9038 for B2 and 1.8121 for B3. Taken together, the T/Bz and X/Ebz ratio results suggest that these fresh emissions correspond to vehicular emission from mobile sources.

E. Pearson Correlation and Principal Component Analysis (PCA)

Meteorological analysis and T/B ratios and X/Ebz ratios suggest that autumn BTEX levels at both sites under study were influenced in a dominant way by vehicular traffic emissions (see sections 4.2, 4.3 and 4.4). However, PCA analysis can reveal more detailed information about the behavior of the studied pollutants. While BTEX ratios are used as markers of fresh, local traffic emissions, different gasoline formulations can result in different T/B ratios. These ratios must therefore be used with caution. In addition, low values for the T/B ratio may indicate the presence of other benzene sources in this area in addition to motor vehicle emissions. Therefore, it is necessary to investigate the relationship among BTEX and meteorological parameters in order to elucidate the possible source for the BTEX. Pearson correlation matrixes were constructed for each sampling period (B1, B2 and B3) for SS1 (Tables 2-4) and for SS2 (Tables 5-7). A good mutual correlation among the species indicates that they might primarily originated from the same source and a good mutual correlation between Ebz and X indicates that they might possibly originated from gasoline vehicles and gasoline stations [34-36]. A low correlation factor indicates towards the spiking of BTEX from some additional sources apart from vehicular inputs.

Table 2 Pearson correlation matrix for SS1 during B1 sampling period

Variables	Benzene	Toluene	Ethylbenzene	p-Xylene	Wind speed	Wind direction	Relative Humidity	Temperature	Barometric Pressure
Benzene	1	0.069	-0.137	-0.528	0.048	-0.095	0.244	-0.151	0.406
Toluene	0.069	1	-0.339	-0.015	0.168	-0.257	0.026	0.170	0.060
Ethylbenzene	-0.137	-0.339	1	0.125	0.173	0.073	0.252	-0.211	-0.330
p-Xylene	-0.528	-0.015	0.125	1	-0.159	-0.231	-0.170	0.493	-0.445
Wind speed	0.048	0.168	0.173	-0.159	1	-0.225	**0.830**	**-0.802**	-0.080
Wind direction	-0.095	-0.257	0.073	-0.231	-0.225	1	0.112	-0.001	0.572
Relative Humidity	0.244	0.026	0.252	-0.170	**0.830**	0.112	1	**-0.750**	0.169
Temperature	-0.151	0.170	-0.211	0.493	**-0.802**	-0.001	**-0.750**	1	0.095
Barometric Pressure	0.406	0.060	-0.330	-0.445	-0.080	0.572	0.169	0.095	1

For SS1 during B1 sampling period low correlations between benzene and the rest of BTEX were found, indicating that excepting benzene, the other studied BTEX could have their origin in sources different from vehicular emissions. It was found a good correlation between relative humidity and wind speed (0.830) and a good negative correlation between temperature and relative humidity (-0.750).

Table 3 Pearson correlation matrix for SS1 during B2 sampling period

Variables	Benzene	Toluene	Ethylbenzene	p-Xylene	Wind speed	Wind direction	Relative Humidity	Temperature	Barometric Pressure
Benzene	**1**	-0.079	**-0.661**	-0.464	0.535	0.046	0.015	0.441	0.562
Toluene	-0.079	**1**	0.345	**0.645**	-0.450	-0.282	0.185	-0.014	0.183
Ethylbenzene	**-0.661**	0.345	**1**	**0.865**	-0.318	-0.106	0.157	-0.135	-0.458
p-Xylene	-0.464	**0.645**	**0.865**	**1**	-0.308	-0.258	0.288	-0.087	-0.356
Wind speed	0.535	-0.450	-0.318	-0.308	**1**	0.506	-0.108	0.293	-0.078
Wind direction	0.046	-0.282	-0.106	-0.258	0.506	**1**	**-0.613**	0.551	-0.321
Relative Humidity	0.015	0.185	0.157	0.288	-0.108	**-0.613**	**1**	-0.461	0.355
Temperature	0.441	-0.014	-0.135	-0.087	0.293	0.551	-0.461	**1**	0.037
Barometric Pressure	0.562	0.183	-0.458	-0.356	-0.078	-0.321	0.355	0.037	**1**

Toluene-p-Xylene and Ethylbenzene-p-Xylene showed good correlations during the midday sampling period for SS1 (B2) (Table 3), 0.645 and 0.865, respectively. Benzene and Ethylbenzene showed a significant negative correlation (-0.661) indicating that Ethylbenzene was probably precursor of benzene. Wind direction showed a significant negative correlation with relative humidity.

Table 4 Pearson correlation matrix for SS1 during B3 sampling period

Variables	Benzene	Toluene	Ethylbenzene	p-Xylene	Wind speed	Wind direction	Relative Humidity	Temperature	Barometric Pressure
Benzene	**1**	0.374	-0.130	0.031	-0.484	-0.416	**0.627**	0.393	**0.606**
Toluene	0.374	**1**	-0.209	-0.545	-0.226	-0.533	0.395	-0.103	**0.641**
Ethylbenzene	-0.130	-0.209	**1**	**0.639**	-0.128	0.077	-0.060	0.010	-0.096
p-Xylene	0.031	-0.545	**0.639**	**1**	-0.150	0.203	0.023	0.108	-0.456
Wind speed	-0.484	-0.226	-0.128	-0.150	**1**	**0.616**	-0.493	-0.055	-0.169
Wind direction	-0.416	-0.533	0.077	0.203	**0.616**	**1**	**-0.746**	0.513	-0.483
Relative Humidity	**0.627**	0.395	-0.060	0.023	-0.493	**-0.746**	**1**	-0.341	0.319
Temperature	0.393	-0.103	0.010	0.108	-0.055	0.513	-0.341	**1**	0.174
Barometric Pressure	**0.606**	**0.641**	-0.096	-0.456	-0.169	-0.483	0.319	0.174	**1**

In Table 4 it can be observed that during the afternoon for SS1, benzene had good correlation with relative humidity (0.627), whereas toluene showed good correlation with barometric pressure (0.606). Ethylbenzene and p-xylene once again showed good correlation (0.639) indicating that these pollutants probably had common sources. Good correlations between wind speed-wind direction, barometric pressure and benzene were found.

Table 5 Pearson correlation matrix for SS2 during B1 sampling period

Variables	Benzene	Toluene	Ethyl-benzene	p-xylene	Wind Speed	Wind Direction	Relative Humidity	Temperature	Barometric Pressure
Benzene	**1**	**0.839**	0.495	**0.765**	-0.085	-0.459	-0.374	0.179	**-0.761**
Toluene	**0.839**	**1**	0.164	**0.742**	-0.252	**-0.630**	-0.234	-0.088	**-0.864**
Ethylbenzene	0.495	0.164	**1**	0.060	-0.160	0.085	0.060	0.170	-0.198
p-xylene	**0.765**	**0.742**	0.060	**1**	-0.007	-0.511	-0.339	0.032	**-0.628**
Wind Speed	-0.085	-0.252	-0.160	-0.007	**1**	0.209	**-0.806**	**0.865**	0.329
Wind Direction	-0.459	**-0.630**	0.085	-0.511	0.209	**1**	0.087	0.068	**0.603**
Relative Humidity	-0.374	-0.234	0.060	-0.339	**-0.806**	0.087	**1**	**-0.741**	0.132
Temperature	0.179	-0.088	0.170	0.032	**0.865**	0.068	**-0.741**	**1**	0.178
Barometric Pressure	**-0.761**	**-0.864**	-0.198	**-0.628**	0.329	**0.603**	0.132	0.178	**1**

For SS2 during morning sampling period (B1)(Table 5), B, T and X probably had sources in common because they showed a strong positive correlation among each other. Wind speed had a good negative correlation with Relative humidity and a strong positive correlation with temperature. Barometric pressure had a negative strong correlation with B, T and X. Ebz did not show correlation with the rest of BTEX, suggesting that this compound could be originated from different sources.

Table 6 Pearson correlation matrix for SS2 during B2 sampling period

Variables	Benzene	Toluene	Ethyl-benzene	p-xylene	Wind Speed	Wind Direction	Relative Humidity	Temperature	Barometric Pressure
Benzene	**1**	0.469	-0.034	0.196	-0.184	-0.031	-0.104	-0.299	-0.218
Toluene	0.469	**1**	0.476	**0.688**	0.086	0.060	-0.300	-0.195	**-0.652**
Ethylbenzene	-0.034	0.476	**1**	**0.821**	0.566	0.059	0.140	0.119	-0.209
p-xylene	0.196	**0.688**	**0.821**	**1**	0.222	0.072	-0.156	0.247	-0.491
Wind Speed	-0.184	0.086	0.566	0.222	**1**	0.485	-0.112	0.300	-0.023
Wind Direction	-0.031	0.060	0.059	0.072	0.485	**1**	**-0.609**	0.543	-0.326
Relative Humidity	-0.104	-0.300	0.140	-0.156	-0.112	**-0.609**	**1**	-0.462	0.331
Temperature	-0.299	-0.195	0.119	0.247	0.300	0.543	-0.462	**1**	0.057
Barometric Pressure	-0.218	**-0.652**	-0.209	-0.491	-0.023	-0.326	0.331	0.057	**1**

During the midday sampling period (B2) for SS2 (Table 6), T and X showed a good correlation (0.688) indicating that at least partially they could be originated from common sources. Ebz and X had a strong positive correlation (0.821) indicating that these BTEX could have the same sources. Relative humidity-wind direction and Barometric pressure-Toluene showed negative moderate correlation between each other.

Table 7 Pearson correlation matrix for SS2 during B3 sampling period

Variables	Benzene	Toluene	Ethyl-benzene	p-Xylene	Wind speed	Wind direction	Relative humidity	Temperature	Barometric Pressure
Benzene	**1**	0.304	0.122	-0.102	0.210	0.403	-0.081	0.007	-0.264
Toluene	0.304	**1**	-0.090	0.467	-0.153	0.399	-0.403	-0.238	**-0.605**
Ethyl-benzene	0.122	-0.090	**1**	-0.280	0.151	0.236	-0.001	0.106	-0.429
p-Xylene	-0.102	0.467	-0.280	**1**	-0.480	0.041	-0.294	0.034	0.108
Wind speed	0.210	-0.153	0.151	-0.480	**1**	0.339	-0.477	-0.012	-0.049
Wind direction	0.403	0.399	0.236	0.041	0.339	**1**	**-0.636**	0.399	-0.543
Relative humidity	-0.081	-0.403	-0.001	-0.294	-0.477	**-0.636**	**1**	-0.344	0.292
Temperature	0.007	-0.238	0.106	0.034	-0.012	0.399	-0.344	**1**	0.196
Barometric Pressure	-0.264	**-0.605**	-0.429	0.108	-0.049	-0.543	0.292	0.196	**1**

During the afternoon sampling period for SS2 (Table 7), BTEX did not correlate among each other, suggesting that during this period all aromatic hydrocarbons could have different and diverse sources. Wind direction showed a moderate negative correlation with relative humidity during this period. In addition, a Principal component analysis (PCA) was used to study the variability patterns present in this multivariate data set. A PCA model was developed for air pollutants levels (benzene, toluene, ethylbenzene and p-xylene) and the meteorological parameters (temperature: T, barometric pressure: P, relative humidity: RH, wind direction: WD, and wind speed: WS). Figures 9-11 shows the PCA biplot obtained for the morning (B1), midday (B2) and afternoon (B3) sampling periods during autumn season for SS1. PCA for the morning sampling period (B1) gave two principal components (F1 and F2) expressing about 55.60% of the total variance. In figure 9 it can be observed that B was influenced by wind direction, it means that this pollutant was probably transported from the highway Veracruz-Mexico. Ebz and X did not show correlation between each other indicating that they probably had their origin in different sources.

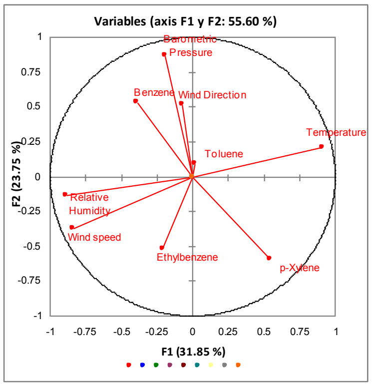


Fig. 9 Multivariate set Biplot from the PCA for B1 sampling period for SS1.

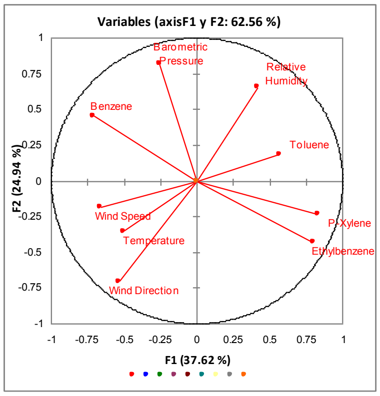


Fig. 10 Multivariate set Biplot from the PCA for B2 sampling period for SS1.

In figure 10 it can be observed that ethylbenzene and p-xylene had a good relation indicating that they probably had common sources. Toluene had a negative correlation with wind speed, indicating that this pollutant was probably dispersed by wind effects. Temperature and relative humidity, once again showed a negative correlation.

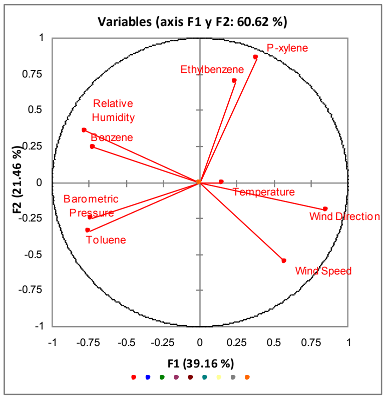


Fig. 11 Multivariate set Biplot from the PCA for B3 sampling period for SS1.

In Figure 11 it can be observed that ethylbenzene and p-xylene showed a good relation each other and that benzene and toluene had good correlation with barometric pressure, whereas, Benzene and relative humidity showed a good relation too. Figures 12-14 showed the PCA analysis for the SS2 sampling site during the morning, midday and afternoon sampling periods (B1, B2 and B3, respectively).

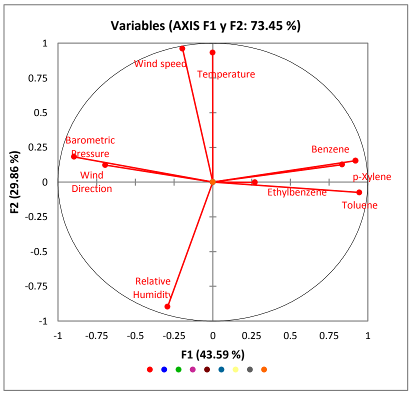


Fig. 12 Multivariate set Biplot from the PCA for B1 sampling period for SS2.

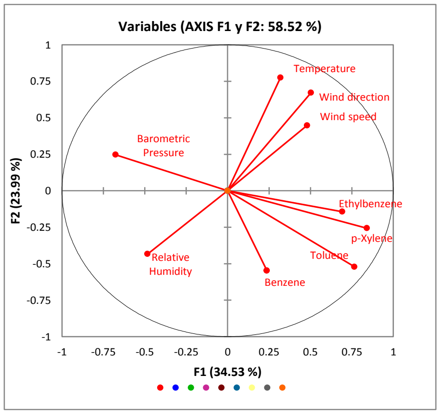


Fig. 13 Multivariate set Biplot from the PCA for B2 sampling period for SS2.

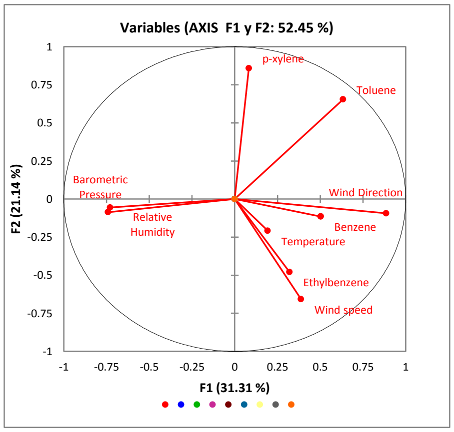


Fig. 14 Multivariate set Biplot from the PCA for B3 sampling period for SS2.

Two components were necessary to express 73.45% of the total variance (Figure 12) for SS2 during the morning sampling period (B1). F1 group included three BTEX (B, T and X). F2 group included Wind speed, Temperature and Relative humidity. For the midday sampling period (B2) for SS2 (Figure 13), two components expressed 58.520% of the total variance, whereas, considering three components, the percentage of the total variance was 76.566%. F1 group included T, Ebz and X; whereas F2 grouped to Wind direction and Temperature. During this period, Relative humidity showed a negative correlation with all measured pollutants. This behavior suggest that high concentrations of water vapor partially remove pollutants from the atmosphere by means of chemical reaction (acid rain) or condensation (promoting deposition) [36]. Two components (F1+F2) expressed 52.454% of the total variance for the multivariate data set during B3 for SS2 (Figure 14) and three components (F1+F2+F3) contributed to 69.417% of the total variance. X and Temperature showed good correlation, indicating that this hydrocarbon could be originated from evaporative emissions.

F. Assessment of the carcinogenic risk levels of benzene

Daily exposure E (mg/kg per day) of an individual by inhalation can be calculated as (according to Zhang et al. [25]):

$$E = C \ x \ Ira \ x \ Da/Bwa \quad (1)$$

Where C (mg m^{-3}) is the mean concentration of benzene, Ira is the inhalation rate of an adult (0.83 m^3 h^{-1}) [39], Da is the exposure duration of an adult (24 h/day) and Bwa is the body weight of an adult (65 kg) [39]. The lifetime cancer risk (LTCR) is then calculated as:

$$LTCR = E \ x \ SF \quad (2)$$

Where SF is the slope factor of inhalation unit risk for toxics when the exposure-carcinogenic effect is considered as linear. Here, it was adopted a SF value of 2.98 E-02 kg day/mg for benzene by USEPA [26]. The non-cancer risk of benzene is measured by the hazard quotient (HQ):

$$HQ = C/\ Rfc \quad (3)$$

Where C is the yearly average daily received concentration and Rfc is the inhalation reference concentration, which is 0.03 mg m^{-3} for benzene [40].

The average daily exposure (E) for SS1 and SS2 were of 22.83 E-03 and 27.93 E-03 mg/kg per day, respectively. The lifetime cancer risk (LTCR) was of 68 E-05 and 83.23 E-05 for SS1 and SS2, respectively. These values exceeded the acceptable LTCR value of 1 E-06 for adults according to USEPA [26]. This implies that the potential cancer risk due to ambient benzene exposure should be a health concern in the urban region of Orizaba, Veracruz, Mexico. For the non-cancer risk, the calculated HQs were in the range of 2.483-3.038. According to USEPA, if the HQ value is greater than 1, the pollutant represents cancer risk [26].

V. CONCLUSION

Benzene was the dominant BTEX for both sampling sites. BTEX levels were strongly influenced by winds blowing from the S and SSE where an important highway is located (highway Mexico-Veracruz), indicating that BTEX were mainly originated from vehicular sources. Benzene concentrations were similar to those reported in other large cities like Great Cairo and Algeriers, whereas, the levels of Ethylbenzene were similar to those reported for Seoul, Korea. p-Xylene and Toluene showed a clear diurnal pattern but benzene and ethylbenzene did not show significant differences between the different sampling periods for sampling site 1 (SS1). For the second sampling site (SS2), the highest concentrations of benzene, toluene and ethyl benzene were found during the midday sampling period. T/B and X/Ebz ratios showed that benzene was influenced by fresh vehicular emissions. PCA analysis showed that all BTEX probably were transported from the highways 180 and 190 during B1 and B2 sampling periods, whereas, additional sources beyond traffic related emissions could influence the levels of ethylbenzene and p-xylene during the afternoon sampling period (B3). The found results for benzene levels are higher than those previously observed in the US and Europe and the lifetime cancer risk due to exposure to ambient benzene in both study sites exceeded the acceptable level reported by USEPA. Health concerns of benzene exposure should be seriously considered in this region and it should be set an ambient air quality standard for benzene. We recommend to investigate the relationship between ambient benzene levels and the incidence of different kinds of cancer in this region.

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