

BTEX Atmospheric Levels and Health Risk in an Urban Site in Ciudad del Carmen, Campeche

RAMÍREZ-LARA E.¹, CERÓN-BRETÓN J. G.², CERÓN-BRETÓN R. M.²,
LÓPEZ-CHUKEN U. J.¹, VICHIQUE-MORALES A.², UC-CHI M. P.²,
HERNÁNDEZ-LÓPEZ G.², SOLIS-CANUL J. A.², LARA-SEVERINO R. C.³,
RANGEL-MARRÓN M.², ROBLES-HEREDIA J. C.²

¹Universidad Autónoma de Nuevo León, Facultad de Ciencias Químicas, Av. Universidad s/n, Ciudad Universitaria, C.P. 66455, San Nicolás de los Garza, Nuevo León, MEXICO

²Universidad Autónoma del Carmen, Facultad de Química, Calle 56 No. 4, Esq. Ave. Concordia, C.P. 24180, Ciudad del Carmen, Campeche, MEXICO

³Universidad Autónoma del Carmen, Facultad de Ciencias de la Salud, Av. Central s/n, Mundo Maya, Ciudad del Carmen, Campeche, MEXICO

Abstract: Benzene, toluene, ethylbenzene, and xylenes (BTEX) were measured in ambient air in an urban site of Ciudad del Carmen, Campeche during spring 2022. Samples were collected during the morning (from 07:00 to 08:00 h), midday (from 14:00 to 15:00 h) and afternoon (from 18:00 to 19:00 h) using glass tubes packed with activated carbon, at a controlled air flow of 1.5 L/min. Samples were analyzed by gas chromatography with flame ionization detection. The relative abundance in ambient air of BTEX was the following: benzene (9.197 $\mu\text{g}/\text{m}^3$) > toluene (8.953 $\mu\text{g}/\text{m}^3$) > xylenes (7.789 $\mu\text{g}/\text{m}^3$) > ethylbenzene (7.538 $\mu\text{g}/\text{m}^3$). The statistical analysis revealed that BTEX compounds had strong correlations between each other, indicating that they were originated from common sources. From the meteorological analysis, it was found that the prevailing winds blew from the east and southeast, indicating that vehicular emissions coming from avenues located in these directions may have contributed to the BTEX levels in the study site. Principal component analysis and BTEX ratios (T/B and X/Ebz) revealed that vehicular emissions and fresh local air masses influenced the BTEX concentrations during the study period. From the health risk analysis, cancer risk coefficients exceeded the acceptable level (1×10^{-6}), thus exposed population may be at a possible risk of developing cancer in the lifetime due to the inhalation of BTEX at the measured concentrations. These results will be a useful tool for local authorities in order to establish control measures focused on the reduction of BTEX emissions and the improvement of the air quality in the study area.

Key words: BTEX, gas chromatography, correlations, health risk.

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1 Introduction

Air pollution has become a topic of interest for the scientific community and society due to different studies have related a poor air quality with harmful health effects such as respiratory diseases and some types of cancer, [1].

Common factors such as the use of fossil fuels and coal-fired power plants, dependence on private transport motor vehicles, inefficient use of energy in buildings, and the use of biomass for cooking and heating, produce so-called Volatile Organic

Compounds (VOCs), [2]. There is a sub-group of compounds within the VOCs group called BTEX, which includes Benzene and their alkyl-derivatives: Toluene, Ethylbenzene and Xylenes. These compounds can be used as markers of damage to human health, considering both carcinogenic and non-carcinogenic effects (respiratory and cardiovascular diseases). In addition, BTEX is key compounds due to they provide information on the degree of exposure of the human body to the harmful effects of VOCs

and they are precursors of the reactions that lead to the generation of tropospheric ozone (under appropriate meteorological conditions), [3].

Benzene is a highly toxic compound, becoming myelotoxic and inducer of leukemia in humans, besides that it has been identified as a human carcinogen by the International Agency for Research on Cancer, so the WHO and the United States Environmental Protection Agency (EPA) do not recommend any safe level of exposure, [4]. Considering the risks and consequences of exposure to these pollutants, it is important to include the monitoring of atmospheric BTEX in order to assess air quality present in the atmosphere of urban areas. However, in Mexico, most of the monitoring stations of the air quality monitoring network do not measure BTEX levels in ambient air, except in some sites of Mexico City. In addition, there is no Mexican standard that regulates the maximum permissible levels in ambient air of BTEX compounds. The present study is focused on determining the levels of BTEX in ambient air in an urban site in Ciudad del Carmen, Campeche during the spring 2022 and the potential risk (cancer and non-cancer) to health in the exposed population.

2 Methodology

2.1 Site Description

Ciudad del Carmen is an island of sedimentary origin located in the southeast of Mexico within an area named Sonda de Campeche located in the south of the Mexican Gulf. This city is an important center of processing and distribution of oil and gas in Mexico. The sampling was carried out within the facilities of the Autonomous University of Carmen located in the downtown area at 18.645502° N and -91.817140° W, during the spring season (from June 6 to 10, 2022). This period was selected due to it corresponds to the dry season, when the lowest wind speeds usually occur, so that the dispersion of pollutants is limited and it may result in higher concentrations of these air pollutants and a greater risk in the population. Three samples were collected per day, the first sampling period was performed during the morning (from 07:00 to 8:00 h), the second one during the midday (from 14:00 to 15:00 h) and the third sampling period was carried out during the afternoon (from 18:00 to 19:00 h). Temperature, pressure, wind direction, solar radiation and

relative humidity were registered during each sampling period.

2.2 BTEX Sampling in Ambient Air

Samples were collected from June 6 to 10, 2022 to determine BTEX concentrations (benzene, toluene, ethylbenzene and p-xylene) in ambient air by active sampling by passing air through glass tubes packed with activated carbon (226-01 Anasorb CSC), at a constant and controlled flow of 1.5 L/min by means of an SKC vacuum pump model PCXR4 [4]. The sampling lasted 1.0 hours for each sample considering three different periods according to the population activity observed in the city: during the morning (B1: 07:00 to 08:00 h), noon (B2: 14:00 to 15:00), afternoon (B3: 18:00 to 19:00 h).

2.3 Gas Chromatography - Flame Ionization Detection Analysis (GC-FID).

Collected samples were desorbed with 1 mL of chromatographic grade carbon disulfide. The samples were analyzed based on the method "Determination of aromatic hydrocarbons (benzene, toluene, ethylbenzene, p-xylene) in air-Adsorption method in activated carbon/gas chromatography MTA/MA-030/A92, of the National Institute of Safety and Hygiene of Spain. The chromatographic analysis of the collected samples was carried out in the Gas Chromatography Research Laboratory of the Chemistry Faculty of the Autonomous University of Carmen. A Thermoscientific brand gas chromatograph, model TRACE GC Ultra Gas Chromatographs was used in splitless mode in order to analyze the desorbed samples. This system was coupled to a flame ionization detector using extra dry air and ultra-high purity hydrogen. Ultra-pure nitrogen was used as a carrier gas and a 30 m x 0.32 mm ID capillary column was used to carry out the separation (fused silica methyl type and with a film thickness of 0.5 µm).

2.4 Statistical Analysis

Normality analyses were performed to determine whether parametric or non-parametric statistics were applicable. Hypothesis tests were applied to determine if there were significant differences in BTEX concentrations at different sampling periods (Levine Test and Bartlett Test). A bivariate analyses (Pearson correlation analysis) was carried out in order to find relations between the measured variables. A multivariate analysis

(Principal Component Analysis: PCA) was carried out in order to explain the variance and for uncovering the structure of the data set. This method is commonly used in environmental studies in order to identify patterns in data. A PCA analysis let us to create new variables (principal component) which are linear combinations of the original variables. The results of PCA are resumed as scores and loading vectors usually represented as bi-plots of the two principal components, revealing relations among the observations (measured variables). The software used was XLSTAT for Excel version 2016.

2.5 Meteorological Analysis

The analysis of the frequency of occurrence of the winds (direction and speed) was carried out per day during the sampling period. The daily wind roses were constructed using the Wind Rose statistical tool from NOAA (Air Resource Laboratory: ARL). Figure 1 shows a typical wind rose representative of the sampling period. It can be observed that the prevailing winds blew from E and SE, with wind speed ranging from 1 to 4 m/s, indicating that sources located in these directions could contribute to the measured BTEX levels. An important avenue with high vehicular flow (the Peripheral Avenue) is located to the east, which communicates the city (Carmen Island) with the mainland towards the State of Tabasco. On the other hand, to the southeast is 31st Avenue, which crosses the city from east to west, being the main transportation route on the island. Both avenues constitute two of the main ways of population mobility with heavy vehicular traffic during the peak hours (during the morning when the population moves to their workplaces, and during the afternoon when the flow of vehicles is towards homes).

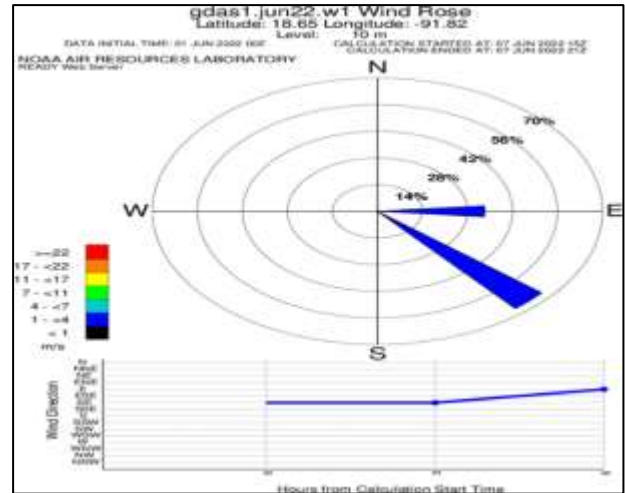


Fig. 1: Wind rose representative for the sampling period at the study site.

2.6 Health Risk Analysis

The carcinogenic potential of benzene is widely known, [5]. The European Union recommends an annual limit of 5 µg/m for benzene in ambient air and the Minimal Risk Level (MRL of 1 in 10,000), while the EPA sets a value of 4.0 ppbv for this pollutant, [6]. This study used the methodology proposed by EPA, [5] in order to determine the daily exposure (E), the lifetime cancer risk and the non-cancer potential risk coefficients, LTCR and HQ, respectively. The individual's daily inhalation exposure can be calculated as :

$$E = \frac{C \times IRa \times Da}{BW} \quad (1)$$

Where: E = is the daily inhalation exposure in mg/kg per day, C = is the average concentration of benzene in mg/m³, IRa = is the inhalation rate for an adult (0.83 m³/h), Da = is the duration of exposure in an outdoor ambient according to the typical activities (being 24 and 16 hr/day, for adults and children, respectively), [5-6]. BW = is the weight of the body (being 65 and 36 kg for adults and children, respectively). The lifetime cancer risk (LTCR) is calculated as:

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

Where: LTCR = is the lifetime cancer risk, SF = is the slope factor (kg day/mg). The slope factor of the inhalation unit risk for toxic substances when the exposure-carcinogenic effect is considered linear, being 2.98 E⁻⁰². kg day/mg for benzene, [6]. The determined values were compared with

the permissible limit values established by the EPA (1×10^{-6}) and the WHO (1×10^{-5}).

The non-carcinogenic risk of BTEX was calculated as a hazard quotient (HQ) according to the following equation:

$$HQ = \frac{C}{RfC} \quad (3)$$

Where C is the average daily received concentration and RfC represents the inhalation reference concentration of specific air pollutants: 0.03, 5, 1 and 0.1 mg/m^3 for benzene, toluene, ethylbenzene and p-xylene, respectively. An $HQ > 1.0$ indicates that long term exposure may cause adverse non-cancer health effects (respiratory and cardiovascular diseases). An $HQ < 1.0$ is considered as acceptable level according to the recommended values established by EPA and OMS.

3 Results and Discussion

3.1 BTEX Concentrations in Ambient Air at the Study Site

Figures 2 and 3 show the parametric statistics and boxplot BTEX at the study site during the summer season. The relative abundance of BTEX in ambient air was the following: benzene ($9.197 \mu\text{g}/\text{m}^3$) > toluene ($8.953 \mu\text{g}/\text{m}^3$) > xylenes ($7.789 \mu\text{g}/\text{m}^3$) > ethylbenzene ($7.538 \mu\text{g}/\text{m}^3$). According to figures 2 and 3 it can be seen that the BTEX had a diurnal pattern, showing higher concentration values during the midday and afternoon and with lower concentration values during the morning. This behavior was expected, since the greatest mobility of the population in this area occurs during these periods, since commercial areas, banks and restaurants are located in this zone. The results are comparable to those obtained at other study sites (Table 1). As can be seen, benzene and ethylbenzene showed higher concentrations than those registered in the study site during 2012, [8]. Toluene and xylenes showed a decrease in their levels compared to those obtained 10 years earlier, [8]. Benzene concentrations in this study were comparable to the values reported in Beijing, China, [11] but higher than those reported for León, [9] and Mexico City, [10] (Table 1). Toluene presented lower values than those reported in León, [9] and Mexico City, [10] according to Table 1.

Ethylbenzene presented concentrations higher than Beijing, China, [11] but lower than reported for the city of León, Guanajuato, [9]. Finally, xylenes in this study presented higher concentrations than those reported for the cities of Beijing, [11] and León in Guanajuato, [9] (Table 1).

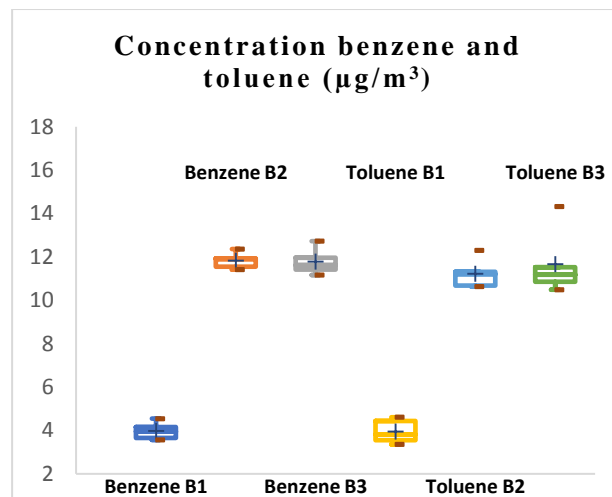


Fig. 2: Parametric statistics and boxplot for benzene and toluene concentrations at the study site during the summer season.

B1: sampling period from 07:00 to 08:00 h; B2: sampling period from 14:00 to 15:00 h; B3: sampling period from 18:00 to 19:00 h. + is the mean value, - represents maximum and minimum values, the central horizontal bars are the medians. The lower and upper limits of the box are the first and third quartiles.

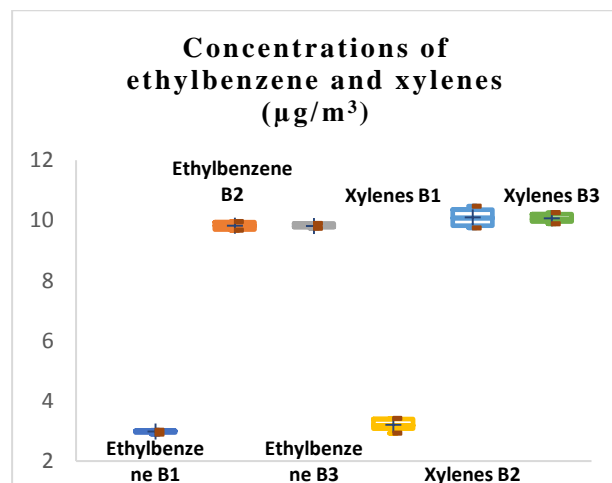


Fig. 3: Parametric statistics and boxplot for ethylbenzene and xylenes concentrations at the study site during the summer season.

B1: sampling period from 07:00 to 08:00 h; B2: sampling period from 14:00 to 15:00 h; B3: sampling period from 18:00 to 19:00 h. + is the mean value, - represents maximum and minimum values, the central horizontal bars are the medians. The lower and upper limits of the box are the first and third quartiles.

horizontal bars are the medians. The lower and upper limits of the box are the first and third quartiles.

Table 1. Comparison of results found for BTEX in Ciudad del Carmen with other studies

Place	Benzene $\mu\text{g}/\text{m}^3$	Toluene $\mu\text{g}/\text{m}^3$	Etilbenceno $\mu\text{g}/\text{m}^3$	Xylenes $\mu\text{g}/\text{m}^3$	Reference
Ciudad del Carmen, 2022	9.19	8.95	7.55	7.78	This study
Ciudad del Carmen, 2012	5.42	11.23	3.97	8.32	Cerón <i>et al.</i> [8]
Leon, Guanajuato, 2018	1.73	11.85	11.86	3.31	Cerón <i>et al.</i> [9]
Mexico City, 2002	3.67	17.63	-	-	Baez <i>et al.</i> [10]
Beijing, China, 2012	9.2	1.39	0.42	1.27	Zhang <i>et al.</i> [11]

The highest concentrations of benzene and ethylbenzene were observed during the midday period (B2) with average concentration values of 11.836 and 9.820 $\mu\text{g}/\text{m}^3$, respectively. While toluene and xylenes showed higher average concentration values during the afternoon period (B3) with values of 11.675 and 10.097 $\mu\text{g}/\text{m}^3$, respectively. The lowest average concentration values for BTEX were found during the morning sampling period (B1) with values of 3.977, 3.956, 2.978 and 3.201 for benzene, toluene, ethylbenzene and xylenes, respectively.

The BTEX concentrations showed a normal distribution, which was confirmed by applying the Shapiro-Wilk (W) test. For this reason, it was decided to apply parametric statistics to the data set. Although a diurnal pattern was observed in BTEX concentrations (with higher average values during midday and afternoon), after applying the Levene (F) and Bartlett (Chi square) tests, it was confirmed that these differences were not significant at a significance level of $\alpha = 0.05$. This allows us to infer that BTEX levels at the study site were homogeneously distributed, with influence from local sources (vehicular traffic). The Pearson correlation matrix (Table 2) shows that all BTEX had positive linear correlations (correlation coefficients >0.97), indicating that they could be originated from the same sources.

Table 2. Pearson Correlation Matrix of the measured variables

	B	T	EBZ	X	DV	TE	HR	P	SR
B	1								
T	0.986	1							
EBZ	0.995	0.971	1						
X	0.997	0.976	0.999	1					
DV	0.252	0.279	0.222	0.237	1				
TE	-0.240	-0.279	-0.221	-0.235	-0.703	1			
HR	0.239	0.271	0.232	0.238	0.425	-0.923	1		
P	0.495	0.551	0.478	0.478	0.312	-0.408	0.459	1	
SR	-0.184	-0.184	-0.186	-0.188	-0.432	0.821	-0.808	-0.077	1

Bold values are different from 0 with a significance level $\alpha=0.05$; B: Benzene; T: Toluene; EBZ: Ethylbenzene; X: Xylenes; DV: wind direction. TE: temperature, RH: relative humidity, P: atmospheric pressure, SR: solar radiation.

A significant moderate correlation was found between toluene and atmospheric pressure (0.551), indicating that this compound could be influenced by high pressure systems in the study site. A significant linear correlation was found between temperature and solar radiation (0.821). Significant negative correlations were recorded between relative humidity and temperature (-0.923) and between relative humidity and solar radiation (-0.808), indicating that increases in moisture content result in decreases in the temperature and are also associated with a reduction in solar radiation levels due to greater cloud cover.

Figure 4 and Table 3 show the results of the Principal Component Analysis (PCA). From the PCA applied to the data set, two principal components, F1 and F2 were obtained, which together contributed with 82.555% to the total variability of the data. Figure 4 shows the bi-plot of the main components F1 and F2, observing a strong relationship between the BTEX measured (see the vectors located at the upper right quadrant). A strong correlation was also observed between temperature and solar radiation (see vectors located at the upper left quadrant) and strong negative correlations were observed between the pair's temperature-solar radiation and wind direction-relative humidity (see vector located diametrically opposed in the bi-plot).

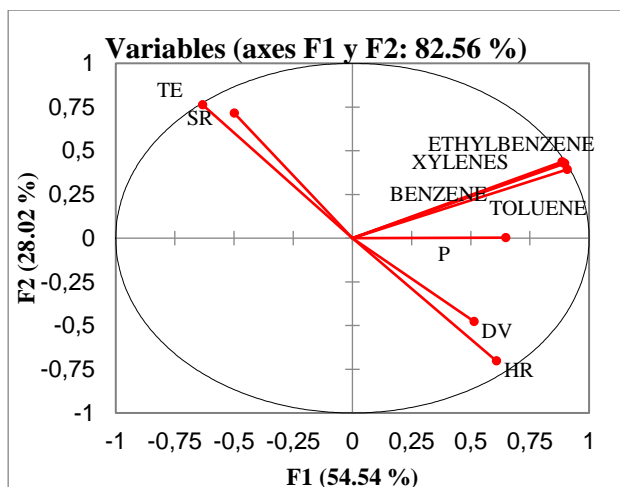


Fig. 4: Bi-plot of the principal components found from the PCA Analysis.

Note: DV: wind direction; TE: temperature; RH, relative humidity; P, atmospheric pressure; SR: solar radiation

Table 3 shows the factor loadings for the measured variables and it was possible to identify three groups of correlated variables: F1, F2 and F3. F1 shows a first group of interrelated variables, which includes benzene, toluene, ethylbenzene, and xylenes. F2 shows a second group of variables highly correlated, which includes temperature, relative humidity, and solar radiation. F3 only included Wind Direction. This variable was not correlated with the rest of variables, indicating that the air pollutants in the study site were only influenced by local sources and that the regional transport was negligible.

Table 3. PCA Factor loadings for the measured variables.

	F1	F2	F3	F4
BENZENE	0.804	0.182	0.012	0.001
TOLUENE	0.824	0.154	0.002	0.000
ETHYLBENZENE	0.784	0.191	0.018	0.000
XYLENES	0.794	0.184	0.017	0.001
DV	0.264	0.227	0.063	0.444
TE	0.400	0.580	0.001	0.000
HR	0.370	0.494	0.000	0.108
P	0.419	0.000	0.480	0.085
SR	0.249	0.509	0.185	0.008

Note: The bold values correspond for each variable to the factor for which the square cosine is the largest DV: wind direction, TE: temperature, RH: relative humidity, P: atmospheric pressure, SR: solar radiation.

3.2 BTEX Ratios

BTEX Ratios are commonly used to infer the emission sources (vehicular, area or industrial sources) and to know the grade of photochemical processing of the air masses that contain BTEX. So, from the BTEX ratios, it is possible to determine if the emissions come from mobile sources or from area or industrial sources (T/B ratio) and if BTEX come from fresh or aged air masses (X/E ratio). The T/B ratios are used as indicators of vehicular traffic emissions, due to benzene and toluene are commonly constituents of gasoline (toluene content is 3 to 4 times higher than the benzene content), for this reason during the combustion process they are emitted into the atmosphere by the exhausts of motor vehicles. Values of T/B ratio between 2 and 3 or lower have been reported in various urban areas of the world. Values of this ratio >3 indicate that BTEX levels can be associated with sources beyond vehicular, for example: industrial facilities, area sources such as evaporative emissions, automotive paint shops, food cooking processes, screen printing workshops, dry cleaners, among many others, [12]. The xylene/ethylbenzene ratio (X/Eb) is commonly used as an indicator of the photochemical age of air masses at a given site. Values greater than 3.8 indicate that BTEX comes from old air masses and values less than 3.8 indicate that BTEX comes from fresh air masses (recent emissions), [13]. This reason is related to the atmospheric lifetime of these pollutants in the air. Low values of this ratio indicate that air masses are fresh (recent emissions), whereas, high values of this ratio are an indicator that the air masses are aged (with a high grade of photochemical processing). Values for X/Eb between 3.8 and 4.4 have been reported for fresh gasoline emissions, [14, 15]. The T/B ratio showed values ranging from 0.892 to 1.126 with an average value of 0.976, indicating that BTEX levels at the study site were under the influence of vehicular-type emissions. On the other hand, the concentration ratio (X/Eb) showed values in a range from 0.974 to 1.129 with an average value of 1.043. The values obtained indicate that the air masses containing BTEX at the study site were fresh emissions (recent emissions from local sources).

3.3 Health Risk Assessment

Table 4 shows the carcinogenic and non-carcinogenic risk coefficients (LTCR and HQ,

respectively) to which the population may be exposed by inhalation of BTEX at the measured concentrations. It can be observed that the LTCR values exceeded the permissible limits established by the EPA and WHO (1×10^{-6} and 1×10^{-5} , respectively), being this level of risk higher in children. The non-cancer (HQ) risk coefficients did not exceed the maximum permissible level established by EPA and WHO (1.0), indicating that the risk of developing respiratory and cardiovascular diseases due BTEX inhalation at the study site is low.

Table 4. Cancer and non-cancer risk coefficients for concentrations measured at the study site.

Cancer Risk Coefficient: LTCR for Adult population	
Pollutant	Average
Benzene	8.17×10^{-05}
Cancer Risk Coefficient: LTCR for Child population	
Pollutant	Average
Benzene	1.55×10^{-04}
Non-cancer risk coefficients (HQ)	
Pollutant	Average
Benzene	0.3070
Toluene	0.0017
Ethylbenzene	0.0075
Xylenes	0.0079

4 Conclusions

The dominant BTEX in the ambient air of the study site was benzene ($9.197 \mu\text{g}/\text{m}^3$) followed by toluene ($8.953 \mu\text{g}/\text{m}^3$). All measured BTEX showed a clear diurnal pattern with higher concentration values during the midday and afternoon sampling periods, due to greater mobility by the population in the study area, resulting in higher vehicular-type emissions. From the Pearson correlation and Principal Component Analysis, it was possible to confirm that all BTEX were probably originated from common sources, since they showed strong correlations between each other. From the meteorological analysis it was found that the prevailing winds blew from the E and SE, indicating that vehicular emissions from avenues located in these directions could contribute to the levels of BTEX measured. The T/B and X/Eb ratios showed that BTEX concentrations were influenced by vehicular-type emissions and local fresh air masses. From the health risk assessment, it was found that there is a

possible risk of developing cancer in the lifetime at the measured concentrations, being more critical for the child population. The level of risk of developing cardiovascular and respiratory diseases from BTEX inhalation is low. It is important that environmental authorities in Mexico considering the inclusion of BTEX within the National emissions inventory, in order to identify the main sources of hydrocarbons, considering industrial, natural, mobile and area sources, with the aim of developing control measures of BTEX emissions that improve the air quality in the study area. This work provides information about BTEX concentration distribution in an urban area of Ciudad del Carmen, Campeche. However, it is necessary to include more monitoring points in this area that considering a more extensive sampling period that covers the different seasons along the year (dry season, rainy season and the Norths season), as well as, the monitoring of other air pollutants such as ozone, sulphur dioxide, nitrogen oxides, PM10, PM2.5, among others.

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